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Microfibers generated from the laundering of cotton, rayon and polyester based fabrics and their aquatic biodegradation

Marielis C. Zambrano[a], Joel J. Pawlak[a], Jesse Daystar[b,d], Mary Ankeny[b], Jay J. Cheng[c], Richard A. Venditti[a,*]

[a] Department of Forest Biomaterials, College of Natural Resources, North Carolina State University, Raleigh, NC 27695-8005, United States
[b] Cotton Incorporated, Cary, NC 27513, United States
[c] Department of Biological and Agricultural Engineering, North Carolina State University, Raleigh, NC 27695-7625, United States
[d] Nicholas School of the Environment, Duke University, Durham, NC 27708, United States

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ABSTRACT

The effect of fiber type (cotton, polyester, and rayon), temperature, and use of detergent on the number of microfibers released during laundering of knitted fabrics were studied during accelerated laboratory washing (Launder-Ometer) and home laundering experiments. Polyester and cellulose-based fabrics all shed significant amounts of microfibers and shedding levels were increased with higher water temperature and detergent use. Cellulose-based fabrics released more microfibers (0.2–4 mg/g fabric) during accelerated laundering than polyester (0.1–1 mg/g fabric). Using well-controlled aquatic biodegradation experiments it was shown that cotton and rayon microfibers are expected to degrade in natural aquatic aerobic environments whereas polyester microfibers are expected to persist in the environment for long periods of time.

1. Introduction

In the last 60 years, the production of resin and fibers has increased significantly from two million metric tons (Mt) in 1950 to around 380 Mt. in 2015 (Geyer et al., 2017; Plastics Europe, 2017; Statista, n.d.). Geyer et al. (2017) estimated that 8300 Mt. of plastics have been produced up to 2015 generating approximately 6300 Mt. of plastic waste from which 9% have been recycled, 12% incinerated, and 79% accumulated in landfills and natural environments. The accumulation of plastics in the environment is a subject of concern for industries, governments, and communities (Rochman et al., 2013; Thevenon et al., 2011; Thompson et al., 2010), especially in water sources (Thompson et al., 2004).

The presence of plastics in water bodies can be the origin of different problems such as aesthetic issues, entanglement and suffocation of marine animals in plastic nets, plastic ingestion by the fauna, adsorption of pollutants and pathogens in plastics particles and transportation in the ecosystems (Moore, 2008; Thevenon et al., 2011).

Plastics found in the environment may be classified according to their size and source. Macroplastics are big pieces (> 5 mm) generated from poor waste management strategies. Particles below 5 mm in size are commonly called microplastics (Boucher and Friot, 2017; Eriksen et al., 2014; Moore, 2008; Thevenon et al., 2011; Wagner et al., 2014). In general, microplastics are classified into two forms. Primary microplastics are discharged to the environment at the micro size; these can be plastics manufactured in the micro size such as scrubbing agents or pellets, or particles produced from the abrasion during wear and use of plastic goods such as tires and synthetic textiles (Boucher and Friot, 2017). Secondary microplastics are produced from the fragmentation of mismanaged plastic waste in the environment (Boucher and Friot, 2017). Nevertheless, there is no consensus about the microplastics definition in the scientific community. After a comprehensive review of reported methods for describing and identifying microplastics, Frias and Nash (2019) proposed a new definition of microplastics that considers their physical and chemical properties, size, and origin: "*Microplastics are any synthetic solid particle or polymeric matrix, with regular or irregular shape and with size ranging from 1 μm to 5 mm, of either primary or secondary manufacturing origin, which are insoluble in water*". In that study nanoplastics were considered < 1 μm.

Microplastics are floating in the world's oceans, rivers, lakes, and are depositing in sediments (Browne et al., 2011; Eriksen et al., 2014; GESAMP, 2015; Miller et al., 2017; Thompson et al., 2010; Wagner et al., 2014). It has been estimated that a minimum of 5.25 trillion plastic particles weighing about 270,000 tons are floating in the world's oceans and 93% of these plastics particles are in the micro size range (Eriksen et al., 2014). This represents ~ 0.1% of the world's plastic

* Corresponding author.
E-mail address: richardv@ncsu.edu (R.A. Venditti).

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annual production (Eriksen et al., 2014).

Available studies have demonstrated that marine fauna is susceptible to microplastics ingestion (Thevenon et al., 2011; Wagner et al., 2014). The presence of anthropogenic debris in seafood for human consumption has been observed (Miranda and de Carvalho-Souza, 2016; Rochman et al., 2015). There is also evidence of microplastic ingestion by humans (Schwabl, 2018). Microplastics have been observed even in commercial food-grade salts (Kim et al., 2018). Consequently, microplastic pollution is raising concerns regarding human health.

Even though the microplastics are relatively inert, due their large surface-to-volume ratio and chemical composition, they can adsorb pollutants and pathogens and transfer them via ingestion to other trophic levels, even to humans through the food chain (Egbeocha et al., 2018; GESAMP, 2015; Rummel et al., 2017; Wagner et al., 2014; Wang et al., 2018). It has been shown that microplastics sampled from different environments adsorbed toxic compounds such as polychlorinated biphenyls (PCB), dichloro diphenyl trichloroethane (DTT), hexachlorocyclohexane (HCH), polycyclic aromatic hydrocarbons (PAHs), polybrominated diphenyl ethers (PBDE), hexabromocyclododecane (HBCD), heavy metals, nonylphenols (NP), and perfluoroalkyl substances (Akhbarizadeh et al., 2017; Chen et al., 2018; Holmes et al., 2014; Llorca et al., 2014; Mato et al., 2001; Vedolin et al., 2018; Wang et al., 2018). Several studies indicated that microplastics and macroplastic debris distributed at various concentrations in aquatic ecosystems around the world affect the growth, development, behavior, reproduction, and mortality of aquatic animals (Chae and An, 2017). However, other authors consider that at environmentally relevant concentrations, microplastics have not been shown to have harmful effects to aquatic fauna and do not represent a significant exposure route for toxic chemical compared to prey consumption in aquatic birds or organisms (Burton, 2017; Henry et al., 2019).

There are many different sources of primary microplastics and one of the main challenges is to prevent the generation and leakage of these particles to the environment from their origin (Boucher and Friot, 2017; GESAMP, 2015). Although wastewater treatment plants (WWTPs) have shown good efficiency in microplastic removal, they are not designed for that purpose (Lares et al., 2018; Magnusson and Norén, 2014; Talvitie et al., 2015). Microplastics, mainly particles and fibers smaller than 100 μm, have been observed in wastewater effluents at low concentration (Browne et al., 2011; Lares et al., 2018; Magnusson and Norén, 2014; McCormick et al., 2014; Mintenig et al., 2017; Talvitie et al., 2015, 2017a, 2017b; Wolff et al., 2018). However, the large volumes of effluents discharged every day to the water bodies and the use of sewage sludge (where most of the microplastics are retained) as soil amendment/fertilizer represent an important source of microplastics introduced into the environment (Lares et al., 2018; Mintenig et al., 2017; Nizzetto et al., 2016; Talvitie et al., 2017b). These microplastics are found mainly in the fiber-like form in some effluents (Lares et al., 2018; Mason et al., 2016; Mintenig et al., 2017; Talvitie et al., 2015, 2017a, 2017b). These fibers are predominantly made of polyester, and more specifically, polyethylene terephthalate (PET) (Lares et al., 2018; Mintenig et al., 2017; Talvitie et al., 2017a; Wolff et al., 2018; Ziajahromi et al., 2017). Nevertheless, some studies also indicate that cellulose-based fibers are present in the wastewater effluents (Lares et al., 2018; Talvitie et al., 2017b, 2015; Ziajahromi et al., 2017).

All of these fibers are common in the textile industry and it is believed that they enter the WWTPs from the effluent of the washing machines (Browne et al., 2011; McCormick et al., 2014; Thompson et al., 2010). In addition, in less developed countries, it is common to discharge home laundering effluents directly into the environment (Boucher and Friot, 2017).

The International Union for Conservation of Nature (IUCN) (Boucher and Friot, 2017), estimated that between 0.8 and 2.5 Mt./year of primary microplastics are released into the ocean. From this, it is estimated that 35% are micro-size fibers released from textiles during laundering (Boucher and Friot, 2017). Fibers at the micro-size are commonly named microfibers by the environmental science community. Microfibers could be made of synthetic polymers, in that case, they will also be microplastics. Natural fibers also represent a large proportion of the textile industry and the fate of these microfibers are largely unknown in published literature (Henry et al., 2019; Ladewig et al., 2015; Mishra et al., 2019; Salvador Cesa et al., 2017). The term “microfiber” is also used in the textile industry to refer to fabrics made of fine polyester or polyamines fibers, < 1 denier (mass in grams of 9000 m) and a fiber cross-section smaller than 10 μm (Henry et al., 2019; Salvador Cesa et al., 2017). To avoid confusion in this study, the term microfiber will refer exclusively to the synthetic, artificial, and natural fibers (< 5 mm) released from fabrics during laundering.

Several studies have quantified the number of microfibers released during home laundering, however, there is lack of standardization in the methods used and the metrics that are needed to incorporate a reliable value in environmental sustainability assessments (Henry et al., 2019; Salvador Cesa et al., 2017).

Browne et al. (2011) observed that polyester garments (blankets, fleeces, and shirts) can shed > 1900 fibers per wash. All garments released > 100 fibers per liter of effluent, with > 180% more from fleeces than other construction types. Carney Almroth et al. (2018) reported, using a laboratory scale washing machine, that fleece and microfleece synthetic fabrics shed the greatest amount of fibers, up to 7360 fibers $m^{-2} L^{-1}$ in one wash, indicating that the fabric construction plays a major role in the shedding ability of fabrics. Likewise, Napper and Thompson (2016) observed that 6 kg of synthetic material fabrics (polyester, polyester-cotton blend, and acrylic fabrics) could release about 140,000 to 700,000 fibers per wash in commercial home laundering washing machines. Another study showed that the recovered microfiber mass per garment tested, jackets or sweaters made of polyester fleece or nylon shell with nonwoven polyester insulation, ranged from approximately 0 to 2 g, exceeding 0.3% of the unwashed garment mass (Hartline et al., 2016).

