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# Examining the release of synthetic microfibres to the environment via two major pathways: Atmospheric deposition and treated wastewater effluent

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1 **Examining the release of synthetic microfibres to the environment via two**  
2 **major pathways: atmospheric deposition and treated wastewater effluent**

3

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5

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11

12 **Abstract**

13

14 Research on the discharge of synthetic microfibres to aquatic environments has  
15 typically focused on laundering, where fibres can be discharged via wastewater  
16 effluent. However emerging research suggests that microfibres generated during the  
17 wear of textiles in normal use could present a major, additional, pathway for  
18 microfibre pollution to the environment. This study aimed to quantify and compare  
19 the quantities of microfibre entering the marine environment via both these  
20 pathways; wastewater discharge and atmospheric deposition. Areas of high and low  
21 population density were also evaluated. Samples were collected in and around two  
22 British cities (Bristol and Plymouth) both of which are located on tidal waters. Fibres  
23 originating from the atmosphere were deposited at an average rate of 81.6 fibres m<sup>2</sup>  
24 d<sup>-1</sup> across urban and rural areas. Treated wastewater effluent contained on an  
25 average 0.03 synthetic fibres L<sup>-1</sup>. Based on our results we predict ~20,000-500,000

26 microfibrres could be discharged per day from the Wastewater Treatment Plants  
27 studied. When the two pathways were compared. Atmospheric deposition of  
28 synthetic microfibrres appeared the dominant pathway, releasing fibres at a rate  
29 several orders of magnitude greater than via treated wastewater effluent. Potential  
30 options to reduce the release of microfibrres to the environment are discussed and  
31 we conclude that intervention at the textile design stage presents the most effective  
32 approach. In order to guide policy intervention to inform the Plastics Treaty UNEA  
33 5.2, future work should focus on understanding which permutations of textile design  
34 have the greatest influence fibre shedding, during both everyday use and laundering.

35

### 36 **Key words**

37 Marine pollution, microplastics, synthetic fibres, atmospheric pollution, wastewater  
38 effluent, Plastics Treaty.

39

### 40 **Introduction**

41

42 Plastics in the microplastic size range (<5 mm) are prevalent environmental  
43 contaminants (Alimi *et al.*, 2018; Horton *et al.*, 2017; Napper *et al.*, 2021a;  
44 Thompson *et al.*, 2004; Wright *et al.*, 2020) of substantial public and scientific  
45 concern. The majority of studies have focused on microplastics in aquatic systems,  
46 primarily the marine environment (Duis and Coors, 2016; Horton *et al.*, 2017), where  
47 estimates suggest there could be 5.25 trillion plastic particles at the ocean surface  
48 and of those, 92% are microplastics (Eriksen *et al.*, 2014). However, fewer have  
49 traced and quantified microplastics along pathways or at points of entry to aquatic  
50 environments.

51

52 Microplastic waste can originate from a variety of different land-based sources such  
53 as leakage from wastewater treatment plants (WWTPs) (Kay *et al.*, 2018; Murphy *et*  
54 *al.*, 2016) and atmospheric pollution (De Falco *et al.*, 2020; Dris *et al.*, 2015; Wright  
55 *et al.*, 2020). Microplastics can then be transported to the marine environment by a  
56 variety of different mechanisms. Freshwater systems often connect inland and  
57 coastal communities to the ocean, (Miller *et al.*, 2017; Napper *et al.*, 2021b; Rech *et*  
58 *al.*, 2014; Schmidt *et al.*, 2017; Seo and Park, 2020; Weideman *et al.*, 2020).

59 Subsequently, microplastic pollution has been found to be highly abundant in coastal  
60 habitats, especially estuaries which provide unique and complex environments  
61 located between oceans and river mouths (Harris, 2020; Hitchcock and Mitrovic,  
62 2019). There is also growing recognition that atmospheric deposition is an important  
63 vector for the transportation of microplastics (Dris *et al.*, 2017; Napper *et al.*, 2020;  
64 Stanton *et al.*, 2019; Wright *et al.*, 2020), even into remote regions far from emission  
65 sources (Allen *et al.*, 2019; Brahney *et al.*, 2020; Roblin *et al.*, 2020).

66

67 Once in the environment, microplastics can be ingested by a range of organisms  
68 (Anastasopoulou *et al.*, 2013; Barnes *et al.*, 2009; Maaghloud *et al.*, 2020; Scherer *et*  
69 *al.*, 2017). Laboratory studies indicate they can, in some cases, interfere with feeding  
70 capacity (Cole *et al.*, 2015) and cause internal damage or toxicological effects in  
71 certain species (Lahive *et al.*, 2019; Powell *et al.*, 2010; Sussarellu *et al.*, 2016). In  
72 addition, organic and inorganic contaminants may adhere to and accumulate on  
73 microplastics surface, which may lead to negative effects to biota (Alimi *et al.*, 2018;  
74 Wang *et al.*, 2018). There is uncertainty about the specific extent and magnitude of  
75 the harm of microplastic pollution in the environment; however, there is a general

76 consensus, microplastic pollution is accumulating in the environment and that unless  
77 mitigation measures are implemented we could see wide-scale ecological harm in  
78 the natural environment within the next 50 – 100 years (SAPEA, 2019).

