

# Quantification and Morphological Characterization of Microfibers Emitted from Textile Washing

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1	Quantification and morphological characterization of microfibers emitted from textile
2	washing
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11	Abstract
12	Microplastics are a subject of growing interest as they are a potential threat for living organisms.
13	Textile microfibers (MFs) are an important microplastics sub-group that have been reported as
14	a major source of microplastics release into the environment. This pollution occurs mainly
15	during the washing of synthetic garments. However, standardized methods to quantify and
16	characterize these MFs are scarce. This study proposes a new analytical protocol to characterize
17	these MFs in number and size by means of filtration techniques, optical and electronic
18	microscopy and automatic image post-processing. This approach was developed and validated
19	on effluents from washing machines produced in different conditions (5 different garments,
20	sequential cycles, and presence or not of detergent). Among the analyzed effluents, it was found
21	that 40 to 75% of microfibers have a length comprised between 50 and 200 $\mu$ m, with average
22	microfiber diameters ranging from 8 to 17 $\mu$ m depending on the type of textile. The emission
23	range of microfibers was estimated to be between 220 000 to 2 820 000 microfibers per kg of
24	textile depending on the type of garment and the washing conditions. The counting method
25	developed is adapted to a certain range of textiles, such as 100% polyester fleece jackets (PET-
26	1), 100% smooth polyester T-shirt (PET-2) and 100% acrylic sweater (PAN), and is not affected

27	by the presence of detergent. The proposed method of characterization of these MFs lengths
28	can also be extrapolated to the counting of other objects that have a similar morphology to the
29	analyzed fibers. Hence, it can be helpful to develop new testing capture technologies and, thus,
30	contribute to the enhancement of filtering techniques of several pollutants.
31	
32	Highlights
33	• New method for quantitative determination of textile microfibers release.
34	• Automatic counting and acquisition of detailed length distributions of microfibers.
35	• Most microfibers (40 to 75%) are between 50 and 200 µm long.
36	• Average microfiber diameters ranging from 8 to $17 \mu m$ .
37	
38	Graphical abstract
	3



40 Keywords: Microplastic; Filtration; Microscopy; Counting method; Length distribution.

#### 42 1. Introduction

Due to attractive properties such as high durability, high versatility and low cost, plastics are
widely used in day-to-day life and their production has increased exponentially since the mid20th century, exceeding 300 million tons produced per year [1], [2]. The textile industry
accounts for an important part of this share, with more than 80 million tons of synthetic fibers
produced in 2020 – a value largely driven by the production of polyester [3],[4].

48 The abrasion of synthetic textiles during washing leads to shedding and releasing of plastic fragments due to the mechanical stress of the textile fibers [5],[6]: over 700 000 synthetic 49 50 microfibers (lengths < 5 mm) can be generated during a simple domestic wash [7], [8]. Despite 51 a relatively high retention efficiency of microplastics in conventional wastewater treatment 52 plants (WWTP), microfibers are still the most abundant item remaining in discharged effluents 53 (up to 83%) [9]. Moreover, most of the plastic microfibers retained in WWTP end up in sludges 54 during the first steps of the treatment [10], which are in many countries spread on agricultural 55 lands for fertilization purposes [11], [12].

The ecotoxicology of microplastics has been demonstrated in many areas of environmental sciences [13], which rises an urgent necessity of developing regulatory measurement analysis and technologies for microfibers capturing [14]. Since the distribution of the microfibers lengths is a fundamental parameter for designing and testing these devices, accurate microfibers characterization method is essential to evaluate their performances.

Filtration is still a conventional method to measure the weight and estimate the amount of microfibers released by washing machines [7], [8], [15]–[22]. It can be coupled to additional characterization techniques to improve the quantification of these microfibers such as optical microscopy [18], [19], [23]–[28], scanning electron microscopy [23], [27], [29], Fourier transform infrared spectroscopy [19], [30]–[32], and Raman spectroscopy [31]–[35]. However, a comparison between the results of the literature is not straightforward due to the absence of a standard protocol. Moreover, the length distribution profiles are seldom estimated [30], [36],[23].

