



Importance of Water-Volume on the Release of Microplastic Fibers from Laundry

Kelly, Max R.; Lant, Neil J.; Kurr, Martyn; Burgess, J. Grant

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The Importance of Water-Volume on the Release of Microplastic Fibres from Laundry

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1 **The Importance of Water-Volume on the Release of**
2 **Microplastic Fibres from Laundry**

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5 Mr. Max Richard Kelly[†], Dr. Neil Joseph Lant[‡], Dr. Martyn Kurr[†], Professor

6

James Grant Burgess^{*,†}

7

8 [†]School of Natural and Environmental Sciences, Ridley Building, Newcastle

9

University, Newcastle upon Tyne, NE1 7RU, United Kingdom

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[‡]Procter and Gamble, Newcastle Innovation Centre, Whitley Road,

11

Longbenton, Newcastle upon Tyne, NE12 9TS, United Kingdom

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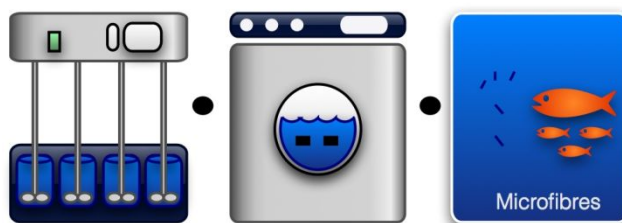
15 ***S Supporting Information***

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*Corresponding author: grant.burgess@newcastle.ac.uk

17 ABSTRACT:

18 The influence of laundry washing parameters on the release of microfibrils (MF) from
19 polyester textiles was studied. These fibres are an important type of microplastic pollution.
20 However, the factors which affect MF release during laundry, are poorly understood and
21 more rigorous methods for quantifying this release are needed. A novel method was
22 therefore developed using a tergotometer with eight (1000 mL) washing vessels and the
23 CIE Lab colour space measure of lightness (L^*). L^* was related to the mass of released MFs by
24 creating a calibration curve to quantify the amounts of MFs released from textiles during
25 washing. This method was used to investigate the effect of water-volume, agitation,
26 temperature, and duration of the wash on MF release. Counter-intuitively, increased water-
27 volume, characteristic of European 'delicate' cycles, resulted in the greatest release of MFs.
28 Full-scale testing was then carried out using domestic washing machines with real consumer
29 cycles to determine the effect of cycle type on MF release. In the first wash, delicate wash
30 cycles released 800,000 more MFs (94 mg/kg) per wash than a lower water-volume standard
31 wash and also increased MF release in subsequent washing cycles ($P < 0.05$). These results
32 indicate that a high water-volume-to-fabric ratio is the most influential factor for MF release,
33 rather than agitation as previously thought. Therefore consumers can reduce MF release by
34 avoiding high water-volume washes (delicate cycles), transitioning to appliances that use a
35 lower water-volume (North American high-efficiency washing machines), and ensuring full
36 wash loads are used.



37

38 INTRODUCTION

39 Alongside climate change and the overexploitation of natural resources, plastic pollution is
40 one of the most problematic anthropogenic impacts on the environment.¹ The environmental
41 consequences of meso- (5-20 mm) and macro- (>20 mm) plastic² pollution for large marine
42 organisms have been well-documented.³⁻⁶ More recently, the impacts of microplastic (<5
43 mm)⁷ pollution are also being investigated.⁸⁻¹⁰

44 Primary microplastics include manufactured microplastics such as, cosmetic microbeads¹¹
45 and textile MFs,¹² whereas secondary microplastics result from the breakdown of larger
46 plastic debris.¹³ Synthetic MFs can be ingested by a range of marine life including
47 commercially available fish and bivalves,¹⁴⁻¹⁵ crustaceans,¹⁶ non-commercial fish,¹⁷⁻¹⁸ birds,¹⁹
48 and worms.²⁰ Once ingested, MFs can lead to reduced food consumption and energy
49 availability,²¹ as well as increased mortality, at least in the laboratory.²²⁻²³ MFs have a global
50 distribution from rivers²⁴ to the ocean surface,²⁵ and are found to pollute even the deepest
51 ocean trenches.²⁶⁻²⁷ Major sources of global primary microplastic pollution include car tyres²⁸
52 and synthetic MFs from clothing,²⁹⁻³² which can enter the environment through waste water
53 treatment plants from laundry of synthetic textiles.³³⁻³⁴

54 Laundering textiles can release 500,000³⁵ to over six million³⁶ MFs for synthetic garments
55 and up to 13 million MFs from cotton garments per wash.³⁷ Over 42 million tonnes of
56 synthetic fibres are produced each year by the clothing industry³⁸ with polyester dominating
57 production (approximately 80%).³⁹⁻⁴⁰ In addition to synthetic MFs, anthropogenic natural
58 fibres are also released from laundering and can persist in and pollute aquatic
59 environments.⁴¹⁻⁴³ Therefore, it is important to target the laundry process to try and reduce
60 its impact on the environment.

61 To understand the factors that affect the release of MFs during laundry, reliable and
62 reproducible methods for their quantification are needed. A wide range of methods including
63 laboratory-scale^{36, 44-46} to full-scale washing machines^{32, 35, 37, 47-50} or a combination of both⁵¹⁻
64 ⁵² have been used to study MF release during laundry, leading to large disparities in the
65 literature and a general lack of understanding of the mechanisms of MF release. Methods
66 often do not reflect real domestic laundry conditions; for example, the use of steel balls
67 during washing,^{36, 44-46, 51-52} is unlikely to represent real world textile interactions. For
68 quantification of MF release, optical^{44, 46, 49} and electron microscopy,^{36, 51} and binary image
69 analysis⁴⁵ have been used. Microscopy can require scaling which incorrectly assumes MFs are
70 homogenously distributed across filters (used to collect MFs) leading to significant
71 inaccuracies. Binary image analysis⁴⁵ does not account for overlapping fibres, resulting in an
72 underestimation of fibre quantities. Consequently, it is difficult to make comparisons
73 between these studies. With larger scale studies the variability of methods is also
74 pronounced. When investigating the effect of repeated washing cycles as a proxy for garment
75 age on MF release, Sillanpää and Sainio,³⁷ reported a roughly 90% decrease in MF release in
76 the latter cycles. Conversely, Hartline et al.⁴⁷ reported that older garments release more MFs
77 when using a 24 hour continuous wash cycle to represent garment aging. Similarly, there are
78 mixed observations on the effects of detergent on MF release. Napper and Thompson³⁵ found
79 that the presence of detergent generally increased MF release, in line with De Falco et al.³⁶ In
80 contrast, Pirc et al.⁴⁸ reported detergent had no significant effect on MF release.

