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Microfibre release to water, via laundering, and to air, via everyday use: a comparison between polyester clothing with differing textile parameters

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ABSTRACT. Textiles are one of the major sources of microplastic pollution to aquatic environments and have also been reported in dry and wet atmospheric deposition. There is still a lack of information on the direct release of microfibres from garments to the air and on the influence of textile characteristics including structure, type of yarn and twist. The present study examined microfibre emissions directly to the air and to water as a consequence of laundering. Polyester garments with different textile characteristics were examined including: various material compositions, fabric structure, yarn twist, fibre type, and hairiness. Scaling up our data indicate release of microfibres per person per year to the air is of a similar order of magnitude to that released to wastewater by laundering. The lowest releases to both air and water were recorded for a garment with a very compact woven structure and highly twisted yarns made of continuous filaments, compared with those with a looser structure (knitted, short staple fibres, lower twist). Our results demonstrate for the first time that direct release of microfibres from garments to air as a consequence of wear is of equal importance to releases to water. Currently there is considerable interest in interventions focused on capture from wastewater. However, our results suggest more effective interventions are likely result from changes in textile design that could reduce emissions to both air and water.

1. Introduction

Microplastic pollution has become a high profile topic. The term first came into popular use following research in the marine environment[1] but it has become clear that microplastics are also present in freshwater[2] and terrestrial[3] systems, as well as urban dust[4]. Microplastics are defined as plastic particles smaller than 5 mm, and are further divided, on the basis of their origin, as “primary” microplastics if they are intentionally produced either for direct use or as precursors to other products, and “secondary” microplastics if they are formed in the environment from breakdown of larger plastic materials.[5,6]

According to desk based evaluation conducted by the International Union for conservative of Nature (IUCN), washing of synthetic textiles could contribute 35% of the release of primary microplastics to the oceans.[7] However, this estimation is based on releases of primary microplastics via laundering alone and the release to the atmosphere as a consequence of wearing clothes is not included. Hence, the overall importance of textiles as a source of microplastics could be underestimated. Irrespective of the pathway via air or water it is important to note that, during laundering and wearing, not only microfibres of synthetic origin (microplastic of fibrous shape < 5 mm) could be released, but also microfibres of artificial or natural origin. Natural fibres can be modified during manufacturing processes, for example by reconstitution into semi-synthetic cellulosic materials as well as by bleaching, dyeing, finishing. The persistence and potential impacts of these varying forms of fibres in the environment is not fully understood.

There is clear evidence of widespread microfibre accumulation in aquatic environments, including shorelines[8], rivers[9], oceans[10] as well as the Arctic[11,12] and deep sea[13,14]. Microfibre ingestion has also been reported for a variety of aquatic organisms, including oysters[15], fish[16,17], and turtles[18]. More recently, microfibres have been found in terrestrial environments. For example, microfibres accounted for more than the 50% of the total microplastics detected in farmland around Shangai,[19] for 92% of the microplastics in samples from the Chai River valley, China,[20] and represented the largest proportion of microplastics also in soils from rice-fish experimental stations in Shanghai.[21] Recent studies have also indicated the presence of microfibre contamination in the air[22,23] and it has been estimated that between 3 and 10 tons of microfibres are deposited by atmospheric fallout every year in Paris alone, with 29% constituted by fibres of petrochemical nature[24]. The presence of microfibres in the air has also been investigated indoors[25,26], in an intercity terminal and a university campus in Turkey[27]. The results of these works confirmed the presence of synthetic microfibres in the indoor and outdoor environments, but the prevalent microfibres were artificial and natural ones. Concentrations of microfibres in the range 1.0-60.0 microfibres/m^3 and deposition rates between 1586 and 11,130 microfibres/day/m^2, were documented indoor.[25] The quantity of microfibres in the atmosphere shows considerable temporal and spatial variability.[27] Human exposure to microplastics via ingestion of contaminated seafood and exposure via household microfibres fallout during a meal, has also been examined, showing that microplastic ingestion by humans via consumption of mussels ranged between 123 particles/year/capita in the UK to 4620 particles/years/capita in countries with a higher shellfish consumption, whereas microfibre exposure during a meal via dust fallout in a household was found to be much greater, with 13,731-68,415 particles/years/capita.[28] Exposure to microfibres could lead to inhalation, as already observed for natural and synthetic fibres found in human lungs by Pauly et al.[29] Synthetic fragments and fibres, as well as non-synthetic particles of protein and cellulose origin, were founded inhaled by a Breathing Thermal Manikin, in a set of simulation experiments in indoor environments.[30]

Several studies have investigated the release of microfibres to wastewater from the washing of synthetic clothes, using different washing procedures, and estimated that thousands of microfibres could be released by a single household wash of 5-6 kg.[31,32,33] It has been suggested that textile parameters such as material composition of the fabric[31], type of yarn[32] and textile construction[34,35] could all influence release. However, very little information is available on these parameters and more research is needed to provide information to guide interventions to reduce microfibre emission. The present work aims to assess the influence of several parameters on release. Furthermore, we will quantify release of microfibres during everyday use and wear of clothing. The three main objectives were: (1) determine the amount of microfibres released from garments during washing, and (2) compare this with the amount shed directly to the air due to their wearing, (3) examine the extent to which microfibre release both to air and water is influenced by textile characteristics. Four polyester garments were selected with different textile characteristics, including material composition (neat or blend), fabric structure (woven or knitted), yarn twist (high or low), fibre type (staple or filament), and hairiness (high or low). Microfibre release to water and air was assessed by using a household washing machine and tests to quantify the release of fibres from wearing clothes.