This work aims at contributing to the progress in microfiber quantification by presenting a new protocol of characterization based on microscopy and automatic image post-processing techniques, which can provide a distribution of the microfibers in number and length. The results obtained in this study can also be helpful for the design of future devices to capture these microfibers and for evaluating environmental impact.

74

#### 75 2. Material and methods

#### 76 2.1. Textiles and washing protocol

For this study, five different types of brand-new commercial garments were selected: 100%
polyester fleece jacket (PET-1), 100% smooth polyester T-shirt (PET-2), 88% polyester/12%
polyamide (PET/PA), 100% cotton (CO) towels, and 100% acrylic sweater (PAN). These
garments were all black or navy-blue to ensure an easier characterization of the microfibers by
optical microscopy. Other characteristics related to manufacturing stages (i.e. fabric type, fabric
weight per unit of area, fabric thickness and yarn type) were also evaluated (Table A1).

83 The washing machine used in this study was a Miele W5794 (2010) with a nominal capacity of 84 7 kg of dry laundry. This washing machine was selected for its high level of reproducibility. All washing experiments were carried out using 3 kg of dry garment, a temperature of 30°C, a 85 86 spin cycle of 1200 rpm and a washing cycle adapted to the type of fabric (synthetic or natural). 87 For the trial with detergent, 57.8 g of a commercial detergent (commercial liquid Ariel Power 88 Original from Procter & Gamble France) was added. Prior to each test with a new garment type, 89 a rinsing cycle was performed on the washing machine to minimize contamination. For 90 successive washings, garments were dried between each washing on a hanger in air for a minimum of 4 hours. For each experiment, the entire water effluent (around 70 L) was collected 91

92 in a stainless-steel vessel. This tank was homogenized to hold a representative water sample of93 around 1 L (glass bottle) for analyses and was rinsed between each test.

94

#### 95 2.2. Filtration

96 Filtration aims at quantifying particles mass and deposit them on a surface that is best adapted 97 to be characterized by microscopy. The water samples obtained from the wash tests were 98 filtered through a 0.7  $\mu$ m glass fiber filter (VWR I516-0345) at room temperature. The filtration 99 system used was a Setaclean Total Sediment Tester. All filters were dried at 60°C before and 100 after filtration during 1 hour to avoid humidity. All masses were measured after this drying 101 procedure with a Mettler Toledo AG204 scale (± 0.1 mg) and were normalized to the mass of 102 garment employed for each wash.

103 The volume to be filtered was adapted to the microfiber concentration of the washing effluents: 104 200 mL for all tests performed with 100% polyester fleece jacket (PET-1) without detergent 105 (including successive washes), and 20 mL for the remaining tests. It must be large enough to 106 allow measuring the deposited mass (limit of quantification of 0.1 mg on the scale) and small 107 enough to limit the entanglement of microfibers that complicates the counting method. To avoid 108 cross contamination, the filtration system was rinsed with demineralized water after each test. 109 Water samples were manually agitated before filtration to ensure a good homogeneity of the 110 solution. Two filtrations were performed for each wash test and both filters were then 111 characterized by microscopy to evaluate the repeatability of the sampling and analytical 112 method. All results presented in this study are the average of both analyses.

113

#### 114 **2.3.** Microscopic characterization

Scanning electron microscopy (SEM) analysis were performed as complementary characterization to verify the detailed morphology of initial garment fibers (before washing) and microfibers released in water effluents and further deposited on glass fiber filters. This technique allowed to precisely measure fiber diameters and identify possible by-products (other than microfibers) that could have been released through the washing process and retained by the filtration method. For this study, the analyses were performed with a Nova NanoSEM 450 FEI scanning electron microscope using a GAD (Gaseous Analytical Detector) detector adapted for low vacuum analysis. The characterization parameters were defined as an acceleration voltage of 5 kV, spot size of 3, diaphragm of 30  $\mu$ m, working distance of 6 mm and chamber pressure between 30 and 50 Pa.