81 In addition to detergent and garment age, other studies have investigated fabric type,^{35, 37,}
82 ^{49-50, 52} filter size,^{46-47, 50-51} water hardness,³⁶ fabric softener,^{35-36, 48} temperature,^{35-36, 45, 49, 52}
83 and type of washing machine^{32, 47, 49} with equally variable results (Table S1). However, factors
84 affecting hydrodynamic forces on textiles such as water-volume, have not been studied. High

85 water-volume and lower levels of drum rotation (mechanical agitation) are characteristic
86 features of a “delicate” wash cycle in European-style front-loading washing machines
87 designed to protect sensitive garments from mechanical damage such as pilling.⁵³ In the US,
88 a delicate cycle also uses a lower agitation and spin speeds. However these machines often
89 use twice the water-volume (64 L in the main wash) compared to high-efficiency machines.⁵⁴⁻
90 ⁵⁵ Given the high levels of public concern about microplastics, best practices for mitigating MF
91 release are increasingly being provided by consumer organisations and media groups but
92 often without robust scientific data.⁵⁶⁻⁵⁷ For example, consumers are being encouraged to use
93 delicate washes to reduce MF release, with no practical evidence to support it.^{53, 56-57}

94 Therefore, in order to provide data which might be useful in justifying positive changes in
95 consumer behaviour, we developed a novel small-scale method to accurately quantify MF
96 release using a measure of lightness from black to white (L^*).⁵⁸ Measurement of colour was
97 preferred over microscopy to quantify MFs, as released MFs can be very small and can form
98 clusters on the surface of filter paper making them difficult to count due to overlapping fibres.
99 By using L^* , the concentration of black MFs more accurately correlates with colour as more
100 fibres result in a darker value and therefore overlapping fibres can be accounted for. L^* offers
101 a very precise measure of MFs as L^* is calculated for every pixel across the filter image. By
102 relating L^* to known masses of MFs using a calibration curve, the mass of released MFs can
103 be experimentally measured. This method was then used to investigate the effect of water-
104 volume, agitation, temperature, and wash duration on the release of polyester MFs and to
105 then confirm if these experimental observations were relevant to real consumer domestic
106 washing cycles. We hypothesised that different washing cycles would release different
107 amounts of MFs.

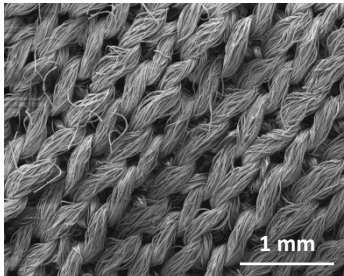
108 MATERIALS AND METHODS

109 A novel small-scale method was developed (Figure 1) using a tergotometer, which is a
 110 benchtop device comprising of eight (1000 mL) washing vessels that simulate full-scale
 111 domestic washing⁵⁹⁻⁶¹ (Copley, Nottingham, U.K.) (Figure S1) and was used to characterise the
 112 parameters affecting MF release. For large-scale studies, different washing cycles were
 113 performed using four front-loading washing machines (Miele[®], model: W3622) to determine
 114 the effects of real consumer cycles on MF release. Tap water (Northumbrian Water, United
 115 Kingdom) was used with a water hardness ranging from 113 – 128 mg/L (concentration of
 116 cations) throughout all testing.

117 **Textile.** In all testing, black 100% textured polyester T-shirts (Fruit of the Loom[®], code:
 118 61390) were used (Table 1). For the small-scale studies, the T-shirts were cut into 5x5 cm
 119 swatches using a laser cutter (HPC laser Ltd, model: LS1290) to seal the edges and prevent
 120 uncontrolled MF release from the cut edge, removing the need for serging (overlocking).

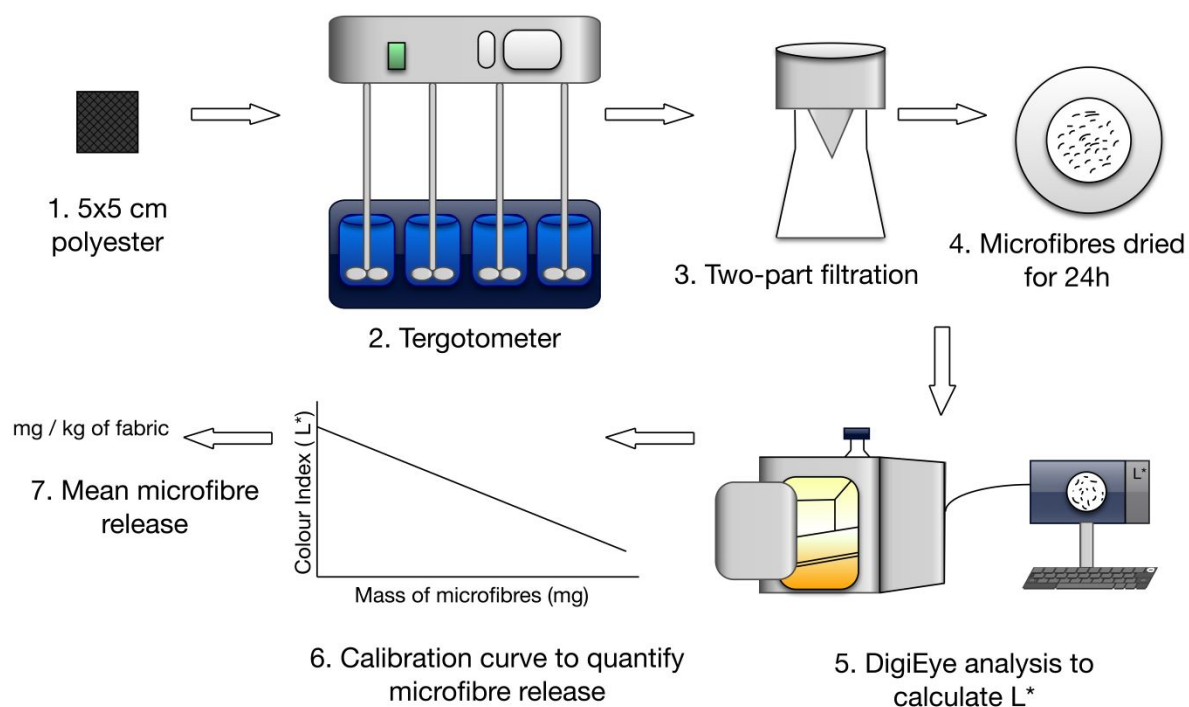
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122 **Table 1.** Physical properties of the textile.