2. Materials & Methods

2.1. Materials

Four different polyester commercial garments were used. These included a 100% green polyester blouse, a 100% blue polyester t-shirt, a 100% black polyester dress, and a 50%:50% pink polyester/cotton sweatshirt. Four replicates of each garment were used for investigating the release of microfibres during washing and four separate replicates were used for investigating release of microfibres during wear. All garments were purchased in the same size (Large).

Before testing, the garments were pre-washed to eliminate loose fibres, impurities and the presence of other type of fibres. All replicates of the same garment type were washed in a Whirlpool WWDC6400 washing machine (40 °C, 1200 rpm, 1h), using a commercial liquid laundry detergent in the dose recommended by the manufacturer, whose composition is reported in the Supporting Information (SI). Cross-contamination of microfibres between washes was prevented as described in the QA/QC section of the SI. Cotton lab coats and nitrile gloves were worn during all the experimental work.

2.2. Textile characteristics of tested garments

The polyester garments were selected considering five textile parameters: (1) material composition, (2) textile structure, (3) yarn twist, (4) fibre length and (5) hairiness. The work focused on polyester garments but, since polyester is widely used in blends with cotton, the effect of neat versus blended polyester fabrics on microfibres release was also studied. The composition of the fabrics reported on the label was confirmed by Fourier transform infrared spectroscopy (FTIR). Spectra were acquired by means of a Hyperion 1000 microscope (Bruker) coupled to an IFS 66 spectrometer (Bruker), using 16 scans and a resolution of 4 cm^{-1}, over the range 4000–400 cm^{-1}.The obtained spectra were compared to a spectral database of synthetic polymers (Bruker I26933 Synthetic fibres ATR library).

Usually, textiles have two main structures: woven and knitted. The woven structure is made of two sets of yarns interlaced, the warp runs in a lengthways direction and the weft runs in a widthway direction; the knitted is obtained by interlacing loops of yarn.[36] The yarn can be constituted by staple fibres, of comparatively short length, and filaments, which are fibres of indefinite length.[37] The yarn twist provides structural integrity to the yarn and is defined as the number of turns present in a unit length of yarn.[38] The fabric structure and yarns of the selected garments was analysed using a Leica M205 FA light microscope (Leica Microsystem) and a field-emission scanning electron microscope (SEM) Quanta 200 FEG (FEI) operating in high vacuum mode, using an accelerating voltage ranging between 15 and 20 kV and a secondary electron detector (Everhart-Thornley detector). Before SEM analysis the samples were sputter-coated with gold–palladium. SEM micrographs were also used to measure the twisting. The yarn twist (turn per meter, t/m), was measured by using equation 1:

$$T = \frac{tan\theta}{\pi d} \qquad \text{Equation 1}$$

where θ is the angle formed by the fibre in the yarn with the yarn axis, d is the diameter in meters.[39] The level of twist was classified in this way: no twist means that no torsion is present in the yarn; low twist indicates values up to 500 t/m; moderate twist refers to a twist value up to 1000 t/m; high twist was used to comment value above 1000 t/m.

Lastly, the hairiness is defined as the presence of small fibres that protrude from the main yarn core[40] and basically could be high or low depending whether the yarn is made of staple fibres or continuous filaments, respectively. Hairiness was evaluated through the observation of the yarns under the light microscope.[41]Its value was expressed as number of protruding fibre ends per meter of yarn (n/m). The level of hairiness was classified as low for values up to 500 n/m, and high for values above 1000 n/m. The values obtained were in line with the type of fibres composing the yarns of the fabrics. In fact, yarns made of short staple fibres presented greater hairiness than those made of continuous filaments.

Figure S1 in the Supporting Information (SI) reports the optical and scanning electron micrographs of the surfaces of each selected garment and the yarns constituting them. The textile parameters analysed for each garment as reported in Table 1, and were used to codify the samples.

Table 1. Code and textile parameters of the selected garments.