125 Glass fiber filters from the filtration method were also characterized via an optical microscope 126 (OM) Olympus BX51 equipped with 6 objective lenses (5 polychromatic and 1 immersive). 127 The images were taken with a  $\times$  2.5 objective lens in reflective mode with polarizing filter. The 128 chosen exposure time was 16 ms with a gain of 16 dB. Images of size 2080×1544 were taken 129 at a resolution of 2.73 µm/pixel. The protocol for image acquisition consists in randomly taking 130 15 images of the area of the filter covered by the microfibers (about  $11.34 \text{ cm}^2$ ). These 15 131 images correspond to about 32% of the total surface of the filter. The values obtained can then 132 be extrapolated to the entire surface of the filter and the volume of the filtered solution. Fig. 1 shows an example of a surface analyzed using optical microscopy. It is important that no blurred 133 134 areas are shown in the analysis since it could impact the post-processing steps.



Fig. 1. Filter surface area analyzed by optical microscopy.

#### 136 **2.4.** Counting Method

In order to extract the number and size of the filtered microfibers, a post-processing protocol
was applied on the acquired images using the plug im! platform, which is an image processing
platform in open access for the scientific community that is capable of providing advanced
functionalities [37].

A set of classical post-processing image techniques was used to segment the OM images. At first, a set of nonlinear transformations was used since the contrast between the microfibers and the filters was clearly visible. First, a grey level segmentation operation was performed to binarize the image and extract the pixels corresponding solely to the microfibers. This segmentation is based on Otsu's method [38] that calculates the optimal threshold separating two classes on the histogram of pixel intensities by minimizing their intra-class variance. Finally, the negative of these segmented images was calculated.

A length criteria opening [39] allows to clean the binary images of all connected components below a predefined length threshold. The segmentation step may generate connected components related to residual noise or to particles not considered as textile microfibers with length smaller than 50  $\mu$ m, i.e., 18 pixels considering image resolution. This value was determined after the identification and sizing of non-fiber particles (amorphous deposit, particles) by SEM that were dominantly characterized by sizes below 50  $\mu$ m. Hence, the microfibers concerned by this method have a minimum size of 50  $\mu$ m.

A binary morphological operation was performed to keep only connected components completely contained in the image and remove all those in contact with the border of the image, since in this case the full length of the microfibers is impossible to quantify. Then, a morphological analysis algorithm [40] was used to determine the geodesic length of each connected component in the resulting binary image, which renders the length of each fiber in pixel. This value can then be converted to metric units (pixel size of 2.73 μm/pixel).

All these image processing steps are automated and can be performed in batch mode. Fig. 2 shows an example of the image processing used to quantify microfibers. This set of data processing allows quantifying the total number of microfibers, as well as their distribution in number and size.



OM image

1) Histogram segmentation

2) Negative



4) Border kill







5) Morphological analysis



Fig. 2. The several steps of the counting method processed within the plug im! platform.

165

## 166 **2.5. Microfiber weight estimation**

167 The results obtained by the quantification method can be used to calculate the microfibers mass.

168 These values can then be compared to the results obtained by filtration. The SEM images were

169 analyzed with ImageJ [41] to measure the diameter of the fibers before washing and after being 170 retained by the filter. Thirty measurements per fabric type were performed to determine the 171 average diameter D of the fibers, as well as their deviation. Results of average diameter are 172 presented in Table A2. The diameter was assumed to be homogeneous along the fibers. The 173 mass of the released microfibers was then calculated using the Eq. 1:

174

175 
$$M_f = \rho \sum_{i=50\mu m}^{5mm} N_i \cdot \left(\frac{\pi . D^2}{4}\right) \cdot L_i$$
 Eq. (1)

176

177 where  $\rho$ : density of the material;

- $M_{\rm f}$ : total mass of released microfibers;
- 179 N<sub>i</sub>: total number of microfibers released in class i of length L<sub>i</sub> counted by plug im!;
- 180 D: average diameter of microfibers measured by ImageJ;

181 L<sub>i</sub>: length of microfibers considered in class i and measured by plug im!.

For this equation, the length distributions of microfibers obtained by the counting method were divided into 101 classes of 50  $\mu$ m width. Polymer densities were taken from Cesa et al.[17]: 1.365 g.cm<sup>-3</sup> for polyester, 1.150 g.cm<sup>-3</sup> for acrylic.