79

80 Synthetic microfibres are often the most commonly reported form of microplastics in  
81 the environment; from soil to aquatic systems (e.g. oceans, rivers, shorelines and  
82 lakes) (Auta *et al.*, 2017; Biginagwa *et al.*, 2016; Horton *et al.*, 2017; Napper *et al.*,  
83 2021b; Thompson *et al.*, 2004; Woodall *et al.*, 2014). Approximately 63% of textile  
84 fibres produced are synthetic (e.g., polyester, nylon) (The Fibre Year, 2018), and  
85 over 42 million tonnes of synthetic fibres are produced each year by the clothing  
86 industry (Carr, 2017) with polyester dominating production (approximately 80 %)  
87 (Krifa and Stevens, 2016; L'Abbate *et al.*, 2018). As such, Boucher and Friot (2017)  
88 estimated that of all primary microplastics in the world's oceans, 35% arise from  
89 laundry of synthetic textiles, an estimated 2 - 13 million tons per year globally  
90 (Boucher and Friot, 2017; Mishra *et al.*, 2019).

91

92 Microfibres can be released from clothing during the washing process due to  
93 mechanical stresses (Belzagui *et al.*, 2019; Cesa *et al.*, 2020; De Falco *et al.*, 2018;  
94 Napper and Thompson, 2016). As a consequence, it has recently been estimated  
95 that over 6,000,000 microfibres could be released from an average domestic 6 kg  
96 wash (De Falco *et al.*, 2018). Laundry effluent can be released directly to the  
97 environment or it enters municipal wastewater treatment plants where the majority of  
98 microplastics detected are reportedly microfibres (Gies *et al.*, 2018; Gündoğdu *et al.*,  
99 2018; Leslie *et al.*, 2017). To date, the majority of research has focused on the  
100 marine environment with treated wastewater effluent commonly reported as a major

101 pathway for microfibre contamination attributable to the laundering of textiles  
102 (Belzagui *et al.*, 2019; Cesa *et al.*, 2020; De Falco *et al.*, 2018; Napper and  
103 Thompson, 2016). However, several studies have reported that considerable  
104 quantities of microfibrils from clothing pass to aquatic environments via atmospheric  
105 deposition (Napper *et al.*, 2020; Wright *et al.*, 2020). Research by De Falco *et al.*,  
106 (2020), estimated the quantity of plastic microfibrils released into the atmosphere  
107 directly as a consequence of wearing clothes compared to washing clothes. For  
108 polyester clothing, the study estimated that one person could emit approximately  
109  $2.98 \times 10^8$  microfibrils per year to water by washing, and  $1.03 \times 10^9$  microfibrils per  
110 year to the atmosphere by wearing polyester garments. Atmospheric deposition  
111 rates for microplastics (predominately fibres) have also been studied in urban areas  
112 which range from  $10 \text{ m}^2 \text{ d}^{-1}$  (Gdynia, Poland; (Szewc *et al.*, 2021)) to  $771 \text{ m}^2 \text{ d}^{-1}$   
113 (Central London, England; Wright *et al.*, 2020), and for remote regions from  $12 \text{ m}^2 \text{ d}^{-1}$   
114 (Mount Derak, Iran; Abbasi and Turner, 2021)  $365 \text{ to } \text{m}^2 \text{ d}^{-1}$  (French Pyrenees;  
115 Allen *et al.*, 2019). Such findings imply that previous estimations of microfibre  
116 pollution entering the environment are likely to be underestimated.

117

118 Although there is good understanding of the prevalence of microplastic waste in the  
119 marine environment and potential impacts, there is less clarity on the environmental  
120 pathways and underlying causes. This presents a major barrier to implementing  
121 solutions (Galloway *et al.*, 2020; Napper and Thompson, 2020) which will be of  
122 particular importance to inform UNEA 5.2 – the Plastics Treaty. Therefore, the aim of  
123 this study was to characterize and quantify microfibre release and loadings to the  
124 marine environment via two major pathways; treated wastewater effluent and  
125 deposition from the atmosphere. Additionally, we compared synthetic fibre loadings

126 from the atmosphere between urban (densely populated) and rural areas (sparsely  
127 populated).

128

## 129 **2.0 Method**

130

### 131 **2.1 Environmental sampling**

132

133 To quantify synthetic microfibres at their points of entry to marine and estuarine  
134 environments, two pathways were sampled: deposition from the atmosphere and  
135 discharge of treated wastewater effluent. All sampling was conducted in and around  
136 two coastal cities in the South-West of England (U.K.) between October 2018 and  
137 March 2019. The two cities (Plymouth and Bristol) provided generality to the results  
138 and were representative of typical coastal catchments in the UK. For both cities there  
139 were no specific assumptions relating to the locations; consequently, results are not  
140 presented according to location. The population densities of Plymouth city and  
141 Bristol city are 3,300 people per km<sup>2</sup> and 3,892 people per km<sup>2</sup> respectively, while  
142 the total population of Bristol is almost double that of Plymouth; 465,900 (Bristol City  
143 Council, 2022) and 263,070 (Plymouth City Council, 2019).

144

#### 145 **2.1.1 Atmospheric Deposition from Urban and Rural Environments**

146

147 Atmospheric deposition samples were collected at ground level within urban and  
148 rural environments, characterised by densely and sparsely populated areas and by  
149 land use classification. From each city (Plymouth and Bristol), two sites of each  
150 environment were selected (8 locations in total) based upon access and permission

151 to sample on private land to avoid potential tampering of samples from members of  
152 the public. Each site was sampled on two separate occasions and six replicates  
153 were collected on each occasion.