Type of garment	Structure	Yarn	Twist	Hairiness	CODE
100% polyester blouse	woven	continuous filaments	warp 2458 t/m weft 875 t/m	warp 96 n/m weft 173 n/m	PES-Woven-Filament
100% polyester t-shirt	knitted	continuous filaments	no twist	59 n/m	PES-Knit-Filament

100% polyester dress	knitted	short staple fibres	631 t/m	3754 n/m	PES-Knit-Staple
50 %:50% polyester/cotton sweatshirt	knitted	short staple fibres	470 t/m	3426 n/m	PES/COT-Knit-Staple

Due to its mixed composition of cotton and polyester, PET/COT-Knit-Staple was further analysed by SEM to understand how cotton and polyester fibres were combined together in the yarns. In Figure S2, the obtained SEM micrographs are reported and it is clearly detectable the presence in the yarn of cotton fibres, with the typical twisted ribbon form, and of polyester fibres with a cylindrical and smooth surface.[42] The two types of fibres were mixed together in each single yarn.

2.3. Release of microfibres from synthetic clothes during washing

Release of microfibres from the selected garments due to laundering was evaluated. Washing tests were performed using a Bosch washing machine serie 4 varioperfect WLG24225it with the following program for synthetics at 40°C, 1 h 47 min and 1200 rpm. The choice of the washing cycle is described in the SI. The commercial liquid detergent, whose composition was detailed in the SI, was used in the dose recommended by the supplier. Each garment was washed alone, with four replicates for each garment type. A total of 16 washing trials were performed.

The analytical procedure[35] adopted to determine the amount of microfibres consisted in a multistep filtration procedure as detailed in the SI, using filters with 400, 60, 20 and 5 µm pore size. The weight of microfibre recovered on the different pore size filters, was normalized for the washing load, obtaining W in mg/kg. The mean value of the total mass of microfibres per kilogram of washed fabric for each type of garment, W_a, and the standard deviation (SD), were calculated. QA/QC measures applied to prevent cross-contamination of microfibres between washes and among the different filtrations are described in the SI. Since PES/COT-Knit-Staple was made of a blend of cotton and polyester, further analyses were performed on the released microfibres in order to assess if they were of synthetic or cellulosic nature. Thermogravimetric analysis was performed on approximately 5 mg of microfibres recovered form 400 µm and 60 µm pore size filters as well as on a neat sample, about 5 mg, cut from a new sweatshirt (not pre-washed). Samples were placed in an open platinum pan and heated from 30 to 800 °C at the rate of 10 °C min^{-1} under nitrogen atmosphere (flow rate: 40 mL min^{-1}) in a Pyris 1 TGA (Perkin–Elmer).

The dimensions of the collected microfibres were determined using the procedure reported in the SI. For each test, the number of microfibres released to wastewater by each garment, N, was estimated according to Equation 2:

$$N_w = \frac{\frac{W}{\rho}}{\pi \cdot \frac{D^2}{4} \cdot L} \qquad \text{Equation 2}$$

where W is the amount of microfibres in grams released by the washed garment, ρ is the density of the material, L and D are average length and diameter, respectively, of the released fibres.[31] For the PES/COT-Knit-Staple garment, the formula took into account the data obtained from TGA analysis. The numbers of polyester and cotton microfibres released were first calculated separately, considering in the formula the masses (provided by the TGA analysis) and the mean dimensions (obtained from optical microscopy and SEM analysis) of the microfibres released by the two different materials. The two values were then summed to obtain the overall number of microfibres released by PES/COT-Knit-Staple. Each value of N was normalized for the weight of the corresponding washed garment, obtaining the number of microfibres/gram, in order to compare more easily the number of microfibres released to water with those released to air. Finally, the mean value of the total number of microfibres per gram of washed fabric for each type of garment, and the SD, were calculated.

2.4. Release of microfibres from synthetic clothes to air

An experimental procedure was developed to quantify the number of microfibres released to the air from synthetic clothes. Tests with volunteers wearing the selected garments, were carried out in a closed room of 4 m^2 floor area. The room had room had a desk with a height of 85 cm, no windows, no other sources of air flow. The floor was covered using cardboard and paper tape. Before testing, the room was deep cleaned using liquid soap, water and a handheld vacuum cleaner, white cotton cloths were used during the cleaning of the room. All the operators involved in the cleaning and tests, wore polypropylene boilersuits and shoe covers so that microfibre contamination could be eliminated from the results. To assess the cleanliness of the room prior to the trials, 8 polystyrene Petri dishes (9 cm of diameter) each lined with dampened filters papers (Whatman n. 1), were left in the room for 10 days, following a similar procedure reported elsewhere[43]. The Petri were then observed under the Leica M205 FA light microscope. The observation revealed the presence of only one microfibre in one of the Petri, so the room was considered cleaned. Based on the approach used to collect dust,[44] and taking into account recent works on airborne contamination[25,28,43], Petri dishes were used to capture the microfibres released during the tests. Dampened filter papers were used in preference to adhesive tape, so as not affect the FTIR analysis.[28,43] Four adult volunteers were selected and performed the test procedure (approved by the Ethics Committee of the University of Plymouth). Each person separately tested each of the 4 garment types, resulting in a total of 16 tests. Other than the garment under test, the volunteers wore white leggings made of 100% cotton. Each volunteer, positioned at the centre of the room, performed a specific sequence of movements that was selected to simulate a mix of real life activities. The duration of the sequence was set to 20 min as a compromise between a reasonable time to allow microfibres to deposit and acceptable time for the volunteers to remain in the closed room. A detailed description of the sequence of movements is reported in the SI. 8 Petri dishes lined with filter papers were placed at a distance of about 60 cm from the volunteer. To avoid cross-contamination between two consecutive tests, the room was cleaned by an operator wearing 100% cotton clothes under a boilersuit, and by using a handheld vacuum cleaner. QA/QC measures are reported in the SI.