185

#### 186 **2.6. Microfiber average length**

187 The average microfiber length (in number) can be derived from microfibers length distributions188 using the Eq. 2:

189

190

 $L_{avg} = \frac{\sum_{i=50\mu m}^{5mm} N_i \cdot L_i}{N}$  Eq. (2)

191

192 where N is the total number of released microfibers counted by plug im!.

#### 194 **3.** Results and Discussion

#### 195 **3.1. Preliminary characterization before washing**

196 The five different garments were analyzed by SEM prior to washing. The SEM micrographs 197 (Fig. A.1) show different shape and configuration of fibers depending on the type of fabric. The 198 organization of the macroscopic fibers is not well defined except for those from the 100% 199 smooth polyester T-shirt (PET-2). PET fibers from fleece jacket (PET-1) or T-shirt (PET-2) 200 show a smooth surface and a constant cylindrical section. The same morphology is found for 201 acrylic fibers (PAN). In the case of PET/PA textile, 2 fibers morphologies can be distinguished 202 corresponding to PET and PA, with the latter having a flatter morphology (ribbon shape). 203 Acrylic fibers (PAN) have the largest diameter (16.99 µm) among the tested textiles (Table 204 A.2). The cotton fibers are morphologically different from the synthetic ones presenting a 205 twisted ribbon shape well-known for this fabric [42].

206

#### 207 **3.2. Results after washing**

208 In order to test the applicability of the developed analytical procedure, a total of 10 washing 209 experiments were performed. The first washing of PET-1 textile was repeated twice (PET-1 C1 210 and PET-1 C1-R) to verify the repeatability of the washing protocol. It should be mentioned 211 that these tests were made on two different PET-1 garment lots. The impact of the number of 212 washing cycles was investigated on the PET-1 garment up to 4 washes (PET-1 C1, C2, C3 and 213 C4). The effect of the detergent was also tested during the first washing of PET -1 (PET-1 C1-214 D). Finally, the five different textiles types were washed once (PET-1 C1, PET/PA C1, PET-2 215 C1, PAN C1, CO C1).

Table 1 and Fig. 3 and Fig. 4 show the results obtained by the developed analytical protocol. The counted filtered microfibers are expressed with respect to the mass of the initial dry garment. One should note that all results presented in this study are average values obtained from the filtration duplicates performed for each experiment. Moreover, it was decided to set a minimum length of 50 µm for the identification of microfibers by the counting method (optical
microscopy + post-processing image techniques) since the SEM observations did not reveal the
presence of microfibers with a length below this value. One possible explanation is that an
elongated shape object such as a fiber would not easily undergo a mechanical rupture at a low
length to diameter ratio [43].

Table 1	Main	Results.
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			N°	Filtration	plug im!	Eq. (2)	Eq. (1)
	Type of		washing	[]	[]	Lavg	[]
Test Tag	fabric	Detergent	cycles	mg.kg <sup>-1</sup>	µfibers.kg <sup>-1</sup>	μm	mg.kg <sup>-1</sup>
PET-1 C1	PET-1	-	1	485	690 000	194	87
PET/PA C1	PET/PA	-	1	1520	ND*	ND*	ND*
PET-2 C1	PET-2	-	1	428	1 570 000	241	149
PAN C1	PAN	-	1	339	1 390 000	181	273
CO C1	СО	-	1	305	ND*	ND*	ND*
PET-1 C1-D	PET-1	57.8 g	1	1751	2 820 000	160	294
PET-1 C1-R	PET-1	-	1	289	670 000	229	100
PET-1 C2	PET-1	-	2	72	530 000	237	82
PET-1 C3	PET-1	-	3	59	330 000	285	61
PET-1 C4	PET-1	-	4	53	220 000	301	43

\*ND: Not determined due to high level of entanglement of microfibers.



(c)



(d)



Fig. 3. Filtration results – a) repeatability test, b) effect of detergent, c) effect of number of
washing cycles, d) effect of fabric type – PET-1: 100% polyester fleece jacket, PET-2: 100%





Fig. 4. Microfibers length distributions – a) repeatability test, b) effect of detergent, c) effect
of the number of washing cycles, d) effect of fabric type – PET-1: 100% polyester fleece
jacket, PET-2: 100% smooth polyester T-shirt, PAN: 100% acrylic sweater – horizontal axis
was cut at 1050 µm for visualization purposes.