154

155 Sampling consisted of straight sided glass dishes (surface area 0.0177 m<sup>2</sup>) placed at  
156 ground level for 24 hours to collect atmospheric fallout. Given the sampling period  
157 and the collecting surface area, the atmospheric fallout is expressed as a number of  
158 microfibrils deposited per square meter per day. Each dish contained deionised  
159 water (~1 L) which captured any atmospheric fallout on the meniscus. The deionised  
160 water was treated by reverse osmosis before passing through ion-exchanged resin  
161 and filtered to 0.2 µm. On completion, the deionised water was poured through a  
162 glass funnel into 1 L glass Duran bottles on site. Each dish was then rinsed three  
163 times with filtered ion exchanged deionised water and added to the sample.

164 Atmospheric sampling was conducted during periods of dry weather to avoid loss of  
165 sample due to overflowing or splashing. All urban atmospheric deposition sites were  
166 located in and around the city centre, while rural sites were by necessity located in  
167 areas surrounding the city. In Bristol, rural sites were located within a ~ 10 km radius  
168 to the west and northwest of the city, and in Plymouth within a ~25 km radius with  
169 sites to the west and northeast of the city.

170

### 171 **2.1.2 Wastewater Treatment Plants**

172

173 For both locations (Plymouth and Bristol), treated effluent was collected in three  
174 replicate 10 L samples from two WWTPs on two separate occasions (4 locations in  
175 total). WWTP one served a population of 60,000 people, with a 22 km<sup>2</sup> catchment



176 and tertiary level treatment. WWTP two served 65,000 people, with a 20 km<sup>2</sup>  
177 catchment and tertiary level treatment. WWTP three served 2,808 people, with a  
178 1.23km<sup>2</sup> catchment and secondary level treatment. WWTP four served 18,471  
179 people, with a 10.73 km<sup>2</sup> catchment and secondary level treatment. See SI for  
180 specific details of treatment at each plant. The WWTPs sampled received both foul  
181 water and surface water drainage. WWTPs sampled in Bristol were located in the  
182 southeast and northeast of the city. WWTPs sampled in Plymouth were located in  
183 the east and northwest of the city. The WWTPs sampled primarily served residential  
184 areas.

185

## 186 **2.2 Laboratory Analysis**

187

188 All samples were vacuum filtered onto Whatman cellulose nitrate membrane filter  
189 papers  $\leq 12 \mu\text{m}$ . Due to the large volumes of liquid and high content of suspended  
190 solids, treated wastewater samples were first passed through 30  $\mu\text{m}$  (stainless steel)  
191 and 12  $\mu\text{m}$  (clear nylon) meshes. The contents of each mesh was rinsed into a  
192 beaker with deionised water before vacuum filtering.

193

194 All filter papers were then examined using an LED Microtec light microscope and any  
195 potential synthetic microfibres (minimum particle size  $> 20 \mu\text{m}$ ) were removed onto a  
196 blank Whatman glass microfibre 1.6  $\mu\text{m}$  filter paper using the criteria employed by  
197 Hidalgo-Ruz *et al.* (2012). Sub-sampling was employed when the abundance of  
198 microfibres was very dense; 50% of the filter paper surface was analysed. Images of  
199 the isolated microfibres were taken by using LEICA M205C light microscope and  
200 used to approximately measure length using ImageJ. Colour was also recorded.

201

202 Polymer identification was performed via Fourier-Transform Infrared Spectroscopy  
203 (FTIR) in transmission mode with a Hyperion 1000 microscope coupled to a Vertex  
204 70 spectrometer (Bruker). For each sample, the spectra were recorded with 32  
205 scans in the region of 4000 to 600 cm. Spectra obtained were compared against a  
206 spectral database of synthetic polymers (BPAD polymer & synthetic microfibres  
207 ATR) and additionally analysed visually to confirm polymer matches where  
208 necessary. If a sample contained >10 microfibres, a sub-sample of 10 random  
209 microfibres were selected to be analysed by FTIR. To prevent bias in particle  
210 selection, each filter paper with the extracted fibres was divided into 8 sections. A  
211 random number generator was used to determine the section and the order from  
212 which to take each particle (from left to right). If there was not enough in the selected  
213 section, this continued to be repeated until 10 fibres were reached. Relative  
214 proportions of different polymer types were then adjusted to give an approximation  
215 for the whole sample. In this paper the term microfibre will refer exclusively to  
216 microfibres that are <5 mm by their longest dimension.

217

218 While many studies choose to include regenerated cellulose microfibres (e.g. rayon)  
219 in their abundance estimates, (e.g. Frias *et al.*, 2016; Gies *et al.*, 2018; Neves *et al.*,  
220 2015; Peng *et al.*, 2018; Woodall *et al.*, 2014) there remains uncertainty in the ability  
221 to confidently differentiate regenerated cellulose microfibres from naturally occurring  
222 cellulose since they have almost identical spectra (Lusher *et al.*, 2014; Blumenröder  
223 *et al.*, 2017; Martin *et al.*, 2017). Therefore, our analysis does not include microfibres  
224 characterised as 'rayon' by FT-IR. This may result in the overall abundance of

225 microfibrils in this study appearing lower compared to previous estimates where  
226 rayon was included.