Textile	Structure	Yarn	Mass (g/m ²)	SEM
100% Polyester	Knit	Filament	140	

124 **Small-scale washing procedure and microfibre collection.** Textile swatches (20 ± 0.10 g,
125 measured to two decimal places) were washed in the tergotometer steel pots with bi-
126 directional mechanical agitation (Copley, Cat. No. 6401+6403). Tests consisted of four
127 treatments washed over four cycles with four washing runs per cycle, resulting in four
128 treatment repeats for each cycle (Table 2 and 3; Figure S2). Agitation (RPM) and water-
129 volume were tested to determine the impact of the two parameters characteristic of
130 domestic delicate cycles that use an increased water-volume-to-fabric (mass) ratio. All
131 treatments were undertaken at 30°C with 0.5 mL Ariel® liquid for one hour including a single
132 three-minute rinse using the same volume of water as the main wash (Table 2). The use of
133 the same amounts of detergent was carried out deliberately to ensure the two methods were
134 qualitatively similar; generally in domestic use, the same volumes of detergent are added to
135 the washing machine independent of cycle type.⁶² Separately, wash temperature and
136 duration were tested to understand the effects of a cold, quick cycle compared to a longer,
137 warmer cycle on MF release. This was undertaken at 200 RPM in 300 mL of water with 0.5 mL
138 Ariel® liquid, also including a three-minute rinse (Table 3). The treatments were rotated
139 between pots after each wash to eliminate any potential bias. To avoid contamination, the
140 steel pots and arms of the tergotometer were thoroughly washed twice with deionised water
141 before and in-between each washing run to remove any residual fibres. Both tests included a
142 treatment of 0.5 mL Ariel® liquid at 30°C for one hour in 300 mL of water and 200 RPM as a
143 control. The wash and rinse water from each pot was transferred separately through a clean
144 stainless-steel funnel into separate collection containers (2 L) free of plastic particles
145 (confirmed by filtration onto Whatman® 541 filter paper, G.E. Life Sciences, Little Chalfont,
146 U.K).

147 **Filtration.** The same filtration method was used throughout. The wash water was filtered
148 using a vacuum pump in two stages. Firstly, through a 20 μm CellMicroSieve[®] (BioDesign Inc.,
149 Carmel, N.Y., U.S.A.) collecting the MFs on the surface. This was required to remove excess
150 dye and detergent that was found to have previously interfered with MF quantification. In
151 addition, the use of the larger (25 cm in diameter) CellMicroSieve[®] reduced the effect of
152 clogging issues reported in previous methods.^{32, 46, 50} The MFs were then re-suspended in
153 clean water (1 L glass beaker) before a second filtration step onto white, 22 μm pore size
154 Whatman[®] 541 filter paper held using a Büchner funnel. The filter paper was placed in a 140
155 mm diameter circular petri dish with the lid closed (VWR, code: 391-1503) to prevent dust
156 settling, that might cause contamination of the analysis, and left for 24 hours to dry at 50°C.



157
158

Figure 1. The small-scale method.

159 **Small-scale microfibre quantification.** Each filter (n = 64) was imaged using DigiEye® image
160 capture machinery/software⁶³ (VeriVide Ltd, Leicester, U.K.) to calculate an L^* value (defined
161 below). This system uses a DSLR camera (shutter speed 1/2.5, aperture width 7.1 mm) to take
162 an image in a controlled D65 illumination cabinet, subsequently viewed on a calibrated LCD
163 monitor using Engauge Digitizer chart v 3.5. The camera was calibrated using the Digitizer
164 chart characterising the camera RGB signal response to the CIE specification under fixed
165 lighting conditions in the illumination cabinet.⁶⁴ A fixed mask (area of analysis) was set over
166 the filter image using the 'fixed circle' tool with a radius of 750 pixels. The L^* of each pixel is
167 calculated and the overall average was cross-correlated to a calibration curve (Figure 2) which
168 was made to calculate the mass of released MFs. Using mass, the number of released MFs
169 could then be estimated.

170 **Table 2.** Treatment types for small-scale test 1: investigating agitation and water-volume^a

Treatment	Volume (mL)	Revolutions per minute (RPM)
A	300	200
B	600	200
C	300	100
D	600	100

^aWashes were carried out at 30°C for 60 minutes with 0.5 mL Ariel® liquid.

171

172 **Table 3.** Treatment types for small-scale test 2: investigating temperature and wash duration^a

Treatment	Temperature (°C)	Wash duration (minutes)
E	30	60
F	30	15
G	15	60
H	15	15

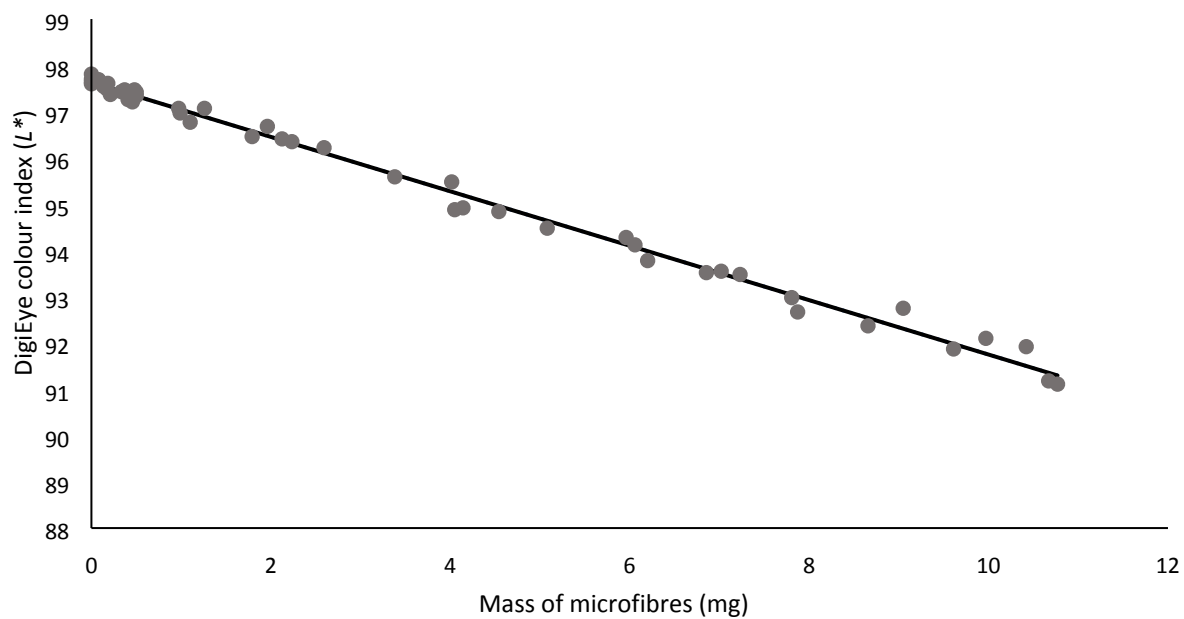
^aWashes were carried out at 200 RPM in 300 mL of water with 0.5 mL Ariel® liquid.