Hernandez et al. (2017) showed in their study of polyester fabrics that the use of detergent during home laundering is the aspect that has the greatest effect on the generation of microfibers. Approximately 75% more microfibers were released when using detergent regardless of type (liquid, powder, or surfactant) and dosage, 0.025 vs 0.1 mg fibers/g textile washed, without and with detergent, respectively (Hernandez et al., 2017). De Falco et al. (2018) observed a similar tendency with polyester and polypropylene woven and knitted fabrics. The use of detergents promoted an increase of microfibers released from fabrics during laundering; in this case, the powder detergent induced more microfiber shedding than the liquid one in domestic washing and, in general, products for industrial washing have a higher impact than domestic washing detergents (De Falco et al., 2018).

De Falco et al. (2018) saw a 35% decrease in microfiber shedding when using a softener. Nevertheless, the influence of temperature and softener use in the microfiber generation have been less consistent and significant between studies than the effect of detergent use (De Falco et al., 2018; Hernandez et al., 2017; Napper and Thompson, 2016; Pirc et al., 2016).

When comparing washing machine types, it was observed that top-load machines shed more fiber mass than front-load machines (Hartline et al., 2016). Additionally, Hartline et al. (2016) reported that mechanically aged-fabrics release more fibers than new fabrics. Nevertheless, sequential washing experiments have shown that the number of microfibers released during laundering is generally reduced with increased washing cycles until reaching a constant level (Carney Almroth et al., 2018; Napper and Thompson, 2016; Pirc et al., 2016; Sillanpää and Sainio, 2017).

To our knowledge, there has been only one study that investigates the effect of the type of polymer yarn material used in textiles with the same fabric constructions. However, natural fibers were not included in

this study, only synthetic fibers such as polyester, acrylic, and nylon were compared (Carney Almroth et al., 2018). In most research, the use of different fabric constructions and/or textile goods studied does not allow a fair comparison of the fiber polymer composition effect on microfiber generation.

Sillanpää and Sainio (2017) reported the mass, 0.12 to 0.33% w/w per mass of fabric washed, and number, 2.1×10^5 to 1.3×10^7 fibers per kg of fabric washed, of microfibers released during sequential home laundering washing cycles of polyester and cotton fabrics of different constructions. The results of this study indicate that cotton fabrics have a higher tendency than polyester fabrics to shed fibers during washing, however, the differences in fabric constructions make it difficult to derive a final conclusion from this data (Sillanpää and Sainio, 2017).

In the research reported herein of fabric shedding during laundering, different fiber polymer materials were studied all with the same knit construction. Accelerated laboratory laundering and home laundering experiments were performed and the shed microfiber quantity and characteristics were determined. Polyester and cotton knitted fabrics were evaluated since they represent the greatest portion of the global fiber production for natural and synthetic fibers, respectively (Mills, 2011). As well, a rayon knitted fabric was included in this study as a representation of the regenerated cellulosic fibers in the market. In addition, methods to predict fiber shedding based on fabric and yarn mechanical properties are proposed and evaluated herein.

Another important dimension to the laundering microfiber issue is their fate in the environment. Surprisingly, little is known about the biodegradation of textile fibers in aquatic environments. Most degradation studies have been focused on biodegradable and synthetic polymers intended for packaging applications in composting environments (Eubeler et al., 2009, 2010; Karamanlioglu and Robson, 2013; Lešinský et al., 2005; Lucas et al., 2008; Pagga et al., 1996; Starnecker and Menner, 1996). There have been no studies on aquatic biodegradation of textile fibers known to the authors of this study. In addition, there is an emerging concern about the role of natural and artificial fibers on the distribution of chemical pollutants in aquatic environments (Ladewig et al., 2015). Therefore, this research includes a study of the aquatic biodegradation of the yarns of the major textile fibers, cotton, polyester, rayon, and polyester-cotton blends as a starting point to differentiate the effect of fiber polymeric material on the fate of the microfibers in aquatic environments.

2. Materials and methods

2.1. Fabrics and yarns

Four interlock fabrics without finishing were manufactured by Cotton Incorporated to perform the laundering experiments; the spun yarns contained 100% cotton, 100% rayon, 100% polyester, and 50%/50% polyester/cotton (Table S1). The weft knitted interlock construction was made on a 24-cut circular knitting machine (24 needles/in.). Spun yarns from staple fibers with a size of 40/1 Ne (English Cotton Count, 40 × 840 yards of one single yarn weight 1 pound) were used to knit the fabrics (Table S2). As pre-treatment, all fabrics were scoured to remove impurities from the fibers such as wax, fats, pectin, proteins, and organic acids, and improve their wettability. Additionally, fabrics containing cotton were also bleached. These fabrics were dyed with different colors to assess cross-contamination. The spun yarns cut in 1 cm length were used in the biodegradation experiments. These yarns were pre-treated as were the fabrics and did not contain finishes or dyes.

2.2. Accelerated laundering experiments

2.2.1. Washing protocol

The behavior of the fabrics during laundering was studied at different conditions using the AATCC (American Association of Textile Chemist and Colorists) standard SDL Atlas Launder-Ometer (South Carolina, USA) with metal canisters of 550 mL capacity. The fabrics were cut in square pieces of size 4in × 4in using an ATOM Swing Arm Clicker Presses model SE20C (Italy). The edges were secured using the 1150 MDA Serger Machine Brand Bernina (Thurgau, Switzerland) with white 100% polyester yarn brand Excell to avoid errors by the excessive release of microfibers through the edges of the fabrics, these edges were approximately 12% of the total sample weight. Each piece of fabric weighed approximately 2–3 g. A cleaning cycle was performed on the fabric pieces. In each metal canister 25 metals balls (6 mm of diameter), 150 ml of deionized (DI) water and the fabric sample were added. The cleaning cycle of the fabric was done for 16 min at the temperature of interest. The cleaning wash is a very important step in this procedure; it allows the removal of fibers and impurities present in the fabrics from the manufacturing process.

Cold (25 °C) and warm (44 °C) laundering experiments were conducted with deionized (DI) water and detergent solution (AATCC Monograph 6-2016, 2017). According to the conditions studied, 25 metals balls (6 mm diameter), 150 ml of DI water or detergent solution and the fabric sample were added to a 550 ml metal canister and tumbled for 16 min. The detergent solution was prepared with 1. 47 g of the 2003 AATCC Standard Reference Liquid Laundry Detergent with optical brightener (AATCC Monograph 2-2005, 2017) dissolved in one liter of DI water (AATCC Test Method 135-2015, 2017). For each condition and fabric type, eight to ten canisters were prepared for the Launder-Ometer. In addition, a canister with the cleaning agent and the metal balls but with no fabric was used as a blank during the filtrate analysis.

2.2.2. Laundering filtrate analysis

After the washing protocol, all the liquid (150 mL) inside each canister was recovered in individual glass bottles of 16 oz. capacity, eight to ten replicates were collected for each fabric type and condition. The mass of microfibers released during laundering was recovered by filtration using Whatman glass microfiber filter papers grade GF/C, 47 mm and 1.2 μm particle retention, whereas the quantification of individual fibers and their length and width distributions were obtained with the HiRes Fiber Quality Analyzer (FQA), OpTest Equipment Inc. (Ontario, Canada). These analyses were also made on the liquid collected from the canisters without fabrics used in each condition as blanks to account for the potential influence of the washing agent and the background variability.

2.2.3. Statistics

The effect of temperature and use of detergent on the microfiber generation during laundering was evaluated with a linear mixed model using SAS 9.4 and Excel 2016. This model assumes data non-linearity and assesses if the effect of fabric type, temperature, and use of detergent influence the microfibers generation at a significant level $\alpha = 0.05$. The details of this model are described in the Supplementary material (S 1.1.4).

2.3. Home laundering experiments

2.3.1. Washing/drying protocol

The Whirlpool Washing Machine Model WTW57005WO (Michigan, USA) with temperature control system Quick Temp SDL ATLAS (South Carolina, USA) and the Dryer Whirlpool Model WED57005WO (Michigan, USA) were cleaned carefully prior to performing this protocol (Supplementary material section S 1.2.1). The fabrics were cut to obtain samples of 4 lb. (1.8 kg), representative of a large load in a home laundering washing machine (AATCC Test Method 135-2015, 2017). The samples were weighed using an OHAUS Champ bench scale (New Jersey, United States). In addition, the edges of the fabric were secured using the 1150 MDA Serger Machine Brand Bernina (Thurgau, Switzerland) with white 100% polyester yarn brand Excell to avoid errors

by the excessive release of microfibers through the edges of the fabrics. As a first step, each sample was washed with tap water using the normal wash load setting to remove the loose fibers and impurities from the manufacturing process and storage.

Before starting the washing cycle, a circular nylon mesh filtering screen of 20 μm Sefar 03–20/14 of 24 cm of diameter was secured at the discharge pipe of the washing machine to collect the microfibers generated during the laundering process. The temperature control system was configured to have 115 °F (46 °C) during the washing cycle and 80 °F (27 °C) during the rinsing cycle. Once the washing cycle was completed, the fabric sample was transferred to the drying machine. The drying process was set at high temperature (heavy load) for 60 min.

After the first cycle of washing and drying was completed, the procedure was repeated two more times with the same fabric to study how subsequent washings influence the microfibers generation. Samples collected during washing on the nylon mesh were re-dispersed in water prior to further analysis. The nylon mesh was washed five times with 100 mL of DI water to remove the fibers retained and this water was collected in a clean glass beaker (1 L). A new nylon mesh was used in each washing cycle to avoid cross-contamination. The re-dispersed sample was filtered through a Whatman glass microfiber filter paper grade GF/C (47 mm and 1.2 μm particle retention) and the filter paper was dried at 105 °C overnight and weighed.

2.4. Correlations between microfiber shedding and physical testing of yarns and fabrics

The tensile properties of yarns used to knit the fabrics were measured by the single-strand method (ASTM D2256/D2256M − 10, 2015) in the dry and wet states. The tensile tester MTS Q Test 5 (Minnesota, USA) was used to perform the test in 1 Grab 10 in. GL mode. The yarns were conditioned for 24 h prior to testing under the standard atmosphere for textile testing (70 ± 2 °F and 65 ± 2% relative humidity) and the dry tests were performed in the same environment. For the wet state tests, the samples were submerged for 5 min in DI water and loaded immediately in the tensile tester. For the dry state, 15 strands were tested of each yarn sample; and for the wet state, only 10 strands of each yarn sample were evaluated. The initial speed of the machine was adjusted between samples and conditions to achieve a breaking time of 20 ± 3 s during the experiments (Table S5). The Maxi-Martindale Abrasion tester from James H. Heal and Co. Ltd. (Halifax, UK) was used to measure the abrasion resistance of the fabrics used for the laundering experiments following the Martindale Abrasion Tester Method (ASTM D4966 − 12, 1992). Three replicates of each fabric were prepared, circular specimens of 1.5 in (38 mm). Standard wool abrasion fabric was used as an abrasion agent applying a pressure of 9 KPa and 20,000 rubs. The fabrics were conditioned for 24 h prior to testing under standard atmosphere for textile testing (70 ± 2 °F and 65 ± 2% relative humidity) and the test was performed under the same environment. The mass loss of the fabric after the abrasion test was obtained by weighing the sample before and after the test using the balance Voyager Pro OHAUS, precision = 0.1 mg (New Jersey, United States).

2.4.1. Statistics

SAS 9.4 and Excel 2016 software versions were used to assess the differences in breaking load between the types of yarns, in wet and dry state, and in abrasion resistance between the types of fabrics at a significant level $\alpha = 0.05$. Pair comparisons between the types of yarns were made using PROC ANOVA and PROC GLM within conditions (dry and wet state). The PROC MIXED procedure was used to analyze the fabric abrasion resistance data based on a linear mixed model taking as fixed the effect of the type of fabric tested. There is a significant difference between compared variables when the p-value obtained from the tests is < 0.05.

2.5. Aquatic biodegradation of yarns

The aquatic biodegradation of the spun yarns was assessed by measuring the oxygen demand of the yarns in a closed respirometer using as inoculum aerobic microorganisms in the activated sludge from the Neuse River WWTP in Raleigh, NC (ISO 14851:1999, 2005). The activated sludge was added to the flasks within 72 h after collection. The concentration of suspended solids in the activated sludge was measured (ISO 11923: 1997, 1997) and the inoculum was added to the flasks to achieve a concentration of 30 mg of solids per liter of testing medium to simulate biodegradation in natural environments. For the preparation of each flask, a magnetic stirrer bar was inserted in each bottle with the appropriate amounts of test medium and inoculum to have 400 mL of testing medium in each flask. The standard test medium (nutrients) for the ISO 14851 was used.

The level of biodegradation was determined by comparing the oxygen uptake (Ou) with the theoretical oxygen demand (ThOD) of the spun yarns. Microcrystalline Cellulose (MCC), 20 μm powder from Aldrich Chemistry, was used as reference material to check the inoculum activity during the test.

After measurement and adjustment of the pH to 7, the flasks were aerated using aquarium pumps to achieve oxygen saturation in the medium for 15 min at the beginning of the experiment and after each measurement. The dissolved oxygen (DO) concentration after saturation was measured. The DO in the medium was measured with an Orion Star™ A123 Dissolved Oxygen Portable Meter (Thermo Fisher Scientific, Massachusetts, USA) and the pH meter Accumet XL150 from Fisher Scientific (New Hampshire, USA). The DO meter was calibrated prior measurments with moisture saturated air (100% Relative Humidity) and the pH meter was calibrated using buffer solutions (pH 4, 7, and 10) from Fisher Chemicals (New Hampshire, USA). The flasks were closed tightly, incubated in a water bath at a constant temperature (25 °C) and stirred at 300 rpm. The test medium was allowed one week of incubation to let the microorganisms use the organic matter already present in the activated sludge and allow the stabilization of the inoculum to the test medium. After this incubation period, the test materials (spun yarns and MCC) were added to each flask.

Every week, or more frequently depending on oxygen depletion rates, the concentration of DO in each flask was measured and the pH adjusted to 7. The flasks were then aerated to saturate the test medium with oxygen and maintain the aerobic environment for the biodegradation process. After saturation, the DO in each flask was measured. This procedure was performed to account for the oxygen consumption during the course of the experiment.

Based on the standard method, this test should be considered valid if the degree of biodegradation of the reference material is $> 60\%$ and the biological oxygen demand (BOD) of the blank did not exceed 60 mg/L at the end of the test (ISO 14851:1999, 2005).

Several samples were collected at the end of the experiment. The liquid portion was separated from the solids remaining by filtration using Whatman glass microfiber filter papers grade GF/C (47 mm and 1.2 μm particle retention). The liquid portion was used to measure the nitrification interferences to account for the oxygen consumed by nitrification reactions. In the solid portion, scanning electron microscope (SEM) images were taken and elemental analysis and Fourier-transform infrared spectroscopy (FTIR) were performed and compared with the same analyses on test materials before the biodegradation experiment.

2.5.1. Statistics

SAS 9.4 and Excel 2016 software versions were used to assess the differences in biodegradation between the types of yarns. Pair comparisons between the types of yarns were made using a non-parametric model for ANOVA (Wilcoxon Model) with a significance level $\alpha = 0.05$. There is a significant difference between the variables compared when the p-value obtained from the test is < 0.05.

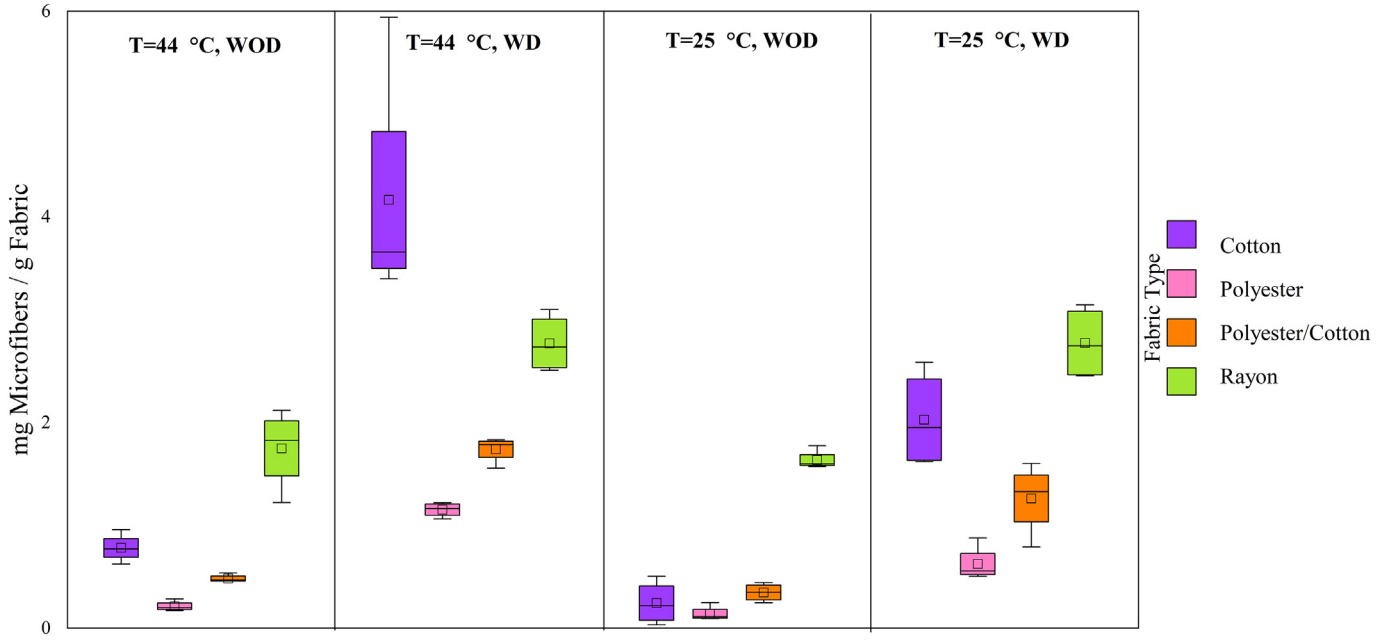

Fig. 1. Distribution of the mass of microfibers generated during accelerated laundering per mass of fabric washed at different conditions; washing with detergent (WD), washing without detergent (WOD), and different temperatures. The different types of fabrics are shown in different colors: purple for cotton, pink for polyester, green for rayon, and orange for the polyester/cotton blend. The length of the box represents the interquartile range (IQR, the range between the 25th and 75th percentiles), the mean is represented by a square and the median by a horizontal line in the box, and the vertical lines (whiskers) represent the range within 1.5 IQR. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

3. Results and discussion

3.1. Microfibers generation during accelerated laundering at small scale

3.1.1. Effect of fabric material

All fabric types released significant amounts of microfibers, with rayon, cotton, and polyester/cotton fabrics of the same knit construction releasing significantly more microfibers during accelerated washing than polyester fabrics (Fig. 1). Similar behavior was observed by Sillanpää and Sainio (2017) in domestic washing, after 5 sequential washes, cotton fabrics released up to 1.0×10^6 fibers per kg of fabric washed whereas polyester fabrics shed up to 5.0×10^5 fibers per kg of fabric washed.

The result reported here for the microfibers released during accelerated laundering of blended knitted fabric (50%/50% polyester/cotton) versus 100% polyester knitted fabric is different than the behavior described by Napper and Thompson (2016). They showed that 65% polyester/35% cotton jumper fabrics released significantly fewer fibers than polyester and acrylic jumper fabrics. These jumper fabrics were not all made in the same way as they were purchased from commercial retailers and were not specified as the same brands. This suggests that the influence in fabric and yarn construction and proportion of the components of the blend play a major role in the microfibers generation during laundering and should not be generalized. The generation of microfibers with length between 25 and 200 μm had the same or higher number magnitude than the microfibers released from the fabrics during laundering with length > 200 μm, at the conditions of this study (data shown in Fig. S10).

The influence of fabric structure was also reported elsewhere (Carney Almroth et al., 2018; De Falco et al., 2018). Carney Almroth et al. (2018) observed that polyester fleece fabrics shed significantly more fibers than knit fabrics (polyester, nylon, and acrylic), about 1200 and 9 fibers per 100 cm^2 of fabric, respectively. Whereas, De Falco et al. (2018) noted the highest release of microfibers from woven polyester with respect to knit polyester and woven polypropylene.

3.1.2. Effect of detergent use

The effect of the washing solution on the mass of microfibers released during accelerated laundering is also presented in Fig. 1. All materials at 25 °C and at 44 °C showed an increased mass of microfibers released during the laundering with detergent. There was a statistically significant increase (p-value < 0.05) in the microfibers mass released when the detergent solution was used as a washing agent for all fabric types at 44 °C (Fig. S9, A). For cotton, rayon, and polyester/cotton fabrics, the increase in microfibers generated by the presence of detergent during accelerated laundering was statistically significant (p-value < 0.05) also at 25 °C (Fig. S9, B).

Similarly, the data obtained using the FQA (Fig. 2) shows the same tendency as the gravimetric measurements, the number of microfibers released during laundering are increased with detergent and this increase is statistically significant (p-value < 0.05) for all but the rayon fabric (Fig. 2, A). Generally, the main influence of the detergent in the washing solution is attributed to the generation of microfibers with 25–200 μm in length, which outnumber, from a count basis, the microfibers with length > 200 μm, Fig. 2.

In fact, several options have been proposed to reduce the number of microfibers released into the effluent wash water, such as The GUPPYFRIEND washing bag, the Coral ball, and filters that retain the fibers for later disposal (Cora Ball, n.d.; Environmental Enhancements, 2019; Filtrol, n.d.; Guppyfriend, n.d.). Nevertheless, our results show that there is an important portion of microfibers with a size below 200 μm that cannot be addressed by these trapping mechanisms.

McIlwraith et al. (2019) showed that the Lint LUV-R filter that can be installed at the effluent pipe of the washing machine can capture 87% of the fibers released by a 100% polyester fleece blanket during washing and the Coral ball only trapped 26%. The fibers lost in the effluent water were also measured. After using the Coral ball, the length of the fibers did not change significantly compared to the length of the fibers collected from the effluent with no mitigation strategy (1.5 ± 0.5 mm). However, this was not the case when the Lint LUV-R filter was used; the length of the fibers decreased significantly to 0.4 ± 0.1 mm, since these mechanisms cannot trap fibers smaller than

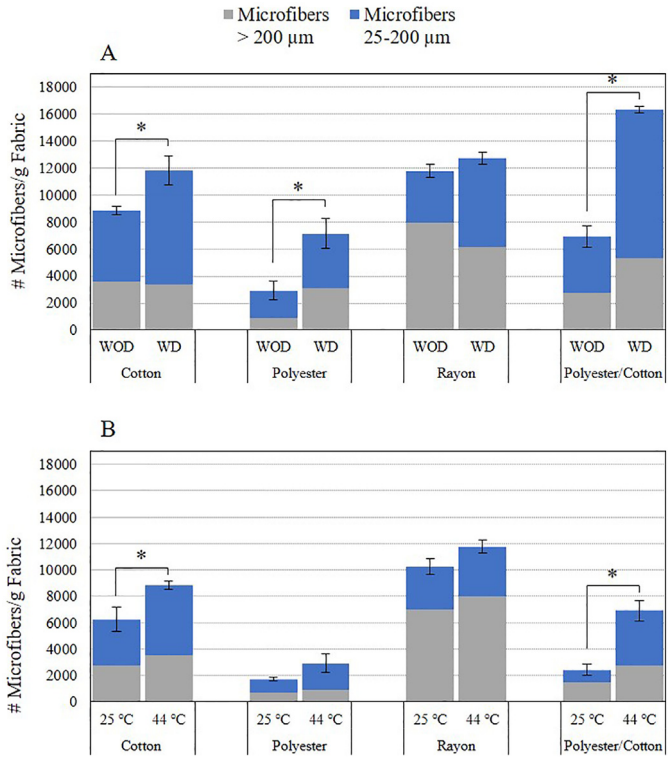

Fig. 2. Number of microfibers generated during accelerated laundering per mass of fabric washed. A - Effect of detergent use on the generation of microfibers at 44 °C. B - Effect of temperature on the generation of microfibers, without detergent (WOD). Error bars represent the standard error (N = 4, except for Polyester at 25 °C with N = 5). Significant differences are indicated by * ($p < 0.05$).

their pore size, in this case, 150 µm (McIlwraith et al., 2019).

According to these results, it can be concluded that the presence of surfactant assists the mobilization process of the broken/loose fibers from the fabric to the washing solution, also reported in previous studies made with polyester and acrylic fabrics during home laundering (Carney Almroth et al., 2018; Hernandez et al., 2017).

3.1.3. Effect of temperature

The effect of temperature on the microfibers generation in the presence of detergent solution and in deionized water is reported in Fig. 1. The mass of microfibers released from the fabrics during laundering increased at a higher temperature for all types of fabrics, nevertheless, the difference was only statistically significant for cotton (p-value < 0.05) (Fig. S9, C and D).

The FQA results were similar to the gravimetric analysis (Fig. 2). In all cases, the total number of microfibers released using detergent solution or DI water increased with higher temperature. These increases were not found to be statistically significant at p-value < 0.05 in some cases (Fig. 2, B). Uncertainty on the effect of temperature on the microfiber generation during laundering has been reported, even at extended washing times (Hernandez et al., 2017; Napper and Thompson, 2016).

In general, the swelling action of water on cellulose-based fibers increases with temperature. The water acts as a plasticizer, lubricating and softening the amorphous regions of the cellulose fibers by the disruption of the hydrogen bonds between the cellulose chains. This causes a reduction in the glass transition temperature of the amorphous regions and a decrease in mechanical properties such as stiffness and resiliency in regenerated fibers such as rayon (Bryant and Walter, 1959). Essentially the increase of temperature promotes the swelling of cellulosic fibers, and as a consequence, the textile structure, generating

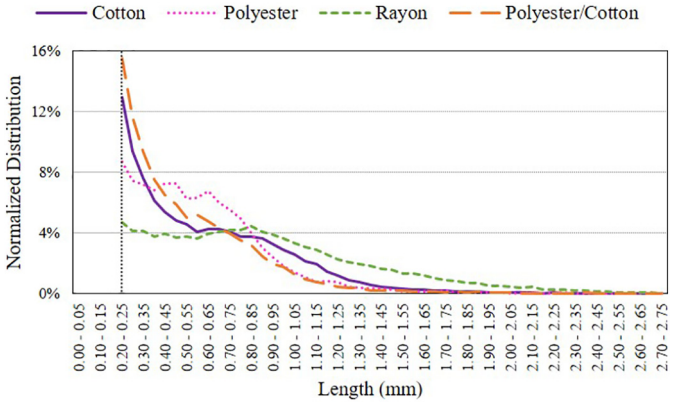

Fig. 3. Fiber Length Distribution of microfibers (T = 44 °C, without detergent). The normalized values are presented based on the percentage of total microfibers counted by the FQA within the size range, 2000 to 19,000 microfibers measured per sample. The vertical dotted lines indicate the FQA detection limit for fiber length.

free space for the mobilization of the broken fibers from the fabric construction. The swollen fabric structure generates expanded material expected to enhance fuzz formation due to the shear forces during washing. Polyester does not swell in water as much as cellulosic fibers and its relatively lower changes in microfiber generation at higher temperatures are in agreement with this (Bryant and Walter, 1959).

3.1.4. Fiber size distribution

The size distribution of the released microfibers collected was measured using the FQA. The FQA only measures the size distribution of fibers with length > 200 µm. The length distribution of the microfibers with length > 200 µm at 44 °C without detergent is presented in Fig. 3. Very few microfibers > 2.5 mm were detected. The length distribution of microfibers released by rayon fabrics was broader than the other fabrics tested. Cotton, polyester, and the blended (polyester/cotton) fabrics released a greater proportion of shorter microfibers than did the rayon.

The width or diameter of the microfibers released is different depending on the fabric material. Cotton and rayon microfibers are thicker than polyester microfibers, Fig. 4. The width distribution of the microfibers generated from polyester/cotton blended fabrics during laundering is bimodal as expected when the two types of microfibers are both being released in the washing solution. Similar size distributions were observed in the microfibers recovered from laundering of

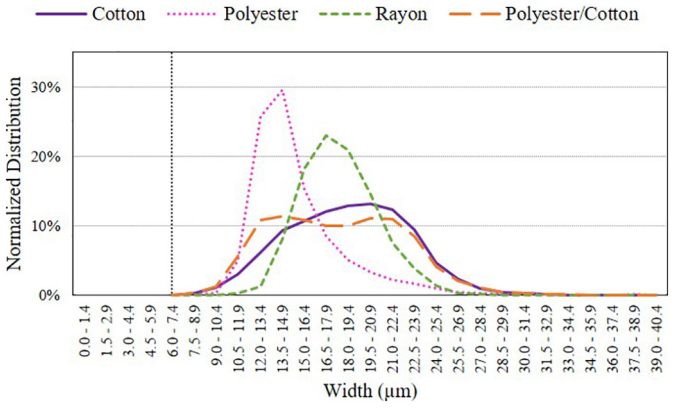

Fig. 4. Fiber Width Distribution (T = 44 °C, without detergent). The normalized values are presented based on the percentage of total microfibers counted by the FQA within the size range, 1000 to 15,000 microfibers measured per sample. The vertical dotted lines indicate the FQA detection limit for fiber length.

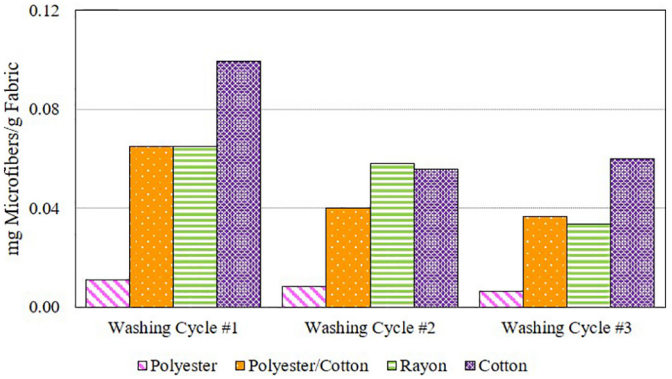

Fig. 5. Mass of microfibers generated during home laundering after subsequent washing per mass of fabric washed. The same fabric sample (N = 1) was washed and dried three times.

fabrics at different temperatures, with or without detergent.

3.2. Microfibers generation during home laundering

In agreement with the accelerated laundering experiments, all fabrics released microfibers with fabrics made of cellulose-based fibers (cotton and rayon) releasing more microfibers than polyester during laundering for all of the three cycles in series, Fig. 5.

There was a consistent decrease in the number of microfibers released with increasing washing cycles, but there was still very significant amounts of microfibers being released during the third cycle. This is in agreement with other studies showing the number of microfibers released during laundering is generally reduced with increased cycles until reaching a constant amount (Carney Almroth et al., 2018; Napper and Thompson, 2016; Pirc et al., 2016; Sillanpää and Sainio, 2017).

In the accelerated laundering experiments, more microfibers were generated per weight of fabric washed than in the home laundering experiments as a result of the more intense mechanical action of the metal balls; approximately forty times more mass was shed. There is general agreement between the accelerated and home laundering experiments for the relative amounts of microfibers shed for the different fabrics with a correlation coefficient of 0.8 (Fig. S19, Supplementary material). Thus, accelerated laboratory laundering can be used as a method to assess the relative difference in shedding capacity of fabrics in a simple, well-controlled way without the burdensome, labor-intensive efforts involved in using the home washing machine that is poorly suited for quantitative analysis. Home laundering machines utilize large quantities of water and have complex washing compartments and inlet and outlet piping and valves that make accurate recovery of microfibers difficult. In addition, the accelerated laundering experiments provide larger quantities of microfibers than home laundering that are easier to capture and measure gravimetrically.

Regarding the size of the microfibers (length > 200 μm), in the home laundering experiments, longer microfibers were obtained than in the accelerated laundering experiments, in agreement with the hypothesis that the lower intensity action during home laundering washing does not break off as many small microfibers (Fig. 6). In addition, for both the accelerated and home laundering experiments, rayon fabrics generated longer microfibers, followed by cotton, polyester/cotton, and polyester, respectively. In addition, SEM images showed that the shedded fibers were approximately the same width as the original fibers in the yarns in the fabrics prior to washing for both accelerated and home laundering washing (Figs. S20 and S21).

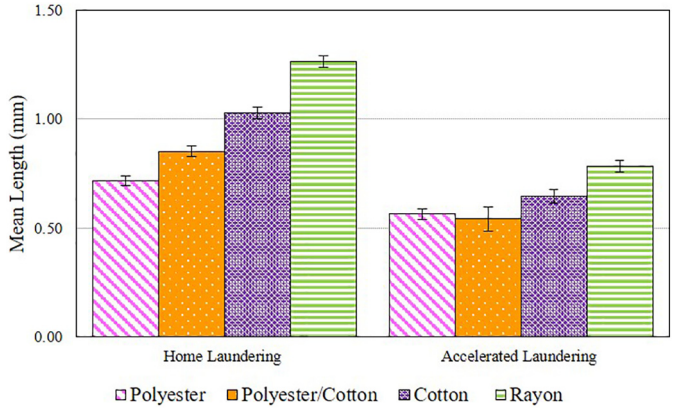

Fig. 6. Mean length of microfibers (> 200 μm) released during home laundering and accelerated laundering (T = 44 °C, with detergent). The mean values are presented based on the number of fibers measured by the FQA, 2000 to 5000 microfibers per sample. Error bars represent a 95% confidence level.

3.3. Predictive tests for the microfibers generation during laundering

Certainly, microfiber generation during laundering involves many factors such as fabric type and geometry (woven, knit, or nonwoven), yarn type (twist, evenness, hairiness, and number of fibers), processing history (spinning, knitting or weaving, scouring, bleaching, dyeing, finishing, and drying processes), and physicochemical properties of the fibers (Hernandez et al., 2017).

In this study, all the above-mentioned variables were the same except the physicochemical properties of the fibers in the yarns, thus, to understand why some knitted fabrics released more microfibers than others, the microfiber generation process during laundering was compared using the schematic model for pilling formation of fabrics. This process is composed of several steps: fuzz formation (a fiber with a loose end extending out of the fabric), swelling, fibrillation, pill formation (entanglement of the fibers), and pill wear off (Geology, 1966; Okubayashi et al., 2005; Okubayashi and Bechtold, 2005). In this case, it is proposed that the shedding capacity of the fabrics depends mainly on the fuzz formation step and how easy these fibers are broken by the mechanical action of the washing machine before forming the pill (Fig. 7). The breakage of the staple fibers is the main contribution to microfiber formation and release. In this study, most shed microfibers were < 2 mm long (Figs. 3 and 6) whereas the staple fibers are much longer. Generally, natural staple fibers are classified as short staple fibers (length between 25 and 60 mm) and long staple fibers (length > 60 mm) (Elhawary, 2015). For cotton fibers, the US Cotton Fiber Chart 2017/2018 reported the length for US cotton fibers ranging from 1.11 in (28.19 mm) to 1.17 in (29.72 mm) (Cotton Incorporated, n.d.). Additionally, synthetic or semi-synthetic fibers are generally uniformly cut in lengths higher than 32 mm for staple fiber applications (Barnet, n.d.).

The fuzz formation step depends on the friction, shape, thickness, stiffness, and abrasion resistance of the fibers (Geology, 1966; Okubayashi et al., 2005; Okubayashi and Bechtold, 2005). Therefore, to assess how easy the fuzz forms and the fibers break, some mechanical properties were evaluated: the abrasion resistance of the fabrics and the tensile breaking load and hairiness of the same yarn types used to construct these fabrics.

The tensile breaking load of the yarns is presented in Table 1 for the dry and wet states. Polyester, which does not swell significantly in water, showed similar breaking loads for both the wet and dry states. The polyester yarns presented significantly higher breaking load compared to the other yarns, explaining to some extent why polyester fabrics released fewer microfibers than the other fabrics studied.

In contrast to polyester, rayon and cotton yarns had different behaviors in the dry and wet states. In the wet state, rayon yarns were

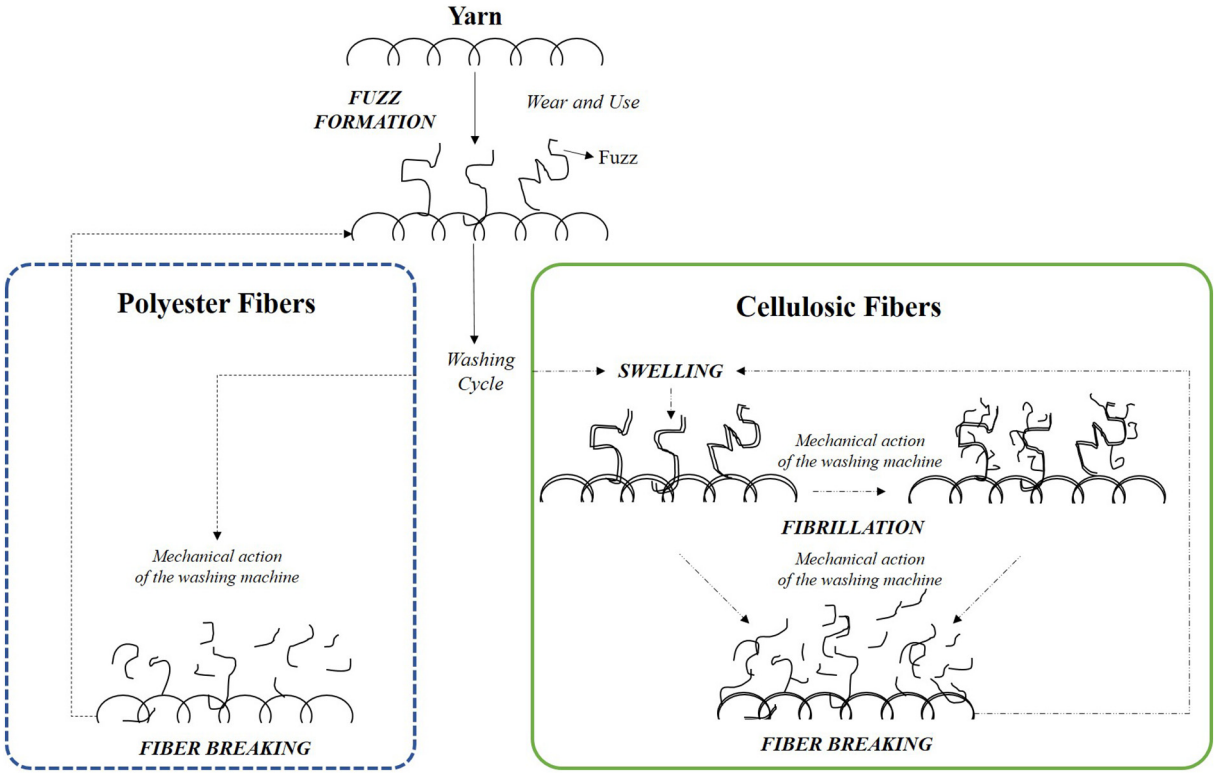

Fig. 7. Proposed mechanism for the microfibers release during laundering from fabrics and yarns made of polyester fibers (blue dashed line, left) and cellulosic fibers (green solid line, right). The loose fibers (fuzz) come out of the textile structure during wear and use (FUZZ FORMATION). Then, these fibers are broken in the washing cycle by the mechanical action of the washing machine, FIBER BREAKING. In the presence of water and detergent, cellulosic fibers swell (SWELLING), due to the mechanical action of the washing cycle these fibers could also be fibrillated (FIBRILLATION) and broken (FIBER BREAKING). Adapted from Okubayashi et al., 2005 and Okubayashi and Bechtold, 2005. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

weaker while cotton yarns were stronger compared against the respective dry yarns. In fact, cotton fibers have higher crystallinity and molecular weight than rayon fibers. The cellulose chains in the non-crystalline regions of the cotton fibers tend to rearrange and orient if a force is applied increasing their tensile strength (Hajime et al., 2000).

Additionally, the crystallites in the cotton in the wet state are maintained as a reinforcing component. In rayon, there is a lower crystallinity and thus a lower wet strength of the yarns. According to the results of the tensile tests, cotton yarns were significantly stronger than rayon yarns when wet. However, the microfibers generation of the cotton and rayon fabrics were similar and not predicted by differences in the tensile strength results.

Other parameters to consider are the hairiness and evenness of the yarns which are critical measurements for the quality of the yarn and also affect the microfiber generation during laundering. The characteristics of the yarns used in this study are presented in Table S2 (Supplementary material). They have similar yarn count and twist; the main differences are due to their strength properties, discussed above, and their evenness and hairiness.

Evenness is a measure of how uniform the yarn is in terms of mass and cross-section variation per unit length (ASTM D1425/D1425M—14, 2014; Hasler and Honegger, 1954; Slater, 1986; Soares et al., 2008). Higher coefficient of variation percents are an indication of uneveness, poor appearance and quality and lower yarn strength. In this case, the yarn samples have similar evenness except for the 50/50 polyester/cotton yarn that has a higher coefficient of variation.

Moreover, hairiness is a measurement of the amount of free fibers loops and ends protruding from the yarn surface at a certain distance. High hairiness affects the appearance of textiles products and increases the surface friction of yarn and fabrics and their pilling tendency, therefore, it can affect the microfibers generation during laundering (Barella, 1957, 1983; Krupincová and Meloun, 2013; Majumdar, 2010; Manich and Barella, 1997; Yuvaraj and Nayar, 2012). The hairiness depends on several factors such as the twisting levels and the diameter

Table 1
Breaking Load of Spun Yarns measured by the single-strand method (ASTM D2256/D2256M − 10, 2015).

Sample	Break load (N)				Strain at break (%)			
	Dry state N = 15		Wet state N = 10		Dry state N = 15		Wet state N = 10	
	Mean	Std Dev	Mean	Std Dev	Mean	Std Dev	Mean	Std Dev
Cotton yarns dry state	1.8053	0.1655	2.2742	0.1912	5.6944	0.554	8.6700	0.7837
Polyester yarns dry state	4.3364	0.3176	4.4316	0.1800	10.7539	0.5252	10.5975	0.6643
Polyester/cotton yarns dry state	2.2028	0.1414	2.2804	0.2320	13.1837	0.9946	14.1328	0.9308
Rayon yarns dry state	1.7077	0.1113	0.9884	0.1097	15.3462	1.1053	15.3316	1.7243

SD – Standard Deviation.

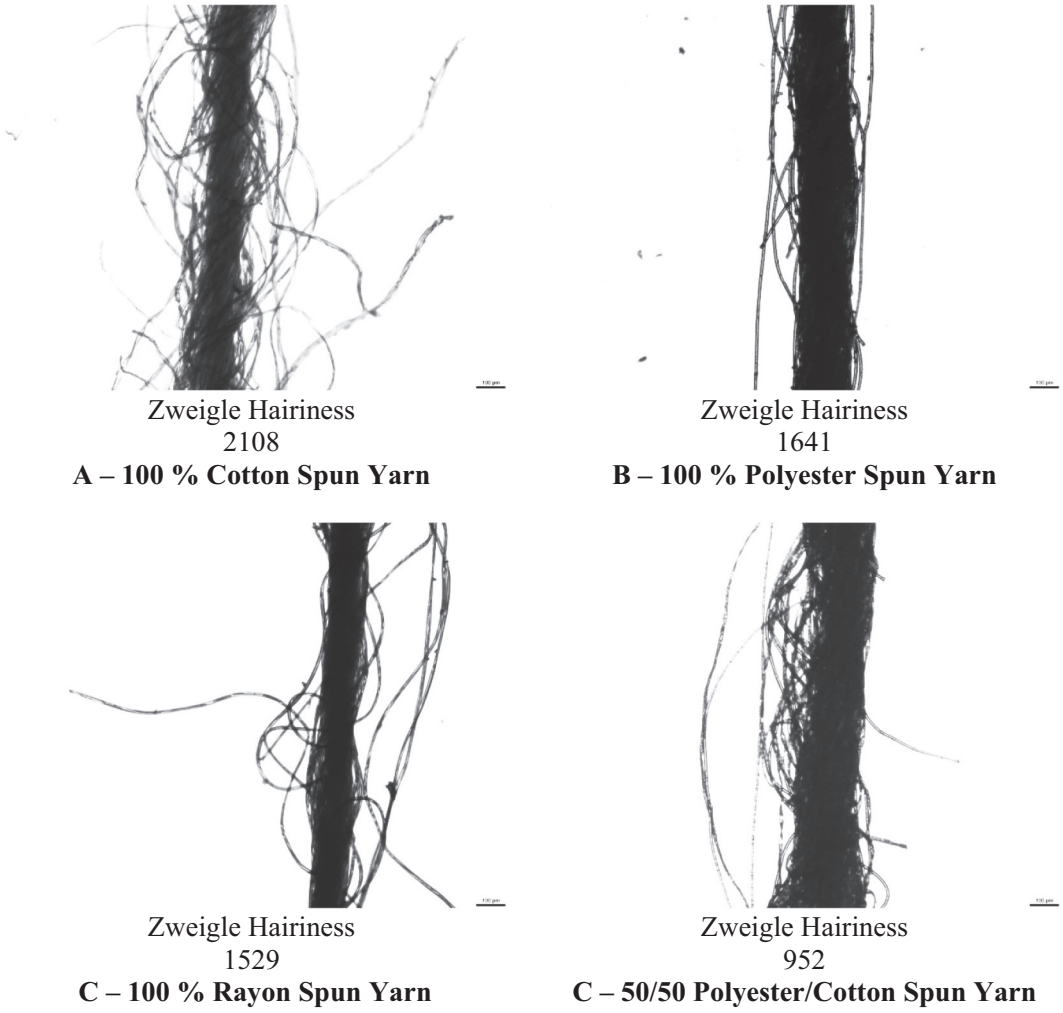

Fig. 8. Selected optical microscope images of the spun yarns. The Zweigle hairiness data for each yarn is indicated.

of the yarn, and the rigidity, elongation, tenacity, and length of the fibers (Barella, 1957, 1983; Majumdar, 2010).

The hairiness measurement also provides an indication of which yarns have more tendency to form fuzz on the surface. The Zweigle Hairiness parameter indicates the number of fibers protruding 3 mm or more from the yarn surface in 100 m of yarn (Krupincová and Meloun, 2013). The hairiness of the yarns used to knit the fabrics in this study is presented in Fig. 8. In decreasing order, 100% cotton spun yarn has higher hairiness followed by 100% polyester spun yarns, 100% rayon spun yarns, and 50/50 polyester/cotton spun yarns. In general terms, it can be said that the similarity in microfibers generation between rayon and cotton fibers may be related to the high hairiness of cotton and low tensile strength of rayon yarns. Polyester yarns have relatively similar hairiness to rayon but the fibers are much stronger, which does not generate a large number of microfibers during laundering. In the case of 50/50 polyester/cotton, the hairiness is low but the variation in evenness is considerably high which can lead to poor abrasion resistance.

Moreover, the abrasion resistance of the knitted fabrics was studied using the Martindale Abrasion Tester. The weight loss of the fabrics was determined after 20,000 cycles of the wool abrasion fabric scraping the surface of the fabrics, reflecting the resistance of the fabric to shed mass under the mechanical action of the washing and drying cycles. The results presented in Table 2 show that rayon knitted fabrics are less resistant to abrasion followed by cotton, polyester/cotton and polyester knitted fabrics with a significant difference between them.

The breaking load of the yarns and the abrasion resistance of the fabrics in this study correlate with the number of microfibers released during laundering (Figs. 9 and 10). These correlations exist for all conditions but at different levels (Figs. S22 and S23).

Table 2
Abrasion resistance of knitted fabrics measured as the weight loss of the fabrics by the action of the Martindale Abrasion Tester (ASTM D4966 – 12, 1992).

Sample	Weight loss	SD
Rayon knitted fabrics	5.63%	0.28
Cotton knitted fabrics	3.34%	0.22
Polyester/cotton knitted fabrics	1.63%	0.12
Polyester knitted fabrics	0.11%	0.13

SD – Standard Deviation, N = 3.

In summary, it is expected that fabrics with higher abrasion resistance, low hairiness, and higher yarn breaking strength would have a lower tendency to form fuzz and/or release microfibers during the mechanical action of washing.

3.4. Aquatic biodegradation of yarns

The aquatic biodegradability of the microfibers shed in the environment via home laundering provides a good indication of their persistence in the environment. The aquatic biodegradability was studied following the ISO standard method for ultimate aerobic biodegradability of plastic materials in aqueous mediums (ISO 14851:1999,

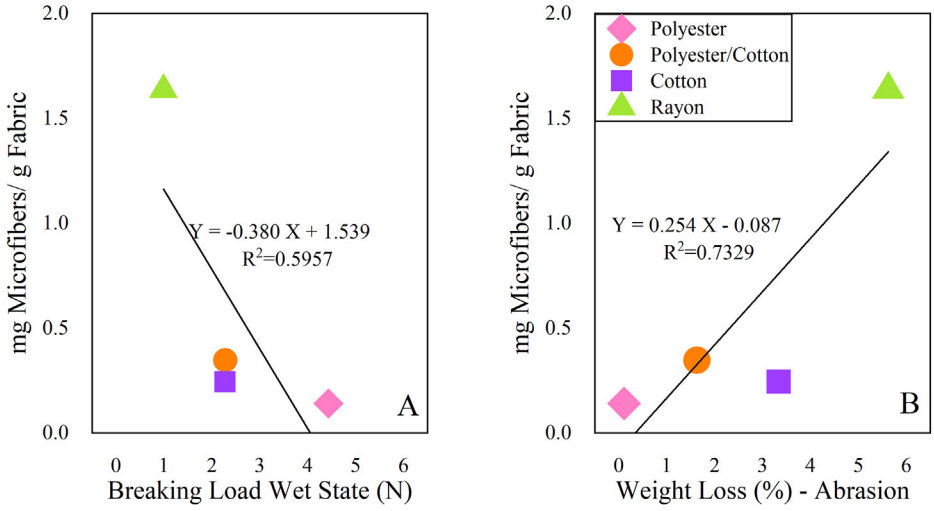

Fig. 9. Mass of microfibers generated during accelerated laundering (T = 25 °C, without detergent) vs A - Breaking Load of Yarns in Wet State and B - Weight Loss of the fabrics after 20,000 cycles in the Martindale abrasion tester. Mean values are shown for all the variables.

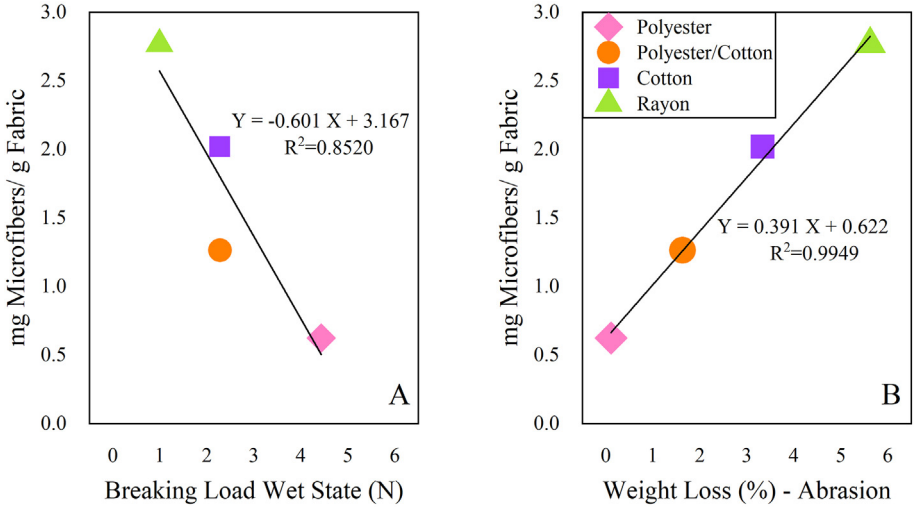

Fig. 10. Mass of microfibers generated during accelerated laundering (T = 25 °C, with detergent) vs A - Breaking Load of Yarns in Wet State and B - Weight Loss of the fabrics after 20,000 cycles in the Martindale abrasion tester. Mean values are shown for all the variables.

2005) using the yarns used to knit the fabrics and a low concentration of activated sludge solids to simulate natural aquatic environments. The yarns have fibers with similar width to the shedded microfibers during laundering and as such are reasonable materials to evaluate for relative biodegradability. The percentage of biodegradation was measured comparing the oxygen uptake of the system with the theoretical oxygen demand ThOD calculated according to the total organic carbon in each sample. The nitrification interferences during the experiments were not significant compared to the oxygen consumption due to biodegradation of the polymeric materials studied. At the end of the experiment, the reference material (microcrystalline cellulose) reached 84% degradation (Fig. 11) indicating the inoculum was working and the BOD of the blank was 12 mg/L, it did not exceed 60 mg/L, as required by a valid experiment (ISO 14851:1999, 2005).

The biodegradation curves of the materials over the 243 days are shown in Fig. 11. Only the polyester material had fully reached the plateau phase; the other materials including the microcrystalline cellulose were continuing to consume oxygen, albeit a slow rate towards the end of the experiment (Fig. 11). The percentage of biodegradation after the 243 days, subtracting the nitrification interferences (oxygen consumed in nitrification reactions), was 75.90 ± 12.35% for cotton yarns, 62.21 ± 13.29% for rayon yarns, 39.76 ± 3.52% for 50/50 Polyester/Cotton Yarns, and 4.05 ± 0.75% for polyester yarns. The results are as expected, the cellulose-based materials are a readily available source of carbon for the microorganisms. The polyester, on the other hand, being hydrophobic and non-sugar based is not degraded by the microorganisms (Li et al., 2010).

There is no pass/fail criteria for aquatic biodegradability in the ISO method 14851 and it is generally understood that there does not exist a clear biodegradability threshold to certify a material as biodegradable in aquatic and marine environments (European Bioplastics, 2018). Nevertheless, the tests were performed under valid conditions based on the method criteria, 60% of degradation in the biodegradable reference material (microcrystalline cellulose) and < 60 mg/L of oxygen consumption occurred at the end of the test (ISO 14851:1999, 2005).

The withdrawn standard for non-floating biodegradable plastics in the marine environment (ASTM D7081-05, 2005) specified that to consider a plastic product biodegradable in marine environments > 30% of the organic carbon must be converted to carbon dioxide (CO_2) within 180 days at 30 °C with satisfactory disintegration after 12 weeks and 90% of biodegradation in an active environment such as composting. However, until now there is no replacement for this standard clarifying these requirements but clearly in this experiment for all materials, except for polyester, > 30% of the carbon was converted to

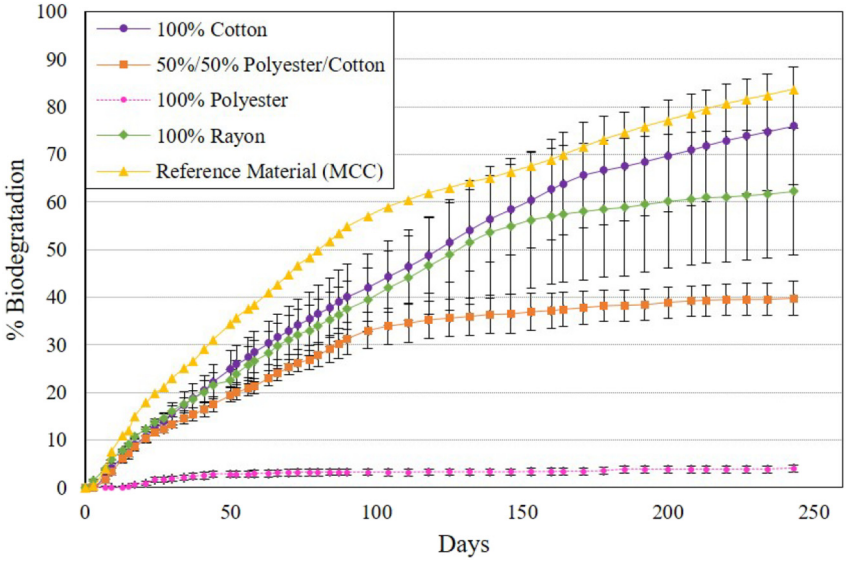

Fig. 11. Biodegradation curves of the textile yarns used to knit the fabrics used for the laundering experiments. The percentage of biodegradation is based on the oxygen uptake versus the theoretical oxygen demand. The error bars represent the standard error (n = 3 for all materials, except MCC with n = 1).

CO_2 (Fig. 11).

With respect to the biodegradation of cellulosic fibers, rayon is expected to have a higher biodegradability than cotton due to the differences in crystallinity, orientation and moisture regain of these fibers; rayon has lower crystallinity and higher moisture regain than cotton due to the antiparallel configuration of cellulose II within the rayon fiber structure (Battista, 1950; Howsman, 1949; Niu et al., 2012; Park et al., 2004). The amorphous regions are easier to degrade, chemically and enzymatically than crystalline regions in polymeric materials (Mochizuki and Hirami, 1997). Nevertheless, in this study, the difference in the biodegradation percentage between the three types of cellulosic materials was not significant. In the cellulosic samples, the variability of the data could be related to the changes in the levels of oxygen in the flasks that affect the behavior of the microorganisms. The oxygen supply was not constant during the experiment, every week or as needed the bottles were saturated with oxygen for 15 min using an aquarium pump and air stones to diffuse the air in the test medium to reach around 8 mg/L of dissolved oxygen. The differences in the oxygen levels represent the oxygen consumed during the biodegradation process to convert the organic carbon to CO_2.

The curves in Fig. 11 indicate that polyester and polyester/cotton yarns had reached more of a plateau than did microcrystalline cellulose, rayon, and cotton. In addition, 50/50 polyester/cotton yarns reached a plateau phase at about 40% of biodegradation, as would be expected from a weighted average of the polyester and cotton degradation results. As expected, only the cotton in the blend degraded during the experiment, confirmed by FTIR (section S 2.4.4, Supplementary material).

Polyester yarns did not degrade under the test conditions. The biodegradation of polyester and cotton fabrics have also been tested under the action of natural soil under aerobic laboratory controlled conditions and a large scale composting facility (Li et al., 2010). After 90 days in natural soil, cotton fabrics without finishing agents achieved around 23% of biodegradation and polyester fabrics only 13% (Li et al., 2010). In composting, the cotton fabrics weight loss was 55% and polyester fabrics only 20% in 90 days (Li et al., 2010). Moreover, the normal microbial communities in the environment do not attach to polyester (Bajpai et al., 2011). It has been shown that bacterial adherence in polyester fabrics was low compared to cotton and polyester/cotton blended fabrics (Bajpai et al., 2011). In addition, polyester is less susceptible to fragmentation of the structure by hydrolysis due to its high hydrophobicity and low moisture regain (Fuzek, 1985; Li et al., 2010).

Herein, cotton spun yarns were completely disintegrated during biodegradation. The rayon sample showed evidence of some partially degraded small fibers in the suspension at the end of the experiment. In the case of polyester, the yarns did not undergo significant changes. The 50/50 cotton/polyester blended yarn shows no intact cotton fibers after biodegradation, in agreement with the FTIR results. Images of the residual solids and initial yarns are shown in the Supplementary material (Figs. S29 and S30).

Despite the variation in the biodegradation data, these results are in line with the behavior expected, that cellulose-based fibers are biodegradable. Cellulose-based fabrics release more microfibers in laundering but they degrade readily in aerobic aquatic environments. More research is needed to understand the flow of microfibers and improve the removal efficiency in the WWTPs.

3.5. Environmental implications of microfibers from laundering

The efficiency of WWTPs in microplastics removal has been reported to be higher than 98% (Lares et al., 2018; Magnusson and Norén, 2014; Talvitie et al., 2015). The microplastics are mainly retained in the sewage sludge, nevertheless, most of these results were reported with microplastics > 250 μm being analyzed (Lares et al., 2018; Magnusson and Norén, 2014), only Talvitie et al. (2015) evaluated particles and fibers > 20 μm in size.

Most of the microplastics (particles and fibers) are removed in primary and secondary treatment (clarification, sedimentation, and aerobic bioreactors or activated sludge treatment) (Talvitie et al., 2017b, 2015). Traditional tertiary treatments such as gravity sand filters, or biological filtration are very efficient to further remove microplastics after secondary treatment (Carr et al., 2016; Mintenig et al., 2017; Talvitie et al., 2017b, 2015). In addition, advanced technologies such as membrane bioreactors (0.4 μm pore size), rapid sand filters, dissolved air flotation, and disc filters as tertiary treatments have the potential to trap up to 99.9% of the microplastics escaping the secondary treatment (Lares et al., 2018; Talvitie et al., 2017a). Nevertheless, WWTPs are not designed to capture microplastics and these advanced technologies are still not implemented in most of the processes.

Without a doubt, WWTPs are a route for microplastics entering aquatic environments. Despite the high removal efficiency, the high volume of effluents discharged constantly to aquatic environments

contribute significantly to the microplastic pollution problem. Talvitie et al. (2017b) estimated, on average, 1.97×10^8 "microlitter" particles per day are discharged into the Baltic Sea from wastewater effluent. Taking into consideration the annual discharges of 12 WWTPs in Lower Saxony (Germany), Mintenig et al. (2017) predicted a total discharge of 9×10^7 to 4×10^9 microplastics (particles and fibers) per WWTP annually. In the US, Mason et al. (2016) estimated that the daily discharges from a municipal WWTP ranged from around 50,000 up to nearly 15 million particles. Likewise, Lares et al. (2018) forecasted that approximately 1.0×10^7 and 4.6×10^8 microplastics were discharged daily with the final effluent and digested sludge from the Kenklaveronniemi WWTP (Finland), respectively. These numbers vary depending on the location, type of effluents, demand and capacity, the population in the area, weather conditions, and type of treatment. In addition, there is a lack of standardization on the methods used to collect and identify these microplastics from the different points in the wastewater treatment process that could cause over or underestimation of these values. Another problem that needs to be considered is the transfer of microplastics to agricultural soils by the use of WWTPs sludge as fertilizer. In Europe, it was estimated that 63,000–430,000 tons of microplastics are applied to farmlands per year (Nizzetto et al., 2016). The consequences in agricultural sustainability and human health of this activity are also not well understood (Nizzetto et al., 2016).

In some wastewater effluents, microfibers represent an important proportion of the microplastics found in the wastewater effluents (Lares et al., 2018; Mason et al., 2016; Mintenig et al., 2017; Talvitie et al., 2015, 2017a, 2017b). In general, it has been reported that these microfibers are predominantly made of polyester, and more specifically, polyethylene terephthalate (PET) (Lares et al., 2018; Mintenig et al., 2017; Talvitie et al., 2017a; Wolff et al., 2018; Ziajahromi et al., 2017).

In most of the WWTP assessments on microplastics, the removal efficiency of cellulose-based fibers has been ignored. However, cellulose-based fibers have been observed in the effluents of WWTPs (Lares et al., 2018; Talvitie et al., 2017b, 2015; Ziajahromi et al., 2017). Talvitie et al. (2017b) found that 30% of the microlitter collected from the wastewater effluent was fibers and from that 66% was comprised of cellulose-based fibers and 33% of the fibers were polyester fibers (PET).

Moreover, in this study, we observed that around 50% of the microfibers released during laundering were below 200 μm in size in all the fabric types evaluated and that these particles still have the potential to escape the wastewater treatment process. The smallest size fractions are the most common microplastics (particles and fibers) observed in the effluents of the WWTPs (10 μm – 100 μm) (Mintenig et al., 2017; Talvitie et al., 2017a, 2017b; Wolff et al., 2018). Nevertheless, another study that analyzes larger microplastics (> 250 μm), including fibers, also reported that > 50% of the microplastic recovered in the effluents were smaller than 1 mm (Lares et al., 2018). In addition, Wolff et al. (2018) reported that the microfibers recovered from the WWTP effluents ranged between 100 μm and 1000 μm in length similar to the results herein (Fig. 4).

Similarly, many studies that have been focused on the ecotoxicity of micro and nanoplastics in marine and freshwater ecosystems overlap with the size of the microfibers observed in our study, nevertheless cellulose-based fibers were generally not considered (Chae and An, 2017). These studies observed various effects on the growth, development, behavior, reproduction, and mortality of aquatic animals (Chae and An, 2017). However, there is a lot of uncertainty about the significance of these toxicity studies due to (1) the differences between simulated lab conditions and environmentally relevant concentrations of micro and nanoplastics, (2) the unknown long-term effects of these small plastics on aquatic organisms, and (3) the possible effects on human health from trophic transfer of micro and nanoplastics through the food chain (Burton, 2017; Chae and An, 2017). Kosuth et al. (2018) estimated that the average person ingests 5,800 synthetic particles, such as synthetic microfibers, a year from tap water, beer, and sea salt alone. However, only one study examined microplastics in stool samples (Schwabl, 2018) with particles between 50 and 500 μm in size observed in all the samples evaluated, 18 to 172 particles per 10 g of stool. No fibers were reported, however, the microplastics were mainly comprised of polypropylene (62.8%) and polyethylene terephthalate (17%) (Schwabl, 2018).

The sorption mechanisms between microplastics/microfibers and chemicals are controlled by the physical-chemical properties of the materials, the nature of chemicals, temperature, and solution chemistry (pH, salinity, etc.) (Wang et al., 2018). Synthetic microfibers such as polyester being low surface energy materials will interact more strongly with non-polar organic chemicals in a stronger way than will cellulose-based microfibers. Natural based fibers have distinct surface chemistries and have a different role in the environment; they may adsorb different chemicals than do synthetic fibers (Grancaric et al., 2005). It is therefore important to study microfibers throughout their life cycle, including both the generation and environmental fate of these materials.

4. Conclusions

The shedding mechanism of microfibers from textiles and their biodegradability are fundamental to understand how to reduce the presence of these fibers in aquatic environments. In summary, all fiber types released significant amounts of microfibers during laundering, however, cellulose-based fabrics released more microfibers than polyester with the same fabric structures. The fiber and yarn physicochemical properties play a major role in the microfiber generation. Fabrics with higher abrasion resistance, low hairiness, and higher yarn breaking strength have a lower tendency to form fuzz and/or release microfibers during the mechanical action of washing. From the laundering parameters studied, the use of detergent increases the generation of microfibers, the surfactant promotes the mobilization process of fibers from the fabric to the washing solution. In general, accelerated laundering experiments results correlated with home laundering experiments but accelerated laundering experiments were easier to control and had better sensitivity and repeatability. Despite the fact that the cellulose-based fabrics shed more microfibers than the polyester fabric, cotton and rayon fibers degrade in aquatic conditions whereas polyester fibers do not and are expected to persist in the environment for a long time. More research is needed to understand the fate of textile microfibers and their additives in the environment.

Notes

The authors declare no competing financial interest.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.marpolbul.2019.02.062.

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