227

228 For quality assurance and quality control, procedural blanks were collected. This  
229 involved the entire methodology being repeated but without any exposure to the  
230 environment and deionised water being used as the sample. Minimal contamination  
231 was reported from the procedural blanks ( $\bar{x} \pm SE$ ;  $0.22 \pm 0.15$  microfibrils per litre  
232 from 9 procedural blank replicates). Additionally, all laboratory analysis was  
233 conducted in a purpose-built laboratory; which had controlled access, and where  
234 blanks were collected every 30 minutes to quantify any potential contamination  
235 originating from the laboratory. Cotton laboratory coats were worn at all times to  
236 minimise contamination from synthetic clothing. Glass or stainless steel laboratory  
237 ware was used wherever possible and was thoroughly rinsed with filtered  $1.6 \mu\text{m}$   
238 Milli-Q water before use. Minimal contamination was reported from the laboratory  
239 blanks (3 plastic microfibrils found in total;  $\sim 0.14 \pm 0.10$  microfibrils per sample).

240

241 Analysis of variance (ANOVA) was used to compare synthetic fibre discharge  
242 between wastewater treatment plants (where city and date are random factors).  
243 Homogeneity of variance was assessed prior to ANOVA and transformations  
244 applied, if appropriate. For atmospheric samples, a binomial model was used to test  
245 effects of urban vs. rural environments (as a fixed factor). P-values for fixed factors  
246 (site and date) were derived from likelihood-ratio chi-squared tests. It is not possible  
247 to Tukey Test a model with random factors so planned contrasts was applied to  
248 achieve same end. Standard Error (SE) of the mean was used for all analysis.

249

## 250 **2.5 Comparisons between pathways**

251

252 The units required to quantify synthetic microfibres within treated wastewater effluent  
253 (microfibres L<sup>-1</sup>) and within atmospheric fallout (microfibres m<sup>2</sup> d<sup>-1</sup>) are by necessity  
254 different, making a quantitative comparison between the two pathways challenging.

255 To compare the release of fibres between the two pathways, emissions of fibres  
256 within treated wastewater effluent were scaled from microplastics per litre to  
257 microplastics a day based upon the volume of treated effluent released per day (L)  
258 from each plant, averaged over each weather season. For atmospheric data, the  
259 average number of synthetic fibres deposited over a m<sup>2</sup> per day across all urban and  
260 rural sites was scaled to the catchment size each WWTP served. This enabled  
261 conclusions to be drawn about the relative importance of the various pathways  
262 examined.

263

## 264 **3.0 Results**

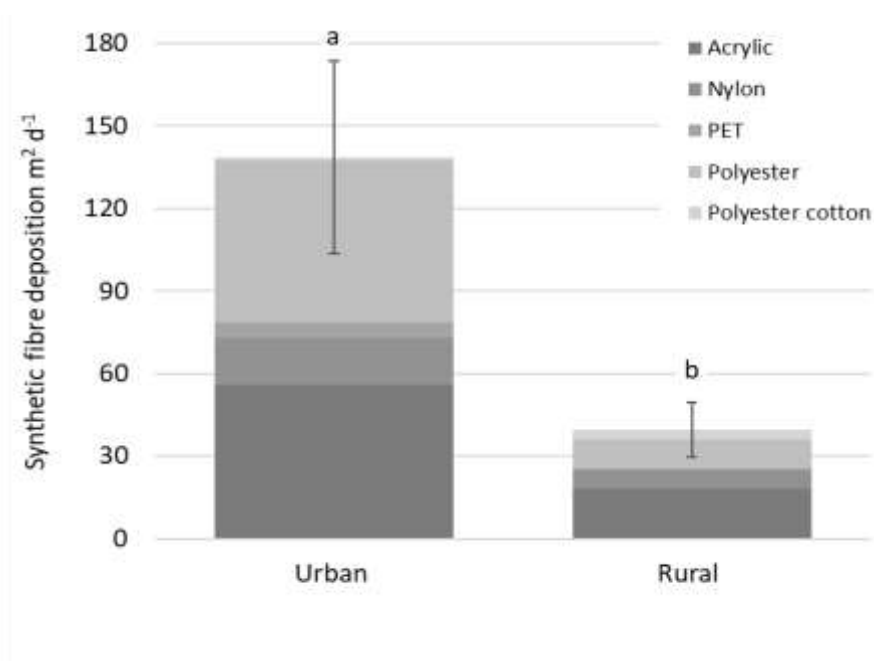
265

266 Synthetic microfibres were detected in 46% of atmospheric deposition samples, and  
267 38% of treated wastewater effluent samples, indicating both can serve as pathways  
268 into the marine environment. A total of 133 synthetic microfibre particles were  
269 identified by FTIR. Two additional fragments (polyethylene) were also found in an  
270 atmospheric deposition rural sample, but not included in analysis as the study's  
271 focus is on synthetic microfibres. Results presented are averaged across both  
272 locations.

273

274 Across both locations, and urban and rural environments, atmospheric deposition of  
275 synthetic microfibres was recorded at an average rate of  $81.6 \pm 10$  microfibres  $m^2 d^{-1}$   
276 ( $\bar{x} \pm SE$ ). Urban environments had an average deposition rate of  $123.2 \pm 30.8$   
277 microfibres  $m^2 d^{-1}$ ; the highest site with 403 microfibres  $m^2 d^{-1}$ . Rural environments  
278 had an average deposition rate of  $40.1 \pm 10$  microfibres  $m^2 d^{-1}$ . Urban samples  
279 contained a significantly higher number of microfibres than rural sites (planned  
280 contrasts,  $p < 0.05$  (Fig. 1A).