#### 243 **3.2.1.** Repeatability tests (PET-1 C1 and PET-1 C1-R)

From Table 1, it can be seen that the number of microfibers released is near those of the
reference (PET-1 C1) and the repeatability tests (PET-1 C1-R): respectively 690 000 and 670
000 µfibers.kg<sup>-1</sup> of garment. Length distributions are also relatively similar (Fig. 4a),
confirming the repeatability of these washing and sampling protocols with respect to
microfibers release.

249 Around 65 to 70% of the microfibers are smaller than 200 µm long and only 5% are longer than 250 600 µm for both tests (Fig. 4a), while the length of the longest microfiber was 3.25 mm for the 251 reference test (PET-1 C1) and 3.4 mm for the repeatability test (PET-1 C1-R). From these two 252 distributions, one can calculate average microfiber lengths of respectively 194 and 229 µm. 253 These values correspond to 16 to 19 times the diameter of PET-1 microfibers determined by 254 SEM (12.3  $\mu$ m). This is a much smaller average length than what has been reported by Napper 255 and Thompson [7] but more consistent with recent works that used dedicated techniques to 256 access information on microfibers length [30],[36],[23]. For instance, Zambrano et al. [30] 257 analyzed PET microfibers length distributions in washing machine effluents with a commercial instrument (HiRes Fiber Quality Analyzer). They determined that the most abundant length 258 259 population was 200 µm which was the detection limit of their analytical equipment. Very few 260 microfibers above 2.5 mm were detected by these authors.

261 Despite the good repeatability in terms of microfibers release, a significant difference in mass was found by the filtration method (Table 1) between the PET-1 C1 and PET-1 C1-R 262 experiments – a particle concentration (>0.7  $\mu$ m) of 485 mg.kg<sup>-1</sup> for the first was larger than 263 the 289 mg.kg<sup>-1</sup> for the latter. This difference is significant with respect to the uncertainty of 264 265 the filtration method (Fig. 2a). In addition, one can notice from Table 1 that the calculated 266 microfibers mass obtained from the length distributions using Eq. 1 is lower than the mass 267 obtained by filtration: the reference test microfibers seem to account for less than 20wt% of the 268 total mass obtained by filtration, which suggests the presence of non-fiber material release. This

269 was confirmed by SEM imaging (Fig. 5) that shows an amorphous deposit on the surface of the filters. This deposition is more present on PET-1 C1 which is likely related to the higher mass 270 271 obtained by filtration. This additional material release could be related to textile treatment such 272 as dyeing or finishing – these processes use various chemicals such as dyes, biocides or stain 273 repellents which could release residual quantities during garment washing. [44]. The 274 repeatability test was carried out with another garment lot and, since residual quantities of 275 chemicals may differ from one lot to another, it could explain the large difference in the mass 276 filtration results obtained.







Fig. 5. SEM observations of the amorphous deposit present at the surface of glass fibers filters
(0.7 μm porosity) after the filtration of water samples from (a) PET-1 C1 and (b) PET-1 C1-R
experiments.

### 280 **3.2.2.** Effect of the number of washing cycles (PET-1 C1, C2, C3 and C4)

Table 1 shows a large decrease in mass recovered by filtration between the first (PET-1 C1, 485 mg.kg<sup>-1</sup>) and the second cycle (PET-1 C2, 72 mg.kg<sup>-1</sup>). This phenomenon is mainly due to the reduction of materials other than microfibers since their calculated mass from length distributions (Eq. 1) is only reduced by 5 mg.kg<sup>-1</sup>. This hypothesis is confirmed by SEM observation (Fig. A.2) that shows the absence of the amorphous deposit after the second cycle. Moreover, calculated microfibers masses are very close to those obtained by filtration for cycles 2, 3 and 4 (Table 1). This shows that textile emissions are mainly composed of microfibers after the first cycle. Even if filtration results are always to be taken with caution for the quantification of microfibers, latter observation tends to validate (for washing cycle > 1) the common literature approach that directly assimilates filtration mass to microfiber mass [16]-[20].

The decrease of microfibers with the number of cycles observed in the present study (see Table 1) is in agreement with other works [7],[19],[20],[15]. For instance, Pirc et al.[19] showed that the emissions from the washing of a polyester fleece (no additive) initially decreased from 0.016 wt% (161 mg.kg<sup>-1</sup>) to 0.00239 wt% (23.9 mg.kg<sup>-1</sup>) for the fourth cycle and then stabilized at approximately 0.0011 wt% (11 mg.kg<sup>-1</sup>) after 8-10 cycles. The large openings of the filter (200 x 200  $\mu$ m) used to evaluate the mass of microfibers could explain the lower values obtained compared to the present work.

Regarding results from the counting method (Table 1), one can also see the decrease of 298 microfibers release from the first (690 000 µfibers.kg<sup>-1</sup>) to the fourth cycle (220 000 µfibers.kg<sup>-1</sup>) 299 300 <sup>1</sup>) of PET-1 garment. Fig. 4c shows the effect of the number of washing cycles on microfibers 301 distribution. The reduction of small microfibers (<200 µm) is strongly related with the number 302 of cycles whereas emissions of long microfibers (>600 µm) are relatively less impacted. This 303 shows the initial presence of small loose microfibers that may be simply attached to the textile 304 surface rather than an inherent part of the matrix. One consequence is that the average length 305 tends to increase with the number of cycles (from 194 µm for the first cycle to 301 µm for the 306 cycle number 4).

307

#### 308 3.2.3 Textile effect (PET-1 C1, PET-2 C1, PET/PA C1, PAN C1 and CO C1)

Filtration results ranged from 305 to 1520 mg.kg<sup>-1</sup> for the first washing of the five different fabric types tested in this study (Table 1). The lowest mass was obtained for the cotton towels (CO) and the highest for the 88% polyester/12% polyamide towels (PET/PA). Similarly to PET-1 garment, particles other than microfibers can be identified on the surface of the filter by SEM (Fig. A.3). These filtration results should not be interpreted as microfibers mass. The mass obtained by Eq.1 (Table 1) and based on microfibers distributions determined by the counting
method is a better evaluation of microfiber mass.

For PET/PA and CO garment, it was not possible to perform the counting method due to a high level of entanglement of the microfibers. Fig. 6 illustrates this phenomenon on PET/PA (left) and CO (right) textiles. Despite dilution and specific ultrasound treatment, the entanglement was still incompatible with the post-processing image techniques used in this study. For other textiles, the counting method was performed without any issue.

321 Microfiber emissions for PET-1, PET-2 and PAN garments during the first washing were respectively found to be 690 000, 1 570 000 and 1 390 000 microfibers per kg, which 322 correspond to 87, 149 and 273 mg.kg<sup>-1</sup> respectively (Eq. 1). A higher release of microfibers was 323 also found by Sillanpää and Sainio [20] for a polyester T-shirt compared to a polyester fleece 324 325 garment. However, Almroth et al. [45] determined that polyester fleece fabrics shed the greatest 326 amount of microfibers while Napper and Thompson [7] found that microfibers emissions from acrylic garments are higher than those of polyester. The variety in analytical methods, washing 327 328 conditions and garment types is probably responsible for the discrepancy between studies and 329 highlights the difficulty to conduct a rigorous comparison between literature results in the 330 absence of normalized procedures.

Fig. 4d, shows similarly shaped distributed microfibers for the three garment types. Microfibers smaller than 200  $\mu$ m in length represent 73, 67 and 78% of the distribution for PET-1, PET-2 and PAN, respectively. This agrees with Hernandez et al. [23] that showed that the overall microplastic fiber length profile remains similar regardless of the wash condition or fabric structure, with the vast majority of fibers ranging between 100 and 800  $\mu$ m in length.

336

337

#### (a) PET/PA C1

(b) CO C1



339 Fig. 6. Optical microscopy images of the surface of glass fibers filters after the filtration of wastewater samples from (a) PET/PA C1 test and (b) CO C1 test.