The microfibres released during the tests and collected on the filter papers, were observed through optical microscopy to allow a quick evaluation of the microfibres present, as already reported in other works.[25,28,43]For their identification different criteria were taken into account, such as the colour and shape of the original fibres from the garments, and are reported in detail in the SI.[25,42] In details, the surface of the filter papers was observed and counted by using a Leica M205 FA light microscope (Leica Microsystem) and analysed by Image J to measure their dimensions.

For each test, the number of microfibres per unit area, N_t, was calculated by using Equation 3:

$$N_t = \sum_{i=1}^{8} n_i / 8a \qquad \text{Equation 3}$$

with n_i the number of microfibres counted in each filter paper, a is the area of the Petri dish. The total number of microfibres released in the room per gram of worn fabric, was determined by using Equation 4:

$$N_a = \frac{N_t}{W_t} \cdot A \qquad \text{Equation 4}$$

where A is the area of the room (4 m^2) and W_t the weight in grams of the garment worn in the test. Then, the mean value of the total number of microfibres per gram of worn fabric for each type of garment, and the SD, was calculated. A mean size was calculated for length and diameter of the released microfibres, based on the measurements of 100 microfibres for PES-Knit-Filament, PES-

Knit-Staple and PES/COT-Knit-Staple, and 10 microfibres for PES-Woven-Filament since the number of microfibres released by this garment was much smaller than those of the others. To confirm the chemical composition of the counted microfibres, FTIR spectroscopy was applied as detailed in the SI.

2.5. Statistics

Statistical analysis of the amount and number of microfibres released to water and air, respectively, was carried out by using IBM® SPSS® Statistics software. One-way Analysis of Variance (ANOVA) with a Student–Newman–Keuls (SNK) post hoc test, Welch ANOVA with a Games-Howell post hoc test, two-sample t-test and Mann-Whitney U (MWU) were applied. A 5% significance level was used for all statistical tests; p values <0.05 indicate significant difference among the data. More details are reported in the SI.

3. Results

3.1. Microplastic release to water

PES/COT-Knit-Staple released the greatest quantity of mg of microfibres per kg fabric of 1054 ± 158, while PES-Woven-Filament fabrics released the lowest one of 128 ± 62. PES-Knit-Filament and PES-Knit-Staple released 296 ± 36 mg of microfibres per kg fabric and 244 ± 25 mg of microfibres per kg fabric, respectively. The mass of microfibres recovered on 400 µm, 60 µm, 20 µm pore size filters and the concentration calculated dividing the mass of microfibres recovered on the 5 µm pore size filter for the volume filtered (mg/L), for each garment type, are reported in Figure S3 and discussed in the SI. It is to be highlighted that the largest quantity of microfibres was recovered on the filter with a 60 µm pore size for all garments except PES-Woven-Filament, for which the greatest aliquot was recovered on the 20 µm filter.

Since the PES/COT-Knit-Staple garment was made of a blend of 50%:50% polyester/cotton, in order to understand the composition of the microfibres released during the washing tests and recovered during the filtration of the wastewater, a thermogravimetric investigation was carried out on the microfibres accumulated on 400 and 60 µm pore size filters. The thermogravimetric curves are reported in Figure S1d and discussed in the SI.[45,46] The results of this analysis indicate that around the 80% of the fibres released from PES/COT-K-S to water were of cotton.

Microscopy analysis of the microfibres recovered on each filter was used to examine their dimensions. The length of the microfibres recovered on the different pore size filters is reported in Figure S1c. PES-Knit-Filament released microfibres had an average length of 610 ± 480 µm and diameter of 16 ± 3 µm; PES-Woven-Filament microfibres had a mean length of 760 ± 690 µm and diameter of 15 ± 2 µm; in the case of PES-Knit-Staple the average length and diameter were 796 ± 604 µm and 16 ± 2 µm, respectively; microfibres released from PES/COT-Knit-Staple had a mean length of 720 ± 742 µm and a mean diameter of 18 ± 6 µm. Regarding this last garment, the average length and diameter of cotton fibres were 889 ± 835 µm and 19 ± 6 µm respectively, whereas those of polyester fibres were 420 ± 395 µm and 15 ± 4 µm. Notwithstanding that the polyester microfibres released from PES/COT-Knit-Staple were significantly smaller than the cotton ones (MWU: U=706, p=0.00), the greater release of cotton microfibres could be due to the different chemical compositions of the fibres composing the yarn. In fact, the higher hydrophilicity of cellulosic fibres could influence the wettability of these fibres[47] during the washing process possibly increasing their release from the fabric. In addition, it should be considered that, during laudering, the wet abrasion of cotton is high since the fibres could swell and fibrils could easily be broken due to mechanical action and physical forces of the washing process.[48]