281



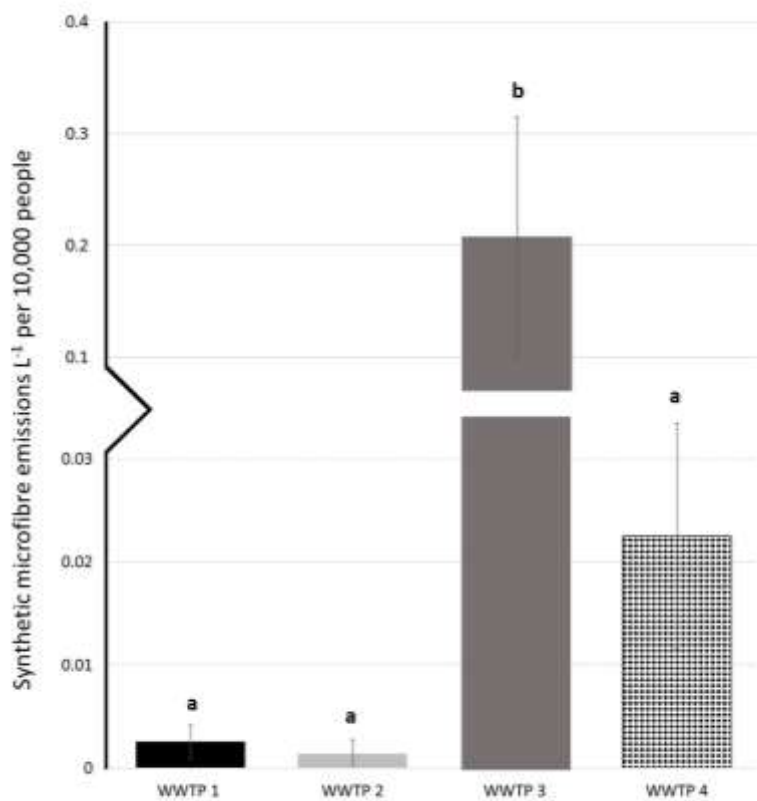
282

283 **Figure 1.** Deposition of synthetic microfibres in urban and rural environments ( $m^2 d^{-1}$ )  
284 by polymer type. Mean  $\pm$  standard error. Letters a and b denote categories that  
285 are statistically different.

286

287 Synthetic microfibres were discharged within treated wastewater effluent at an  
288 average abundance of  $0.03 \pm 0.01 L^{-1}$  ( $\bar{x} \pm SE$ ). No significant differences in absolute  
289 emissions of synthetic microfibres between treatment plants were observed  
290 (ANOVA,  $df = 3$ ,  $p > 0.05$ ). However, when microfibre emissions were normalised by

291 the population served (and scaled to 10,000 people) significant differences were  
292 apparent with plant 3 releasing significantly greater quantities than the other plants  
293 examined (ANOVA,  $df = 3$   $p = <0.01$ ) (Fig. 2). The same pattern was also observed  
294 when emissions were scaled to the catchment served (synthetic fibre emissions  $\text{km}^2$ ,  
295 ANOVA,  $df = 3$ ,  $p = <0.01$ ). Because Cochran's revealed heteroscedasticity (after  
296 applying a  $\log^{+1}$  transform), a conservative approach was taken and only  $p$ -values  
297  $>0.01$  were considered significant.



298  
299 **Figure 2.** The mean abundance of synthetic microfibrines recorded in final effluent  
300 from four wastewater treatment plants ( $\text{L}^{-1}$ ), normalised by serving population and  
301 scaled to 10,000 people. Within each plot letters a, and b denote categories that are  
302 statistically different. Error bars represent standard error.

303

304 The average fibre measured  $413 \pm 42 \mu\text{m}$  in length ( $n = 133$ ); on average  $424 \pm 48$   
305  $\mu\text{m}$  ( $n = 115$ ) within atmospheric deposition samples, and  $348 \pm 52$  ( $n = 18$ ) in  
306 treated wastewater effluent. Black and blue were the most dominant colour (72%).  
307 Other colours present (red, green, grey, yellow, brown, clear, pink, purple, and white)  
308 each accounted for less than 10% of total abundance.

309

310 The most dominant polymers were acrylic (45%), polyester (34%), and nylon (12%),  
311 while the remaining polymers including polyester-cotton blend, polyurethane,  
312 polypropylene, and polyethylene all contributed less than 5% to the total. Although  
313 regenerated cellulose microfibrils (e.g. rayon) were not included in the formal  
314 analysis, the majority of microfibrils extracted from the various sources were  
315 identified by FTIR as 'rayon'. This accounted for 90% of total atmospheric fallout and  
316 just less than half of those from treated wastewater effluent samples.

317

318 It is worth considering that microfibrils identified by the FTIR as 'rayon' were diverse  
319 in colour. Although they were dominated by black and blue microfibrils (78%) they  
320 also included yellow, pink, red, green and purple microfibrils. These colours are  
321 unlikely to be naturally occurring and could indicate release of fibres from dyed  
322 'semi-synthetic' fabrics.

323

324 In the regions examined, microfibre deposition from the atmosphere ( $81.6 \pm 10$  fibres  
325  $\text{m}^2 \text{d}^{-1}$ ) was observed as the dominant pathway in comparison with treated  
326 wastewater effluent ( $0.03 \pm 0.01$  fibres  $\text{L}^{-1}$ ) emitting fewer microfibrils than  
327 atmospheric transport by several orders of magnitude. For example, WWTP 1  
328 served a catchment area of  $22 \text{ km}^2$ , multiplied by the average daily deposition of

329 fibres (81.6 fibres m<sup>2</sup>) equating to 1795200000 fibres a day. Plant 1 releases  
330 approximately 17260000 L of treated effluent a day, multiplied by the average  
331 number of fibres released per litre (0.03 fibres/L) equates to 517800 fibres a day.  
332 Hence, considerably fewer fibres reached these estuaries via treated effluent than  
333 via atmospheric deposition. The same pattern was observed for the other three  
334 plants examined, see SI for more details.

335

#### 336 **4.0 Discussion**

337

338 While a small number of studies have reported atmospheric deposition of microfibres  
339 (Allen *et al.*, 2019; Dris *et al.*, 2016; Wright *et al.*, 2020), attributing fibre pollution in  
340 remote environments to atmospheric transportation (Allen *et al.*, 2019; Brahney *et*  
341 *al.*, 2020; Napper *et al.*, 2020), and recorded microplastics in discharge from waste  
342 water treatment effluent (Gies *et al.*, 2018; Murphy *et al.*, 2016; Talvitie *et al.*, 2017;  
343 Ziajahromi *et al.*, 2016) this study presents data on both, collected in tandem,  
344 enabling a novel comparison on the relative importance of these pathways.

345

346 With regards to atmospheric deposition, the deposition rate of microfibres recorded  
347 within this study sits within existing estimates (Dris *et al.*, 2016; Wright *et al.*, 2020;  
348 Truong *et al.*, 2021). Previous research in urban environments by Dris *et al.* (2015)  
349 reported 29 – 280 MP m<sup>-2</sup> d<sup>-1</sup> from atmospheric fallout in Paris (France) where more  
350 than 90% of the microplastics observed were microfibres. A similar study by Wright  
351 *et al.* (2020) showed deposition rates in London (UK) ranged from 575 to 1,008  
352 microplastics m<sup>-2</sup> d<sup>-1</sup>, with fibrous microplastics accounting for the majority (92%). In  
353 Vietnam, Truong *et al.* (2021) reported an atmospheric fallout rate (dominated by



354 microfibrés) in the range of 71 – 917 m<sup>2</sup> d<sup>-1</sup>. In the present study urban environments  
355 recorded an average of 81.6 plastic microfibrés m<sup>2</sup> d<sup>-1</sup>, the highest deposition rate  
356 being 430 plastic microfibrés m<sup>2</sup> d<sup>-1</sup> at an urban site. For rural environments, Allen *et*  
357 *al.* (2019) sampled atmospheric microplastic deposition in the remote French  
358 Pyrenees mountains and found 365 microplastics m<sup>2</sup> d<sup>-1</sup>; whereas this study found  
359 an average of 42 synthetic microfibrés m<sup>2</sup> d<sup>-1</sup> in rural environments.

360

361 These variations between studies are most likely due to differences in the  
362 environments examined, the field and laboratory methods employed, and the  
363 inclusion, or exclusion of rayon fibres. For example, atmospheric deposition of  
364 microfibrés has previously been examined at height (on a rooftop) with the use of a  
365 funnel or rain gauge (Dris *et al.*, 2016; Stanton *et al.*, 2019; Truong *et al.*, 2021;  
366 Wright *et al.*, 2020). Whereas this study quantified atmospheric fallout at ground  
367 level, to best mimic deposition to surface water in the marine environment.

368 Additionally, our overall estimations of the prevalence of synthetic microfibrés in the  
369 environment may appear lower than other studies which choose not to eliminate  
370 microfibrés identified as regenerated cellulose microfibrés (e.g. rayon). Rayon is  
371 often reported as a common polymer type for microplastics in both freshwater and  
372 marine samples (Lindeque *et al.*, 2020; Nan *et al.*, 2020; Park *et al.*, 2020) and is  
373 mainly used in clothing or personal care products (Comnea-Stancu *et al.*, 2017; Frias  
374 *et al.*, 2016). Lastly, sampling was conducted largely during colder months where  
375 footfall and therefore release of fibres may vary from warmer summer months.

376

377 A key pattern in our study was densely populated urban environments having a  
378 significantly greater rate of atmospheric deposition than rural environments.

379 Population density can be considered as an indicator of human activity in an area  
380 (i.e. where the majority of microfibres originate from textiles) (Wright *et al.*, 2020) and  
381 has previously been reported in other studies to correlate with deposition of  
382 microplastics from the atmosphere (Dris *et al.*, 2016; Stanton *et al.*, 2019; Truong *et*  
383 *al.*, 2021). Although, as highlighted by Wright *et al.* (2020) the opposite has also  
384 been observed, with higher microplastics deposition rates being recorded in areas  
385 with lower population densities (London) (Wright *et al.*, 2020) than higher population  
386 density areas (Paris) (Dris *et al.*, 2016). This is likely attributable to increasing footfall  
387 associated with commuters and tourists etc., increasing activity and indicating other  
388 influencing factors (Wright *et al.*, 2020).