### 340

#### 341 3.2.4 **Detergent effect (PET-1 C1 and PET-1 C1-D)**

342 Detergents may contribute to the mass of filtering residues because of the insoluble inorganic 343 compounds they contain [46]. These compounds will account in mass determination by 344 filtration method and can also create a thick layer that may induce an error (underestimation) 345 in the counting method since the microfibers can be partially or completely embedded in the 346 detergent compounds.

From Table 1, one can see that a high value of 1751 mg.kg<sup>-1</sup> is obtained for the filtration result 347 with detergent (PET-1 C1-D) which is probably due to the presence of water insoluble 348 compounds at the surface of the filter. SEM observations confirm the presence of a thick layer 349 350 on the surface of the filter (Fig. A.2).

351 Detergent was found to cause a drastic increase of around 300% in the number of microfibers 352 emitted during the first cycle (see Table 1, 2 820 000 for PET-1 C1-D versus 690 000 for PET-353 1 C1). This is even more relevant for small microfibers (<200 µm), as shown in Fig. 4b, which 354 tends to decrease the average microfiber length.

355 Several studies have studied the impact of detergent on microfibers emissions, but no clear 356 conclusion seem to emerge. Hernandez et al. [23], showed that among different parameters such as the number of cycles or the washing temperature, the use of detergent appeared to impact the 357

release of microfibers the most. Falco et al. [46] came to a similar conclusion for polyester and 358 359 polypropylene fabrics. Carney Almroth et al. [45] showed that the use of detergent increase the 360 microfibers release for 3 out of 4 of their tested fabrics. However, Pirc et al [19] mentioned that 361 the use of detergent and softener did not significantly influence emission and Cesa et al. [17] 362 even recommend the use of detergent to diminish microfibers pollution. These inconsistencies 363 in conclusions are likely due to the different methodologies used in each study (analytical 364 method, washing conditions, garment type, detergent type etc...) and highlight the need for 365 setting standards in the community.

366

#### 367 **3.2.5** Design of microfibers capture technologies and environmental aspects

To prevent microfiber pollution, it is important to develop technologies that will efficiently retain these particles during textile washing. Currently they are no regulation already in force with respect to microfibers capture, but, for instance, the French government has recently voted a law that will impose the implementation of a microfibers capturing device for all new professional and domestic washing machines by 2025 [14]. So far, no retention efficiency target has officially been set, but one should expect ambitious goals to effectively limit this pollution and justify the deployment of such technologies.

375 The analytical procedure developed in present study allows to access the detailed morphology 376 of microfibers released from textile washing. Therefore, it is a tool to better apprehend the 377 object size that must be captured by the microfibers capturing device. Fig. 7 presents the 378 microfibers cumulative length distributions (in number and in mass) for the different 379 experiments conducted in present work. If one considers a target of 90% retention efficiency in 380 number, from Fig. 7a it can be inferred that the future system should be able to stop microfibers 381 with a length lower than 100 µm and probably close to 50 µm. If the mass is chosen as the 382 criterion, then the 90% efficiency would lead to consider a minimum microfiber length to be 383 retained of around 100 µm based on Fig. 7b. One should also keep in mind that the small

384 diameter of microfibers (8 to 16  $\mu$ m, see Table. A.2) must be accounted in the design of the 385 retention technology. Indeed, depending on the approach angle, a microfiber can pass a filter 386 through its diameter section rather than its longitudinal section.

387 Several microfibers filter technologies are available at commercial stage for domestic washing machines such as PlanetCare [47], Lint-LUV [48], GuppyFriend [49] or Xfiltra<sup>TM</sup> [50]. These 388 389 technologies were tested and characterized by Napper et al. [22]. The largest pore size was 390 found for the filters Lint LUV-R (2 pore sizes: 285 µm and 175 µm) and PlanetCare (pore size 391 of 200 µm). Low microfiber retention efficiencies were found for these two technologies (<30% 392 in mass [22]) which seems consistent with the small dimensions of microfiber described in the 393 present work. The bag device (Guppyfriend) and the Xfiltra filter showed higher efficiency (54 and 78% in mass respectively [22]) due to their smaller pore size of respectively 50 and 60µm. 394 395 This confirms the necessity of a fine filtration technology to effectively retain microfibers from 396 washing machines.