174 **Calculating L^* .** L^* is a correlate of the perceived lightness of an object in the specified
175 illuminant defined by the International Commission on Illumination.⁵⁸ It is proportional to the
176 luminance of the sample. This value was obtained through the DigiEye[®] software by
177 calculating the RGB values of every pixel across the entire filter image and taking an overall
178 mean. The RGB values are then converted to XYZ D65/10 (measured XYZ values for D65
179 illuminant a 10-degree observer). This is the amount of red, green, and blue response of the
180 light sensitive cone cells of the eye needed to match the colour in the specified illuminant. L^*
181 is measured using the XYZ directly. The XYZ D65/10 is converted to L^* using equation 1.⁶⁵

$$L^* = 116 \left(\frac{Y}{100} \right)^{\frac{1}{3}} - 16 \quad (1)$$

184 Where Y is a measure of the luminance scaled to 100. Therefore, a white that perfectly
185 reflected the light source would have a Y value of 100 and a perfect black would have a Y
186 value of 0. The L^* value therefore lies between 0-100 which represents a scale from black (L^*
187 = 0) to white ($L^* = 100$). This value can therefore be used as a proxy for the mass of MFs on a
188 filter, as the colour measurement is governed by the concentration of MFs.

189 **Calibration curve.** A calibration curve was created by generating MFs from washing the
190 textiles in the tergotometer before filtration (see above) onto filter paper and dried for 24
191 hours at 50°C. Clusters of MFs were then removed from the filter paper with forceps and
192 weighed using a thermogravimetric analyser, discovery model (TA Instruments, New Castle,
193 U.S.A.), a microbalance which accurately records the mass as a function of time (\approx five
194 minutes) and temperature (\approx 24° C) to four decimal places. Mass of MFs ranged from 0-11
195 mg. The clusters of MFs were then suspended in clean water (1 L glass beaker) and filtered
196 onto new filter papers. Each filter paper ($n = 49$) was then imaged with DigiEye[®] and the L^*



213

214 **Figure 2.** Microfibre calibration curve (n = 49).

215 **Full-scale washing procedure.** Four treatments were tested (Table 4) over four cycles with
216 four washing runs per cycle and no drying in-between cycles. For each washing run, all
217 treatment types were tested in four separate identical washing machines resulting in four
218 repeats of each treatment being washed per cycle. All treatments apart from treatment I
219 (Table 4) contained 35 mL Ariel® liquid to test the effect of detergent on MF release. Each
220 single washing load consisted of 10 T-shirts (1.5 ± 0.01 kg). Before testing, all washing
221 machines were initially cleaned with a high temperature (95°C) extended wash (130 minutes)
222 ensuring no MFs were present (confirmed by filtration, as above). The waste water pipe was
223 also cleaned following this to remove any residual fibres. After each washing run, the
224 machines were cleaned by running a further 'wash out' cycle (cold express cycle) with no load
225 to collect any residual fibres that were also filtered. The treatment type was then rotated to
226 a different machine to eliminate potential bias. Water was collected for filtration and analysis
227 on cycles one and four only; for cycles two and three, the wash water was discarded.

228 **Full-scale microfibre collection and quantification.** The wash water from each washing
229 machine was collected directly from the outflow pipe and stored in plastic containers (25 L)
230 cleaned of MFs and any residual particles with hot water (95°C), and filtered as above. The
231 experimental filters (n = 32) containing the MFs were weighed on a microbalance (AE ADAM®,
232 Milton Keynes, U.K.) to four decimal places. To account for the change in filter mass after
233 drying, ten blank filters were washed and weighed before and after drying, the change in mass
234 ($\approx 1\%$) was averaged and applied to the mass of each recorded filter from the investigation.
235 The average mass (mg) of fibres released per kg of textile washed was then calculated.

236 **Quality assurance testing.** To determine the amount of fibres remaining in the washing
237 machine after a washing run, additional testing of 'blank' washes was carried out using the
238 same textile in navy. Treatment II (Table 4) washing parameters were used on ten T-shirts in

239 triplicate using three identical washing machines (same model used in full-scale washing).
240 MFs were collected from the wash and wash out step. An additional cycle (treatment II) was
241 then carried out with no fabric present and any residual MFs were filtered and weighed.

242 **Statistical analysis.** Data was checked for homoscedasticity using Levene's tests. Treatments
243 in the small-scale method met the assumption of homogeneous variance but treatments in
244 the full-scale method were Log_{10} transformed to meet this assumption. Data from both
245 investigations were analysed using two-way ANOVA with 'treatment' (type of wash; 8 in the
246 small-scale, 4 in the full-scale) and 'cycle' (number of times that fabric had been washed;
247 either 1 or 4) as fixed, orthogonal factors. An alpha level of 0.05 was adopted, and *Tukey's*
248 post-hoc analyses were used to compare means of significant interactions or main-effects.
249 Results are presented as mean \pm standard error. All MF release rates are provided (Table S2).

250 **Table 4.** Treatment types for full-scale testing^{a,b}

Treatment	Detergent	Cycle	Parameters of the cycle
I	None	<i>Cotton short cycle</i>	85 minutes, 1600 RPM, 36 L, 30°C
II	35 mL Ariel® liquid	<i>Cotton short cycle</i>	85 minutes, 1600 RPM, 36 L, 30°C
III	35 mL Ariel® liquid	<i>Cold Express</i>	30 minutes, 1600 RPM, 30 L, 13-15°C
IV	35 mL Ariel® liquid	<i>Delicate cycle</i>	59 minutes, 600 RPM, 69 L, 30°C

^aThe cold express cycle uses un-heated water resulting in the small variation in temperature, whereas the water in the 30°C cycles is heated during the wash.

^bThe water-volumes provided are the total water-volume for the entire cycle.