389

390 The presence of synthetic microfibres, albeit in lower concentrations, in sparsely  
391 populated areas (rural) indicates that these particles have the potential to be aerially  
392 transported, as was also evidenced by Allen *et al.* (2019) and Bergmann *et al.*  
393 (2019). These studies predicted that microfibres have the ability to travel tens of  
394 kilometres before settling, and fibre sizes for this study fell within the size range for  
395 regional transport (Brahney *et al.*, 2020; Roblin *et al.*, 2020) subsequently, increasing  
396 their likelihood of entering the marine environment and polluting environments even  
397 when emitted in locations far removed from their final point of deposition. It is worth  
398 noting that atmospheric transport also includes microplastic particles serially  
399 resuspended from the ground at limited height before being re-deposited; fibres  
400 recorded in this study will likely feature a mixture of both.

401

402 Due to the nature of the sampling set up, atmospheric sampling was conducted  
403 during periods of dry weather. Stanton *et al.* (2019) reported fibre deposition during

404 dry and wet conditions, concluding influences other than rainfall have a part in  
405 atmospheric fallout of fibres. Likewise, Dris *et al.* (2016) reported no significant  
406 correlation between atmospheric fallout of microplastics and daily rainfall, but  
407 observed a greater fallout during periods of wet weather than dry or low rainfall  
408 periods, noting rainfall to be contributing temporal factor. Similarly, Germanov *et al.*  
409 (2019) reported that plastic abundance was up to ~ 44 times higher in the wet than  
410 the dry monsoon seasons in three coastal locations. Therefore, it is possible that  
411 deposition rates may be slightly increased upon those reported here during  
412 precipitation events.

413

414 For WWTP effluent, the discharge of synthetic microfibres ( $0.03 \text{ L}^{-1}$ ) appeared  
415 consistent with other studies, such as two WWTPs in the U.S.A. at  $0.02 \text{ L}^{-1}$  and  $0.05$   
416  $\text{L}^{-1}$  (Dyachenko *et al.*, 2017; Mason *et al.*, 2016). However, the concentrations are  
417 still relatively low compared to the majority of studies including  $0.25 \text{ L}^{-1}$  in the U.K.  
418 (Murphy *et al.*, 2016),  $70 \text{ L}^{-1}$  in Russia and  $90 \text{ L}^{-1}$  in the U.S.A (Carr *et al.*, 2016).  
419 This concurs with previous studies which report high microplastic retention in WWTP  
420 ranging from 66 to 99%, where sludge is expected to be the final fate of retained  
421 MPs (Carr *et al.*, 2016; Gies *et al.*, 2018; Habib *et al.*, 1998; Magnusson and Norén,  
422 2014; Mintenig *et al.*, 2017; Talvitie *et al.*, 2017; Ziajahromi *et al.*, 2016). The  
423 resultant sludge is often however returned to the land as a fertilizer which could be  
424 mobilised during rainfall events, providing a further pathway for microfibres to be  
425 released to aquatic environments (Corradini *et al.*, 2019; Gies *et al.*, 2018;  
426 Kirchmann *et al.*, 2017).

427

428 The variations in emissions and retention efficiencies for WWTP between these  
429 studies likely occurred due to differences in the treatment employed, loads entering  
430 the plant, treatment capacities, as well as variations in the methodological sampling  
431 approach (e.g. limit of detection). Despite the observed low concentrations of fibre  
432 emissions from treated wastewater effluent, the large volumes of effluent exiting  
433 each plant daily still equate to a substantial quantity of synthetic microfibres entering  
434 aquatic environments. For example, it has been estimated that a secondary  
435 wastewater treatment plant that serves a 650,000 population (Glasgow, UK) with a  
436 removal efficiency of 98% could release 65 million microplastic particles every day  
437 (Murphy *et al.*, 2016). A wastewater plant with a lower retention efficiency (84%) and  
438 a greater population equivalent (1,200,000) could discharge up to 160 million  
439 particles per day in its treated effluent (Magni *et al.*, 2019). Based on results from the  
440 4 WWTPs sampled in this study, we predict between ~20,000 and 500,000  
441 microfibres per plant could be discharged to receiving waters daily.

442

443 For both atmospheric deposition and treated wastewater effluent samples, acrylic  
444 was the most common synthetic polymer recovered in this study, followed by  
445 polyester, a pattern which does not reflect that of the textile market (polyester two-  
446 fold that of acrylic). The densities of acrylic and polyester ( $1.185 \text{ g cm}^{-3}$  and  $1.23 -$   
447  $1.38 \text{ g cm}^{-3}$  respectively), which affects their transport potential particularly when in  
448 water, are similar do not appear to explain this disparity. However, previous work by  
449 Napper and Thompson (2016) has suggested that acrylic sheds more microfibres  
450 than polyester during laundering. It can be assumed that similar factors such as fibre  
451 type, textile construction methods, and garment design that effect shedding rates  
452 during laundering (De Falco *et al.*, 2018) also impact shedding during normal use.

453

454 We also report that the most common microfibre colours for both atmospheric  
455 deposition and treated wastewater effluent was blue and black, which is consistent  
456 with findings from other studies and may in part be attributed to their greater visibility  
457 during enumeration. For atmospheric deposition studies, Welsh *et al.* (2022)  
458 reported that blue and red made up 84% of all microfibres, making them the most  
459 prevalent colours and Stanton *et al.* (2019) found that black and grey were the most  
460 common colours (47%) followed by blue (24%). For wastewater effluent, research by  
461 Ben-David *et al.* (2021) found that (irrespective of treatment stage or mesh size) the  
462 retrieved microfibres were predominantly black (50–85%), with blue being the  
463 second most common (10–20%). Blue is reported to be the population's favourite  
464 colour in the United Kingdom, which may be reflected in a greater quantity of people  
465 who choose to wear blue apparel (Jordan, 2015). However, as opposed to other less  
466 vivid colours, eye-catching hues like red and blue may be more readily identified  
467 during visual identification and potentially overreported (Hidalgo-Ruz *et al.*, 2012;  
468 Dris *et al.*, 2015; Zhang *et al.*, 2020). Additionally, as they are more difficult to spot,  
469 translucent or uncoloured microplastics may also be overlooked (Dris *et al.*, 2015).