The weight of microfibres released by each garment type, was converted to a number of microfibres released by applying a formula with the mean dimensions of the released fibres, and the density of the material. The average numbers of microfibres released per gram of washed fabrics, per each type of textile, are reported in Figure 1a. The amount of microfibre released by the four different fabrics were compared through statistical analysis with Welch ANOVA (Table S1 in the SI), that detected significant difference in the quantities released ($F(3,6)=49.29$, $p=0.00$). Games-Howell post hoc test revealed a significantly greater release of microfibres from PES/COT-Knit-Staple (3898 ± 467 microfibres/g of fabric), than the releases from all the other garments ($p<0.05$ in all cases). Comparing PES/COT-Knit-Staple with PES-Knit-Staple, that has the same textile parameters (knitted structure, short staple fibres), the difference in the release for these two fabrics is related to the presence of cotton in the blend of PES/COT-Knit-Staple, as supported by the TGA analysis. PES-Knit-Filament (1747 ± 193 microfibres/g of fabric) released significantly more microfibres than both PES-Woven-Filament (709 ± 343 microfibres/g of fabric, $p=0.014$) and PES-Knit-Staple (1128 ± 111 microfibres/g of fabric, $p=0.011$). All three garments are made of 100% polyester, but with differences in textile parameters. PES-Knit-Filament and PES-Knit-Staple are both arranged in a knitted structure, but the first has yarns made of continuous filaments no twisted and with low hairiness, whereas the second has yarns made of short staple fibres moderately twisted and with high hairiness. Since the hairiness of PES-Knit-Filament is lower than PES-Knit-Staple, the greater release of PES-Knit-Filament could be ascribable to the absence of the twist in its yarns that could have favored the release of microfibres from the fabric. Moreover, it should be taken into account that the minimun lengths detected for PES-Knit-Filament and PES-

Knit-Staple were 71 μm and 87 μm, respectively. Then, this contradictory result on the release from PES-Knit-Filament, could be due to the release from PES-Knit-Staple of microfibres with dimensions lower than about 70 μm, not observable with the filtration and identification methods applied in these experiments. Instead, the differences in the release between PES-Knit-Filament and PES-Woven-Filament could be found in their different textiles structures (knitted vs. woven). In general, PES-Woven-Filament was the fabric that released the lowest amount of microfibres, but it was found not significantly lower than PES-Knit-Staple (p=0.248), indicating that a woven structure, with more twisted yarns, plays a role in reducing the amount of microfibres released during washing.

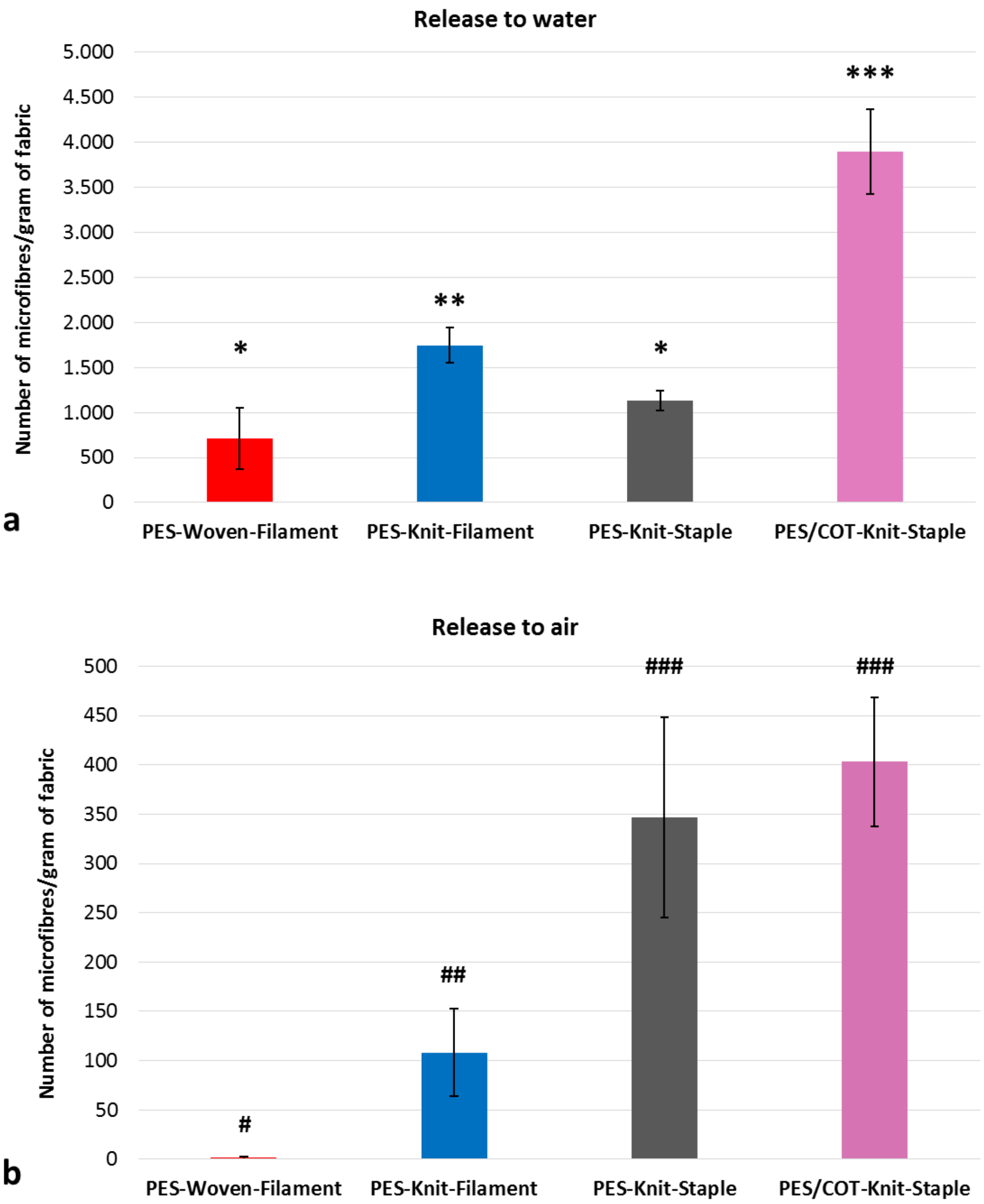

Figure 1. a) Release to water: number of microfibres released per gram of washed fabric from 100% polyester blouse PES-Woven-Filament, 100% polyester t-shirt PES-Knit-Filament, 100% polyester dress PES-Knit-Staple, 50% polyester/50% cotton sweatshirt PES/COT-Knit-Staple, (n=4); b) Release to air: number of microfibres per gram of worn fabric, released to air by wearing the 4 selected garments (n=4); Different symbols denote significant differences among the number of microfibres released by each type of fabric to water (*) and air (#).

3.2 Microplastic release to air

The number of microfibres released per gram of worn fabric for each kind of tested garment are reported in Figure 1b, together with the SD. Statistical analysis (Table S2) performed on these values confirmed that the number of microfibres released to air during the wearing of the garments differs significantly depending on the type of fabric (ANOVA-SNK: F(3,12)=35.45, p=0.00) and follow the order PES-Woven-Filament < PES-Knit-Filament < PES-Knit-Staple < PES/COT-Knit-Staple. No significant difference was found between the values of the two knitted fabrics with short staple fibres PES/COT-Knit-Staple (403 ± 65 microfibres/g of fabric) made of 50%:50% polyester/cotton, and PES-Knit-Staple (347 ± 102 microfibres/g of fabric) made of 100% polyester (ANOVA-SNK: p=0.24). A significant difference (ANOVA-SNK: p>0.05) was found between the number of microfibres released by the two fabrics constituted by continuous filaments, PES-Knit-Filament (108 ± 44 microfibres/g of fabric) and PES-Woven-Filament (1 ± 1 microfibres/g of fabric), confirming a mitigating effect of the woven structure. These results also indicate that the fabrics made of short staple fibres, PES-Knit-Staple and PES/COT-Knit-Staple, released more microfibres to air than those released by fabrics made of continuous filaments, PES-Woven-Filament and PES-Knit-Filament (ANOVA-SNK: p>0.05). Moreover, PES/COT-Knit-Staple and PES-Knit-Staple have also yarns poorly twisted whereas PES-Woven-Filament yarns presented the greatest twist. Regarding PES-Knit-Filament, the effect of the twist is less clear in this case since its yarns have no twist but maybe the fact that they are made of continuous filaments, could be responsible for the low release of microfibre and thus the fibre length had a mitigating effect. Mean fibre dimensions, length and diameter, were calculated analyzing the optical micrographs of the microfibres recovered in the Petri dishes. Microfibres released by PES-Knit-Filament were characterized by a length of 1036 ± 393 µm and diameter of 18 ± 4 µm; the dimensions of microfibres released by PES-Knit-Staple were length of 1023 ± 467 μm and diameter of 18 ± 3 μm; the microfibres released by PES/COT-Knit-Staple were 1024 ± 1008 μm in length and 21 ± 6 μm in diameter. Microfibres released by PES-Woven-Filament had diameter of 15 ± 4 μm and length of 494 ± 15 μm, significantly smaller than the lengths of both PES-Knit-Filament (t-test: t(28)=8.61, p=0.00) and PES-Knit-Staple (t-test: t(37)=7.81, p=0.00). It is interesting to note that microfibres released by both PES-Knit-Filament and PES-Knit-Staple have length not significantly different (t-test: (198)=0.21, p=0.83) even if the release for the latter was significantly higher (ANOVA-SNK, p>0.05). The comparison of the average length of the microfibres released to both media by each garment, showed that the length was greater for the microfibres released to air than water for all garments, except for PES-Woven-Filament where the contrary occurs.

The high SD of the length of PES/COT-Knit-Staple microfibres, measured among the 100 microfibres analysed to determine mean length and diameter, could be due to the difference between the length of cotton and polyester staple fibres. The staple length of a synthetic fibre is controlled by the manufacturer, so they may be all the same length or they consist of a mixture of fibres of different lengths blended in known proportions. In the case of a natural fibre, staple length is a much less easily defined characteristic of any batch of fibre, which basically consist of fibres varying in length over a wide range.[47]

FTIR analyses of subsamples of the microfibres collected in the Petri dishes during the tests revealed that they were all polyester for the polyester garments PES-Knit-Filament, PES-Woven-Filament and PES-Knit-Staple. Alternatively, in the case of PES/COT-Knit-Staple, results pointed out that only 1 of the 32 analysed fibres were polyester, while the others were all of cellulosic nature. Such result was foreseen during the inspection of filters under light microscope since the microfibres observed had all the characteristics of cotton fibres.[34]

4. Discussion

All the textiles examined released measurable quantities of fibres as a consequence of both laundering and everyday wear. The polyester/cotton blend garment showed the greatest release to both media, with a majority of microfibres, 80 % (in water) and 97 % (in air), being identified as cotton. This appears to indicate that garments with a polyester/cotton blend composition tend to release more cotton microfibres than synthetic ones, a finding in line with previous works[35,49,50]. However, there is no scientific consensus on microfibre release from cotton, since other works reported that polyester/cotton blended garments release less[31] or found no clear comparisons between the releases of polyester and cotton textiles[51]. Several studies have already reported the presence of cotton fibres in aquatic environments[52,53,54], ingested by fish[55] as well as in the atmosphere[19,25,27,56]. The occurrence of natural microfibres in different environment highlights that natural fabrics could shed more microfibres than synthetic ones. This could be due to the material composition and textile characteristics of natural fabrics. This data allows to hypotize a possible underestimation of the exposure of human to microfibres, since microfibres of natural origin are often not taken into account. Therefore, that calls for further research on the release, fate and impact of cellulosic microfibres in the environment.

PES-Knit-Filament and PES-Knit-Staple garments have the same knitted structure but the first was composed of continuous filaments with no twist and low hairiness, whereas the second of short staple fibres with moderate twist and high hairiness. Short staple fibres and high hairiness, have been found responsible for a greater release of microfibres during washing in a previous investigation[35] since the short fibres can more easily slip away due to the mechanical actions of wearing and moving, as also supposed by the mechanism of fibre release reported by Zambrano et al.[49]. However, PES-Knit-Filament released significantly less microfibres to air than PES-Knit-

Staple, but it behaves oppositely during the washing process, calling for further studies to assess if this outcome is due to limits in fibre length detection in washing tests (as discussed in the Results section) or if the release of microfibres during washing could have more complex release mechanisms and variables than that to air. PES-Woven-Filament was the garment responsible for the overall lowest numbers of microfibres released to both media. The reasons for this behavior lay in the textile parameter of PES-Woven-Filament, which is made of continuous filaments highly twisted and arranged in a woven structure. In fact, the releases of PES-Woven-Filament to air and water were significantly lower than those of PES-Knit-Filament, whose yarns were also made by continuous filaments but were arranged in a knitted structure with no twist. This could be due to the presence of high twisted yarns arranged in a woven structure, resulting in a more compact textile with respect to knit fabrics which are typically softer and more flexible.[57]

Comparing the overall quantities of microfibres released during washing to those reported by De Falco, et al.[35] of 49-308 g per kg of washed fabric, the release for the present study was much higher (128-1054 mg/kg of washed fabric). A possible explanation could be that washing tests carried out with only one garment result in greater wettability of the fabric that could enhance the mobility of microfibres that detach from the yarns. In fact, a recent work has pointed out that washing programs with a high water-volume-to-fabric ratio are responsible for a greater release of microfibres.[58] The outcomes of the present work indicate that in order to reduce microfibre emissions to water and air, the optimal textile parameters are, wherever possible, woven structure, yarns made of continuous filaments highly twisted, low hairiness. The selected parameters are in line with conclusions by Carney Almroth et al.,[34] who found that tightly constructed yarns (i.e. with high twist) are to be preferred to reduce microfibre release. Cesa et al.[59] also suggested that parameters responsible for fibres cohesion (i.e yarn twist, fibres size and regularity) avoid possible propagation of fibres. Zambrano et al.[49] highlighted that fabrics with lower hairiness, higher abrasion resistance and yarn strength released less microfibres. Furthermore, a previous work[20] found that woven polyester fabrics released more than knitted ones but this finding was correlated to the yarn type, that was made of short staple fibres in the woven structure, whereas it was composed by continuous filaments in the knitted fabric. This scenario points out how, despite differences in the method used in the different papers, there seems to be consensus among the scientific community that some parameters have a direct effect on microfibre release.

