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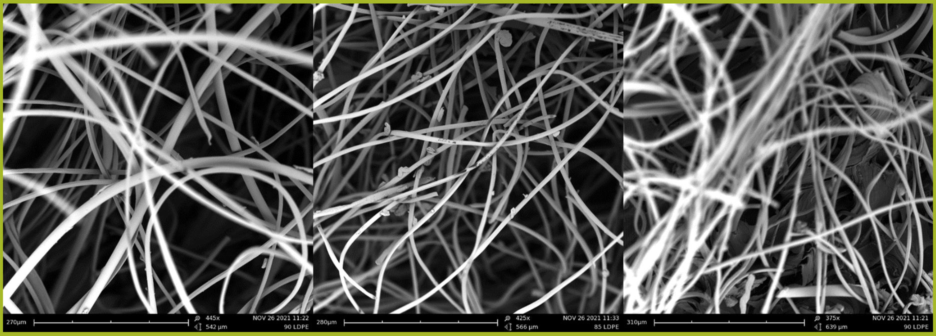
Microplastics and microfibers pollution: study of their environmental issues and evaluation of reduction alternatives



Francisco Belzagui Elder

UNIVERSITAT POLITÈCNICA DE CATALUNYA
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DOCTORATS
INDUSTRIALS

PhD program in Environmental Engineering

Microplastics and microfibers pollution: study of their environmental issues and evaluation of reduction alternatives

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Terrassa, 2022

Francisco Belzagui, *in memoriam*

This work is the compilation of the research carried out by my Ph.D. student, Francisco Belzagui Elder, during his pre-doctoral stage in the Environmental Engineering research group (ENMA) of the Institute of Textile Research and Industrial Cooperation of Terrassa (INTEXTER) that belong to the Polytechnic University of Catalonia (UPC).

Francisco Belzagui obtained an AGAUR grant for an industrial doctorate in 2018, co-founded by INDITEX since January 2019. He carried out his research for more than 3 years, until January 2022. He planned to deposit his thesis in February of this year and defend it in April.

Unfortunately, he died a few days before the deposit could be made, leaving all the thesis research complete. He published 3 articles in high-impact scientific journals (Q1 of the JCR-WoS), he applied for a Spanish patent and wrote 3 more technical articles, which are currently being reviewed for publication. To complete the thesis document, he only needed to write the acknowledgments, the summary and the general conclusions.

As advisor of the thesis, I have completed these three sections that were missing. In the acknowledgments, I have tried to name the people who I believe have contributed to the development of this thesis. With respect to the summary, I have written it based on the summaries of each thesis publication (6 papers and one patent). Finally, in the conclusions section, I have compiled the conclusions of all the publications. I understand that in this way these two sections (summary and conclusions) include exactly the writing and data that he had already provided in each of part of the thesis.

To end this introductory note, I want to convey to the reader the profound pain I feel for the loss of a person so dear and close to me, with whom I have collaborated very closely during the last 3 years. Francisco was a tireless researcher, intelligent, creative, with a great desire to excel and with a lot of initiative. Our university has lost a great researcher, our research group has lost a good colleague and I have lost a brilliant collaborator and also a friend.

I dedicate this compilation work of Francisco's pre-doctoral research to his beloved family, especially Claudia and Verónika, and also to his friend Aroa. But above all, I want it to be a tribute to himself, to Francisco, in gratitude for everything he contributed and shared with me during these 3 years. R.I.P.

The Ph. D. thesis advisor

Francisco Belzagui, *in memoriam*

Este trabajo es la recopilación de la investigación efectuada por mi doctorando, Francisco Belzagui Elder, durante su etapa predoctoral en el grupo de investigación de Ingeniería del Medio Ambiente (ENMA) del Instituto de Investigación Textil y Cooperación Industrial de Terrassa (INTEXTER) de la "Universitat Politècnica de Catalunya" (UPC).

Francisco Belzagui obtuvo una beca AGAUR de doctorado industrial en 2018, cofinanciada por INDITEX desde enero de 2019. Desarrolló sus investigaciones durante más de 3 años, hasta enero de 2022. Tenía previsto depositar su tesis en febrero de este año y defenderla en el mes de abril.

Desafortunadamente, falleció unos días antes de poder hacer el depósito, dejando toda la investigación de la tesis finalizada. Publicó 3 artículos en revistas científicas de alto impacto (Q1 del JCR-WoS), solicitó una patente española y redactó 3 artículos técnicos más, que están actualmente en fase de revisión por las editoriales para su publicación. Para completar el documento de la tesis, únicamente le faltó redactar los agradecimientos, el resumen y las conclusiones generales.

Como directora de la tesis, he completado estos tres apartados que faltaban. En los agradecimientos, he intentado nombrar a las personas que yo creo que de una manera u otra han contribuido a desarrollar esta tesis. En cuanto al resumen, lo he redactado tomando como base los resúmenes de cada una de las publicaciones de la tesis (6 artículos y una patente). Finalmente, en el apartado de conclusiones, he recopilado las conclusiones de todas las publicaciones. Entiendo que de esta manera lo que yo incluyo en estos dos apartados (resumen y conclusiones) son exactamente los datos que él ya había aportado en cada una de las partes de la tesis.

Para finalizar esta nota introductoria, quiero transmitir al lector el profundo dolor que siento por la pérdida de una persona tan querida y tan cercana, con quien he colaborado muy estrechamente durante estos 3 últimos años. Francisco era un investigador incansable, inteligente, creativo, con un gran afán de superación y con mucha iniciativa. Nuestra universidad ha perdido un gran investigador, nuestro grupo de investigación ha perdido a un buen compañero y yo he perdido a un colaborador brillante y también a un amigo.

Este trabajo de recopilación de las investigaciones predoctorales de Francisco se lo dedico a su querida familia, en especial a Claudia y a Verónika, y también a su amiga Aroa. Pero, sobre todo, quiero que sea un homenaje a él mismo, a Francisco, en agradecimiento por todo lo que aportó y compartió conmigo durante estos 3 años. D.E.P.

La directora de tesis

AGRADECIMIENTOS

Quisiera que estos agradecimientos empezaran de la manera típica: el doctorando agradeciendo a la directora de la tesis su labor como tutora de la misma… Quisiera que empezaran así porque esto significaría que los habría escrito Francisco. Pero desafortunadamente, él ya no está aquí para escribirlos, así que, como directora de esta tesis, voy a intentar asumir ese papel.

En primer lugar, agradecemos al Profesor Martí Crespi que planteara el tema de tesis centrado en la problemática de las microfibras textiles y las gestiones efectuadas con INDITEX para conseguir el doctorado industrial. Y también a la codirectora de esta tesis, Mercè Vilaseca, que tuteló a Francisco desde el inicio de la tesis hasta su jubilación.

Gracias al equipo de Sostenibilidad de INDITEX, a Antonio Álvarez, Luis Coloma y Héctor Alonso, por haber aportado la visión de la industria textil a esta tesis y por haber financiado el contrato de Francisco durante toda su etapa de doctorado industrial.

Gracias a nuestros compañeros del laboratorio de Control de la Contaminación: Montse, Bettina, Valentina y Victor. Todos han contribuido en distintas partes de esta tesis y han hecho posible que este trabajo se realizara. También gracias a Beatriz, por codirigirle su trabajo de la ETAF.

Gracias a Fernando Carrillo por aconsejarle, orientarle y permitirle usar sus equipos para obtener los carriers, así como a todo el personal del laboratorio de tintorería del INTEXTER, en especial a Reme por responder pacientemente a todas sus consultas.

Gracias a Manel Lis y al personal del laboratorio de tensioactivos por facilitarle el uso de sus instalaciones. Al director del INTEXTER, Enric Carrera, y a Francesc Cano, por sus explicaciones sobre la constitución de los hilos.

Gracias a Eva Villalón, Gerard Margalef y Margalida Seguí por orientarle sobre las patentes.

Pero, sobre todo, gracias a la toda la familia de Francisco, en especial a sus padres, Claudia y Roberto, por haberle dado la oportunidad de desarrollar sus proyectos y por haberle brindado siempre su apoyo incondicional; a sus hermanos Verónika, Pablo y Daniel, con quienes se sentía muy unido, por los buenos momentos que le proporcionaron; a su adorada sobrina Antonella, por alegrarle la redacción de los artículos; a sus abuelos de Bolivia: la abuela Baby que le apoyó en cada paso que dio y el abuelo Eduardo, que fue su referente, guía y fuente de inspiración.

También gracias a sus amigos Aroa y Jordi, y a todos los que creyeron en él y le animaron.

Finalmente, gracias a todas las personas que no he nombrado pero que de una manera u otra han contribuido al desarrollo de este trabajo.

Como dice Mafalda (de quien Francisco era gran admirador) ….

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Model ADT 1

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ABSTRACT

The microplastics (MPs) are small fragments (length < 5 mm) of non-chemically modified and/or non-biodegradable polymers. Rough estimations point that there are between 15 to 51 trillion buoyant items of MPs in marine environments and 14 million tons in the top 9 cm of sediments of the world's oceans.

According to the "European Chemical Agency", chemically treated and/or non-biodegradable textile microfibers (MFs) are a type of microplastics with a length to diameter ratio higher than 3 mm and a maximum length of 15 mm. One of the most renowned sources of MFs are those detached from every cycle of a textile article laundering. These are considered one of the most environmentally threatening pollutants as they have a continuous and cumulative entrance to the environment. Due to their small diameter they are more prone to be ingested by organisms, so high concentrations have been found in products for human consumption, as shellfish and tap water.

In this research, the microfibers' detachment rates of finished garments were evaluated. Results showed that MFs detachment ranges between 175 to 560 MFs/g or 30'000 to 465'000 MFs/m2 of garment. In addition, there was a high correlation between the MFs detachment and the textile article superficial density.

As there are still no accurate models to assess the MFs pollution, in this thesis a method to estimate the mass flow of MFs detached from household laundry that reaches aquatic environments was developed. The method considered the following parameters: (1) the detachment rate of microfibers from different textile garments, (2) the volume of laundry effluents, (3) the percentage of municipal water that is treated, (4) the type of used-water treatment applied, and, (5) the proportion of front- versus top-loading washing machines. In this way, 0.28 million tons of microfibers per year were estimated to reach aquatic environments. Moreover, hypothetical situations were simulated to evaluate the reduction of microfibers by modifying some parameters at different levels (consumer, government entities, and industry). It was found that depending on the implanted alternatives, microfibers that reach the aquatic environments could be reduced between 30% to 65%.

On the other hand, the current status of MFs as pollutants is reviewed, discussing possible alternatives from the manufacturing until the final disposition of MFs. There are many alternatives to reduce these pollutants but also gaps that need to be addressed. Some viable solutions to retain them are currently on the market. However, until this thesis was over, there was no single proposal on the destination of the retained MFs.

Hence, in this research a filter has been developed to retain the MFs and a proposal to treat the retained MFs was evaluated, following the circular economy philosophy. Both processes were patented.

The developed filtering system is totally made of recycled polymers. Its performance is higher than 97% of MFs' removal from the washers' effluents with a replacement time for the cartridge from 30 to more than 40 washing cycles. The retained MFs are subsequently immobilized in a polymeric matrix, turning them into a composite. Different proportions of polyester MFs were mixed with low-density polyethylene for immobilization of MFs. Results showed that the optimum composition, which improved some of the tensile mechanical properties, was 10% polyester MFs in the polymeric matrix.

Finally, other sources of MFs were studied, specifically, cigarette butts. These contains the smoked filters (SF) and unsmoked rests of tobacco. SFs are hazardous debris composed of > 15'000 strands that can be detached as MFs. Their detachment rate, acute aquatic toxicity, and the aquatic-, thermooxidative-, and photo-degradability were evaluated. It was found that SFs detach approximately 100 small MFs (< 0.2 mm) per day. About 0.3 million tons of potential MFs might be annually reaching aquatic environments from this source. A significant difference of eco-toxicity and a low degradability rate was found when MFs are present in the leachate generated by the SFs. This implies that MFs from SFs constitute an important source of microplastics, which might partially explain the high concentration of artificial polymers found in the deep-sea sediments.

RESUMEN

Los microplásticos (MPs) son pequeños fragmentos (longitud < 5 mm) de polímeros no modificados químicamente y/o no biodegradables. Las estimaciones aproximadas apuntan a que hay entre 15 y 51 billones de elementos flotantes de MPs en ambientes marinos y 14 millones de toneladas en los 9 cm superiores de sedimentos de los océanos del mundo.

Según la "European Chemical Agency", las microfibras textiles (MFs) tratadas químicamente y/o no biodegradables son un tipo de microplásticos con una relación longitud/diámetro superior a 3 mm y una longitud máxima de 15 mm. Una de las fuentes más conocidas de MFs son las que se desprenden en cada ciclo de lavado de un artículo textil. Estos son considerados uno de los contaminantes más amenazantes para el medio ambiente ya que tienen una entrada continua y acumulativa. Debido a su pequeño diámetro son más propensos a ser ingeridos por organismos, por lo que se han encontrado altas concentraciones en productos para consumo humano, como mariscos y agua corriente.

En esta investigación se evaluaron las tasas de desprendimiento de microfibras de prendas terminadas. Los resultados mostraron que el desprendimiento de MFs oscila entre 175 y 560 MFs/g o entre 30.000 y 465.000 MFs/m^2 de prenda. Además, se encontró una alta correlación entre el desprendimiento de MFs y la densidad superficial del artículo textil.

Aún no existen modelos precisos para evaluar la contaminación por MFs, por lo que en esta tesis se desarrolló un método para estimar el flujo másico de MFs desprendido de los lavados domésticos que llega a los ambientes acuáticos. El método consideró los siguientes parámetros: (1) la tasa de desprendimiento de microfibras de diferentes prendas textiles, (2) el volumen de efluentes de lavado, (3) el porcentaje de agua municipal que se trata, (4) el tipo de tratamiento aplicado a las aguas usadas, y (5) la proporción de lavadoras de carga frontal versus superior. De esta forma, se estima que 0,28 millones de toneladas de microfibras llegan cada año a los medios acuáticos. Además, se simularon situaciones hipotéticas para evaluar la reducción de microfibras modificando algunos parámetros a diferentes niveles (consumidor, entidades gubernamentales e industria). Se encontró que, dependiendo de las alternativas implantadas, las microfibras que llegan a los ambientes acuáticos podrían reducirse entre un 30% a un 65%.

Por otro lado, se revisó el estado actual de las MFs como contaminantes, discutiendo posibles alternativas desde la fabricación hasta la disposición final de las MFs. Hay muchas alternativas para reducir estos contaminantes, pero también lagunas que deben abordarse. Actualmente existen en el mercado algunas soluciones viables para retenerlas. Sin embargo, hasta que finalizó esta tesis, no se encontró ninguna propuesta sobre el destino de las MFs retenidas.

Por ello, en esta investigación se ha desarrollado un filtro para retener las MFs y se ha evaluado una propuesta para el tratamiento de las MFs retenidas, siguiendo la filosofía de la economía circular. Ambos procesos fueron patentados.

El sistema de filtración desarrollado está totalmente fabricado con polímeros reciclados. Su rendimiento es superior al 97% de eliminación de MFs de los efluentes de las lavadoras con un tiempo de reemplazo del cartucho de 30 a más de 40 ciclos de lavado. Las MFs retenidas se inmovilizan posteriormente en una matriz polimérica, convirtiéndolas en un "composite". Se mezclaron diferentes proporciones de MFs de poliéster con polietileno de baja densidad para la inmovilización de las MFs. Los resultados mostraron que la composición óptima, que mejoró algunas de las propiedades mecánicas de tracción, fue un 10 % de MFs de poliéster en la matriz polimérica.

Finalmente, se estudiaron otras fuentes de MFs, concretamente, las colillas de cigarrillos. Estas contienen los filtros fumados (SFs) y restos de tabaco sin fumar. Los SFs son desechos peligrosos compuestos por más de 15 000 fibras que pueden desprenderse como MFs. Se evaluó su tasa de desprendimiento, la toxicidad acuática aguda y la acuática-, termooxidativa- y foto-degradabilidad. Se encontró que los SFs desprenden aproximadamente 100 pequeñas MFs (< 0,2 mm) por día. Alrededor de 0,3 millones de toneladas de MFs potenciales podrían estar llegando anualmente a los ambientes acuáticos desde esta fuente. Cuando las MFs están presentes en el lixiviado generado por los SFs, se obtuvo una diferencia significativa de ecotoxicidad y una baja tasa de degradabilidad. Esto implica que las MFs de los SFs constituyen una fuente importante de microplásticos, lo que podría explicar en parte la alta concentración de polímeros artificiales que se encuentran en los sedimentos marinos en aguas profundas.

RESUM

Els microplàstics (MPs) són petits fragments (longitud < 5 mm) de polímers no modificats químicament i/o no biodegradables. Les estimacions aproximades apunten que hi ha entre 15 i 51 bilions d'elements flotants de MPs en ambients marins i 14 milions de tones als 9 cm superiors dels sediments dels oceans del món.

Segons la "European Chemical Agency", les microfibres tèxtils (MFs) tractades químicament i/o no biodegradables són un tipus de microplàstics amb una relació longitud/diàmetre superior a 3 mm i una longitud màxima de 15 mm. Una de les fonts més conegudes de MFs són les que es desprenen de cada cicle de rentat d'un article tèxtil. Aquests són considerats un dels contaminants més amenaçadors per al medi ambient ja que tenen una entrada contínua i acumulativa. A causa del seu petit diàmetre són més propensos a ser ingerits per organismes, per la qual cosa s'han trobat altes concentracions en productes per a consum humà, tals com marisc i aigua corrent.

En aquesta investigació es van avaluar les taxes de despreniment de microfibres de peces de roba acabades. Els resultats van mostrar que el despreniment de MFs oscil·la entre 175 i 560 MFs/g o entre 30.000 i 465.000 MFs/m^2 de peça. A més, hi va haver una alta correlació entre el despreniment de MPs i la densitat superficial de l'article tèxtil.

Com que encara no hi ha models precisos per avaluar la contaminació per MFs, en aquesta tesi es va desenvolupar un mètode per estimar el flux màssic de MFs procedent del rentat domèstic que arriba als ambients aquàtics. El mètode va considerar els paràmetres següents: (1) la taxa de despreniment de microfibres de diferents peces tèxtils, (2) el volum d'efluents de rentat, (3) el percentatge d'aigua municipal que es tracta, (4) el tipus de tractament aplicat a les aigües usades, i (5) la proporció de rentadores de càrrega frontal versus superior. D'aquesta manera, s'estima que 0,28 milions de tones de microfibres arriben cada any als medis aquàtics. A més, es van simular situacions hipotètiques per avaluar la reducció de microfibres modificant alguns paràmetres a diferents nivells (consumidor, entitats governamentals i indústria). Es va trobar que, depenent de les alternatives implantades, les microfibres que arriben als ambients aquàtics es podrien reduir entre un 30% a un 65%.

D'altra banda, es va revisar l'estat actual de les MFs com a contaminants, discutint possibles alternatives des de la fabricació fins a la disposició final de les MFs. Hi ha moltes alternatives per reduir aquests contaminants, però també llacunes que cal abordar. Actualment existeixen en el mercat algunes solucions viables per retenir-les. Tot i això, fins que va finalitzar aquesta tesi, no es va trobar cap proposta sobre el destí de les MFs retingudes.

Per això, en aquesta investigació s'ha desenvolupat un filtre per retenir les MFs i s'ha avaluat una proposta per al tractament de les MFs retingudes, seguint la filosofia de l'economia circular. Tots dos processos van ser patentats.

El sistema de filtratge està totalment fabricat amb polímers reciclats. El seu rendiment és superior al 97% d'eliminació de MFs dels efluents de les rentadores amb un temps de reemplaçament del cartutx de 30 a més de 40 cicles de rentat. Les MFs retingudes s'immobilitzen posteriorment en una matriu polimèrica, convertint-les en un "composite". Es van barrejar diferents proporcions de MFs de polièster amb polietilè de baixa densitat per a la immobilització de MFs. Els resultats van mostrar que la composició òptima, que va millorar algunes de les propietats mecàniques de tracció, va ser un 10 % de MFs de polièster en la matriu polimèrica.

Finalment, es van estudiar altres fonts de MFs, concretament, les burilles de cigarretes. Aquestes contenen els filtres fumats (SFs) i restes de tabac sense fumar. Els SFs són deixalles perilloses compostes per més de 15 000 fibres que poden desprendre's com MFs. Se'n va avaluar la taxa de despreniment, la toxicitat aquàtica aguda i la aquàtica-, termooxidativa- i foto-degradabilitat. Es va trobar que els SFs desprenen aproximadament 100 MFs petites (< 0,2 mm) per dia. Al voltant de 0,3 milions de tones de MFs potencials podrien estar arribant anualment als ambients aquàtics des d'aquesta font. Es va trobar una diferència significativa d'ecotoxicitat i una baixa taxa de degradabilitat quan les MFs són presents al lixiviat generat pels SFs. Això implica que les MFs dels SFs constitueixen una font important de microplàstics, el que podria explicar en part l'alta concentració de polímers artificials que es troben als sediments marins en aigües profundes.

Chapter 1:

Introduction

1 Introduction

This section aims to provide an insight into information about fibers, textiles, plastics, and microplastics.

1.1 Fibers, yarns, and Textiles

A fiber (fibre in European English spelling) is defined as any product capable of being woven or otherwise made into a fabric [1.1]. Technically, it is defined as units of matter characterized by flexibility, fineness, and a high ratio of length to thickness. Almost all the textile fibers are produced by six polymer types: natural as cellulose and proteins, and synthetic as polyester, polyamide, polyolefin, and vinyl [1.2]. However, all these polymers are also used in a wide variety of industries, e.g., fishery, cigarettes, and hygiene, among others [1.3]. Figure 1.1 shows a summary of the most relevant textiles fibers.

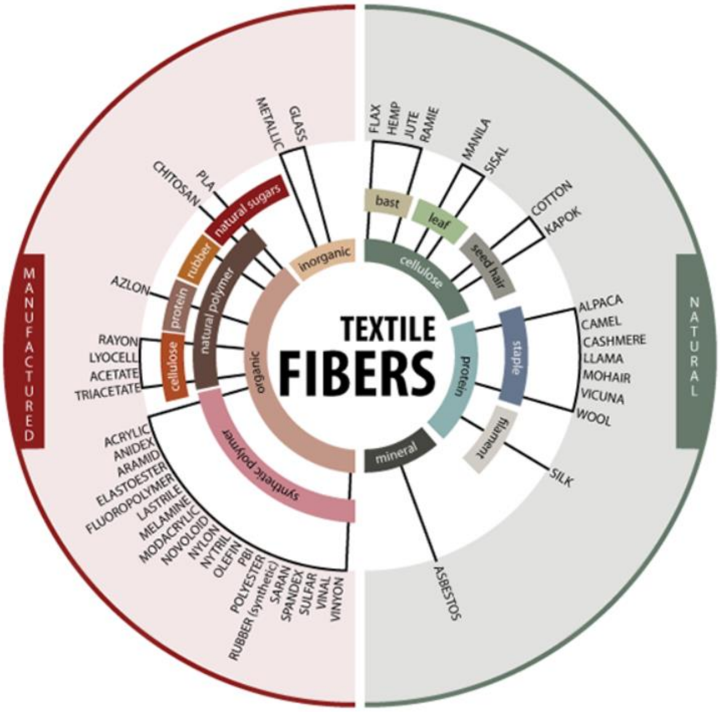

Figure 1.1. Most important textiles fibers and their origin. From [1.4].

On the other hand, textiles articles, and clothing are an essential part of our life. However, due to the "fast fashion" phenomenon, in the last 15 years, clothing manufacturing has almost doubled [1.5, 1.6]. Fast fashion has been defined as "a commercial model based on offering consumers recurrent innovation in the form of low-priced, trend-led products", it is the opposite to the "circular economy" philosophy. Most of the fast-fashion articles are poor quality products with low durability, which turns the article more prone to detach microplastics (a term that is defined later in this chapter). These are focused on cheap and quick purchasing, use, and disposal. Consequently, to bring down the production costs, there is a current lack to attend the associated environmental or social negative impacts [1.5, 1.7]. For instance, this growing clothing offer and demand has a considerable environmental impact as a result of the increased generation of textiles' waste. It has been estimated that the sector's (clothing and footwear) importance contributes to global warming with emissions of 1.3 Gt/year of greenhouse gases [1.8]. In this sense, there is ongoing research to treat this type of waste [1.9, 1.10]. A scheme of the flow of clothing can be seen in Figure 1.2.

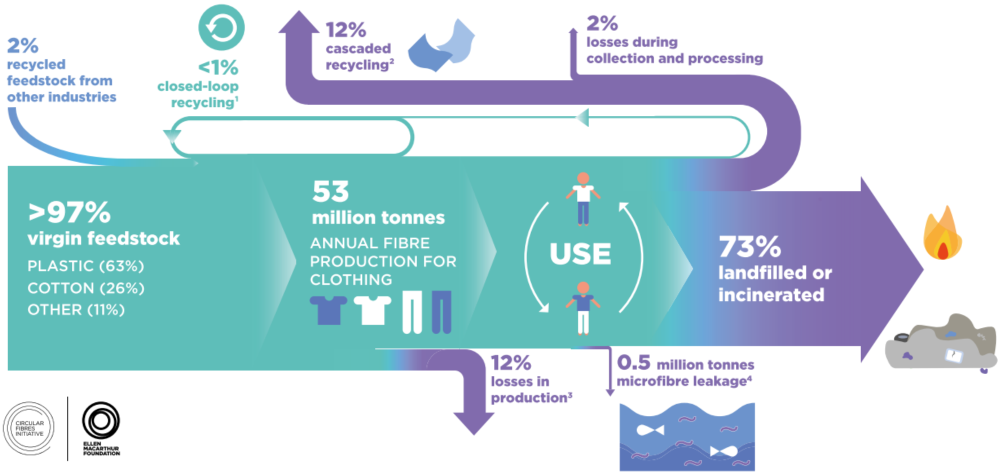

Figure 1.2. Global material flows for clothing in 2015. The microfibers are explained afterward. Figure from [1.6].

The global total textile fiber production in 2015 was estimated at 95 million tons. Within these numbers, synthetic polymers accounted for 65 million tons, where polyester was by far the most manufactured with almost 60 million tons, followed by polypropylene (5 million tons) and polyamide fibers (4.5 million tons) [1.11, 1.12]. Regarding natural fibers, cotton has the higher demand production with 30 million tons. It has to be mentioned that, currently, most of the cotton industry relies on a highly pollutant and environmentally unsustainable production, for instance, cotton production uses 2.5% of the world's farmlands but uses 25% of the world's insecticides consumption and 80% of total water usage in the textile sector [1.13]. Hence, to determine whether synthetic or natural fibers are more sustainable each specific case must be assessed. In Figure 1.3 the production and trend of the main fibers used for clothing are shown.

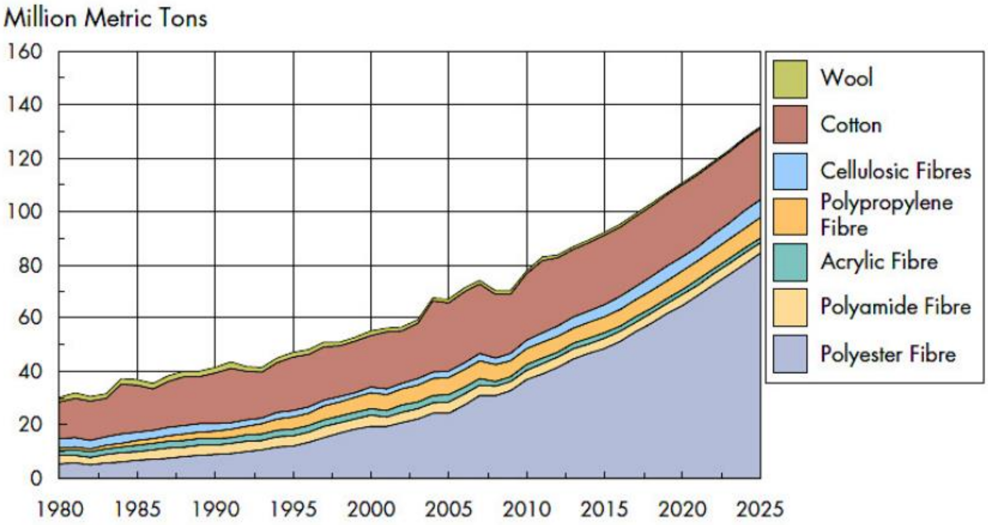

Figure 1.3. Past production and trends for the main fibers used by the textile industry. From [1.14].

As can be seen in Figure 1.3, the evolution of synthetic fibers, mainly polyester, has grown exponentially since about 1995. On the other hand, cotton and other fibers have remained almost unaltered. Some processes involving the manufacturing of synthetic textiles are briefly explained hereafter. For synthetic fibers, the melt spinning process or fiber formation is the most used method for fiber conversion. The extruder is the main part of this process. This technique is capable of producing many synthetic fibers [1.15]. After the spinning process of the fibers, their chainlike molecules are in an amorphous arrangement. Next, fibers are subjected to drawing, stretching, texturing, intermingling, and drying processes. The objective of drawing and stretching is to achieve an increase in the strength of the fibers by improving their orientation and decreasing their elongation at break. This process passes a group of filaments through a pair of rollers to elongate them. Afterward, the texturing process is to increase bulkiness, porosity, softness, and elasticity. In this section flat filaments are distorted to have loops, coils, curl, or crimps along their length. Next, the intermingling, which is a process of imparting inter-filament cohesion by entwining the filaments. The drying or heating process is to give stability to the fibers or yarns [1.16]. If fibers are the final product, a spinning yarn process is used, in which fibers are twisted together to form yarns. Then, the dying process is to give color to the yarns or fabrics. Depending on the process applied, it can be highly pollutant with effluents containing metals, salts, surfactants, and alkaline or acidic conditions [1.17]. However, nowadays, there are more sustainable options like digital printing [1.18]. Continuously, the fabrics are made whether by knitting or weaving processes. The first one produces knitted fabrics and the second woven fabrics. The main difference is that the knitted one has more elasticity. On the other hand, woven fabric can usually be stretched in only one direction [1.19]. Finally, these fabrics are subjected to finishing (mechanical and/or chemical), from where the manufacturing is concluded.

1.2 Plastics

Plastic is a universal term applied for a wide diversity of synthetic or semi-synthetic materials made from petrochemical products, and is a sub-category of a larger class of polymers. Regarding the type of plastic, these can be classified into two main families. Thermoplastics are those that can be melted when heated and hardened when cooled. Hence, these can be recycled in the sense that can be subjected to be reshaped and frozen repeatedly. Examples of thermoplastics are high- and low-density polyethylene, polypropylene, polyvinyl-chloride, and polyethylene terephthalate, among others. On the other hand, thermosets plastics are those that undergo a chemical reaction when heated (mostly combustion), precluding the reshaping of these materials. Examples of thermosets are unsaturated polyesters (like those used in textile articles), acrylic, and polyamide, among others [1.22]. These are employed in a vast and continuously growing range of applications [1.20]. Plastics have undeniable benefits that make them useful materials. For instance, poor water solubility and low biodegradability, which allow the manufacturing of a diverse range of inexpensive, lightweight, strong, durable, and corrosion-resistant products [1.21]. Its worldwide production has grown exponentially during the last decades, going from 1.7 million tons in 1950 to 370 million tons in 2019 [1.22]. However, if synthetic resins used in spinning textile fibers are included, the production rises to more than 400 million tons, a value that matches the global human biomass [1.23]. The production is so relevant that the plastic industry uses 6% of all the oil exploited in the world. Besides, the growth is so marked that it is expected to increase to 20% by 2050.

However, environmentally speaking, the benefits are not well harnessed as plastics are far from being a sustainable material. For instance, only a very small quantity is recycled and the mismanaged plastic waste in the environment is a global growing concern [1.24]. Moreover, the pandemic COVID-19 increased the use of single-use plastic items [1.25]. On the other hand, Europe and other countries have been improving their regulations for plastic use and waste management with, e.g., strategies to tackle single-use plastic items [1.26, 1.27]. In this line, Europe has achieved the recycling and energy recovery from plastic waste surpass the amount of plastic that ends up in landfills [1.28]. Figure 1.4 illustrates this trend for Europe 28+2.

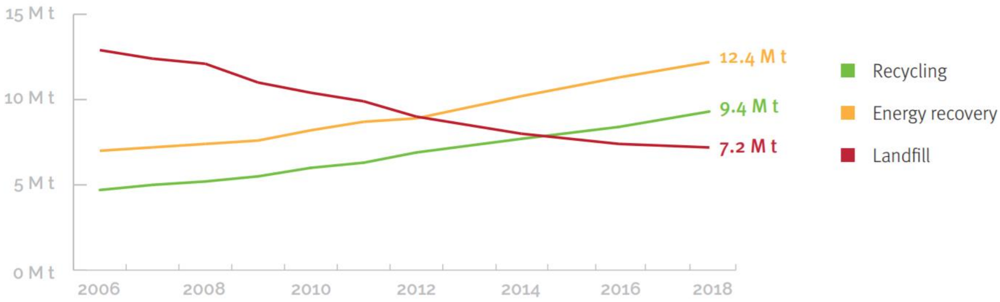

Figure 1.4. Waste treatment options for plastic waste in EU28+2. From [1.28].

1.3 Microplastics

The microplastics (MPs) as pollutants are small fragments (length <5 mm) of non-chemically modified and/or non-biodegradable polymers [1.29]. MPs have been found in every environmental compartment investigated so far (aquatic, terrestrial, atmospheric, and biota). Given the extent of this global pollution, some authors refer to the current period as the "plasticene" or describe the world's ocean as a "plastic soup" [1.30, 1.31]. It has been even suggested that MPs are a key geological indicator of the “Anthropocene” period [1.32]. Rough estimations point that there are between 15 to 51 trillion buoyant items of MPs in marine environments and 14 million tons in the top 9 cm of sediments of the world’s oceans [1.33–1.35]. These pollutants have even reached remote locations far from anthropogenic influence as the Arctic or the Antarctic, meaning that they can be transported by wind or ocean currents [1.36-1.39].

Sources and Types

Sources of MPs can be distinguished between primary, those emitted into the environment in a MPs size range; and secondary, those generated in the environment from physical degradation and fragmentation processes of larger plastic debris. In this way, primary MPs include a wide variety of sources (e.g., microfibers detached from textile garments, plastic pellets, tire dust); while secondary MPs have their origin in discarded plastic garbage and their derivatives into the natural environment that degrades into MPs [1.40, 1.41]. It should be mentioned that the agreement for the definition for MPs isn’t yet concerted. In this sense, some authors apply primary MPs as those that are manufactured in that size (e.g., pellets) and secondary MPs as those that are generated from bigger plastics’ products (e.g., microfibers detached from textile garments). In this thesis, the former definition has been applied.

Regarding the shape of MPs (see Figure 1.5), these are mainly distinguished in four, which can be summarized as:

1 **Fragments:** Have irregular shapes, with rough and broken edges. They usually come from larger plastic pieces such as plastic bottles or food packaging that suffered physical degradation and fragmentation processes.

2 **Fibers:** Have a regular fibrous shape that is almost equally thick throughout their entire length and has a high relation length to thickness. Common sources are fishery activities, the fibers shed from textile articles, or the ones released from cigarette butts. From now on, these will be named “MFs”.

3 **Films:** Have irregular flat shapes. These usually come from the fragmentation of thin plastic items such as bags or adhesive tapes.

4 **Granulated:** Have a spherical shape. These could come from microbeads used in cosmetics or from plastic pellets used in the plastic industry as feedstock.

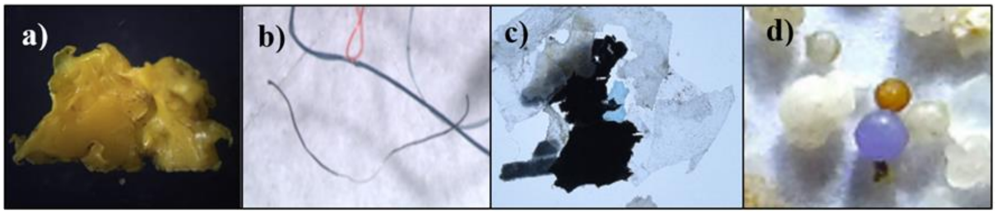

Figure 1.5. Types of microplastics regarding the shape.

MPs are also encountered in an extensive range of colors. Whether the colors of the MPs are a relevant factor in their misidentification and consumption by aquatic organisms is still a matter that is in debate. However, it seems intuitive that some colors might negatively affect because many aquatic species are visual predators. Indeed, current evidence suggests that black and red-colored MPs are the least likely to be ingested by aquatic species, in contrast with blue MPs [1.42, 1.43]. On the other hand, the small size of MPs has been found to be difficult the distinguish to some fishes between their food and the pollutant [1.44].

Effects and Behaviors of MPs

This contamination is fully recognized as ubiquitous; the potential effects have a current lack of knowledge. This is mainly because most of the experimental data encountered come from evaluations where unrealistic (high) concentrations of MPs were used [1.45]. However, it must be recognized that "the dose makes the poison", hence, from an uncertain point, the effects of these particles will become severely noticed. In this sense, MPs poses an intrinsic risk for ecosystems and living organisms basically because they come from a wide range of sources that can contain an extensive variety of added chemicals like phthalates, benzophenones, fungicides, among others [1.46-1.50]. Hence, these particles may behave as vectors for hazardous chemicals or invasive species. It was also published that soil MPs can also contain high concentrations of heavy metals (particularly, Cd, Pb, and Mn) and might pose a potential risk to soil organisms and safety [1.51]. Common heavy metals found in MPs are lead and copper [1.52, 1.53]. It was also noticed that these heavy metals have a weak attachment to the MPs and can be easily released into the aquatic ecosystems [1.54]. More examples of MPs carrying heavy metals have also been reported [1.55–1.57][1.58]. Moreover, there is the concern that MPs' aging increases their adsorption of heavy metals, though added chemicals during the product manufacturing seem to have a greater impact than adsorbed ones [1.59].

Their ingestion has been fully published, from where more than 200 species were found containing MPs in their gastrointestinal tract but also in other parts of their bodies [60]. For instance, these particles have been found in the guts of mussels in one of the most remote areas of South America, the Ushuaia Bay, with a mean occurrence of 9 MPs per mussel [1.61]. Studies in the Mediterranean Sea have also found high amounts of MPs, especially in the bottom of the sea, suggesting that deeper areas and benthic specimens are more exposed to this pollution [1.62]. A bigger concern is the translocation of these pollutants once inside the specimens. For instance, it has been reported that after MPs were ingested by oysters, these were translocated through the digestive gland to the hemolymph. These authors commented that the most plausible explanation for the translocation was via phagocytosis [1.63]. On the other hand, it was also published that, in some species, the excretion time for MPs is the same as for the retention of "real food", suggesting that some species are less prone to be affected by these particles [1.64]. Other impacts are the transport of MPs across the trophic chain, intestinal damage, endocrine disruption, false sensation of satiety, among others [1.65][1.60, 1.66, 1.67]. Yet, further investigation is needed to validate most of the outcomes. All these considerations are summarized in Figure 1.6.

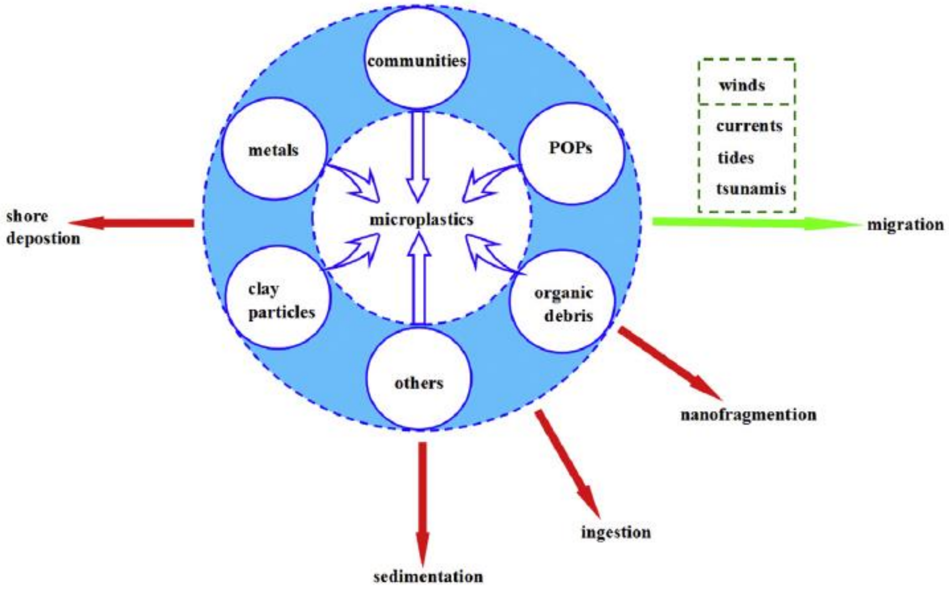

Figure 1.6. Physical and chemical microplastics behaviors [1.68].

Once in the ocean, It has been suggested that the abundance and migration of MPs in seawater are influenced by natural factors, such as ocean currents [1.69]. Even denser than sea-water particles can still be transported by underlying tides [1.68]. There are five identified "hot-spots" around the world where it is known that MPs debris accumulates, i.e., where they tend to migrate (see Figure 1.7). These are the North and South Atlantic, North and South Pacific, and Indian Sea gyres [1.70]. Besides, another hot-spot is the Mediterranean Sea, however, this is mainly a consequence of its enclosed type of system and the high density of population that is around it [1.71, 1.72]. In some cases, remote areas such as the Southwestern Atlantic have been found to concentrate high amounts of MPs, this could be related to intense harbor activities and the use of ropes and fishing nets [1.73].

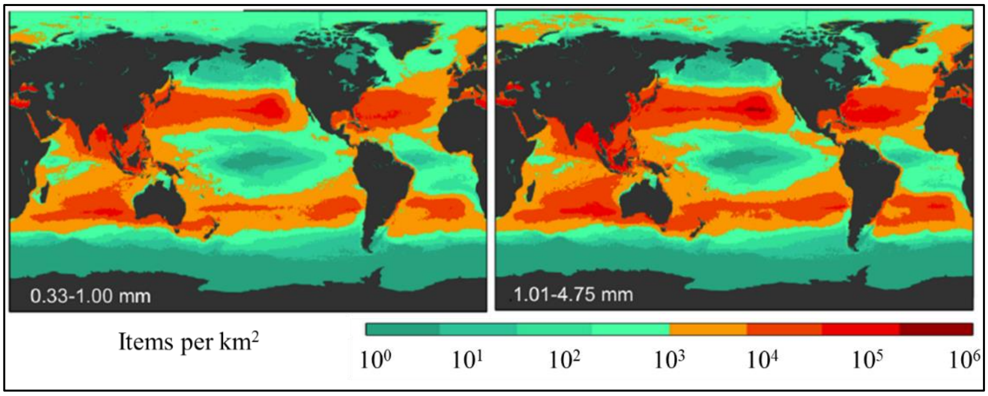

Figure 1.7. Modeled distribution pattern of MPs. Note the "hot-spots" in red. From. [1.70]

An interesting study found that about 90% of all the plastic garbage that flows from the rivers into the oceans comes from only 10 rivers around the globe. Five of these rivers are found in Asia and discharge their waters into the North Pacific Ocean. This makes sense, as the hot-spot corresponding to that ocean is one of the most polluted [1.74]. It is evident that the problem of mismanaged waste should be addressed from its root. However, it could be a practical and short-term solution to implement measurements to stop the transit of the plastic debris that is continuously flowing through those rivers. In this way, an important quantity of secondary MPs could be avoided as there will be less mismanaged debris to generate them.

Microplastics in Products for Human Consumption

MPs have been extensively found in in human-consumption products like fish [1.75-1.78], shellfish [1.79-1.83], drinking water [1.84, 1.85], bottled water [1.86-1.89], fruits, vegetables, table salt [1.90-1.92], honey [1.93], beer [1.94], among others. Other authors have also reported studies with more information on this topic [1.95–1.101]. The origin of the particles found in commercial products is usually difficult to determine as they deteriorate over time. In fishes, concentrations are generally very low, from 1 to 2 items per individual: however, in some species, concentrations up to 75 items per individual were found, especially in mussels, which are usually the most contaminated living species. The reason is that these are filtering-feeding animals; i.e., they filter a large amount of water and retain their “food”. In this line, it`s a fact that we are already ingesting MPs when we feed ourselves. For instance, estimations reported that a European eat up to 11'000 microplastics per year only from shellfish consumption [1.98] Nevertheless, the potential risks for human health either by involuntary consumption or inhalation are still an unknown area of study [1.42].

1.4 Microplastics and Municipal Water Treatment Plants (WTPs)

In municipal or industrial used-WTPs, a proportion of the incoming MPs will be transferred into the sludge throughout the consecutive treatments. It is important to notice that WTPs should not be considered as sources of MPs but a pathway where they can be removed from the liquid stream. Globally, the percentage of used-waters that are not safely treated is estimated at around 20% [1.102, 1.103]. Populations connected to urban WTPs are markedly variated across the countries. For instance, South American and Asian countries treat around 20% to 30% of their municipal waters, while Central European countries have achieved a 97% of treatment coverage [1.104-1.106]. WTPs are considered significant pathways for all types of MPs to aquatic and soil environments [1.107, 1.108]. Yet, as mentioned before, fiber-shaped ones are within the most encountered types in these streams.

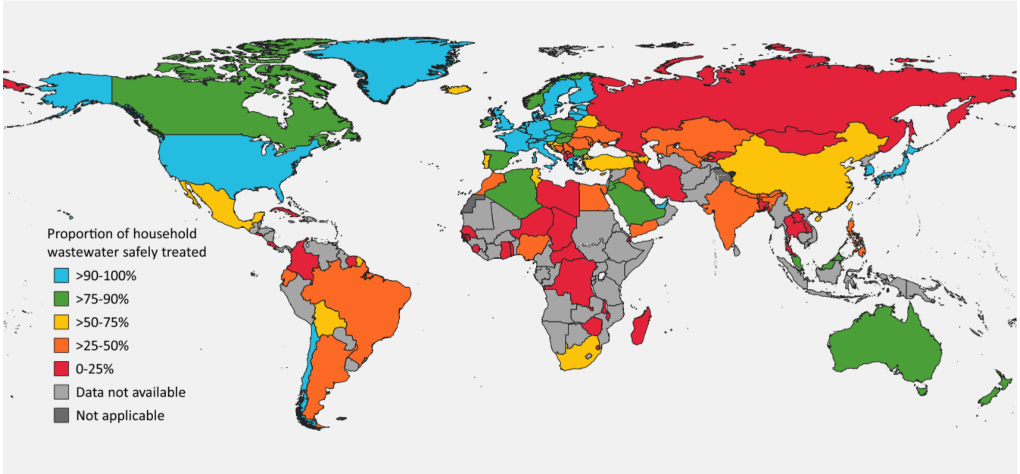

Figure 1.8. Percentage of safely treated used-water from households in 2020 (%).

Regarding the material, polyester MFs usually surpass other types of MPs [1.109–1.113]. Currently, polyester is the top synthetic material used by the textile industry [1.114].

Besides, as pointed out by a series of publications, regardless of the material, each textile article detaches thousands to millions of MFs in every domestic washing cycle [1.115–1.119]. For instance, Alavian Petroody et al. (2020) reported that most of the MPs found in a WTP were in the form of MFs, from which polyester was the most abundant, followed by polyamide and acrylic fibers. These particles have also been found in potable water drinking plants [1.121].

Despite having a relatively high retention efficiency, these facilities treat millions of liters every day, releasing high amounts of MFs [1.113]. Also, as previously explained, the proportion of treated waters is still very low across the world, and it must be noticed that other sources of textile MFs (as hand-washed garments) will be still left aside from these treatments. Equally important is that these particles can still enter the environment via the final disposal of the sludge, as conventional treatments don't remove sludge-based MPs [1.122]. The reported abundance of MPs in the sludge varies from 1 500 to 180 000 particles per kg of dry weight sludge [1.112, 1.123, 1.124]. Hence, MFs might still be dumped into the environment if the sludge is used as, e.g., an agricultural fertilizer [1.125–1.130]. Furthermore, it has been suggested that MPs can be taken by crops and cause multiple adverse effects on plants, furtherly reaching human vegetable or fruits consumption [1.131].

1.5 Microfibers

Within the types of primary MPs, microfibers (MFs) are those with a length to diameter ratio > 3 and a maximum length of 15 mm [1.29]. One of the most renowned sources of MFs are those detached from every cycle of a textile article laundering. In this thesis, it has been estimated that 0.5 million tons are generated only from household laundering, from where more than 50% reaches aquatic environments [1.132]. More details about these pollutants are explained throughout this document.

This doctoral thesis aims to explore mainly into these types of MFs, willing to develop an alternative to reduce their contamination rate into the environment. It must be mentioned that there are alternatives to reduce the generation or to retain the already detached ones [1.133]. However, until this thesis was over, there was no single proposal on the destination of the retained MFs. Hence, one of the main objectives of this work was to develop a practical treatment to retain these pollutants and a proposal to treat the retained MFs. One of the main characteristics of the filtering system is that it is entirely constructed by recycled materials. Moreover, even the filtering media is made of recycled polymers. Both processes were patented and are furtherly explained in Chapters 6 and 7. Figure 1.9 shows examples of MFs that were detached from a domestic machine washing and filtered through 20 μm polyamide filters.

Figure 1.9. Textile microfibers retained by filtering a laundering effluent.

As can be seen in Figure 1.9, these MFs can have different orientations, sizes, colors, etc. However, the main characteristic is their small diameter or large length to diameter ratio, which makes these particles more prone for organisms to ingest them. The MFs detached from domestic washing are the main subject of this thesis. Nonetheless, there are other sources like ropes or cigarette butts. This last source was also evaluated in this thesis and results are shown in chapter 8.

I desire a nice and productive time when you, readers, go inside this thesis.

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Chapter 2:

Objectives

2 Objectives

2.1 Main Objectives

This doctoral thesis has been named "**Microplastics and Microfibers Pollution: Study of their Environmental Issues and Evaluation of Reduction Alternatives**". In this sense, it aims to:

- Delve into the knowledge of this pollution, making a special focus on textile synthetic microfibers,
- Study the alternatives for the reduction of its emission during the domestic laundering process,
- Develop a system to retain these microfibers, and,
- Develop a method to treat these particles.
- Close the loop between the microfibers' detachment and their treatment.

2.2 Specific Objectives

The thesis has been structured according to the following specific objectives:

- Develop a method to quantify the microfibers detached from household laundering. It must have good replicability, at least around 10%. The method must be simple in the sense that no specialized equipment is needed for the quantification.

- Develop a mathematical equation to estimate the number of microfibers reaching aquatic environments. The equation must consider the parameters known until the moment of its development. Also, it has to own the capacity to be used by other scientists.

- Make a critical review on the textiles microfibers as microplastics' pollutants. This document must consider from the manufacturing until the different solutions or alternatives that has being proposed to reduce this contamination. Yet, the parts where microfibers are also detached but have not been studied will not be considered.

- Develop a new system to retain the microfibers in the effluent of different equipment such as washing machines or dryers. The filter must be built from recycled polymers, including the filtering media. In this sense, the circular philosophy can be included in the system.

- Propose a practical alternative to treat the retained microfibers. When this thesis was started, no solutions were proposed. Hence, the objective is to treat microfibers emitted from diffuse points, i.e., domestic washing machines.

- Evaluate other types of microfibers, specifically, those generated from cigarette butts.

2.3 Thesis arrangement

The thesis has been arranged as follows:

- The development of a method to quantify the microfibers detached from household laundering is in Chapter 3. This chapter was published in Environmental Pollution Journal – Elsevier (Q1).

- The development of a mathematical equation to estimate the amount of microfibers reaching aquatic environments is in Chapter 4. The equation and the assessment of its outcomes were published in Environmental Pollution Journal – Elsevier (Q1).

- A critical review of the textiles microfibers as microplastics' pollutants is in Chapter 5. This chapter considers from the manufacturing until the different solutions or alternatives that has being proposed to reduce this contamination. The microfibers that are detached from the daily usage and the final disposition of a given textile article are not considered. The chapter has been sent to Journal of Environmental Management– Elsevier (Q1).

- The development of a new system to retain the microfibers in the effluent of different equipment such as washing machines or dryers is in Chapter 6. The filtering media is constituted by recycled polymeric pellets. The system was patented under a Spanish patent. In addition, a paper with the applications of this filter and the comparison with other marketed filters has been sent for publication (Q1).

- A practical proposition of an alternative to treat the retained microfibers is in Chapter 7. The polymeric filtering media is liquefied and mixed with the microfibers. In this sense, homogeneous and rigid structures can be made. These composites are being tested to find applications for them. The chapter was sent to Journal of Industrial Textiles (Q1). Besides, the treatment is also covered by the Spanish patent previously indicated.

- An evaluation of other types of microfibers, specifically, those generated from cigarette butts is in Chapter 8. The study was executed to give a perspective of this “invisible” but ubiquitous toxic debris. The work was published in Science of the Total Environment (STOTEN) Journal - Elsevier (Q1).

Chapter 3:

Microplastics' Emissions: Microfibers' Detachment from Textile Garments

3 Microplastics' Emissions: Microfibers' Detachment from Textile Garments

This work provides new insights with respect to microplastic pollution. It also establishes a method for the quantification of textile microfibers and recommends comprehensive and comparable units to be used when publishing the results.

This chapter corresponds to the pre-prints of the paper refered below, that has been published in Environmental Pollution (Elsevier Journal, IF = 6.792. Q1, ScienceDirect):

F. Belzagui, M. Crespi, A. Álvarez, C. Gutiérrez-Bouzán, M. Vilaseca. Microplastics' Emissions: Microfibers'Detachment from Textile Garments.*Environmental Pollution, 248 (2019), 1028-1035.*
DOI: 10.1016/j.envpol.2019.02.059

The supplemenary information of the article has been included in the correspoding sections of the chapter.

Abstract

Microplastics (synthetic polymers <5 mm) have been recently recognized as a big environmental concern, as their ubiquity is an undeniable fact. Their wide variety regarding shapes, sizes, and materials turn them into an intrinsically risky pollutant capable of causing several environmental impacts. Textile microfibers (MF) are a microplastic sub-group. These are mostly shed when a normal laundry of any garment takes place. Special attention has been put onto them, as high concentrations have been found in products for human consumption as shellfish and tap water. However, as there is no consensus on the methodologies to quantify and report the results of MFs detached from textile garments, the degree of similarity between published studies is very low. Hence, the aim of this research was to evaluate the microfibers' detachment rates of finished garments and to provide a set of comparable units to report the results. These were found to range between 175 to 560 MF/g or 30'000 to 465'000 MF/m^2 of garment. In addition, there was a high correlation between the MF detachment and the textile article superficial density. Finally, our results were compared with a recent paper that estimated the annual mass flow of MFs to the oceans. This previous publication is 30 times higher when related to the mass but 40 times lower if related to the number of MFs.

Keywords: Microplastic; Microfiber; Pollution; Textile.

3.1 Introduction

The globally widespread plastic pollution is a well-known environmental concern that has been even suggested as an indicator of the *Anthropocene* period [3.1]. Synthetic polymers debris sized at <5 mm, generally defined as microplastics (MPs from now on), have been recently recognized as an important and abundant pollutant [3.2]. MPs occurrence is increasingly growing in freshwaters, terrestrial and atmospheric ecosystems [3.3–3.5], and have even reached remote places far from anthropogenic influence [3.6]. However, the major sink seems to be the marine environment, where these pollutants are ubiquitous, as they are found from the top to the bottom and from the equator to the poles [3.7–3.10]. Last estimations reported 15 to 51 trillion *buoyant* MPs in the oceans, but these are believed to be only the "tip of the iceberg" [3.11,3.12].

The ingestion of these plastic particles by biota is well registered [3.13], especially in marine organisms, where MPs have been identified in all levels of the trophic chain [3.14, 3.15]. However, it also extends to organisms of other ecosystems [3.16, 3.17]. Observed possible impacts in biota are MPs' retention [3.18] and trophic transfer [3.19], reduced vital functions capacity [3.20–3.22], translocation to other organs [3.23], gene exchange [3.24], endocrine disruption [3.25], increased mortality [3.26], bioaccumulation of toxic chemicals [3.27], altered sinking rates for fecal pellets [3.28], etc. MPs are also known to act as vectors for alien species and for hydrophobic contaminants (either added in the plastic manufacturing process or adsorbed once in the environment) [3.29, 3.30], and to alter physical properties of beach sediments [3.31]. Moreover, there is evidence of MPs presence in products for human consumption as seafood [3.32, 3.33], tap and bottled water [3.34], and table salt [3.35]. Nevertheless, the human health risks still remain unclear.

The MPs' sources are usually classified into two main groups (adopted for this work): *Primary MPs* are those emitted to the environment in an MP size range (e.g., textile microfibers, microbeads); and, *Secondary MPs* are those originated once in the environment from the degradation and fragmentation of mismanaged plastic waste [3.2, 3.36]. However, there are still no accurate estimations of the contribution of each MP source; hence, it is necessary to elaborate tools that enable us to achieve a better knowledge of the importance of each contributor.

Textile microfibers (MFs from now on) are detached among every step of a textile article life cycle, especially when its laundry takes place [3.36]. Many publications have reported high concentrations of MFs in aquatic environments [3.37], hence, they appear to be one of the most important primary MPs contributors [3.36, 3.38]. A few studies have proposed different methods to quantify detached MFs [3.37, 3.39–3.44]. These usually applied indirect methodologies, e.g., estimating the amount of MFs from their weight and length. However, the accuracy between these studies is still low, and the units used to express the results are different, making their comparison even more difficult. These methods were tested in our laboratory, but the estimations were not in accordance with our visual quantification of MFs (discussion in section **¡Error! No se encuentra el origen de la referencia.**). Therefore, in this work, it was developed and applied a direct and reliable method to determine the ***microfibers' detachment rates*** (**MFDR** from now on) when washing textile articles. In addition, a relation between the number of MFs and their mass, and also a set of comparable units are provided. Finally, from this quantification, an estimation of the amount of MFs released to the environment is carried out and compared with previously published works.

3.2 Materials and Methods

3.2.1 Materials

Brand new garments of polyester, polyester-elastane, and polyamide-elastane were bought from different fashion stores in Spain and tested. The characteristics of the selected garments are described in Table 3.1 and seen in Figure 3.1.

A front-load conventional washing machine (*FAGOR Innovation F-2810*, Spain) was used. In order to save energy and water, the *superquick* program was chosen (15 minutes, 22 liters of effluent, 1'000 revolutions per minute, ambient temperature). Tap water from *Terrassa* was supplied to the washing machine. A common detergent ("*Bosque Verde*", Spain) was selected, where the quantity applied was in accordance to the specifications written on the container as a function of the weight of the garment and the hardness of the water (349 mg $CaCO_3/L$ [3.45]). An explanation of the operational parameters' selection is described in **¡Error! No se encuentra el origen de la referencia.**.

Table 3.1. Characteristics of the tested garments

Material	Group	Name	Type	Mass [g]	Area [m^2]	Type of Fabric
100% Polyester	F	F1	"Fluffy"	603	1.63	Woven fabrics, with the exception of F4 which is knitted.
		F2		723	1.95	
		F3		643	1.60	
		F4		396	1.08	
		F5		728	1.50	
	P	P1	Shirt (1)	101	0.55	Knitted fabric.
		P2	Nightgown	241	1.50	Woven fabric.
80% Polyester 20% Elastane	PE	PE1	Shirt	134	0.35	Knitted fabric.
		PE2	Gym pants	193	0.57	
		PE3	Jacket	305	0.90	
70% Acrylic 30% Polyamide	PAC	PAC	Woolen cap	74	0.09	Knitted fabric.

(1) Recycled polyester.

Figure 3.1. Tested garments.

Polyamide filters (*Millipore NY20*, Ireland) with a 20 μm pore diameter were used to retain the MFs. The filtering system consisted of a flask connected to a vacuum pump. A stereomicroscope (*Carton Stereo Zoom SC*, Japan) and an electronic microscope or SEM (*PHENOM ProX Desktop*, The Netherlands) were used to analyze the MFs.

3.2.2 Methodology

The developed method is constituted by the following steps:

a) The garments were weighted, measured and characterized with respect to the material before starting the procedure.

b) A commercial washing machine was selected to execute the trials, as laboratory washing machine simulators might not produce the same effects (e.g., the centrifugal operation step is not simulated). An empty washing cycle was done to clean residual dirt between launderings of different garments.

c) One of the selected garments was independently washed 10 times to determine the number of washing cycles required to achieve a stationary situation. According to the result of this test and to other previously published works, the rest of garments were only washed 5 times (also independently).

d) For each washing cycle, the washing effluent was completely collected (22 L) in a closed container. A 10 L sample was taken, while stirring, using a hose assembled at the bottom of the closed container.

e) From the 10 L sample, three smaller aliquots of 10 mL were taken, while stirring, and rinsed up to 100 mL with distilled water to get a more homogeneous MF distribution on the filters. The purpose of such smaller aliquots is to be able to visually count the MFs retained after the filtration, as previous trials showed that major volumes made it impossible due to MFs overlapping.

f) The filters, which were always kept in Petri dishes to avoid contamination, were carefully placed on the filtration system with a clamp and the small water aliquots were filtered. Then, distilled water was used to drag all the MFs retained in the sample collector.

g) Subsequently, the filters were dried at 60 ºC for 24 hours. Once cooled, these were placed under the stereomicroscope where the visual counting of the MFs was done.

h) The background on which the Petri dishes with the filters were posed was alternated between light and dark-colored depending on the color of the garment.

A simple diagram of the described methodology is shown in Figure 3.22:

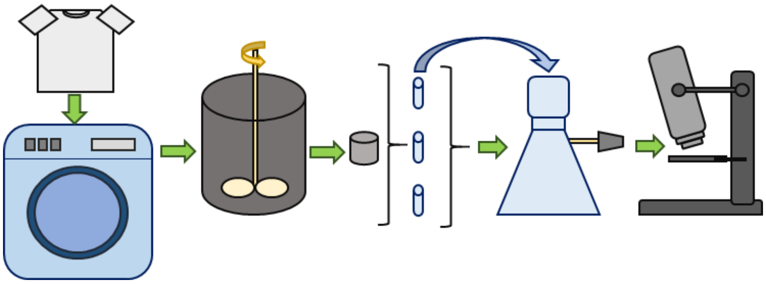


Figure 3.2. Scheme of the developed method.

It must be clarified that the operational parameters were also chosen expecting the minimum MF detachment from the garments. Previous publications have reported that front-loading washing machines [3.41], liquid detergent and lower temperatures [3.44] produce less mechanical stress than their opposites; the same was expected from washing the garments for a short period of time. Still, further work is needed to determine the relevance of these parameters in the MFDRs.

The observed amount of detached MFs will be expressed with respect to the garment weight $(W_G: MF/g)$ and surface $(A_G: MF/m^2)$. Additionally, an evaluation of these results and the superficial density $(SD_G = W_G/A_G)$ of each garment was done, from where a positive correlation was expected. Furthermore, a scanning electron microscope was used to evaluate the MFs' morphology.

3.2.3 Calculations

The repeatability of the method for the quantification of the detached MFs (steps a-h) was evaluated with the average error and the average coefficient of variation of all the samples. The equations used are described in Table 3.2.

Table 3.2. Equations applied to estimate the repeatability of the method.

Applied for	Average Error	Coefficient of Variation
Each garment and each cycle	$E_{gc} = \frac{\sum_i^n \lvert X_i - \bar{X} \rvert}{n \cdot \bar{X}} \cdot 100\%$	$CV_{gc} = \sqrt{\frac{\sum_i^n (X_i - \bar{X})^2}{n-1}} \cdot 100\%$
Each garment and all cycles	$E_g = \frac{\sum_c^m E_{gc}}{m}$	$CV_g = \frac{\sum_c^m CV_{gc}}{m}$
All garments	$\bar{E}_T = \frac{\sum_g^p E_g}{p}$	$\overline{CV}_T = \frac{\sum_g^p CV_g}{p}$

n = Number of samples per garment per cycle; m = Number of cycles per garment
p = Number of garments; $\bar{X} = \sum_i^n X_i/n$ = Average number of MFs per garment per cycle

On the other hand, a relation between the quantity of MFs and their mass, $MF_W\ (MF/mg)$, has been established. This can be obtained from the fiber linear weight, usually named yarn count $C\ (dtex)$, and the MFs' average length, $\bar{L}_{MF}\ (mm/MF)$, by using the following Equation 3.1:

$$MF_W = \frac{1}{C \cdot \bar{L}_{MF}} \quad ; Eq. 3.1$$

A decitex (dtex) is a unit of measurement of the linear weight of a filament textile fiber. It is expressed in grams of filament fiber per 10'000 meters. The Equation 3.2 [3.46–48] used to estimate the linear weight (C) is described hereunder:

$$C = \emptyset^2 \cdot \frac{\pi \cdot \gamma}{400} \quad ; Eq. 3.2$$

Where the average diameter $\emptyset\ (\mu m)$ of the MFs was obtained from SEM observation, and $\gamma\ (g/cm^3)$ is the specific weight of the fiber material. In this study, the specific weight of the polyester (1.38 g/cm^3) was used in all calculations as it is the predominant material.

From Equation 3.1, a table with typical *dtex* values versus a range of MF lengths between 0.1 to 5 mm was plotted to facilitate the estimation of the mass of the MFs, this can be seen in Table 3.3.

Table 3.3. Relations between the linear density (dtex) and the length of a microfiber to estimate the quantity of microfibers in a milligram.

	***dtex* = g/10’000m**					
L MF mm	**0.5**	**1**	**2**	**3**	**4**	**5**
	MF/mg					
0.1	200’000	100’000	50’000	33’333	25’000	20’000
0.5	40’000	20’000	10’000	6’667	5’000	4’000
1.0	20’000	10’000	5’000	3’333	2’500	2’000
1.5	13’333	6’667	3’333	2’222	1’667	1’333
2.0	10’000	5’000	2’500	1’667	1’250	1’000
2.5	8’000	4’000	2’000	1’333	1’000	800
3.0	6’667	3’333	1’667	1’111	833	667
3.5	5’714	2’857	1’429	952	714	571
4.0	5’000	2’500	1’250	833	625	500
4.5	4’444	2’222	1’111	741	556	444
5.0	4’000	2’000	1’000	667	500	400

Finally, MF_W from Equation 3.1 was afterward applied to estimate the mass loss of MFs by multiplying its inverse with the quantity of MFs detached from the garments.

3.3 Results and Discussions

3.3.1 Microfibers’ Detachment Across the Washing Cycles

In order to determine the required number of washing cycles to apply across this work, the garment F1 was washed 10 consecutive times (as indicated in Materials and Methods). It was found that between the 4th and 5th washing cycle the MFDR stabilized (see Figure 3.3). This result is in accordance with the publication of Napper & Thompson (2016). Henceforth, the rest of the garments studied in this work were only submitted to 5 washing cycles. All the observations gathered from the washing trials are included in Table 3.4. As an example, some of them are shown in Figure 3.4, where confidence intervals for each trial are also plotted.

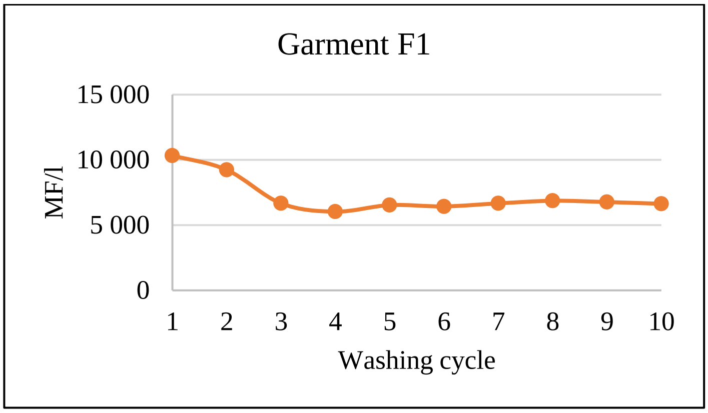


Figure 3.3. MFs detached per liter through 10 continuous washing cycles for the F1 garment.

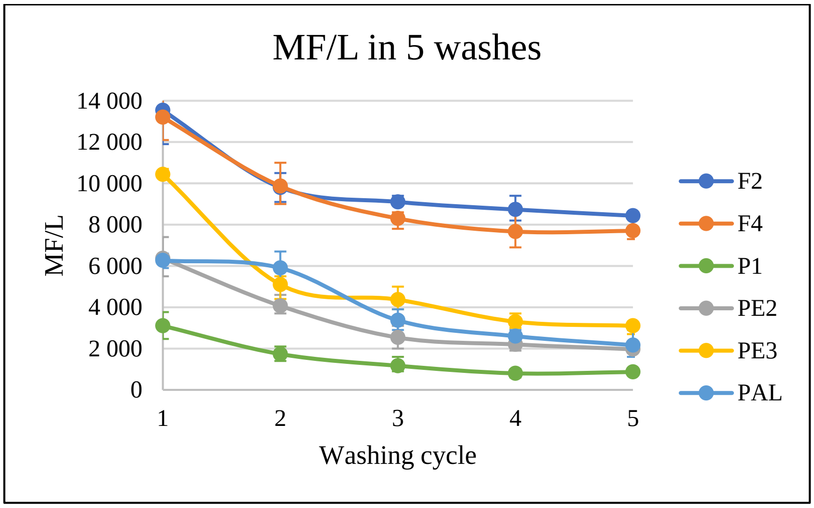
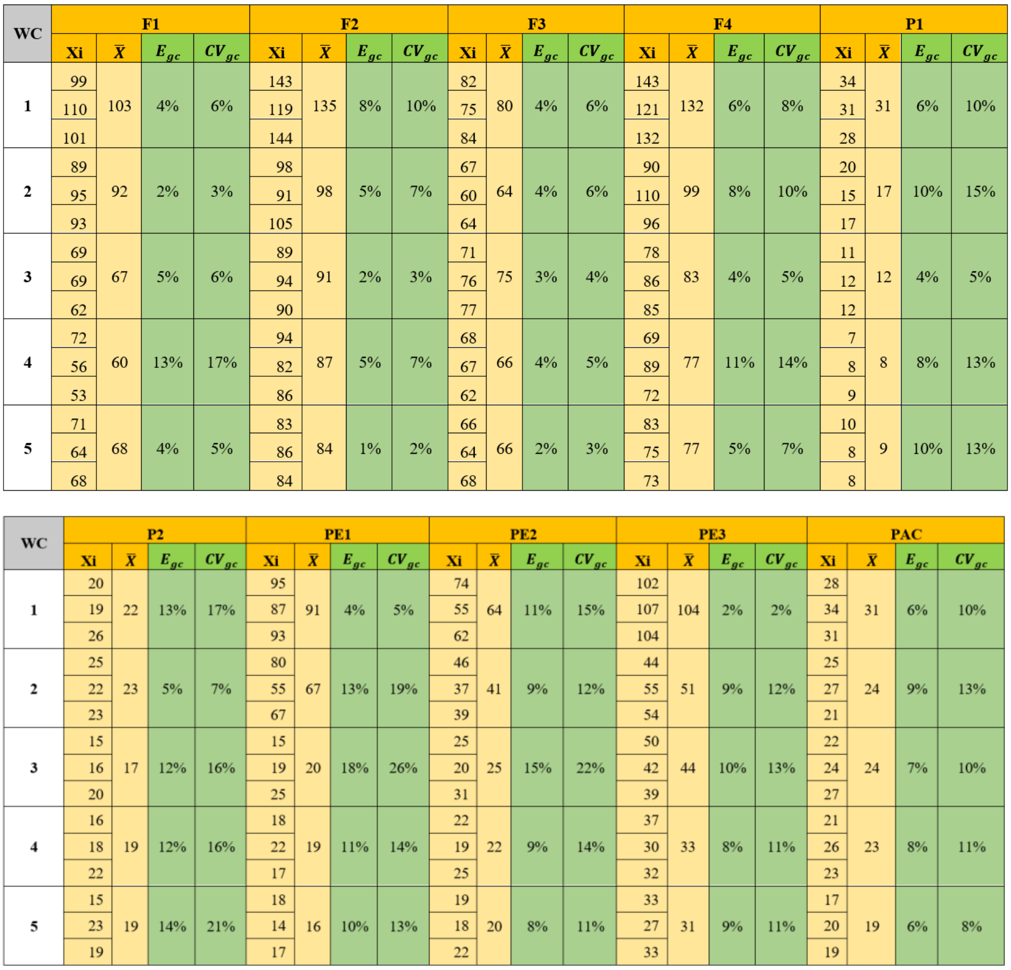


Figure 3.4. The MFs detachment decreases from the 1st to the 5th washing cycle in all tested garments.

From the washing trials observations, it can be noticed that the garments shed more MFs in the first washing cycles, which is probably due to the presence of leftovers from the garment manufacturing process. Hence, the application of MFs' retention mechanisms in industrial stages, as in the textile dyeing and finishing processes, could easily help to reduce a considerable amount of MFs from reaching the environment.

However, no clear trend was found between the garment material and the progressive reduction from the 1st to the 5th wash of the MFs' detached. Fluffy garments reduced the MFDR from 20 to 40%, other polyesters (P1, P2) from 10 to 70%, polyester mixed with elastane (PE1, PE2, PE3) from 70% to 80%, and PAC 40%. Also, the type of fabric (knitted or woven) does not seem to have a relevant influence on MFDR results.

Table 3.4. Data gathered during the experiments and data analysis.



WC	F1				F2				F3				F4				P1			
	Xi	$\bar{X}$	E_{gc}	CV_{gc}	Xi	$\bar{X}$	E_{gc}	CV_{gc}	Xi	$\bar{X}$	E_{gc}	CV_{gc}	Xi	$\bar{X}$	E_{gc}	CV_{gc}	Xi	$\bar{X}$	E_{gc}	CV_{gc}
1	99	103	4%	6%	143	135	8%	10%	82	80	4%	6%	143	132	6%	8%	34	31	6%	10%
	110				119				75				121				31			
	101				144				84				132				28			
2	89	92	2%	3%	98	98	5%	7%	67	64	4%	6%	90	99	8%	10%	20	17	10%	15%
	95				91				60				110				15			
	93				105				64				96				17			
3	69	67	5%	6%	89	91	2%	3%	71	75	3%	4%	78	83	4%	5%	11	12	4%	5%
	69				94				76				86				12			
	62				90				77				85				12			
4	72	60	13%	17%	94	87	5%	7%	68	66	4%	5%	69	77	11%	14%	7	8	8%	13%
	56				82				67				89				8			
	53				86				62				72				9			
5	71	68	4%	5%	83	84	1%	2%	66	66	2%	3%	83	77	5%	7%	10	9	10%	13%
	64				86				64				75				8			
	68				84				68				73				8			

WC	P2				PE1				PE2				PE3				PAC			
	Xi	$\bar{X}$	E_{gc}	CV_{gc}	Xi	$\bar{X}$	E_{gc}	CV_{gc}	Xi	$\bar{X}$	E_{gc}	CV_{gc}	Xi	$\bar{X}$	E_{gc}	CV_{gc}	Xi	$\bar{X}$	E_{gc}	CV_{gc}
1	20	22	13%	17%	95	91	4%	5%	74	64	11%	15%	102	104	2%	2%	28	31	6%	10%
	19				87				55				107				34			
	26				93				62				104				31			
2	25	23	5%	7%	80	67	13%	19%	46	41	9%	12%	44	51	9%	12%	25	24	9%	13%
	22				55				37				55				27			
	23				67				39				54				21			
3	15	17	12%	16%	15	20	18%	26%	25	25	15%	22%	50	44	10%	13%	22	24	7%	10%
	16				19				20				42				24			
	20				25				31				39				27			
4	16	19	12%	16%	18	19	11%	14%	22	22	9%	14%	37	33	8%	11%	21	23	8%	11%
	18				22				19				30				26			
	22				17				25				32				23			
5	15	19	14%	21%	18	16	10%	13%	19	20	8%	11%	33	31	9%	11%	17	19	6%	8%
	23				14				18				27				20			
	19				17				22				33				19			

On the other hand, on the bases of equations described in Table 3.2, the average error (calculated for all samples and 5 washing cycles) was E = 8% and the coefficient of variation was CV = 10% (data in Table 3.4), which indicates that the method has a high repeatability. It should be mentioned that methods proposed in previous publications were tested [3.39–3.41]. Nevertheless, overestimated results were found when comparing the data obtained from those methodologies with the visually counted MFs. A feasible explanation comes from the impurities that were found on the filters and within the MFs. These might come from the detergent, from additives un- or intentionally applied to the garments during the manufacturing process, and/or from the tap water used for the washing machine trials. Hence, the reliability of the method developed and applied in this work is generally higher than that of previously published ones because possible interferences implied in the weighting process of the MFs are eliminated.

3.3.2 Morphological Aspects of Detached MFs (length and shape)

By means of the stereomicroscopic observation, it was found that the length of the detached MFs decreased from the 1st to the 3rd wash in every tested garment. This behavior was also confirmed by determining the trend of the ratio MF/mg, which was seen to increase from the 1st to the 3rd washing cycle. In this way, longer and more MFs are detached in the firsts washing cycles, strengthening the greater ease and effectiveness that the early-stages MFs' retention systems mentioned in 4.3.1 could have.

In the last washing cycle, all MFs were visible under the stereomicroscope. The average length was between 0.2 to 0.4 mm, and the minimum found was of ~ 0.08 mm. However, a smaller fraction < 20 µm to nanoscales might exist but was not evaluated. As an example, the evolution of the trend of the P1 garment MF length across 5 washing cycles is shown in Table 3.5.

Table 3.5. Evolution of the trend of the P1 garment MFs' length across 5 washing cycles.

Wash cycle	**1**	**2**	**3**	**4**	**5**
Length [mm]	< 4	< 2	< 1	< 1	< 1
Mean Length [mm]	1.1	0.9	0.7	0.4	0.4

Furthermore, from SEM observation, two possible causes for the detachment of these MFs were identified. A first group corresponds to MFs already attached or entangled with the fibers' grid of the garments (Figure 3.5A), which have a regular tail-ended shape. In contrast, the other group appears to be MFs that were ripped-off from the fibers' grid (Figure 3.5B) as a consequence of the mechanical stress suffered by the garment throughout the launderings. The garment UV degradation and its use might also debilitate the fibers and facilitate the MFs' ripping [3.49].

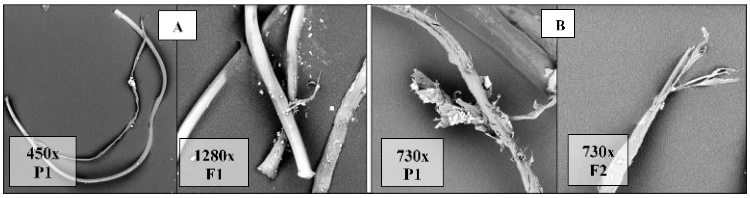


Figure 3.5 (A) Microfibers with a regular end-tailed shape, and (B), microfibers with a ripped-off end-tailed shape.

Hence, the application of a biodegradable coating could help to reduce these MFs by enhancing the grid connections and/or the garments' resistance to mechanical stress.

Finally, as seen in Figure 3.5A and B, there is more material detached from the garments, which are thought to be oligomers from the fiber manufacturing process [3.50]. As these microparticles are also released to the environment, an evaluation of an inclusive definition that contemplates the "total released material to the environment" should be considered in case of a terminology standardization.

3.3.3 Detached MFs

As previously indicated, based on previous publications, the operational parameters described in Materials and Methods were selected to get the lower MFs' detachment conditions for the tested garments. Total MFs detached per garment is plotted in Figure 3.6. The evaluation of the detached MFs is referred to the 5th washing cycle, as it is the point where the MFDR stabilizes.

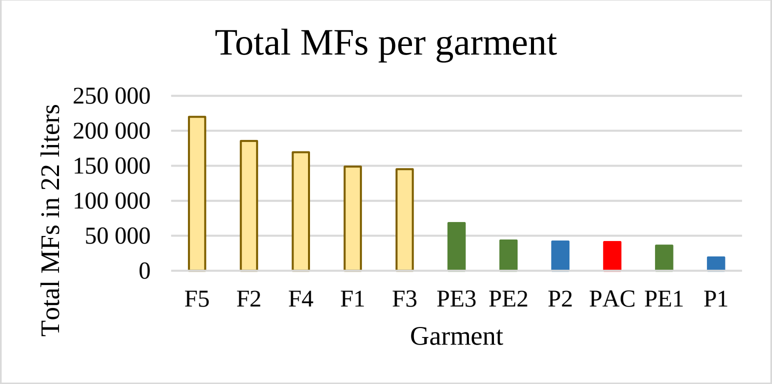


Figure 3.6. Total amount of detached MFs per garment for a total effluent of 22 liters (5th washing cycle).

When considering absolute values, fluffy garments detached the most MFs, followed by a non-obvious pattern of garments. However, absolute values are only useful to appreciate the difference between whole finished garments and to efficiently inform about this issue to the consumers, but should be avoided when the objective is to achieve fundamental conclusions on the MFs' detachment behaviors of the textiles. For this reason, the detachment of MFs was also expressed in two other different units (MF/m² and MF/g) and evaluated with the superficial density of each garment (Figure 3.7A and B):

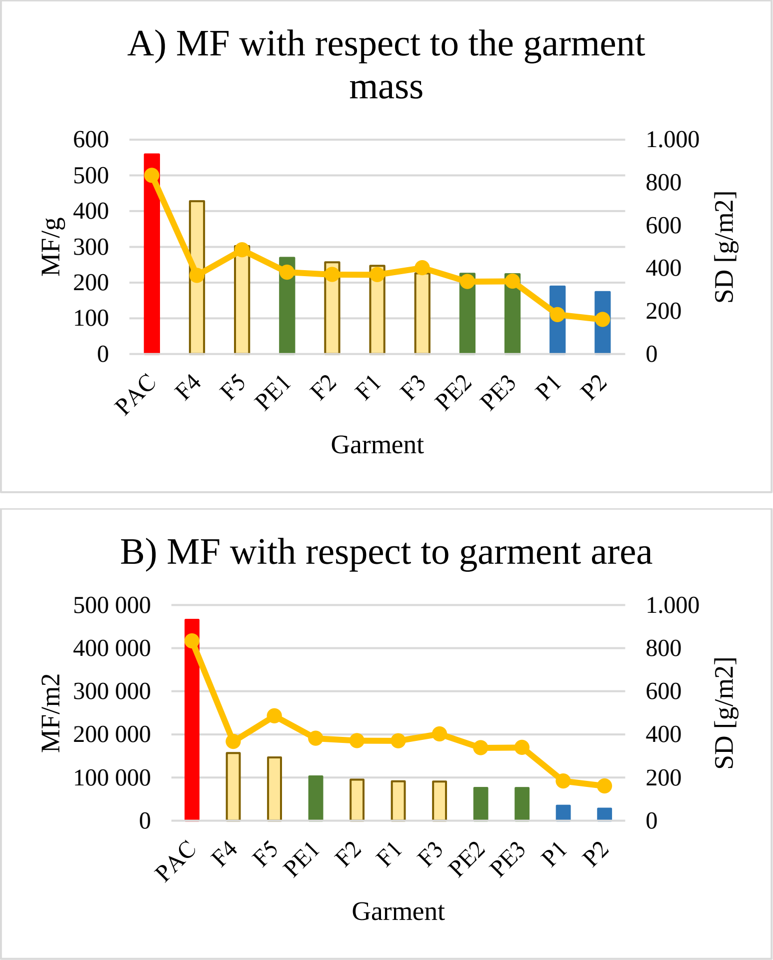


Figure 3.7. The MFDR divided by the garment (A) mass and (B) area. The Superficial Density "SD" of each garment is indicated by yellow points. (Results expressed for a total effluent of 22 liters and taken from the 5th washing cycle).

As expected, both units allow better comparability of the results and should be used when evaluating MFs' detachment trends. Firstly, from Figure 3.7 it can be seen that the acrylic/polyamide garment (PAC) had by far the highest MFDR (560 MF/g or 465’000 MF/m^2), which might be a consequence of the textile type (knitted fabric and a tassel formed by a low twist yarn, which seems to have a major role in the MFDR); whereas fluffy garments situated at the middle, followed by polyester/elastane and other polyesters (175 MF/g or 30’000 MF/m^2). However, it should be noticed that different operational conditions of the washing machine (e.g., washing time, temperature, etc.) than the ones used for our trials might give other results, although the relative detachment rates between garments should remain constant.

In addition, a positive correlation was found between the surface density (SD) and the MFDRs. Adjusted R^2 was 0.71 for MF/g and 0.89 for MF/m^2. Also, the relative MFDRs between the garments remained unchanged when unifying the results with respect to the garment mass or area. This means that although the SD is an important predictor of the MFDR, other factors also influence it, as e.g., the garment material and fabric type. For this reason, it is a purpose for future works to make an exhaustive evaluation of them.

Finally, by using Equation 3-1 and 3-2 with an average length of 0.3 mm and a diameter of 20 μm, results were transformed to mass loss of MFs. In this way, ranges of 2 to 29 mg/garment, 23 to 73 mg/kg of garment and 4 to 61 mg/m^2 of garment were obtained.

As seen in Table 3. 6, when referring to the number of MFs, our results are mostly higher than previous publications, although in some cases they are similar or even lower. However, when related to the weight, our results are always lower. These discordances could be because different methods and factors were used. For instance, particles that are not MFs might have been weighted and reported as MFs. Also, in some cases, the units used to report the results were confusing or even useless. Hence, it is recommended to standardize a method with clear parameters and to unify the observations with respect to the garment area and/or weight (MFs and/or milligrams of MFs per unit of area and weight of the garments). Finally, it should be pointed out that a strict comparison is always affected by the intrinsic inter-laboratory variability, due to factors such as the washing machine and washing cycle, water quality, etc.

Table 3. 6. Previous published works information and comparisons with the present work.

Reference	Comments on the analytical method	Bibliographic results	Our results expressed in the same units
Browne et al., 2011 [3.37]	No clear information of the methodology used. Conservative estimations reported.	130 – 280 MF/L per garment > 1’900 MF/garment	1’500 – 10’000 MF/L per garment > 30’000 MF/garment
Napper & Thompson, 2016 [3.39]	Indirect method ⁽ᵃ⁾. Fibers with a mean length > 5 mm considered.	140’000 – 730’000 MF/6 kg of washed garments 500’000 MF/mg	1’000’000 – 6’500’000 MF/6 kg of washed garments 5’760 – 11’521 MF/mg (b)
Pirc et al., 2016 [3.40]	Indirect method (a). Filters used of 200 μm; mean length considered > 5 mm.	135’000 MF/6 kg of washed garments	1’000’000 – 6’500’000 MF/6 kg of washed garments
Hartline et al., 2016 [3.41]	Indirect method (a).	29 – 431 mg of MF/garment washed (front-load) 1’471 – 2’121 mg of MF/garment washed (top-load)	2 – 29 mg of MF / garment washed (c) (front-load)
Åström 2016 [3.42]	*Gyrowash* to simulate washing machine.	7’360 MF/(m^2 L)	1’200 – 33’000 MF/(m^2 L)
Salvador et al., 2017 [3.43]	Used Napper & Thompson (2016) and Pirc et al. (2016) methods.	184’000 – 250’000 MF/garment	30’000 – 230’000 MF/garment
De Falco et al., 2018 [3.44]	*Linitest* apparatus used to simulate washing machine. Direct quantification.	6’000’000 – 17’700’000 MF/5 kg of washed garments	1’000’000 – 6’500’000 MF/6 kg of washed garments

(a) Indirect method: the quantification is estimated from the weight, the length, and/or the density of the MFs.
(b) Estimated by applying the calculation methodology explained in ¡Error! No se encuentra el origen de la referencia. and using an average MF diameter of 20 μm and a MF length between 0.2 to 0.4 mm.
(c) Same procedure than (b) but using an average MF length of 0.3 mm.

3.3.4 Estimation of the Textile Microfibers' Global Input to the Oceans

The last estimation of the global textile MFs' flow to the environment was published by Boucher & Friot (2017). They approached three scenarios (minimum, central and maximum) based on previously published works. However, the quantity of textile MFs reaching the oceans (Table 3.7) was estimated using data from works in which their purpose was not to evaluate the MFDRs [3.37, 3.51–3.53].

Hence, in this work, the textile MFs' flow to the oceans is re-estimated on the bases of the following assumptions:

a. The approaches of the annual laundry cycles per capita, load per standard wash, and regional availability of wastewater technologies and population are still the ones proposed by Boucher & Friot (2017).

b. The data for Boucher & Friot was obtained by using a MF linear weight of 300 *dtex*. In the present work, the linear weight of the MFs was estimated by applying the methodology described in section 3.3.3. In this way, MFs were considered to have an average length of 0.3 mm and a maximum diameter of 20 μm (Figure 3.8). Applying those values, a linear weight of 4.34 *dtex* was obtained and adopted to proceed with the analysis. This value is consistent with ranges reported for typical polyester filament yarns [3.54].

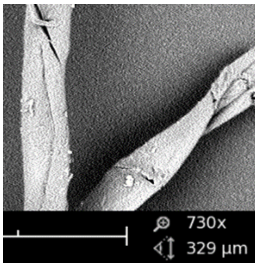


Figure 3.8. Example of typical PES microfiber. The distance between both microfibers is 40 μm, hence, one microfiber has approximately 20 μm.

c. As an uneven number of garments of each group were tested, the same weight was applied to every group in order to homogenize the observations. This was done by calculating the MFDRs' average within the garments of each group (refer to Table 3.1); those outcomes were used to determine the resulting average between the groups, which was considered as the central value. We considered that garment PAC is not a representative sample for the purpose of this analysis, as this type of garment is less frequently used and/or laundered. Hence, it was removed from the data.

Therefore, we recalculated the different scenarios of MFs reaching the oceans. The results are indicated in Table 3.7:

Table 3.7. Recalculated values for textile microfibers reaching the oceans.

Results		**Boucher and Friot (2017)**		**Present Work**	
Values (mg of MFs per kg of garments)	Minimum	300		23	
	Central	900		33	
	Maximum	1'500		56	
Total MFs reaching the oceans using central values		Tons MF/year	MF/year (a)	Tons MF/year	MF/year
		520'000	$3.6\cdot10^{15}$	17'830	$1.4\cdot10^{17}$

(a) Estimated using the 300 ***dtex*** and 5 mm for a MF length assumed in Boucher & Friot's work**.**

From Table 3.7 we can conclude that, based on Boucher & Friot's calculations and with respect to our results, they might have overestimated the mass flow rate of MFs to the oceans. This discrepancy is mainly because the linear weight applied to calculate the mass of the MFs is presumably overestimated. In fact, the calculations of Boucher & Friot derive from using a linear weight of 300 dtex, which is a common value of yarns composed of a group of individual filament fibers [3.46, 3.54]. This factor was firstly applied in a report of the Norwegian Environmental Agency [3.52] and later assumed as appropriate in most of the subsequent publications [3.36, 3.41, 3.51, 3.55]. However, in this particular case, less does not necessarily means better, since using the values of Boucher and Friot a particle flow of $3.6\cdot10^{15}$ MFs/year can be estimated, which is a 2.7% of the $1.4\cdot10^{17}$ MFs/year calculated with our results. Moreover, it should be underlined that the values reported by Boucher & Friot were estimated with an assigned MF length of 5 mm, in contrast with the average of 0.3 mm measured in this work. As a consequence, according to our results, smaller and more easily ingestible MFs are heading towards the oceans.

3.4 Conclusions

A direct and highly reliable method to quantify the detachment of textile microfibers from whole finished garments was developed and applied. In order to normalize the microfiber detachment rates results, comprehensive and comparable results are needed. In this way, we recommend a set of units that give fundamental conclusions of the microfiber detachment with respect to the textile article. In addition, a methodology to estimate the relation between the number of MFs and their mass was developed.

From consecutive washing trials, it was found that the microfiber detachment rate (MFDR) decreases until stabilization is reached in the 5th washing cycle. The MFDR in that point is between 175 to 560 MF/g or 30'000 to 465'000 MF/m^2 of garment. It was also found a high and positive relation ($R^2 = 0.71$ to 0.89) between the MFDR and the superficial density (g/cm^2) of the garment. Transforming the results into units of mass, we estimated a MF loss between 23 to 73 mg/kg of garment or 4 to 61 mg/m^2 of garment.

Moreover, the morphology of the microfibers was analyzed, and two different shapes were found: one group that comes from microfibers that were already loosely entangled with the fibers' grid of the garments, while the other corresponds to microfibers that were ripped-off from the fiber grid as a consequence of the mechanical stress suffered in the launderings. This latter case could be perpetuated by the garment use and its UV degradation. With respect to the microfiber length, it was found that it decreases from the 1st to the 3rd washing cycle. Both findings are helpful to evaluate the applicability of new microfibers' reduction solutions in different steps of the garment life cycle.

Finally, our results were used to re-estimate the mass flow of microfibers to the oceans, which was found to be overestimated by other authors. However, according to our results, the amount of MFs reaching the oceans is $1.4 \cdot 10^{17}$ MFs/year, which is higher than the value obtained when our calculation methodology is applied to the previously published data. This implies that a higher quantity of smaller and more easily ingestible microfibers is heading towards the oceans[1].

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Chapter 4:

Textile Microfibers Reaching Aquatic Environments: A New Estimation Approach

4 Textile Microfibers Reaching Aquatic Environments: A New Estimation Approach

This chapter provides a base model to estimate the mass flow of textile microfibers from household laundry into aquatic environments.

This chapter corresponds to the pre-prints of the following paper, that has been published in Environmental Pollution (Elsevier Journal, IF = 8.071. Q1, ScienceDirect):

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DOI: 10.1016/j.envpol.2020.114889

Some of the supplemenary information of the article has been included in Chapter 10, "Annexes".

Abstract

Textile microfibers are one of the most important sources within primary microplastics. These have raised environmental concerns since its recent identification as pollutants. However, there are still no accurate models to assess their contribution to the microplastic pollution. Hence, in this study, a method to estimate the mass flow of microfibers detached from household laundry that reaches aquatic environments has been developed. The method considers a set of parameters related to the detachment of microfibers, which are, basically: (1) the detachment rate of microfibers from different textile garments, (2) the volume of laundry effluents, (3) the percentage of municipal water that has been treated, (4) the type of used-water treatment applied, and, (5) the proportion of front- versus top-loading washing machines. In this way, 0.28 million tons of microfibers per year were estimated to reach aquatic environments, which is approximately half than the last published valuation. Finally, hypothetical situations were simulated to evaluate the reduction of microfibers by the modification of some of the parameters at different levels (consumer, government entities, and industry). Thus, depending on the implanted alternatives, microfibers that reach the aquatic environments could be reduced between 30% to 65%.

Keywords: Microplastic; Microfiber; Detachment; Pollution; Contamination.

4.1 Introduction

Microplastics (synthetic polymers < 5 mm in diameter) are a mix of pollutants that have been identified in every explored ecosystem. In numerical terms, it has been estimated that there are between 15 to 51 trillion buoyant microplastics in the marine environments [4.1]. However, these represent a minor fraction of the total extent of the MPs polluting the oceans [4.2]. Sources of microplastics (*MPs*) are mainly distinguished between *primary*, those emitted into the environment in a MP size range; and *secondary*, those generated in the environment from degradation and fragmentation processes of larger plastic debris. In this way, primary MPs include a wide variety of sources (e.g., microfibers detached from textile garments, plastic pellets, tire dust); while secondary MPs have their origin in mismanaged plastic garbage [4.3].

Regarding their impacts, its ingestion across the trophic chain is evident, as these particles have been found in at least 200 species [4.4–4.6]. Also, measured effects include MPs' retention, trophic transfer, increased mortality, and endocrine disruption, among others [4.7–4.10]. Moreover, MPs can behave as long-distance vectors for invasive species and hydrophobic contaminants [4.11, 4.12]. Also, these pollutants have been extensively identified in products for human consumption as seafood, tap and bottled water, and table salt [4.13–16]. Nevertheless, human health risks are still an unknown area that needs further investigation.

To understand the causes, impacts, and possible solutions, evaluations concerning the sources' contributions must be done. In this way, first attempts to estimate the flow of textile microfibers (*MF*) to aquatic environments have already been executed. Most renowned estimations concerning MFs have established its flow between 0.2 to 0.5 million tons per year [4.3, 4.17]. However, in a recent publication, it was noticed that an inappropriate factor was being applied in the calculations of previous publications [4.18]. In particular, a fiber linear weight of 300 grams per 10'000 meters (300 dtex) was considered. However, a MF is an individual filament that has a linear weight between 1 to 5 dtex (1-5 g per 10'000 m) [4.19]. Besides, most of the estimations previously reported do not include a full description of the applied criteria, making a difficult task to replicate or update the results.

Henceforth, this research aims to establish a replicable baseline model to estimate the total generation of MFs from household laundering and the fraction of these that reach aquatic environments. The model is applied to several hypothetical scenarios. Results of estimations are discussed and MFs reduction strategies at government, industrial, and consumer levels are evaluated. The main parameters considered to establish the model, are the following: the range of MFs detachment rates per textile garment and washing cycle in a steady-state; worldwide trends of household washing machines (in particular, the proportion of washers' type and volume of laundry water effluents); municipal water treated per world region (specifically, percentage and technologies applied); and proportion of synthetic materials (mainly polyester, acrylic, and polyamide) used in the manufacturing of textile garments.

4.2 Materials and Methods

An extensive literature research was done to develop a new approach to estimate the MFs' flow to aquatic environments. Taking this into account, a research of the data regarding the textile MFs' detachment rates, types of washers and trends of their usage, the MF removal in municipal water treatment plants, and the geographic distribution of these data, was carried out. The washing machine trends, the efficiency of MFs' removal in municipal water treatment plants, and the geographic contribution of both parameters are discussed in sections 4.3.1 and 4.3.2. According to the collected information, the main parameters and values to be included in our calculations were organized and combined to develop the equations shown in section 4.3.3. On the other hand, to evaluate the MFs' contribution on a regional basis, the world was divided into 10 sections, following the criteria commonly used in the literature:

- **North America**: *Aruba, Bermuda, Canada, Cayman Islands, Guam, Puerto Rico, Saint Pierre and Miquelon, United States, U.S. Virgin Islands, British Virgin Islands.*
- **Latin America and the Caribbean***: Antigua and Barbuda, Argentina, Bahamas, Barbados, Belize, Bolivia, Brazil, Chile, Colombia, Costa Rica, Cuba, Dominica, Dominican Republic, Ecuador, El Salvador, French Guyana, Grenada, Guadeloupe, Guatemala, Guyana, Haiti, Honduras, Jamaica, Martinique, Mexico, Montserrat, Netherlands Antilles, Nicaragua, Panama,*

Paraguay, Peru, St. Lucia, St. Vincent / Grenadines, Suriname, Trinidad and Tobago, Uruguay, Venezuela.

- **Europe***: Albania, Austria, Belgium, Bosnia and Herzegovina, Bulgaria, Croatia, Czech Republic, Cyprus, Denmark, Faroe Islands, Finland, France, Germany, Gibraltar, Greece, Greenland, Hungary, Iceland, Ireland, Italy, Liechtenstein, Luxembourg, Macedonia, Malta, Montenegro, Netherlands, Norway, Poland, Portugal, Romania, Serbia (incl. Kosovo), Slovakia, Slovenia Spain, Sweden, Switzerland, Turkey and United Kingdom.*
- **NIS:** *Armenia, Azerbaijan, Belarus, Estonia, Georgia, Kazakhstan, Kyrgyzstan, Latvia, Lithuania, Moldova, Russia, Tajikistan, Turkmenistan, Ukraine, Uzbekistan.*
- **Pacific OECD** (*Australia, Cook Islands, Japan, New Zealand, Niue*) **and South Korea**,
- **Central Asia and China***: Cambodia, China, Hong Kong, Korea (North), Laos, Macau, Mongolia, Vietnam.*
- **South Asia***: Afghanistan, Bangladesh, Bhutan, Fiji, French Polynesia, India, Maldives, Nepal, Pakistan, Sri Lanka.*
- **other Pacific Asia***: American Samoa, Brunei, Burma (Myanmar), Indonesia, Kiribati, Malaysia, Micronesia, Nauru, New Caledonia, Papua New Guinea, Philippines, Salomon Islands, Samoa, Singapore, Taiwan, Thailand, Timor-Leste, Tonga, Vanuatu.*
- **Middle East and North Africa***: Algeria, Bahrain, Egypt, Iran, Iraq, Israel, Jordan, Kuwait, Lebanon, Libya, Morocco, Oman, Palestine, Qatar, Saudi Arabia, Sudan, Syria, Tunisia, United Arab Emirates, Western Sahara, Yemen.*
- **Sub-Saharan Africa:** *Angola, Benin, Botswana, Burkina Faso, Burundi, Cameroon, Cape Verde, Central African Republic, Chad, Comoros, Congo (Brazzaville), Congo (Kinshasa), Cote d´Ivoire, Djibouti, Equatorial Guinea, Eritrea, Ethiopia, Gabon, Gambia, Ghana, Guinea, Guinea-Bissau, Kenya, Lesotho, Liberia, Madagascar, Malawi, Mali, Mauretania, Mauritius, Mozambique, Namibia, Niger, Nigeria, Reunion, Rwanda, São Tomé and Príncipe, Senegal, Seychelles, Sierra Leone, Somalia (incl. Somaliland), South Africa, Swaziland, Tanzania, Togo, Uganda, Zambia, Zimbabwe.*

4.3 Results and Discussions

4.3.1 Washing Cycles Washing Machine Trends

The number of washers and their annual volume of water consumption were calculated for 2020 on the bases of Barthel and Götz (2013) studies. These authors estimated that in 2013 there were 840 million household washing machines, with an annual water effluent of 19.2 billion m^3. From their published tendencies, it can be foreseen that in 2020 it will increase up to 1.1 billion washing machines and a water consumption of 22.2 billion m^3. The new estimation of regional distribution of the washers and their annual water consumption can be seen in Figure 4.1 (a) and (b).

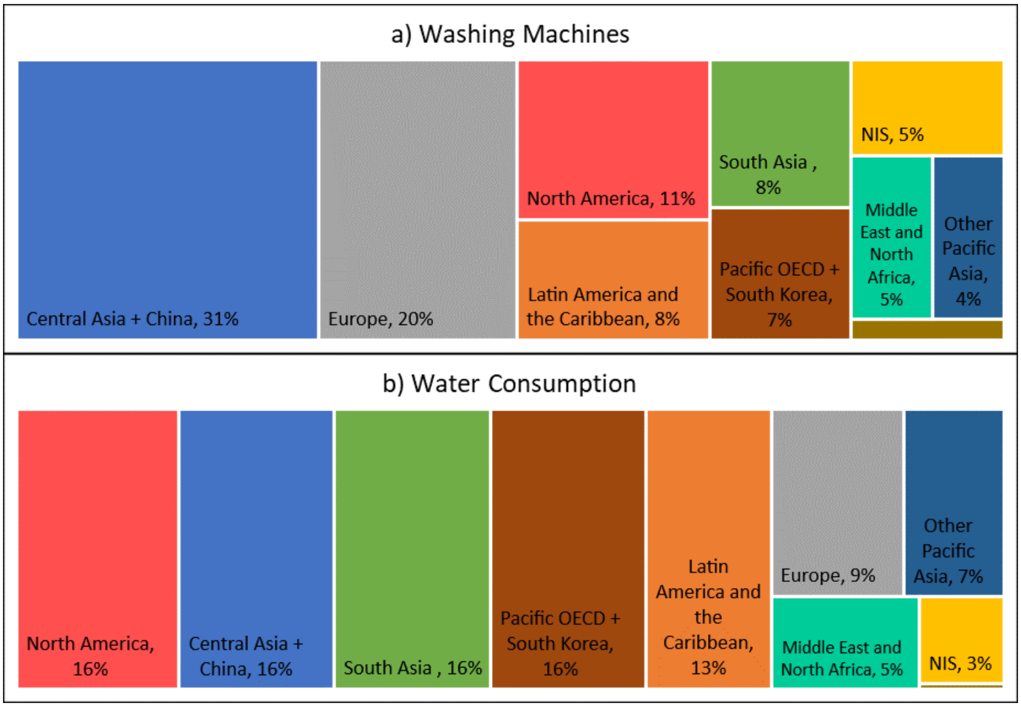

Figure 4.1. Trends for 2020 for (a) Washing machines distributed across the world. And (b) Worldwide water consumption for household launderings. In both cases, the smallest quadrangle corresponds to Sub-Saharan Africa (1% washing machines and 0.2% water consumption).

As seen when comparing Figure 4.1 (a) and (b), there is no direct relation between the number of washing machines and their water effluents. This is mainly a consequence of three aspects: the type of washing machine, the usage of newer and more efficient technologies regarding water and energy consumptions, and also the different regional behaviors on the selection of laundering programs. Based on Barthel and Götz (2013) studies the global yearly average water consumption per washer ($\bar{C}$ from Equation 4.1) was obtained as follows (see Table 4.1 for the data):

$$\bar{C} = \frac{\sum C_i}{\sum W_i} = \frac{22.218 \cdot 10^9 \; {m^3}/{year}}{1.154 \; \cdot 10^9 \; washer} = 19 \, {m^3}/{(year \cdot washer)} \; ; \; Eq.(4.1)$$

Where;

- $\bar{C}$ Global yearly average water consumption per washer.
- C_i Water consumption per region in ${m^3}/{year}$.
- W_i Number of washers per region.

Table 4.1. Washing machine (W) and water consumption (C) in different regions of the world for 2013 and 2020

	2013		2020	
	Washers (W)	C (m^3)	Washers (W)	C (m^3)
North America	107’000’000	3.85E+09	128’400’000	3.65E+09
Latin America and the Caribbean	69’000’000	2.42E+09	96’600’000	2.80E+09
Europe	193’000’000	1.93E+09	231’600’000	2.00E+09
NIS	58’000’000	6.96E+08	61’480’000	6.00E+08
Central Asia + China	247’000’000	3.21E+09	353’210’000	3.50E+09
South Asia	21’000’000	1.01E+09	86’940’000	3.50E+09
Other Pacific Asia	28’000’000	9.52E+08	49’000’000	1.50E+09
Pacific OECD + South Korea	71’000’000	3.55E+09	78’100’000	3.50E+09
Middle East and North Africa	39’000’000	1.05E+09	55’380’000	1.10E+09
Sub-Saharan Africa	6’400’000	1.98E+08	13’120’000	3.50E+07
Worldwide	**840’000’000**	**1.92E+10**	**1’153’830’000**	**22.22E+09**

The global yearly average water consumption for household laundry can be obtained as the quotient of the global discharged water and the total number of washers. This value was estimated at 19 m^3/washer. Hence, the regional efficiency of water consumption can be estimated from its variation to the global average.

In Figure 4.2, these variations are shown as positive when the water consumption is more efficient, and as negative when it is the opposite condition.

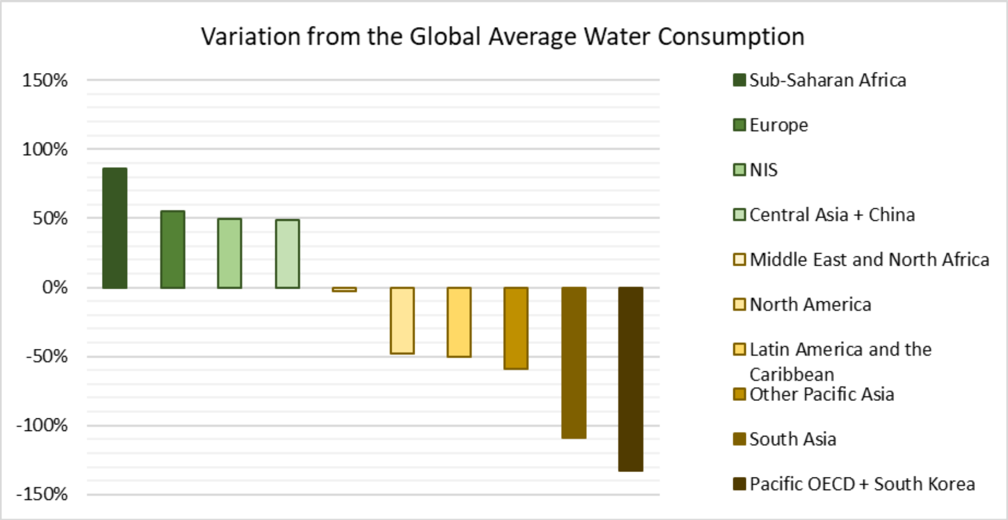

Figure 4.2. Regional variations from the global average use of water for the year 2020 (19 m^3 per washing machine). Positive percentages are more efficient in water consumption.

Concerning the type of washing machines, they can be divided into two groups: front- and top-loading washers (*FL and TL, respectively*). Their proportion across the globe has been only reported for certain regions (see Figure 4.3) [4.21]. In general, traditional top-loading washers use between 2 and 4 times more water than front-loading ones [4.22]. In the present work, a relation of 3 has been assumed, which is the average of both values. The estimation of MFs has been done considering three different scenarios of FL:TL proportions, which are explained in Section 4.3.4.

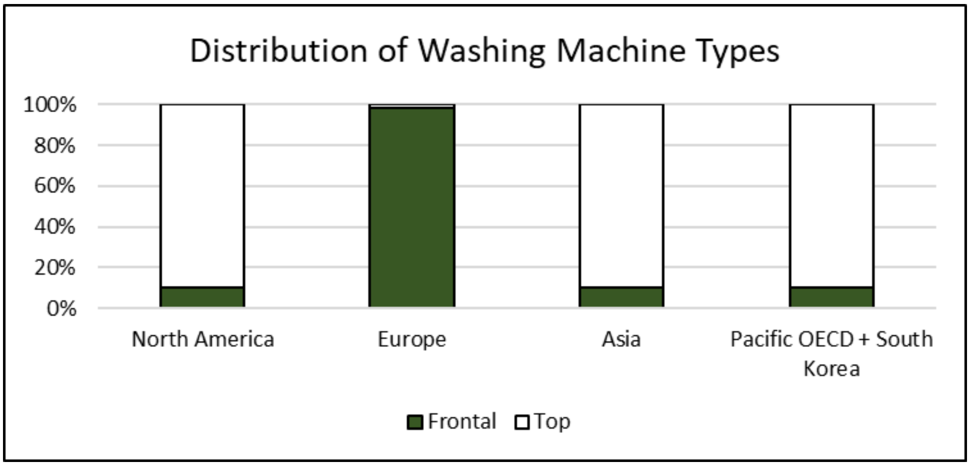

Figure 4.3. Percentage of frontal (FL) versus top-loading (TL) washing machines used in different regions of the world.

As can be seen in Figure 4.2 and Figure 4.3, most regions with high a proportion of top-loading washing machines have an intensive water consumption. On the other hand, Europe is in the opposite situation. It is worth to mention that the washing machine type is an important predictor for the MF detachment rate of a garment. In fact, it has been reported that TL detaches 7 times more MFs than FL per washing cycle [4.23]. Also, despite that the trends show that the washing machine types have been progressively shifting towards FL, the replacement will be slow. Hence, the proportion of FL versus TL is not expected to change in the next years [4.21].

4.3.2 MFs Removed in Used-Water Treatment Plants (UWTP)

As stated in different publications, in the UWTPs' processes the MFs are partially transferred from the liquid to the solid (sludge) stream. Hence, the rate of used-water treated at UWTPs is an important parameter to predict the proportion of MFs that will be discharged into water bodies, especially in those regions where a high percentage of the municipal used-water is treated.

Globally, between 75% to 80% of municipal used-water is discharged untreated into water receptors [4.24, 4.25]. However, the proportion of the population connected to urban UWTPs has a wide variation across the world. For instance, 97% of the municipal water is treated in Central European countries, whereas in Latin America and Asia this value decreases to 20% [4.17, 4.26–4.28]. In this way, the MFs flowing in the effluent of the washing machines will have different fates depending on the existence of a UWTP. In addition, the technologies installed between the regions are also unequal. This is a consequence of the cost of the technologies, as well as the regional economic status and legislation. Advanced treatment facilities are more expensive than primary and secondary processes. Hence, these are largely found in developed regions like Europe or North America [4.24, 4.25, 4.27–4.32]. This can be furtherly seen in Figure 4.4.

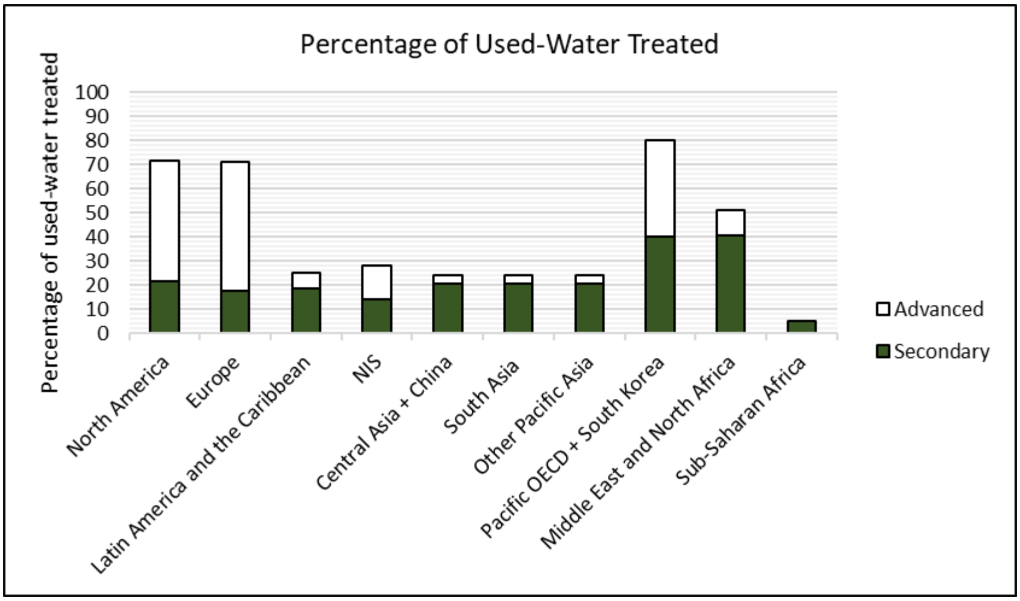

Figure 4.4. Percentage of municipal used-water treated and the proportion of applied technologies in the different regions of the world.

As explained before, the presence of a UWTP will be an important predictor of the fate of a MF. In addition, the removal efficiency from the liquid stream will differ between different technologies. These were estimated at 89% and 98% for secondary and advanced treatments, respectively (see Table 4.2). However, it must be noticed that MPs do not disappear in a UWTP, instead, they are only removed from the liquid and transferred to the solid stream, i.e., to the sludge. Henceforth, the MPs *transfer efficiencies* for different technologies can be seen in Table 4.2.

Table 4.2. Percentage of MPs transferred to the sludge for different used-water treatment technologies.

Reference	**Treatment**		
	Primary	**Secondary**	**Advanced**
Magnusson and Norén 2014 [4.33]	-	-	99.9
Gasperi et al. 2015 [4.34]	-	90.0	-
Talvitie et al. 2015 [4.36]	50.0	97.8	-
Michielssen et al. 2016 [4.37]	84.1	93.8	97.2
	88.4	89.8	99.4
Murphy et al. 2016 [4.38]	78.3	98.4	-
Carr, Liu, and Tesoro 2016 [4.39]	-	-	99.9
Leslie et al. 2017 [4.40]	-	72.0	-
Mintenig et al. 2017 [4.41]	-	-	97.0
Talvitie, Mikola, Setala, et al. 2017 [4.42]	97.0	99.9	99.9
Talvitie, Mikola, Koistinen, et al. 2017 [4.43]	-	-	40.0(1)
	-	-	98.5
	-	-	97.1
	-	-	95.0
	-	-	99.9
Gündoğdu et al. 2018 [4.44]	-	73.0	-
	-	79.0	-
Gies et al. 2018 [4.45]	-	99.0	-
Magni et al. 2019 [4.46]	-	84.0	-
Average (% of transferred MPs to the sludge)	**79.6**	**88.8**	**98.4**

(1) This value is discordant; hence, it was removed from the data when calculating the average MP transfer efficiency.

As seen in Table 4.2, UWTPs provide a significant microplastic reduction from the liquid stream. Nonetheless, given the high volumes that they treat, the remaining amount in the water effluent is still considerable. Also, as MPs are mostly transferred to the sludge, depending on the final disposal of this by-product, these pollutants might still be released into the environment (e.g., as compost) [4.47–4.52]. Indeed, a rough estimation has found that MFs annually dumped with the composted sludge into agricultural lands are between $6.3 \cdot 10^4$ to $4.3 \cdot 10^5$ tons in Europe and $4.4 \cdot 10^4$ to $3.0 \cdot 10^5$ tons in North America [4.53]. However, there is a lack of information regarding the MPs presence and its impacts on terrestrial environments.

Table 4.3 shows the expected percentage of MF's retention in different regions of the world by considering the percentage of treated water and the technologies applied. This information is furtherly used in the equations described afterwards.

Table 4.3. Percentage of municipal used-water treated and type of treatments per region of the world. Also, percentage of MFs retained as a function of the technologies applied.

	Municipal used-water treatment plants (UWTP)					
	Treated water %	Secondary treatment %		Tertiary treatment %		Retention %
Worldwide	20					
North America	72	30	89	70	98	96
Latin America and the Caribbean	25	75		25		91
Europe	71	25		75		96
NIS	28	50		50		94
Central Asia + China	24	85		10		85
South Asia						
Other Pacific Asia						
Pacific OECD + South Korea	80	50		50		94
Middle East and North Africa	51	80		20		91
Sub-Saharan Africa	5	100		0		89

4.3.3 Emission of Microfibers to the Environment: Developed Equations

By considering the parameters explained in the previous sections, an equation has been developed to achieve the following estimations: (a) the quantity of MFs reaching aquatic environments, and (b), the effects that different mitigation strategies could have on the MFs' pollution.

Before introducing this equation, some parameters should be defined. Firstly, a volume distribution factor for each type of washer is required to relate the proportion of front (F) and top (T) loading washers and their volume of effluents. These factors, called "W_F" and "W_T", are obtained from Equations 4.2 (a) and (b):

$$W_F = \frac{P_F \cdot V_F}{(P_F \cdot V_F) + (P_T \cdot V_T)} \quad ; Eq. 4.2a, \quad W_T = \frac{P_T \cdot V_T}{(P_F \cdot V_F) + (P_T \cdot V_T)} \quad ; Eq. 4.2b$$

Where,

- $W_F; W_T$ Volume distribution factor for the effluents of front (F) and top (T) loading washers.
- P_F, P_T Proportion of each type of washer.
- V_F, V_T Average volume of effluent from each type of washer.

Also, an additional factor of MFs detachment between top- and front-loading washers "y" is introduced, which can be calculated from Equation 4.3:

$$y = \frac{M_T}{M_F} \cdot \frac{V_F}{V_T} \quad ; Eq. 4.3$$

Where,

- y Factor of MF detachment between top- and front-loading washers.
- M_F, M_T MFs detached in front (F) and top (T) loading washers.
- V_F, V_T Average volume of effluent from each type of washer.

In this way, Equation 4.4, developed to calculate the annual flux of MFs reaching aquatic environments, is described hereafter:

$$\boldsymbol{F_A} = \overbrace{f_{MF}}^{\text{Microfibers detachment}} \cdot \underbrace{Q_{WM}}_{\text{Laundry effluents}} \cdot \overbrace{[D_{UA} + I_{UA}(1 - R)]}^{\text{Municipal treated waters}} \cdot \underbrace{S}_{\text{Synthetic vs. Natural}} \cdot \underbrace{(W_F + y \cdot W_T)}_{\text{Washing machine}} \quad ; Eq. 4.4$$

Where,

- $\boldsymbol{F_A}$ **Annual flux of MFs reaching aquatic environments, in MF/year.**
- f_{MF} Flux of MFs per liter of water effluent in a front-loading washer, in MF/L.
- Q_{WM} Annual volumetric flow of washing machines effluents, in L/year.
- D_{UA} Proportion of municipal used-water directly discharged to aquatic environments.
- I_{UA} Proportion of treated municipal used-water.
- R Proportion of retained MFs as a function of the existing municipal used-water treatment technologies.

- S Proportion of synthetic versus natural fibers used globally in the manufacture of textile garments.
- $W_F; W_T$ Volume distribution factor for the effluents of front (F) and top (T) loading washers (Eq. 4.2 a and b).
- y Factor of MF detachment rate between top versus front-loading washing machines (Eq. 4.3).
- P_F, P_T Proportion of each type of washer.
- V_F, V_T Average volume of effluent from each type of washer.

When the total mass of MFs generated or emitted from the laundering process has to be estimated (F_E , annual flux of MFs emitted, in MF/year), Equation 4.4 is reduced to equation 4.5:

$$\boldsymbol{F_E} = f_{MF} \cdot Q_{WM} \cdot S \cdot (W_F + y \cdot W_T) \quad ; Eq. 4.5$$

Excluding f_{MF}, S and y, all factors were applied for a determined region of the world. Hence, the sum of all F_E gives the global mass of MFs emitted and the sum of F_A gives those reaching aquatic environments. Also, for the parameter f_{MF}, minimum and maximum values were applied (see Table 4.4).

Also, to express the annual flux of MFs in mass units, the procedure of Belzagui et al. (2019) was applied. According to this method, the linear weight of an individual filament fiber is calculated with Equation 4.6:

$$\boldsymbol{C} = \emptyset^2 \cdot \frac{\pi \cdot \gamma}{400} \quad ; Eq. 4.6$$

Where,

- C Linear weight of the MFs, expressed in *decitex* (1 dtex = 1 g per 10’000 m).
- $\emptyset$ Average diameter of the MFs, in µm (19 µm, see Table 4.4).
- γ Specific weight of the fibers, in g/cm³.

Then, by applying the values of F_E, F_A and C obtained from Equations 4.4, 4.5 and 4.6, the annual mass flux of MFs is estimated with Equation 4.7:

$$\boldsymbol{mF_{MF}} = F_{A\ or\ E} \cdot C \cdot \bar{L}_{MF} \cdot \frac{1}{10^9} \quad ; Eq. 4.7$$

Where,

- mF_{MF} Annual mass flux of MFs, in ton MF/year.
- $\bar{L}_{MF}$ Average length of MFs, in mm (0.343 mm, see Table 4.4).

C Linear weight of the MFs. In this equation, the value is applied in g/m.

4.3.4 Emission of Microfibers to the Environment: Estimations

Initial approaches

The developed equations were applied to estimate the MFs emitted from household laundering and reaching the aquatic environments. With this purpose, the following approaches were made:

- The current proportion of synthetic textile fibers from the overall production is approximately 0.62 [4.54,4.55]; this value corresponds to “S” in Eq. 4.4.

- The volume proportion factors (W_F and W_T) were calculated by means of Equations 4.2 (a) and (b), assuming an average volume of effluent between top- and front-loading washers of 3:1.

$$W_F = \frac{P_F}{P_F + 3P_T} \quad (a); \quad W_T = \frac{3P_T}{P_F + 3P_T} \quad (b)$$

- Top-loading washers detach 2.2 times more MFs per liter than front-loading ones. This value corresponds to “y” in Equation 4.4 and was calculated by applying Hartline et al. (2016)’s values to Equation 4.3.

$$y = \frac{M_T}{M_F} \cdot \frac{V_F}{V_T} = \frac{1`736\, mg}{210\, mg} \cdot \frac{36\, L}{136\, L} = 2.2$$

- An average loading of 75% of a common washing machine was considered [4.21]. This corresponds to 4 kg of garments washed per laundry cycle.

- As there is no information regarding the washer type for some regions, **three different scenarios were established**. In all these scenarios, regions with information maintain their known proportions. However, to include the possible settings, regions without information were considered to have ratios of front versus top-loading (FL:TL) washers of 7:3, 5:5, and 3:7 in scenarios S1, S2, and S3, respectively. See complete data values for P_F and P_T in tables found in Chapter 10, "Annexes".

The minimum and maximum MFs detachment rates, and the physical characteristics of the MFs are shown in Table 4.4.

Table 4.4. Published MFs` detachment rates and their physical characteristics

Characteristic	**Units**	**Belzagui et al. 2019 [4.18]**	**De Falco, Gullo, et al. 2018 [4.56]**
Minimum MF detachment (1)	MF/L	**30'303 (2)**	80'000 (3)
Maximum MF detachment (1)	MF/L	196'970 (2)	**236'000 (3)**
MF length	mm	0.30	0.38
MF diameter	µm	20	18
Average linear weight (4)	dtex	3.8	
Average MFs' mass	MF/mg	**7'587**	

(1) MFs detached from a 4 kg front-loading washing machine.
(2) Calculated from Belzagui et al. (2019) by extrapolating their results to 4 kg of garments load.
(3) Calculated from De Falco, Gullo, et al. (2018) by extrapolating their results to 4 kg of garments load and assuming a washing machine effluent of 60 L.
(4) The specific weight of polyester was used as it is the most produced material. Hence, a value of 1.38 g/cm^3 was applied.
Data in light blue was used for the optimistic and pessimistic MF detachment rates.

Emission of MFs

After applying minimum and maximum MFs' detachment rates in Equations 4.4 and 4.5, a range of values for the three scenarios (S1, S2, and S3) were estimated. Henceforth, on a worldwide base, the total generated MFs from household laundering ranged from 0.47 to 0.49 million tons of MFs per year, or $3.6 \cdot 10^{18}$ to $3.7 \cdot 10^{18}$ MFs particles per year. Scenario

S2 is illustrated in Figure 4.5, where it can be seen that most of the MFs are generated in North American and Asiatic countries.

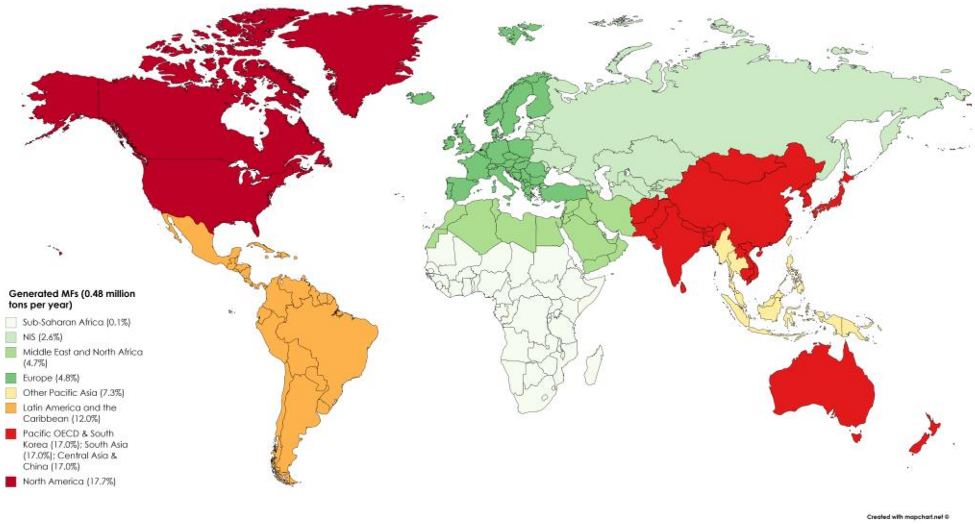

Figure 4.5. Total emitted mass of microfibers from household laundering (scenario 2). The relative contribution of each region is classified into three levels: lower (green), medium (yellow), and higher (red), illustrated with different intensities.

However, the MFs reaching aquatic environments are only those that are not subjected to any UWTP or MF retention system (see Figure 4.6). In this case, the mass flow of MFs flowing to aquatic environments was found to fluctuate between 0.27 to 0.28 million tons of MFs per year. This means that $2.1 \cdot 10^{18}$ to $2.2 \cdot 10^{18}$ MFs particles are being annually discharged into aquatic environments. In Figure 4.6 it can be seen that Asia is the most polluting region in terms of MFs reaching aquatic environments, followed by Latin America and the Caribbean. On the other hand, North America and Pacific OECD & South Korea have a lower impact than expected on water systems due to the application of retention measures. From these data, it can be approximated that the quantity of MFs being retained by UWTPs is 0.20 million tons per year, or around 40% of the total generated MFs. Unfortunately, these MFs will have an uncertain disposal and might still end up as litter.

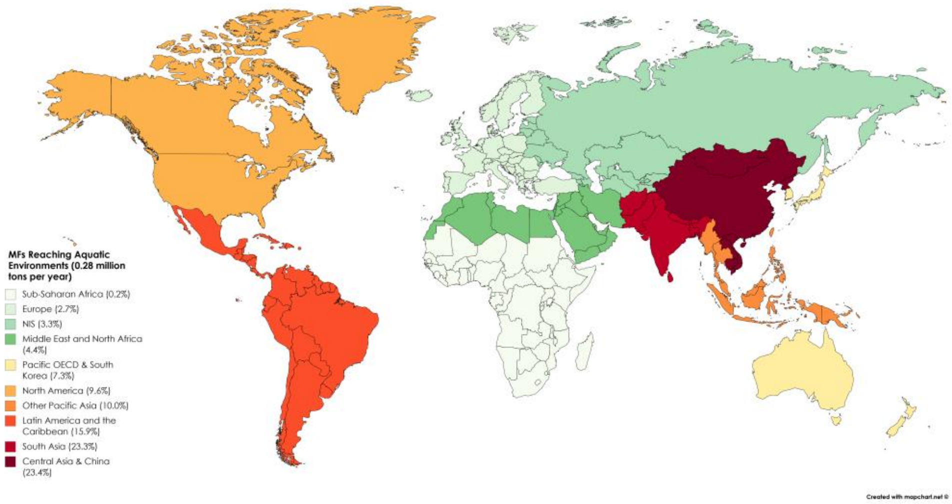

Figure 4.6. Mass flow of microfibers reaching aquatic environments (scenario 2). The relative contribution of each region is classified into three levels: lower (green), medium (yellow), and higher (red), illustrated with different intensities.

The high proportion of MFs that reaches aquatic environments from Asia (65%) is consistent with the considerable load of MPs exported by their rivers [4.57], and with the elevated concentrations of MPs found in the North Pacific ocean [4.1]. Besides, on a regional basis, the order of MF contribution to aquatic environments has no variation in any of the established scenarios. Hence, the simulated variations do not seem to have a major impact on the worldwide analysis. Finally, results were analyzed and the following conclusions were obtained:

- The volume of water discharged from the washers is an important predictor for the MFs' release, as greater volumes can be related to larger laundry programs. Also, the water consumption provides an intrinsic idea of two components: (1) the behavioral trends of every region about the selection of the washing program, partially reflecting the awareness of the consumers concerning the best use of the washing machines; and (2), the type and/or efficiency of the washing machine mostly used.

- Concerning the washing machine type, it was found to be as influential as the other parameters. This was proven by developing a hypothetical situation in which the proportion of washer types in Asia and North America was inverted from the existing front- versus top-loading washers of 0.1:0.9 to 0.9:0.1 (see 4.3.6).

- According to our calculations, about 40% of the worldwide MFs generated in washing machines are retained in UWTPs. A priori, this might seem an unexpected value as only 20% of world municipal waters are treated before the discharge. However, it is because some regions, as North America, generate a high percentage of the MFs but also treat a high proportion of their municipal waters. On the other hand, three observations are worth to be mentioned: (1), the worldwide percentage of treated water is still very low, meaning that most of the washers' effluents are being directly discharged to water receptors; (2), it should be noted that even primary treatments can be used as systems to reduce MFs from reaching aquatic environments. This means that their implementation in regions without UWTPs will considerably reduce the flow of MFs to aquatic environments; and (3), the implementation of UWTPs will not reduce the generation of MFs, and these do not disappear in UWTPs, instead, they are transferred to the sludge. In this way, these particles will be dumped to the soil if the sludge is used as compost. Hence, UWTPs are not a solution to the reduction of *MFs* flowing to the environment.

In Figure 4. 7 each regional change between the MFs emitted and those reaching aquatic environments can be visualized.

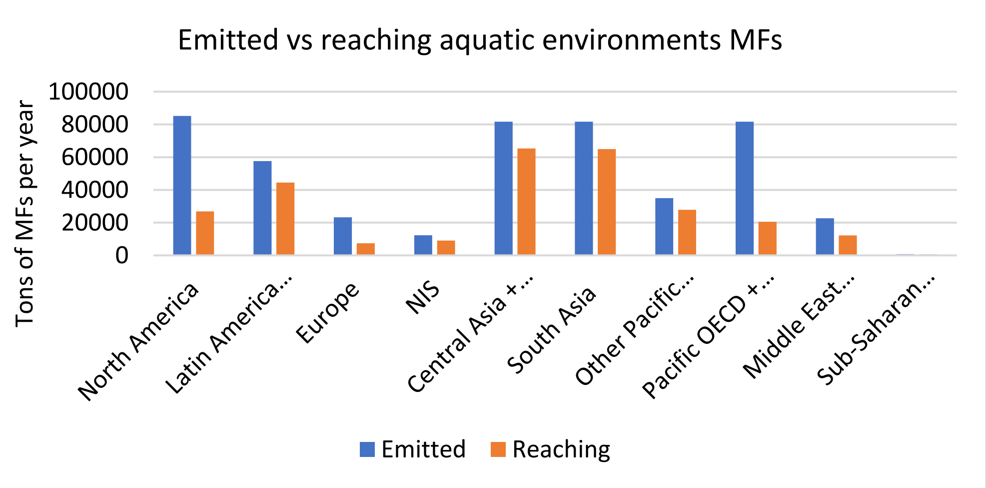

Figure 4. 7. MFs emitted versus those reaching aquatic environments.

Finally, it must be mentioned that regions with low washing machine ownership will also contribute to the pollution of MFs when doing the garments' hand-washing. These MFs are out of the limits of this study, as there is no reliable information available on that specific subject. Further sources of textile MFs that were out of the limits of this study are, for instance: industrial textile processes, drying, and usage of garments, etc. In this way, the expected total quantity of MFs reaching the environments will be higher than the estimations made in this work.

4.3.5 Comparison with Previous Estimations

A previous global estimation of the total quantity of MFs flowing to the oceans was made by Boucher and Friot (2017) [4.3]. An approximation of the equation applied in their work can be found in this section. In that research, the central value was calculated at 0.5 million tons of MFs per year. As seen in Section 4.3.4, the result found in this study is approximately 50% lower than their estimation. In Boucher and Friot (2017)'s methodology, the main approach was based on the number of laundries per capita. In contrast, in this study the volume of effluent from washing machines is used. However, it was found that an incorrect value was applied in one of their parameters. Specifically, a linear weight of 300 dtex was used to calculate the mass of the MFs. As explained before, a common linear weight for a MF is between 1 to 5 grams per 10'000 meters (1 to 5 dtex). If updated information and a correct linear weight for the MFs is applied in Equation 4.8, the mass flow of MFs reaching aquatic environments is estimated at 0.19 million tons per year. As seen, it provides an estimation 30% lower than the one calculated with the equations proposed in this study. In general aspects, the factors considered by Boucher and Friot (2017) can be encompassed in the next equation:

$$MF_{An} = WCP \cdot P \cdot L \cdot MF \cdot x \ \ ; Eq. 4.8$$

Where:

MF_{An}	Annual mass flux of MFs, in ton MF/year.
WCP	Average annual number of laundry cycles per capita.
P	Population.
L	Load per washing cycle (4 kg considered).
MF	MFs detached per kg of garment (mg MF/kg garment).

- x Factor probably referring to the proportion of population owning a washing machine and municipal water treated.

The values applied for estimating the annual mass flux of MFs for the year 2020 are:

- A value of 0.45 was considered for the "x".
- Global 2020 population of 8E+09.
- 244.5 mg of MFs detached per kg of garment.

Observations

- The trends of the world population aren't intrinsically related to the tendencies of the washing machines demand and usage. The global population grew from 7.2 billion to 7.8 billion between 2013 and 2020, which corresponds to an increase of 8%. In that same period, the number of washers increased from 0.84 billion to 1.15 billion, which is an increase of 27% [20,58,59]. The factors influencing these parameters are more complex, as the economic status of a region, the variations in the washing machines costs, etc. In this way, our methodology uses the average effluent per washing machine and per region. Hence, the trends of the consumers respecting the intensity of their washing cycles are indirectly considered, reducing the needs of assumptions.
- Published papers and reports with an inappropriate linear weight of 300 dtex ([3,17,60,61]).
- Despite applying 300 dtex for the microfibers, Boucher and Friot 2017's results were balanced by using an underestimated MF garment detachment of "more than" 1'900 MF/L, from Browne et al. (2011), for their maximum scenario

From these results, some observations and comments regarding the different methodologies need to be done. The type of washers (TL or FL) is an important factor in the MF detachment of a textile garment that needs to be included in these approximations [4.23]. Used-water treatment plants (UWTP) must be also considered when estimating the quantity of MFs reaching aquatic environments. Also, the trends of the world population aren't intrinsically related to the tendencies of the washing machines demand and usage). Hence, by using the volume of effluent per washing machine and region the needs for assumptions are reduced. Finally, studies should be careful with the in the units and the order of magnitude of the parameters applied to do the estimations.

4.3.6 Recommendation for MFs' Reduction

Hereafter, a set of hypothetical situations is presented to estimate the attained improvements from different possible MFs' reduction strategies. In this way, in situations HA, HB, and HC one single parameter was modified, whereas in the situation HD all the modifications were combined. In this sense, new hypothetical situations were created for manufacturers (HA), consumers (HB), and government (HC) levels. A summary of the situations is shown in Figure 4.8, where each central value is compared with scenario S2 (50% of front- and top-loading washing machines in regions without data).

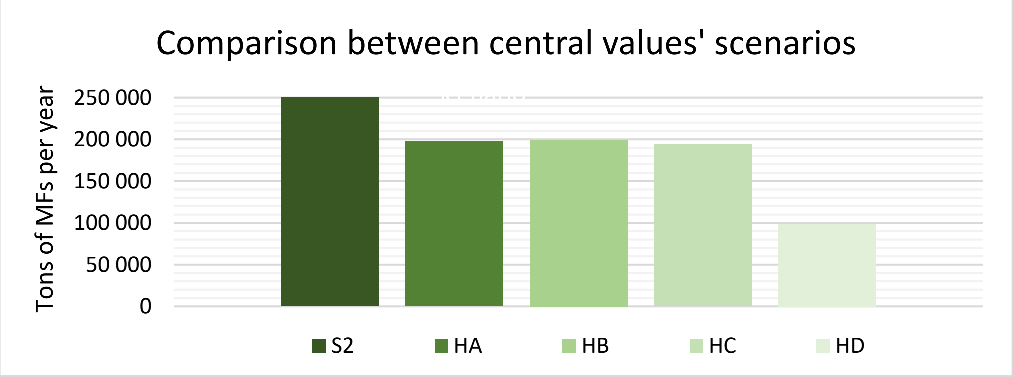

Figure 4.8. MFs released to aquatic environments. Comparison between the central value scenario S2, and the hypothetical situations HA (the type of washing machine), HB (consumers and water usage), HC (water treatment plants), and HD (combined effect).

The hypothetical situations shown in Figure 4.8 are explained hereafter:

Washing Machine Manufacturers (HA – Table in Annexes): As shown before (see Section 4.3.1), regions of Asia and North America have a 0.1:0.9 proportion of front- versus top-loading washers. Hence, a new hypothetical situation HA was considered by inverting them to 0.9 FL versus 0.1 TL. This modification resulted in a global MF reduction of 29%. It must be noticed that changing from TL to FL reduces the detachment of MFs. Hence, this strategy could have a major role in reducing not only MFs from reaching aquatic systems but to the whole environment too. However, other solutions or mitigation strategies are also feasible. For instance, improved designs of TL washing machines that cause less stress to the garments, or the marketing of new washing machines with built-in MFs' filters. Finally, manufacturers should also include in their brochures a qualification category regarding MFs emissions or stress induced to the garments in the washing machines. In this way, consumers could consider this factor when acquiring a new washer.

Consumers (HB – Table in Annexes): The population awareness on the MFs' contamination and their capacity to reduce their contribution are important subjects that must be continuously consolidated. In the last years, social media platforms have been increasingly making publications on this topic [4.58–60]. Also, the words "Microplastic and Microfiber" have been appearing in newspapers and digital screens (e.g., 4.60–4.64). Consumers' contributions related to some MFs' reduction strategies are available on the web. Some examples reported as "better practices" are: washing less but enough, filling up the washing machine, using liquid detergents, selecting colder and quicker laundry settings, among others [4.66,4.67]. In addition, there are commercially accessible capturing MFs technologies, which work by capturing the MFs either inside the washing machine [4.68, 4.69] or in the effluent [4.70, 4.71]. These technologies have accomplished a MF reduction in the washer effluent of 26% to 87% [4.72]. Nevertheless, the final disposal of the retained MFs has not yet been afforded. On the other hand, using more natural than man-made fibers has also been mentioned within the possible solutions. This declaration is controversial, as nowadays most of the cotton industry relies on a highly pollutant and environmentally unsustainable production [4.73]. Hence, there are no justified studies to claim that specific statement.

The hypothetical situation HB was created by decreasing the water consumption in regions with a high consumption rate to the current worldwide average of 19 m^3/washer. Thus, a reduction of 29% on the generation of MFs can be achieved. This measure can be accomplished by instructing consumers to use quicker but adequate laundry programs and/or more efficient washers. It must be highlighted that this strategy will reduce the MFs' generation and, consequently, a decrease in the emission of MFs to the whole environment.

Government Entities and Used-Water Treatment Plants (HC – Table in Annexes): The existence of a UWTP has been demonstrated to play a relevant role to remove MFs from the liquid stream. Situation HC was applied in regions with a low percentage (<50%) of treated water. If these regions were to build enough installations to treat 60% of their municipal used-waters (without making any changes in their current proportion of treatment technologies), a global MF reduction of 31% could be achieved. Nevertheless, further investigation is needed to develop possible treatments for the MFs in the sludge, as this strategy will still introduce MFs into the environment.

As seen in Figure 4.8, a MF reduction of approximately 65% could be achieved in the situation **HD** (**Table in Annexes**) where all strategies of scenarios are combined. Also, each hypothetical situation has a different scope in the reduction of MFs. Situation HC (used-water treatment plants) only avoids MFs from reaching aquatic environments. On the other hand, situations HA (washing machine type) and HB (consumers' usage) could achieve a real MF reduction, as they reduce the generation of these particles. In this way, if only HA and HB scenarios were conducted (**HAB – Table in Annexes**), a MF reduction of approximately 50% could be attained. See complete data values for tables HA, HB, HC, HD, and HAB in Chapter 10, "Annexes".

Changes in the **textile industry** were out of the limits of this study, as there is no reliable data regarding MFs' reduction techniques applied in the manufacturing process of textile articles. However, as can be seen in studies on textile MFs, there is a wide variation on the MFs detachment rates. Hence, the textile industry can play a key factor in the reduction of these pollutants by enhancing their processes and products towards reducing MFs release. Some recommendations have already been published; for instance, a *Life European* project evaluated textile procedures as the spinning, cutting, dyeing, among others. In this way, they compiled a guideline of "better practices" for the textile industry [4.74]. In addition, investigations are working forward for possible techniques to reduce the MF detachment from the garments. For example, a reduction of 90% of the MFs release was obtained by applying pectin, poly-lactic acid, and polybutylene succinate onto polyamide fibers [4.75, 4.76]. Nonetheless, further investigation is required to develop sustainable techniques to avoid or reduce the MF detachment rates from textile articles.

As a recommendation, and based on what was seen throughout this article, there are some important gaps in the input data. Hence, some of the main parameters related to the MFs detachment that are advisable to consider to improve the estimations are: the operational conditions of the washing cycles (temperature, centrifugation, etc.); the physical properties related to the manufacture of garments (type of fabric, torsion, etc.) and the different strategies that will be implemented to reduce the MF detachment or to retain the generated ones. Once the influence of these parameters has been established, the equations proposed in this work to calculate the MFs detachment can be upgraded to obtain more accurate estimations.

4.4 Conclusions

An estimation of the mass flow of microfibers (MFs) to aquatic environments was accomplished by developing a new calculation methodology. The method applies a set of known-parameters that are linked to the MFs' pollution, which are: (1) MFs detachment rate from different textile garments; (2) volumes of laundry effluents; (3) percentage of municipal used-water treated per world region; (4) type of water treatment applied, and (5) proportion of front- versus top-loading washing machines. In this way, different scenarios were studied and a central value of 0,28 million tons per year of MFs was obtained, which is approximately 50% lower than previously published.

On a regional basis, 65% of all the MFs that reach aquatic environments come from Asia. The explanation for this major influence is a combination of the high proportion of top-loading washing machines, an inefficient water-usage in the washing cycles, a low rate of municipal water treated, and a high population density. In contrast, other regions such as Europe have a relatively low contribution to the MFs' pollution, basically, as a consequence of the opposite conditions. On the other hand, when estimating the overall mass of generated MFs in the laundering process, North America gets situated in the first place with 18% of the global MF generation, from where a high proportion of these MFs is retained in municipal water treatment plants.

In addition, three hypothetical situations were analyzed with the attempt to quantify the impacts on the MFs release and to make positive proposals able to be applied at government, industries, and consumer levels. Concerning the washing machine types, the current proportion of front- versus top-loading washers in the Asian region was inverted. In this way, a global MFs release reduction of 29% was accomplished. Regarding the consumers, regions with high consumption of water per laundry were matched to the worldwide average. Thus, the attained MF reduction was of 29%, meaning that it is an efficient and sizeable MF reduction strategy. Additionally, at a governmental level, the evaluation was done by increasing the percentage of treated water in regions with a low used-water treatment rate. By doing so, a global MFs' reduction of 31% MFs was achieved. Finally, if all strategies were combined, a MF reduction of 65% could be achieved. However, it must be noticed that while all measurements decrease MFs from reaching aquatic environments, only modifications in the washer type and washing behaviors (e.g., lower but sufficient washing time) could efficiently reduce the detachment of MFs. Henceforth, major importance should be applied in those strategies that tackle the generation of MFs[2].

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Chapter 5:

Review on alternatives for the reduction of textile microfibers emission to water

5 Review on alternatives for the reduction of textile microfibers emission to water

This chapter is a review on the different alternatives that have been suggested to reduce the textile microfibers pollution. It was published in the *"Journal of Environmental Management" (JENVMAN, Elsevier, IF = 8,910. Q1, JCR-WoS) with the following reference:*

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Highlights

- Microfibers' reduction approached from textile production to water treatments.
- Changing textile manufacturing processes is effective but complicated.
- Improving users' washing habits is a viable and effective option in short-term or medium-term.
- Applying additives or filters to the washers could be an effective short-term alternative.
- Further efforts in water and sludge treatments are required to handle the MFs.

Abstract

The microplastics are considered one of the most threatening pollutants. One of the main concerns is their continuous and cumulative flow to water environments, as they are very difficult to be removed. Microfibers (MFs) are a significant type of MPs, with textile articles as one of the most renowned sources. This review aims to provide the current status of these MFs as pollutants, discussing possible alternatives from the manufacturing until the final disposition of MFs. There are many alternatives to reduce these pollutants from reaching the environment but also gaps that need to be further evaluated and addressed. Besides, it should be noticed that alternatives could be complementary between them. Some viable and non-contaminating solutions to reduce this pollution are currently on the market. Also, one relevant aspect is the final disposition or usage of the retained MFs to avoid them from reaching aquatic environments.

5.1 Introduction

The still under-revision definition of the European Chemical Agency (ECHA) indicates that the microfibers (MFs) are particles with a length to diameter ratio > 3 and a maximum length of 15 mm (ECHA, 2019). If those come from chemically modified and/or non-biodegradable polymers, they are considered as a type of microplastics (MPs). These pollutants have been constantly found contaminating every ecosystem. It has been estimated that there are from 15 to 51 trillion floating MPs in marine environments and 14 million tons in the top 9 cm of sediments of the world's oceans (Barrett et al., 2020; UNEP and GRID-Arendal, 2016; van Sebille et al., 2015). Sources of MPs can be distinguished between primary, those emitted into the environment in a MP size range; and secondary, those generated in the environment from physical degradation and fragmentation processes of larger plastic debris. In this way, primary MPs include a wide variety of sources (e.g., MFs detached from textile garments, plastic pellets, tire dust); while secondary MPs have their origin in mismanaged plastic garbage (Boucher & Friot, 2017). Textile MFs have received significant attention as these have been extensively found across the environment. For instance, Alavian Petroody et al. (2020) reported that most of the MPs found in water treatment plants were MFs, from which polyester MFs were identified at the highest concentration, followed by polyamide and acrylic. This is in line with the main synthetic fibers that are manufactured across the world.

Regarding their impacts, the ingestion across the trophic chain has extensively been reported, as these particles have been found in at least 200 species (Collignon et al., 2012; Fossi et al., 2014; GESAMP, 2015a; Ohkubo et al., 2020; Patterson et al., 2019). Also, some works have found that these particles can potentially have negative effects, such as MPs' retention, trophic transfer, and endocrine disruption, among others (Jemec et al., 2016; Nelms et al., 2018; Rochman et al., 2014; Welden & Cowie, 2016). Moreover, MPs can behave as long-distance vectors for invasive species and hydrophobic compounds (Browne et al., 2013; Mao et al., 2020; Rochman et al., 2013; Ta & Babel, 2020; Turner et al., 2020). Besides, these contaminants have been identified in human-consumption products as seafood, tap and bottled water, fruits, vegetables, and table salt (Abidli et al., 2019; G. Chen et al., 2021; Cox et al., 2019; Oßmann, 2021; Rochman et al., 2015;

Schymanski et al., 2018; Shruti et al., 2020; Teng et al., 2019; Van Cauwenberghe & Janssen, 2014; Yang et al., 2015). Nevertheless, the potential risks for human health are still an unknown area of study (Vital et al., 2021).

This review aims to evaluate the situation of different alternatives that have been suggested to reduce the textile MFs' pollution. The review is divided into four levels, (1) textile manufacturing, (2) garments laundering, (3) used water treatment plants, and (4) gaps that must be treated to close the loop of this specific MFs contamination. Figure 5.1 shows a summary of all that has been included in this document.

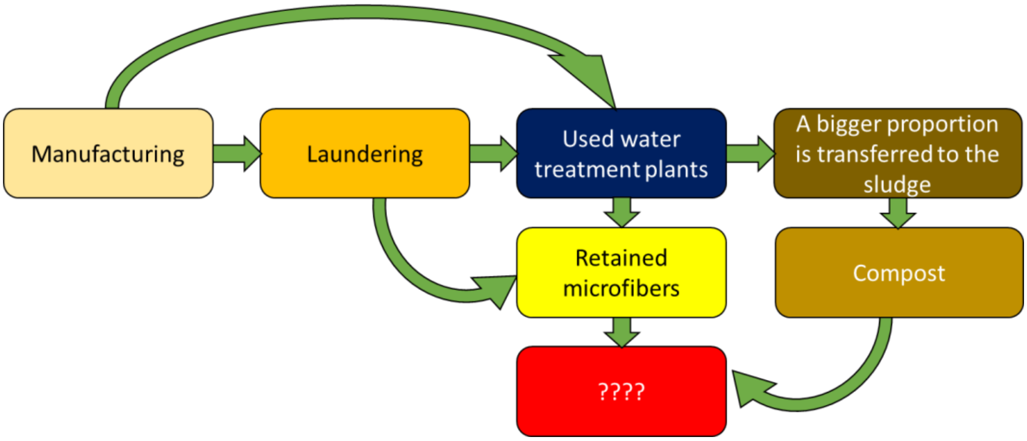

Figure 5.1. Summary of what has been included in this document. No circular trend for the microfibers; a lot of gaps to be studied.

To be more explicit, (1) textile manufacturing is referred to all treatments and proceedings applied in the production of textile articles in general before their use. (2) Garments laundering involves the washing and drying processes. (3) The effluents from laundering processes contemplate what happens when the MFs reach a possible water treatment plant and the effects that these particles have on the treatments. And (4), the gaps that this contamination has, followed by criteria, and requirements that alternatives to reduce this pollution should have. The MFs that can be generated in the daily use or final disposal of a textile article are out of the scope of this study.

5.2 Manufacturing, Dyeing, and Industrial Processes

The textile or apparel industry is the business for the manufacture of fibers, yarns, fabrics, clothing, and articles for home and/or decoration. Textile and clothing activities present different treatments, each with its own qualities and characteristics. These can have enormous variations between them (Bullon et al., 2017). In this sense, there is still a lack of rigor regarding MFs' reduction techniques applied in the manufacturing process of the textile industry. Besides, there are uncountable textile mills spread around the globe. Moreover, developing countries produce 50% of the world's textile exports and 75% of the world's clothing exports. In some of these countries, environmental regulations are not usually a governmental priority and that water treatment plants are scarce (Mara, 2004)(UN-Water, 2021).

Yet, as textiles release MFs during production, use, and at end-of-life disposal, this industry can play an important role in the reduction of MFs by upgrading their processes and their products (Ellen MacArthur Foundation, 2017; Henry et al., 2019). Also, according to Xu et al., 2021, the MFs should be included in the concept of the circular economy, providing a regulatory framework to address this pollution (Xu et al., 2021). However, the textiles and apparel industry accounts for a very high percentage of total manufacturing occupations in many countries where poverty mitigation is a central issue (InfoDev, 2008). This complicates the challenging task to achieve a global adaptation to sustainable techniques that target the reduction of MFs' detachment. Besides, the term "microfiber" is not usually found in books or references for improvements in the sustainability in the textile industry (e.g., Muthu, (2017)). This is an indirect demonstration that textile MFs are not receiving the attention that they should. The trend is more focused onto the customer expectation fulfillment or the currently "fast-fashion" trend and the waste that it generates. This last topic is another problem that must be treated as more than 70% of the annual fiber production for clothing is burnt or disposed in landfills, which involves a great wastage of every kind of resources (water, energy, textiles, etc.) (Rese et al., 2022; Rahman et al., 2022).

There are many examples of improved techniques to enhance productivity. For instance, in recent decades several new spinning systems have been introduced to the industry.

These systems have resulted in important enhancements in yarn productivity but not necessarily in yarn quality (Islam, 2019; Nergis, 2017). Yet, there are some studies that are looking forward sustainable techniques like using wet spinning of fungus to create monofilament yarns (Svensson et al., 2021). The type of fabric (woven, knitted, etc.) also seems to play an important role. For example, De Falco et al. (2018b) reported that woven polyester released the highest number of MFs when compared to knitted polyester and woven polypropylene. Carney Almroth et al. (2018) observed that polyester fleece fabrics shed much more MFs than knitted fabrics made of polyester, acrylic, and polyamide. They also reported that high twisted yarns are less prone to detach MFs, which is in line with the observation exposed in Table 5.1.

On the other hand, in a study conducted by Zambrano et al. (2020), an evaluation of cotton knitted fabrics was done. They reported relevant information that can be furtherly assessed with synthetic fabrics. They found that the treatments applied during textile processing influenced the MFs released during laundering. In this sense, fabrics treated with softeners generated the longest MFs, while durable press and water repellent generated the shortest ones. They pointed out that, in general, fabrics with more abrasion resistance, higher friction coefficient, and less softness (i.e., fuzz or hairiness) reduce the detachment of MFs. In this line, a *Life European* project evaluated textile procedures and published a guideline of "better practices" for any industry relevantly linked to the textile chain and the main barriers to perform them (Mermaids, 2018). The key issues identified were the fiber (fineness, irregularities, length), the yarn (number of plies, twist, count), the fabric (structure, density, processes as dyeing and finishing), and garment washing in factories and their wastewater management. A summary of their observations is listed in Table 5.1:

Table 5.1. Summary of better practices in the textile industry published by Mermaids (2018).

Issue	Better practice	Main barriers
Melt spinning process	- *Adjust to preserve fibers' mechanical properties.* - *Fiber fineness should be increased to decrease yarn tendency to form protruding MF.* - *Fiber irregularities increase the friction between fibers and avoid the release of MFs from the yarn.*	- *Lower temperatures will increase the production time.* - *Yarn modification will alter its characteristics.*
Drawing, stretching, texturing, intermingling and drying	- *Adjust to preserve a good fiber tensile strength. The higher the tensile strength of the yarn the lower the probability of releasing MFs during washing.* - *Length of the fibers should not be too low.*	- *Process modification depends on the client's requirements.*
Spinning (yarn)	- *Continuous fibers detach less MFs than discontinuous or staple.* - *Plied yarns detach fewer MFs than single yarns.* - *High twist yarns detach fewer MFs than yarns with a lower twist.* - *The lower the linear density of the yarn (yarn count) the lower the number of fibers per cross-section the lower the release MFs.*	- *Yarn specification depends on the client's requirements.*
Dyeing	- *Avoid garment dyeing as it releases more MFs than yarn dyeing.*	- The solution is to avoid or reduce dyeing.
Knitting and weaving	- *The yarn carrier velocity of the knitting may be reduced to decrease the damage of the fiber.* - *The quantity or the nature of the sizing agent in the weaving process could be optimized and the velocity of the weft transporter could be reduced.* - *High-density fabrics have a tighter structure than lower ones, reducing the release of MFs.* - *Plain weave fabrics detach fewer MFs than twill weave ones.*	- *Increase of the production time.*
Mechanical finishing	- *The condition in the napping process may be optimized to reduce the mechanical stress on the fabric and its weakening.* - *The cut fibers should be recollected and managed in the factory.* - *Singeing mechanical finishing avoids the MFs formation on the fabric surface.*	- Client requirements. - Difficulties to adjust the equipment.
Finishing	- *Finishing agents capable of protecting the fabric surface can be used in this process.*	- *The agent must be compatible with the other finishing treatments.*
Manufacturing	- *A preliminary washing of the textile article can be done before selling it to remove the MFs generated during textile previous processes.*	- *Textile manufacturing mills may not have the facilities for industrial washing and wastewater treatment.* - *Current water treatments might not be able of collecting the smallest MFs.*

Textile parks also are a source of MFs. For instance, the work of Zhou et al. (2020) explored the MFs' presence in a typical textile park in China. They found that these installations can release as much as 54x10^3 MFs/L in printing and dyeing wastewaters. However, The MFs' generation was seen to significantly vary between mills. Hence, the authors discerned between the treatments that are applied in each textile plant, supporting the idea that the release of MFs is crucially influenced by the raw fabric materials and the conditions and chemical products applied in each industrial process. In this sense, the mill with the most concentration of MFs worked with rayon as raw material, which is usually subjected to heavy treatments like high temperatures and pressures. It has to be mentioned that, under the definition of the ECHA, rayon MFs are considered as MPs. This means that the MFs' pollution could be underestimated because rayon MFs are usually not considered as MPs. The authors also mentioned that textile printing and dyeing might be a more significant source of MFs than domestic washing, which is on the order of half a million tons per year (Belzagui et al., 2020). In this case, the studied mills have their own water treatment plants (WTPs), in which the efficiencies went from 85% to 99% of MFs' removal. The final effluents of these WTPs are subsequently treated in a centralized facility, from where they estimated a MFs' release of 430 billion particles per day.

On the other hand, producing and manufacturing textiles from natural rather than man-made or synthetic fibers has also been mentioned as a possible alternative to reduce the MFs' pollution. This statement is, at least, debatable, as nowadays most of the cotton business is based on a highly pollutant and environmentally unsustainable production (Allary, 2021; Garcia et al., 2019; Maraseni et al., 2010; Rukhaya et al., 2021). For instance, cotton production uses 2.5% of world's farmlands but consumes 25% of world's insecticides and 80% of total water usage in the textile sector (Garcia et al., 2019). Hence, this claim can only be considered as an option in particular cases where it can be proved, for instance, by making a life cycle analysis (LCA) that the declaration is true. Regarding only the MFs' detachment, Salvador et al. (2020) reported that cotton articles shed more MFs than synthetic ones. However, these findings must be treated carefully to avoid misleading and generalized conclusions. In other words, there is a lack of inter-laboratory repeatability to ensure that the outcomes are accurate.

With regards to mixtures of materials, i.e., blends of synthetic and natural polymers, a reduction of MFs detachment might be achieved. In this sense, Napper and Thompson (2016) reported that, independently of the washing treatment, polyester (65%) cotton (35%) blend fabrics detached fewer MFs than garments made only of polyester or acrylic. Conversely, Zambrano et al. (2019) found that polyester (50%) cotton (50%) blends, as well as cotton and rayon ones, detached more MFs than polyester fabrics. This might again suggest that the fabrics' specific manufacturing process and the proportion of materials play an important role in the MFs shedding rate. Besides, polyester, cotton, and their blends receive different dying treatments that could be an important parameter to be furtherly examined. However, mixed materials can hamper the possible subsequent recycling procedure (Herweyers et al., 2020). Hence, it is of special interest to find the balance between the optimal blending proportion and the possibility of future uses or recycling of the discarded textile articles.

Alternatives can also consider the production of more resistant and/or higher quality fabrics. In this line, the quality of the products is an important parameter that should be considered. The current "fast-fashion" business model induces a decrease in social and environmental conditions and produces low-quality garments that are more prone to detach MFs (Peters et al., 2021; Zamani et al., 2017). Hence, manufacturers and consumers can play a key role by investing in garments that are manufactured to last more and detach less MFs (Patagonia, 2018). Additives can be also considered, for instance, a reduction of 90% of the MFs release was obtained by applying pectin, poly-lactic acid, and polybutylene succinate onto polyamide fibers (De Falco et al., 2019; De Falco, Gentile, et al., 2018). Nonetheless, further investigation is required to develop additives to avoid or reduce the MFs' detachment from chemically more stable materials (i.e., polyester) but without using toxic compounds as, e.g., methanol. For instance, Martel et al. (2002) explored the finishing of polyester fabrics with cyclodextrins and polycarboxylic acids. The study had no relation with MFs, however, they found an easy way with non-toxic chemicals to physically but permanently adhere a coating onto polyester fabrics. These types of studies can be assessed to apply them with MFs' reduction purposes.

Many works have studied the coating or surface functionalization of fabrics. These investigations can also be furtherly considered as alternatives to reduce the MFs pollution and stop them from reaching aquatic environments (e.g., Carosio et al., 2014; Chen et al., 2012; Cireli et al., 2007; Glampedaki et al., 2012; Rojas and Azevedo, 2011; Trad et al., 2018). Besides, it is equally important to make the additive or process sustainable. Using toxic or hazardous compounds or highly energetic demanding procedures in the production line could develop worst impacts than the benefits from the reduction of MFs.

5.3 Household and Industrial Washing Machines, Tumble Dryers and Additives

To the best of our knowledge, