2023-01

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Napper, IE

10.1016/j.scitotenv.2022.159317
Science of The Total Environment
Elsevier BV

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

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Abstract

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

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

Key words

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

Introduction

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

Microplastic waste can originate from a variety of different land-based sources such as leakage from wastewater treatment plants (WWTPs) (Kay *et al*., 2018; Murphy *et al*., 2016) and atmospheric pollution (De Falco *et al*., 2020; Dris *et al*., 2015; Wright *et al*., 2020). Microplastics can then be transported to the marine environment by a variety of different mechanisms. Freshwater systems often connect inland and coastal communities to the ocean, (Miller *et al*., 2017; Napper *et al*., 2021b; Rech *et al*., 2014; Schmidt *et al*., 2017; Seo and Park, 2020; Weideman *et al*., 2020). Subsequently, microplastic pollution has been found to be highly abundant in coastal habitats, especially estuaries which provide unique and complex environments located between oceans and river mouths (Harris, 2020; Hitchcock and Mitrovic, 2019). There is also growing recognition that atmospheric deposition is an important vector for the transportation of microplastics (Dris *et al*., 2017; Napper *et al*., 2020; Stanton *et al*., 2019; Wright *et al*., 2020), even into remote regions far from emission sources (Allen *et al*., 2019; Brahney *et al*., 2020; Roblin *et al*., 2020).

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

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

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

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

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

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

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

2.0 Method

2.1 Environmental sampling

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

2.1.1 Atmospheric Deposition from Urban and Rural Environments

Atmospheric deposition samples were collected at ground level within urban and rural environments, characterised by densely and sparsely populated areas and by land use classification. From each city (Plymouth and Bristol), two sites of each 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^2) 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 microfibres deposited per square meter per day. Each dish contained deionised
159 water (~1 L) which captured any atmospheric fallout on the meniscus. The deionized
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 overfilling 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^2 catchment

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

2.2 Laboratory Analysis

All samples were vacuum filtered onto Whatman cellulose nitrate membrane filter papers ≤ 12 µm. Due to the large volumes of liquid and high content of suspended solids, treated wastewater samples were first passed through 30 µm (stainless steel) and 12 µm (clear nylon) meshes. The contents of each mesh was rinsed into a beaker with deionised water before vacuum filtering.

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

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

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

225 microfibres 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 ($\bar{x}$ ± SE; 0.22 ± 0.15 microfibres 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 µm
238 Milli-Q water before use. Minimal contamination was reported from the laboratory
239 blanks (3 plastic microfibres found in total; ~0.14 ± 0.10 microfibres 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

2.5 Comparisons between pathways

The units required to quantify synthetic microfibres within treated wastewater effluent (microfibres L^{-1}) and within atmospheric fallout (microfibres $m^2\ d^{-1}$) are by necessity different, making a quantitative comparison between the two pathways challenging. To compare the release of fibres between the two pathways, emissions of fibres within treated wastewater effluent were scaled from microplastics per litre to microplastics a day based upon the volume of treated effluent released per day (L) from each plant, averaged over each weather season. For atmospheric data, the average number of synthetic fibres deposited over a m^2 per day across all urban and rural sites was scaled to the catchment size each WWTP served. This enabled conclusions to be drawn about the relative importance of the various pathways examined.

3.0 Results

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

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

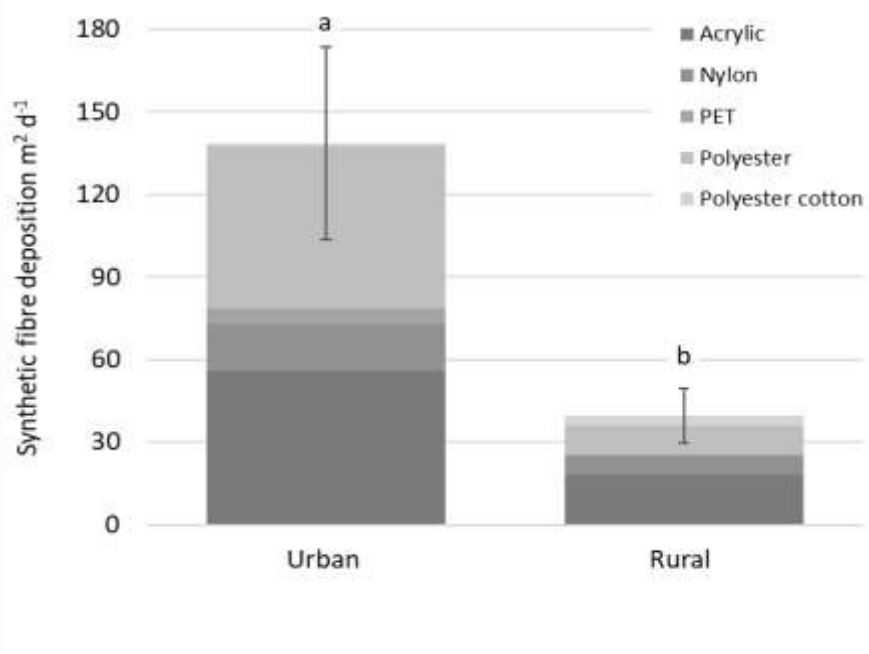

Figure 1. Deposition of synthetic microfibres in urban and rural environments (m^2 d^{-1}) by polymer type. Mean ± standard error. Letters a and b denote categories that are statistically different.

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

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

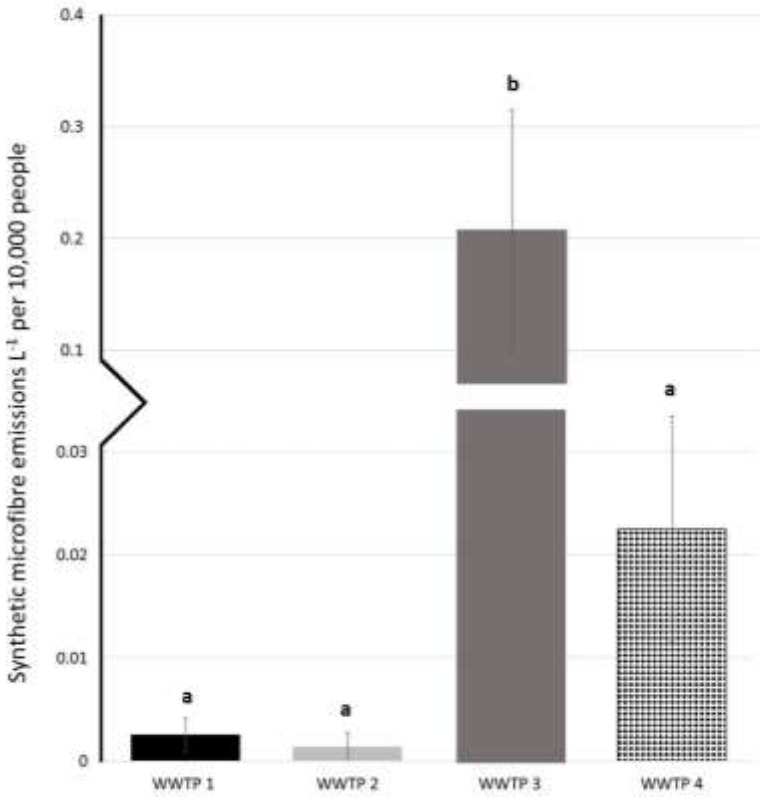

Figure 2. The mean abundance of synthetic microfibres recorded in final effluent from four wastewater treatment plants (L^{-1}), normalised by serving population and scaled to 10,000 people. Within each plot letters a, and b denote categories that are statistically different. Error bars represent standard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

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

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

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

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

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

4.0 Discussion

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

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

354 microfibres) in the range of 71 – 917 m^2 d^{-1}. In the present study urban environments
355 recorded an average of 81.6 plastic microfibres m^2 d^{-1}, the highest deposition rate
356 being 430 plastic microfibres m^2 d^{-1} 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^2 d^{-1}; whereas this study found
359 an average of 42 synthetic microfibres m^2 d^{-1} 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 microfibres 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 microfibres in the
369 environment may appear lower than other studies which choose not to eliminate
370 microfibres identified as regenerated cellulose microfibres (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.

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

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

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

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

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

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

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

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

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

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

Strategies promoted to reduce emissions of synthetic fibre include opting for natural textiles, improvements to wastewater treatment efficiency, fitting fibre-capturing devices to washing machines, and modifications to manufacture and design. Replacing synthetic textiles with natural counterparts would typically be more expensive and the impact of non-synthetic microfibres, which may be synthetically altered to contain dyes and additives (i.e. flame retardants) (Athey *et al*., 2020), accumulating in the environment is currently unknown (Dris *et al*., 2017; Napper and Thompson, 2020). Furthermore, fabrics made from synthetic and natural microfibres often have considerable carbon footprints; the estimated carbon footprint for all polyester and cotton clothing use in the UK during 2009 was 4,750,000 and 15,907,500 (tCO_2e), respectively. This included whether manufactured in or imported to the U.K. (Thomas *et al*., 2012). Hence there would be environmental consequences of a switch to cotton compared to polyester.

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

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

5.0 Conclusion

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

Acknowledgments

This paper has been prepared specifically for *Science of the Total Environment* and is part of a larger study commissioned and funded by the UK Government, Department for Environment, Food and Rural Affairs (ME5435). We thank the landowners and water service companies who allowed access to sites for sampling. We also want to thank the marine biology and ecology technical staff at the University of Plymouth, and the University of Plymouth Electron Microscopy Centre for their support and assistance in this work. RCT was also supported by NERC

grant NE/S003967/1.

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