252 RESULTS AND DISCUSSION

253 **Development of a microfibre release quantification method.** The R^2 value of the calibration
254 curve was very high (0.9937) (Figure 2), showing that L^* is an appropriate proxy for micro-
255 scale estimations of MF release. Previous attempts to quantify MF release in small-scale
256 studies have relied on labour-intensive manual enumeration of MFs using scanning electron
257 microscopy³⁶ or optical microscopy.^{44, 46} To save time when counting large numbers of MFs in
258 microscopy, the filter is sub-sampled and total fibres are estimated by scaling up. De Falco et
259 al.³⁶ accounted for MFs across 55% of the total filter, whereas Almroth et al.⁴⁴ sub-sampled
260 the filter into 16 equal areas. However, MFs do not have a uniform distribution across the
261 filters (see for example, Figures S4 and S5), therefore sub-sampling may not be an accurate
262 way of quantifying the total number of MFs. In contrast, DigiEye[®] images the entire filter,
263 quantifying all MFs. McIlwraith et al.⁶⁶ discuss the advantages of sub-sampling five 50 mL
264 aliquots to remove the need to count all MFs within a wash. However, fibres smaller than 100
265 μm could not be quantified and sub-sampled fibres were excluded from the overall weight of
266 released fibres in the remaining effluent. Similarly, Hernandez et al.⁴⁵ used two-dimensional
267 binary imaging to estimate percentage cover of MFs. When converting in this way however,
268 overlapping MFs are not detected resulting in an underestimation of MFs. When using L^* an
269 accurate measure of MFs is provided as a combination of reflectance, scatter, and adsorption
270 of the fibres are measured. Thus, for a cluster of fibres, some light will be trapped in the
271 spaces and some fibres will cast shadows over others; this results in multiple overlapping
272 fibres being darker than single fibres, thus more accurately measuring clusters. Although
273 testing with a single fabric is unlikely to represent real world laundry, single coloured fibres
274 were used in this study, and also De Falco et al.⁵¹ in small-scale testing, in order to develop an
275 experimental tool which can be used to experimentally investigate factors affecting MF

276 release. To study mixed loads, a range of black garments could be tested; however, to study
277 'real world' loads as part of our future studies, a new calibration curve can be created in the
278 same manner. The method is also applicable to light and dark coloured fibres providing the
279 calibration curve is made with the same textiles used in testing. Transparent fibres would not
280 be as suitable. DigiEye[®] can utilise the full RGB colour space; colour values A* and B*
281 represent green-red and blue-yellow which could be used to quantify individual and total
282 colour of real world laundry loads. Overlapping of mix coloured fibres would also produce a
283 darker colour value than single fibres and could therefore be accounted for; however, there
284 may be small variations in the colour values between different combinations of these
285 overlapping fibres. Microscopy is required to obtain fibre dimensions (Figure S3), which are
286 important when considering environmental, and human health impacts.

287 In addition to the analysis of the MFs, the novel application of the tergotometer does not
288 require the use of steel balls or steel vessels to house the textiles, which may cause unrealistic
289 MF release.^{36, 44-46, 51-52} The tergotometer used here simulates traditional central cone agitator
290 top-loading washing machines. However, the detergent industry also uses them widely as
291 model devices to simulate other types of washing machine through the development and
292 validation of various parameters such as wash duration, agitation level and rotation pattern,
293 and water: fabric ratio. The maximum spin speed (RPM) of the tergotometer arm was selected
294 to understand the effect of high and low agitation. However, the ranges of temperature and
295 duration possible using the tergotometer go beyond the parameters used in this study. In
296 addition, the textiles were laser cut to thermally seal the edges, negating the need for
297 serging.^{36, 45, 49, 52} This is a necessary step when using swatches of textiles as the fabric needs
298 to be cut. Therefore, loose fibres at the cut line may be released more easily than fibres in the

299 yarn structure and provide an inaccurate measure of MF release.⁴⁶ Thermally sealed edges
300 reduce this potential artefact in the results.

301 **Quality assurance testing.** After the wash and wash out cycle during full-scale washing, an
302 additional blank washing cycle was carried out (in triplicate) in order to quantify the level of
303 residual fibres left in the machine. The amount of residual MFs collected during this extra
304 wash was measured to be <3% of the total mass of released MFs during the first cycle (Figure
305 S6). This is within experimental error and therefore negligible.

306 **Delicate wash cycles increase microfibre release.** The effects of water-volume on MF
307 release were then investigated using the small-scale test. Water-volume was tested against
308 mechanical agitation, which relates to the rotation speed of the drum, frequency of
309 directional changes, and length of pauses in the cycle. This revealed significant differences in
310 MF release across the treatments (Table S3; Figure 3). Treatment D used a high water-volume
311 (600 mL) and low agitation (100 RPM), and resulted in a greater release of MFs compared to
312 all treatments except B, which also used 600 mL (Figure 3). These findings highlight that a
313 higher water-volume increases polyester MF release, whereas a higher mechanical agitation
314 does not. These parameters (high water-volume/low agitation) are characteristic features of
315 a 'delicate' wash cycle in European-style front-loading washing machines.⁵³ For US machines,
316 a delicate cycle equivalent will use a lower agitation and spin speed, however the water
317 volume is not always changed. This region has traditionally used larger top-loading machines
318 with a high wash water-volume (64 L in the wash step alone).⁵⁴⁻⁵⁵

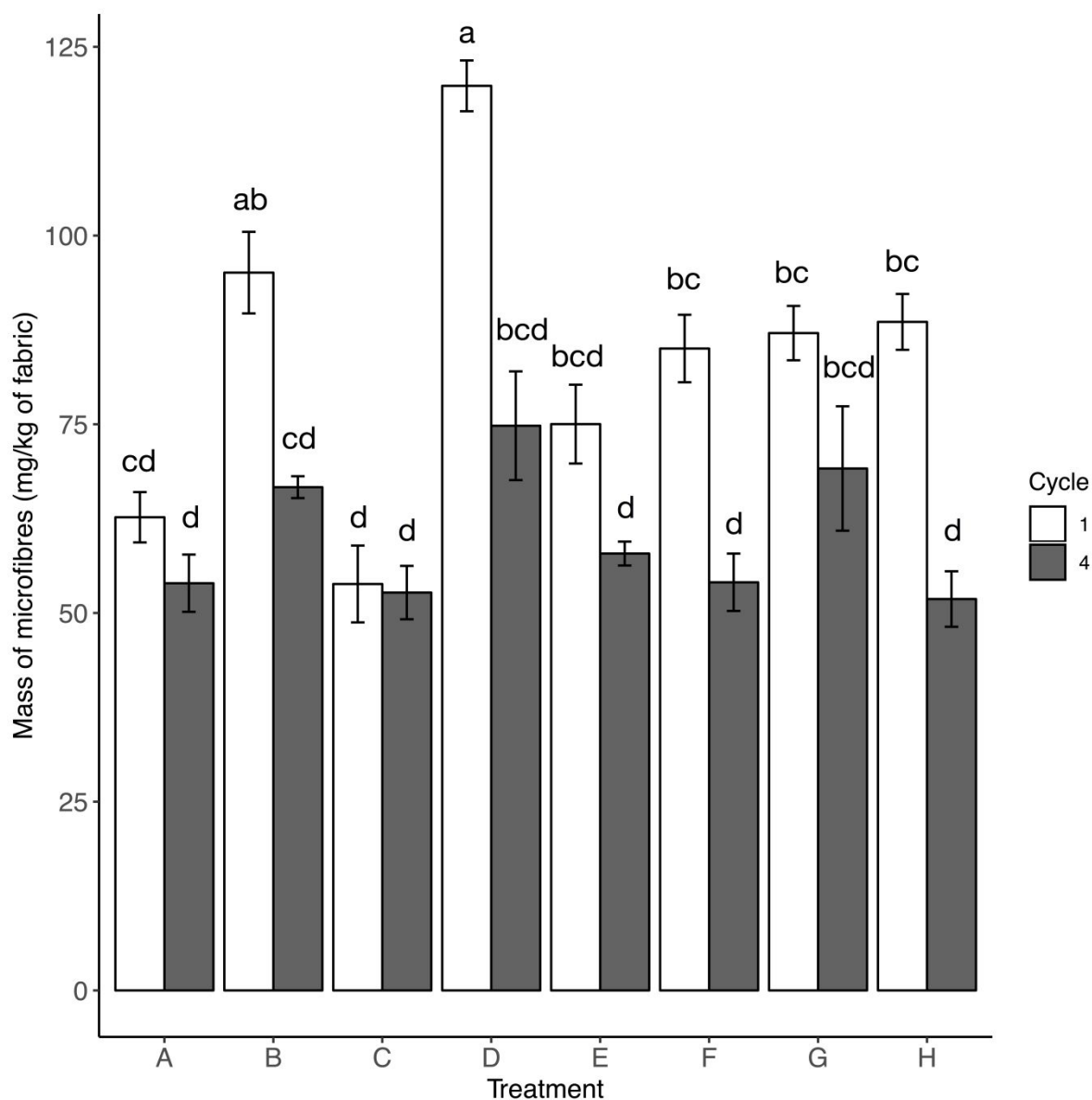
319 The observation that delicate wash-parameters released more MFs than 'normal' washing
320 parameters is somewhat counterintuitive and has not been reported previously. In order to
321 test whether observations made using the tergotometers were reflective of full-size domestic
322 washing machines, an actual delicate wash cycle (treatment IV; Table 4) was then tested and

323 also found to release significantly more MFs compared to other washes (Table S4; Figure 4).
324 A 'cotton short' programme (Table 4) was used as a 'normal' wash as this is one of the most
325 frequently used programmes by consumers with European washing machines.⁶² Therefore,
326 contrary to previous suggestions that higher mechanical agitation increases MF release,^{48, 50,}
327 ⁵⁶⁻⁵⁷ this work provides empirical evidence that water-volume is the more important driver of
328 MF release; reduced agitation still caused greater MF release when higher water-volumes
329 were used. In addition, there was no significant interaction between treatment and cycle
330 number (Table S4), indicating that delicate washes still result in the highest MF release after
331 at least four washes (Figure 4). Further testing is warranted to determine if this pattern
332 continues throughout the whole life of a garment, but also to investigate the effect of water-
333 volume on MF release in 'real world' mixed laundry loads. The physical characteristics of
334 different textiles, such as their structure, can affect the release of MFs.⁵⁰ Therefore additional
335 testing will be needed to determine whether MF release also increases in consumer mixed
336 loads as a result of higher water-volumes across a diverse range of cycle types. Results
337 obtained with the small scale tergotometers are qualitatively similar to results seen in the
338 larger washing machines. This means that the small-scale test method is a useful tool for
339 future more in depth studies on factors which affect MF release during laundry.

340 Delicate cycles may increase MF release due to greater overall hydrodynamic pressure on
341 the textile weave. Individual MFs have a very large surface area to volume ratio, and
342 consequently exhibit a low Reynolds number.⁶⁷⁻⁶⁸ As water passes through and over the
343 fabric, each individual MF will experience extremely large viscous forces, which could act to
344 pluck small fibres from the main textile weave. As delicate cycles also result in high MF release
345 during subsequent washes, hydrodynamic forces may continue to weaken the yarn structure
346 causing more loose fibres to be released from the yarn strand. Delicate washes increased MF

347 release by 114 mg, or approximately 800,000 MFs in the first wash (Table S2). This is
348 concerning since media groups have proposed greater adoption of delicate cycles as a way of
349 reducing MF release, citing ‘mechanical stress’ as an important factor with no experimental
350 evidence.⁵⁶⁻⁵⁷ If the results here are also true for a wider range of textiles then in addition to
351 using lower water-volume washes, switching to commercially available appliances that use
352 lower water-volumes regardless of wash type could also reduce MF release. Hartline et al.⁴⁷
353 reported a reduction in MFs when washing in front-loading machines compared to top-
354 loading machines. The study hypothesised that the central agitator in the top-loader maybe
355 more abrasive than drum rotation in the front-loading machine causing the increase in MF
356 release. However we show, for the first time, this difference could more likely be due to the
357 front-loading machine using a much lower water-volume. The global average annual water
358 consumption for domestic washing is estimated at 19 billion m³, with North America
359 representing the largest share (20%).^{54, 69-70} The use of high wash water-volumes in the
360 popular top-loading machines for North America could be a considerable factor for the high
361 release of 3 million tonnes of MFs each year from the US.^{24, 31} The transition to high-efficiency
362 washing machines that use approximately 50% less water in the main wash⁵⁵ is a necessary
363 step to reduce water and electricity consumption.⁷⁰ These data provide substantial evidence
364 that this conversion would also greatly reduce MF release, and could therefore inform both
365 manufactures and consumers to help reduce the environmental burden. For example, there
366 are an estimated 840 million domestic washing machines worldwide⁶⁹ with consumers not
367 always using a full laundry load.^{62, 70} If each user simply washed their laundry with full wash
368 loads (decreasing water-volume-to-fabric ratio), it would not only have a positive benefit for
369 energy and water consumption by reducing the number of washes, but could also reduce the
370 amount of MFs entering the global environment per wash.

371 Technologies proposed to reduce MF release have included the Lint LUV-R[®] filter and the
372 Cora Ball[®].⁶⁶ However, the implementation of washing machine filters will be challenging and
373 may take additional time to have an impact while the use of the Cora Ball[®] was found to
374 collect much fewer (26%) MFs compared to the filter (87%).⁶⁶ On the other hand, simply
375 reducing the water-volume-to-fabric ratio would have an immediate effect.



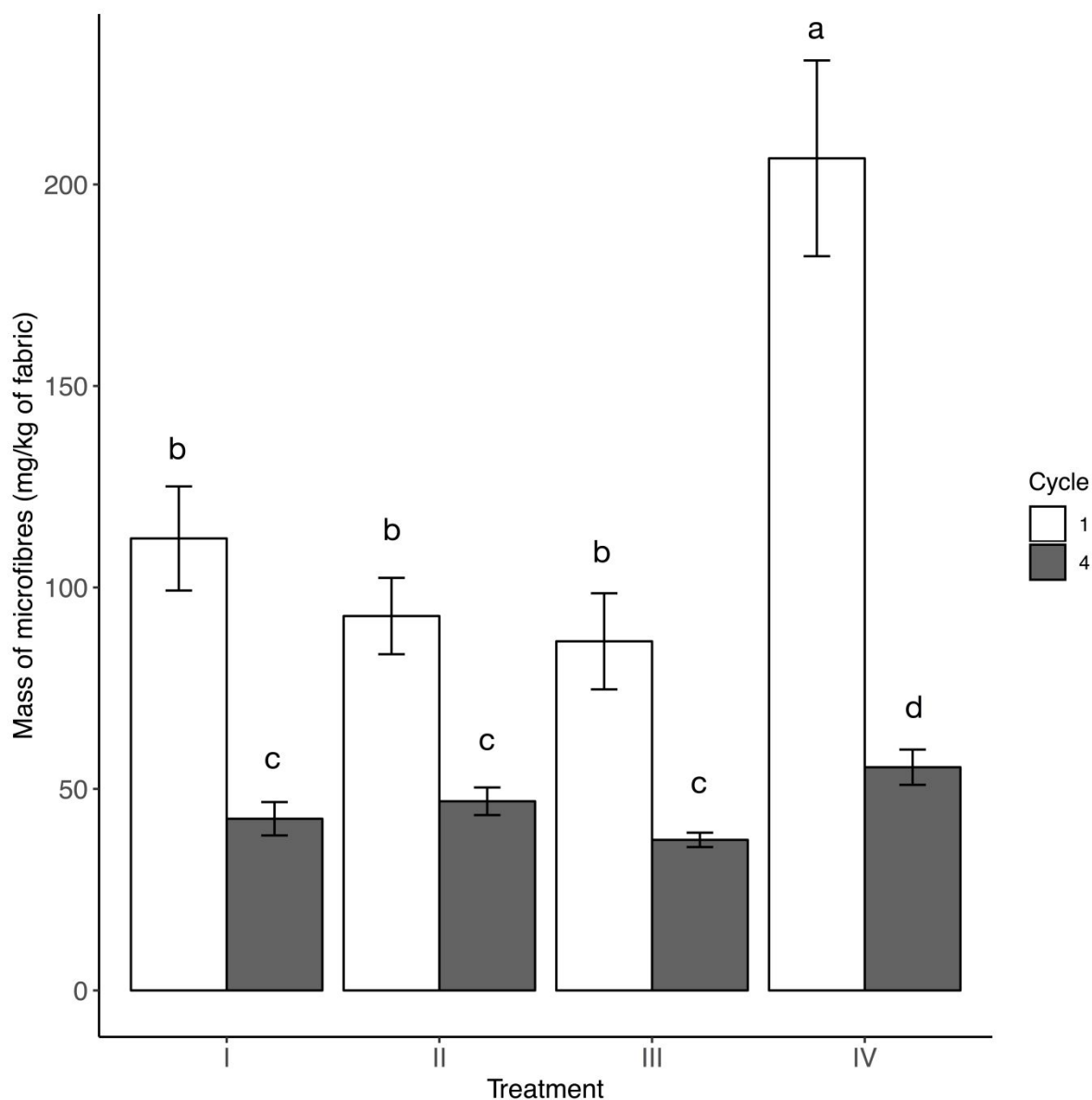
376

377 **Figure 3.** Mean mass (\pm SE) of released microfibres across seven different treatment types,
 378 A (300 mL/200 RPM), B (600 mL/200 RPM), C (300 mL/100 RPM), D (600 mL/100 RPM) all
 379 washed at 30°C for 60 minutes with detergent and treatments E (30°C/60 minutes), F
 380 (30°C/15 minutes), G (15°C/60 minutes), and H (15°C/15 minutes) all washed in 300 mL at
 381 200 RPM with detergent recovered during cycles one and four, for the small-scale
 382 investigation (note. treatments A and E are the same). Groupings based on Tukey's *post-hoc*
 383 analysis ($P > 0.05$); means that do not share a letter are significantly different.

384 **A greater release of microfibrils in the first wash.** After four wash cycles, fewer MFs were
385 typically released compared to cycle one, although this was less pronounced in small-scale
386 tests compared to full-scale washing (Figures 3 and 4). At the small-scale, cycle one in
387 treatments D, B, F, and H released significantly more MFs than cycle four, but for the
388 remaining treatments there was no significant difference. In the full-scale investigation, cycle
389 one ($\bar{x} = 124.37 \pm 14.40$ mg/kg) always resulted in a significantly greater release of MFs
390 compared to cycle four ($\bar{x} = 45.57 \pm 2.43$ mg/kg) (Table S4; Figure 4). The trend of decreasing
391 MF release with increasing wash cycle is documented in the literature (Zambrano et al.⁵²).
392 Sillanpää and Sainio,³⁷ found a large reduction in MF release from cycle one to five, for the
393 majority of garments tested (mixture of polyester and cotton textiles). Napper and
394 Thompson³⁵ also found a decrease in MFs over subsequent cycles with little difference
395 between cycles four and five for acrylic, polyester, and polyester-cotton textiles, which is
396 comparable to Pirc et al.⁴⁸ who reported a large initial spike in MF release for polyester fleece
397 blankets, which then plateaued in the later cycles. De Falco et al.⁵⁰ also reported a plateau in
398 MF release after four washes for polyester fabric, whereas for a garment with a mixture of
399 polyester/cotton/modal, MF release plateaued at cycle ten. Therefore fabric composition and
400 structure also appear to affect MF release as the fabric ages. Thus although there is a
401 possibility that MF release from additional unmonitored cycles two and three maybe higher
402 than the delicate cycle (Figures 3 and 4), this is unlikely. The initial spike in fibre release may
403 be from loose unbroken fibre debris from the yarn interior released in the first cycle. In
404 contrast, Hernandez et al.⁴⁵ found a steady release of 0.025 mg/g regardless of wash cycle;
405 however, the use of steel balls in this study may increase MF release in the later cycles
406 resulting in the consistent release over time. In addition, the method used included a prewash
407 step which could have removed the initial spike of MFs. Hartline et al.⁴⁷ found a 25% increase

408 in MF release for older garments. In this work, the garments were mechanically aged by a
409 continuous washing cycle over 24 hours and this may not simulate the real aging of garments
410 when worn and washed. Therefore additional tests on real consumer loads which have been
411 both worn and washed over longer periods are needed to determine the effects of garment
412 age on MF release. If more MFs are released from the newer garments, particularly in the first
413 cycle, this could be mitigated using a filtered pre-wash after garment manufacturing.

414



415

416 **Figure 4.** Mean mass (\pm SE) of released microfibres across four different treatment types, I
417 (30°C/cotton short/without detergent), II (30°C/cotton short/detergent), III (cold
418 express/detergent), and IV (30°C/delicate/detergent) recovered during cycles one and four,
419 for the full-scale investigation. Groupings based on Tukey's *post-hoc* analysis ($P > 0.05$);
420 means that do not share a letter are significantly different.

421 **The effects of temperature and wash duration.** At the small-scale, there were no significant
422 differences between treatments E, F, G, and H, suggesting the change in temperature and
423 wash duration (15 – 30°C; 15 – 60 minutes) had no impact on MF release (Figure 3; Table 3).
424 This is in agreement with Yang et al.⁴⁹ who found low temperature (between 30°C and 40°C)
425 had no impact on MF release, although higher temperatures (60°C) increased MF release for
426 polyester fabric. Temperatures of 15°C and 30°C were chosen here as studies within the
427 literature have often tested at 30°C and above, therefore, not addressing the effects of
428 ‘colder’ washes. Napper and Thompson³⁵ found temperature was not consistent in affecting
429 MF release, although between 30°C and 40°C there was an unspecified “increase” in
430 polyester MFs released compared to acrylic MFs. De Falco et al.³⁶ found a non-statistically
431 significant increase in MF release at 60°C compared to 40°C in plain weave polyester
432 garments, and Hernandez et al.⁴⁵ also testing polyester garments, found no significant
433 difference in MF release across a wider range of temperatures between 25°C and 80°C. It can
434 be concluded therefore, that temperature is not the most important factor affecting MF
435 release, although increases may occur at higher (60°C) to mid-temperature washes
436 (30/40°C),³⁵⁻³⁶ whereas below 30°C the change in MF release is less pronounced.

437 The 15 minute ‘express’ wash released as many fibres as 60 minute washes (Figure 3) which
438 may indicate that the majority of MFs are released during the first 15 minutes of the wash. In
439 full-scale, the ‘cold express’ was comparable to full-length washes (Figure 4; Table 4). This
440 appears to support the hypothesis that loose MFs are hydro-mechanically ‘plucked’ from the
441 textile opposed to being broken from the weave over the course of the first wash. In the latter
442 cycles more fibres may have to be broken for any subsequent and continued release to be
443 observed. In small-scale treatments F and H (15 minutes) there were significantly fewer MFs
444 released in cycle four in contrast to E and G (60 minutes) where no differences were observed

445 between cycles one and four. Hernandez et al.⁴⁵ initially hypothesised that an increased
446 mechanical agitation due to extended washing times would increase MF release; however,
447 even by extending the wash to eight hours there was no significant increase.⁴⁵ This suggests
448 that mechanical agitation caused by drum rotation and drum speed is not a significant factor
449 affecting MF release within a wash as previously mentioned, although shorter wash
450 programmes may still reduce MF release in subsequent washes. Further studies are needed
451 to establish how individual stages of the wash cycle (main wash, spin, and rinse) impact MF
452 release.

453 **The effect of detergent on microfibre release.** Detergent had no effect on MF release (Table
454 4; Figure 4). This was consistent with Pirc et al.,⁴⁸ although Napper and Thompson,³⁵ observed
455 inconsistencies in MF release in the presence of a bio-detergent. Conversely, Hernandez et
456 al.,⁴⁵ De Falco et al.,³⁶ Almroth et al.,⁴⁴ and Zambrano et al.⁵² found detergent leads to an
457 overall increase in MF release, although these studies used steel balls which could
458 mechanically interact with detergent causing unrealistic MF release. If the steel balls magnify
459 the effects of detergent, perhaps by forcing it into the textile weave or agitating the surfactant
460 so that more bubbles are produced, presumably there is some mechanism by which
461 detergents increase MF release which did not manifest in the present study. Further
462 investigations into detergent type and their interactions with different textiles are probably
463 warranted, but studies should use real-world conditions to keep results relevant.

464 In conclusion, we have developed a method for quantifying MF release in small-scale
465 conditions which qualitatively reflects the outcomes observed in full-sized domestic washing
466 machines. The small- and full-scale method both indicate a higher water-volume increases
467 MF release, temperature and duration have no significant effect, and MF release is greatest
468 in the first cycle. As public awareness of plastic pollution and the overall anthropogenic

469 environmental impact increases, domestic laundry is an important emerging target for
470 reducing the global environmental burden. Changes in domestic laundry behaviour could help
471 to address the UN sustainable development goals (SDG) 14, 'life below water' and 15 'life on
472 land'.⁷¹ This study shows that if consumers can adopt lower water-volume washes or
473 transition to lower water-volume washing machines, and increase wash load size (number of
474 garments per wash), this would prevent substantial quantities of plastic MFs from entering
475 the environment.

476

477 **ASSOCIATED CONTENT**

478 **Supporting Information**

479 Summary of factors affecting microfibre release (Table S1), description and image of the
480 tergotometer (Figure S1), small-scale experimental design (Figure S2), fibre metrics, fibre
481 measurements (Figure S3), all release rates (Table S2), images of released microfibres in the
482 full-scale investigation, cycle one (Figure S4), and cycle four (Figure S5), images of released
483 microfibres from quality assurance testing (Figure S6), Two-Way ANOVA statistics for small-
484 scale (Table S3) and full-scale (Table S4) testing

485

486 **AUTHOR INFORMATION**

487 **Corresponding Author**

488 *E-mail: grant.burgess@newcastle.ac.uk

489

490 **ORCID**

491 Max R. Kelly: 0000-0002-7136-0527

492 Neil J. Lant: 0000-0002-5939-0125

493 Martyn Kurr: 0000-0001-6649-024X

494 J. Grant Burgess: 0000-0002-1491-7341

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503

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