Fig. 7. Microfibers cumulative length distributions – a) in number, b) in mass – PET-1: 100%
polyester fleece jacket, PET-2: 100% smooth polyester T-shirt, PAN: 100% acrylic sweater–
horizontal axis was cut at 1050 µm for visualization purposes.

While the description of the morphological properties of the microplastics delivered by the 401 402 washing of synthetic cloth is essential for the design of capture technologies, it is also of prime 403 importance for the good understanding of their environmental impact. Indeed, the 404 environmental impacts of microplastics are related to their chemical composition, and to the 405 morphology of the individual particles that compose them and that are released in ecosystems. 406 The environmental persistence is, for a specific chemical composition of plastic, directly 407 correlated to the surface to volume ratio of its individual particles. The environmental penalty 408 of a microplastic particle is not only related to the persistence of that particle, but also to its 409 bioavailability to living organisms throughout the food chain, which has been recently shown 410 to be shape dependent [51], [52]. These arguments reinforce the need to characterize in detail 411 the morphology of microplastics in order to better understand and control their impact. The two 412 aspects of capture and environmental impact will certainly be confronted in regulatory 413 measures, so that the technologies reach reasonable remediation levels. In this perspective, the 414 measurement of fibers size distributions should concentrate necessary efforts of normalization.

415

#### 416 4 Conclusions

417 We present in this paper a novel analytical protocol that consists in characterizing microfibers 418 in number and size through filtration techniques, optical and electronic microscopy and 419 automatic image post-processing. The filtration process allowed to estimate the mass of 420 particles larger than 0.7µm while depositing them on a surface that could be easily characterized 421 by microscopy. However, it did not distinguish the mass belonging to the microfibers from the 422 mass coming from other compounds potentially present in solution, especially during the first 423 wash. Scanning electron microscopy provided more precise information on the morphology of 424 all the products retained by the filtration and on the diameter of the microfibers, showing that 425 other amorphous organic compounds are also retained during filtration. However, the nature of 426 these products is currently unknown. Optical microscopy and image post-processing using the 427 plug-im! platform provided reliable and detailed results for counting and size distribution of the428 microfibers emitted from washing machines.

The analytical method was successfully applied on different washing conditions: 3 different synthetic garments (PET-1, PET-2 and PAN), several cycles and in the presence or absence of detergent. The automated image processing was however not successful to characterize cotton and a blend of PET/PA due to a high level of entanglement between the microfibers.

The results obtained by this method provide an alternative to the current characterization protocols. The proposed characterization protocol of microfiber length can also be extrapolated to the counting of several other objects that have a similar morphology to the analyzed fibers. The data can be helpful to the development of new microfibers capture technologies as well as better quantifying the environmental impact of textiles waste, and could pave the way for providing reference methodologies in the future.

439

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- 595 Appendix A
- 596

Table A.1. Garment characteristics.

	<b>Р</b> ГТ_1	PFT_7	ргт/рл	PAN	CO
	1 1 1 - 1	1 1/1 -2		IAN	co
Color	black	black	black	black	navy-blue
Composition	100% PFT	100% PET	88%PET/	100%PAN	100%CO
Composition	100/01/21	100/0121	12%PA	100/01/11	100/000
Garment model	fleece jacket	T-shirt	towel	sweater	towel

Garment Size	Extra Large	Large	-	Medium	-
Garment weight (g)	294	112	327	531	470
Fabric weight per unit area (g.m <sup>-2</sup> )	194	117	166	543	374
Fabric thickness (mm)	1.6	0.4	0.6	4.5	2.4
Fabric type	knitted	warp knit	woven	ribbing knit	terry cloth
Yarn type	Filament	Filament	Filament and	two ply-yarn	two ply-yarn
			staple	(staple)	(staple)





Fig. A.1. SEM micrographs of garment fibers.

600 Table A.2. Average diameter of garment fibers determined by SEM. Average diameter Type of fabric (µm) PET-1 12,32 (± 2,90) 9,59 (± 1,67) PET-2 PET/PA 8,20 (± 2,84) PAN 16,99 (± 4,10) CO 13,21 (± 4,94) 601

ОМ

598

599

SEM



DF1-III OM SEM