Here, we present the first estimation of microfibres released to air directly as a consequence of wearing clothes. Considering other results reported in literature on fibre deposition from the atmosphere, the lengths of fibres (494-1036µm) released from clothing in our experiments resembled that of past studies for indoor air and dust (50-450µm) as well as fibres found in human lung tissues (50-250µm).[25,29,30] Some information of the consequences of chronic high dose exposure to microfibres can be derived from studies on the health of workers of synthetic textile and flock industries, who presented an increased prevalence of the following symptoms: interstitial lung disease, reduced lung capacity, coughing, dyspnoea, wheezing, increased phlegm production, allergic reactions, asthma.[60] Further, due to their hydrophobic nature, textile fibres have the potential to sorb and subsequently release chemical contaminants during wear and washing, including additives, unreacted monomers and environmental pollutants.[61,62] Therefore, more studies are needed to assess the potential exposure and consequent impact of microfibres, both of synthetic and natural origin, on human health.

The data obtained were scaled up to obtain an estimation of the possible number of polyester microfibres that a single person could release per year. In order to scale up the numbers of polyester microfibres released to water and to air, and allow a more direct comparison of the emissions, the following assumptions were made: (1) one person performs 55 laundry cycles per year with an average load of 4 kg of polyester garments per wash18; (2) one person wears 1 kg of polyester garments and performs similar movements simulated during the air tests for 8 h per day; (3) the number of microfibres per gram of fabric released during washing among the values related to the four types of textiles and the same was done for the number of microfibres per gram of fabric released during wearing. Regarding these last assumption, both for washing and wearing, for the PES/COT-Knit-Staple garment only the contribution of polyester fibres was considered, on the basis of TGA analysis for the washing tests (20%) and FTIR spectroscopy for wearing tests (3%). These assumptions lead to data useful for understanding the possible orders of magnitude of polyester microfibre release to water and air, mainly for comparison reasons. Of course, they cannot be representative of the global release of microfibres to both media, since only one type of material, i.e. polyester, was taken into account. The scale up of the data indicate that one person could release a number of polyester microfibres per year of approximately $2.98 \cdot 10^8$ to water by washing, and $1.03 \cdot 10^9$ to air by wearing polyester garments. Of course, these estimations do not take into account the variability of garments actually used by a person. However, the relative magnitude of the two releases is very similar and this highlights that microfibres are not just released from clothing to wastewater but also to the air. Such finding implies that previous estimations of microplastic pollution in world oceans[7], actually underestimated the impact of synthetic textiles on this environmental problem since they did not take into account the amount of synthetic microfibres that can reach the oceans by atmospheric deposition. In fact, atmospheric deposition is likely an important pathway for microfibres to enter soils; yet, the sources, fate and effects of microfibres to terrestrial ecosystems is understudied.[63] Further, there is likely underestimation of microfibre pollution in the environment due to the exclusion of inputs of natural and artificial fibres which this study has shown is an abundant component of microfibres released from clothing (through both washing and wear).

Hence, mitigation actions focused on intercepting microfibre release to water (e.g. washing machine filters, washing conditions, wastewater treatment etc.) will only address part of the issue and a wider approach is therefore needed. Based on the findings of this study and others[32,34,35,49,59], it is clear that textile parameters play a role in influencing microfibre emissions to both water and air. Therefore, improved textile designs utilizing, where possible, textile parameters able to reduce the amount of microfibres released, could be an effective solution to tackle the problem of microfibre emissions. From the work so far it would appear that design should focus on production of fabrics with a compact structure, such as woven, using highly twisted yarns made of continuous filaments, while more loose structures including knitted, short staple fibres and low twist should be avoided when possible. Further investigations should be performed to create a comprehensive set of recommendations to guide industry and policy. Mitigation actions at the textile design stage could also be complemented by application of a finishing treatment that is able to protect the fabric from the chemical and mechanical actions of laundering or from friction and abrasion during wearing, but more research is still needed on this topic.[64,65,66] The combination of these measures, possibly together with fibre capture systems for washing machines[67] could lead to substantial reduction of microfibre pollution from textiles.

ASSOCIATED CONTENT

Supporting Information.

The following files are available free of charge.

Additional information as noted in the text (PDF)

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Notes

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