470

471 The relative importance of the two pathways examined concurs with findings from  
472 the laboratory-based study by De Falco *et al.* (2020) indicating deposition of  
473 synthetic microfibres to the atmosphere during normal use to be far greater than  
474 emissions of fibres originating from wastewater systems as a consequence of  
475 laundering. While discussing the deposition of synthetic microfibres to the  
476 atmosphere, it is worth considering the atmosphere as a potential exposure pathway  
477 to humans and other air breathing animals. Gasperi *et al.* (2018) reports the

478 likelihood of exposure to humans as a function of size, where microfibrils in the  
479 inhalation fraction can be deposited in the upper airways via the mouth or nose, and  
480 microfibrils in the respirable fraction have the potential to reach the lungs. As also  
481 noted by Wright *et al.* (2020), microfibrils present in atmospheric samples are likely  
482 to be deposited in the upper airway, however it is possible that the detection limits of  
483 the analytical process (~ 20 µm) employed in this study limited the identification of  
484 any microfibrils that might be present in the respirable fraction. Consequently, future  
485 work should look to quantifying human exposure to microfibrils via the atmosphere  
486 (Gasperi *et al.*, 2018; Wright *et al.*, 2020).

487

488 Strategies promoted to reduce emissions of synthetic fibre include opting for natural  
489 textiles, improvements to wastewater treatment efficiency, fitting fibre-capturing  
490 devices to washing machines, and modifications to manufacture and design.  
491 Replacing synthetic textiles with natural counterparts would typically be more  
492 expensive and the impact of non-synthetic microfibrils, which may be synthetically  
493 altered to contain dyes and additives (i.e. flame retardants) (Athey *et al.*, 2020),  
494 accumulating in the environment is currently unknown (Dris *et al.*, 2017; Napper and  
495 Thompson, 2020). Furthermore, fabrics made from synthetic and natural microfibrils  
496 often have considerable carbon footprints; the estimated carbon footprint for all  
497 polyester and cotton clothing use in the UK during 2009 was 4,750,000 and  
498 15,907,500 (tCO<sub>2</sub>e), respectively. This included whether manufactured in or imported  
499 to the U.K. (Thomas *et al.*, 2012). Hence there would be environmental  
500 consequences of a switch to cotton compared to polyester.

501

502 Microplastic removal via existing wastewater treatment systems is largely efficient  
503 (>90%) (Carr *et al.*, 2016; Murphy *et al.*, 2016). Upgrading wastewater treatment  
504 plants with more effective filtering systems or retro fitting existing systems could be  
505 hugely expensive (Conley *et al.*, 2019). Furthermore, globally the majority of the  
506 human population are not connected to wastewater treatment systems (ranging  
507 anywhere between 0 – 100% by country; United Nations Environment Statistics,  
508 2011). Additionally, devices fitted to washing machines have been evidenced to  
509 reduce fibre emissions in washing effluent by up to 78% (Napper *et al.*, 2020).  
510 However, these are not yet widely utilised or retrofitted to domestic washing  
511 machines by manufacturers and would not address emissions of microfibres to the  
512 atmosphere as a consequence of wear and tear during normal use.

513

514 Factors such as polymer type, fabric structure, type of yarn and twist have been  
515 shown to be influential in fibre shedding in laundering and to the atmosphere  
516 (Napper and Thompson *et al.*, 2016; De Falco *et al.*, 2020). For example, Napper  
517 and Thompson *et al.* (2016) found acrylic to emit far more microfibres than polyester  
518 or polyester-cotton blend during laundering. Likewise, De Falco *et al.* (2020) reported  
519 compactly woven and highly twisted yarns with continuous filaments release far  
520 fewer microfibres during both laundering and normal use than fabrics with loose  
521 structures. In order to inform policy and industry, focus should be placed on better  
522 understanding what permutations of textile design give rise to the lowest rate of fibre  
523 shedding. Changes in fabric design will likely help reduce shedding during all use  
524 phases: wearing, washing and tumble drying (De Falco *et al.*, 2020; Napper and  
525 Thompson, 2016; Pirc *et al.*, 2016).

526

## 527 **5.0 Conclusion**

528

529 This study provides evidence on synthetic microfibre release to the environment via  
530 atmospheric deposition and treated wastewater effluent, with a strong indication that  
531 atmospheric deposition presents the dominant pathway in coastal river catchments.  
532 This illustrates efforts to mitigate emissions during laundering do nothing to combat  
533 release via the principal route to the environment. In order to inform interventions  
534 such as those required to deliver UNEA 5.2, the focus of future research should shift  
535 on how to mitigate this pollution at source, and track the efficacy of mitigation  
536 methods by improving our understanding of what permutations of textile and yarn  
537 design influence fibre release during laundering and normal use, thereby tackling  
538 fibre pollution via both pathways. It is also recommended that future research takes a  
539 mass balance approach to investigate sources and pathways of synthetic fibres to  
540 the environment.

541

542

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544

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553

554

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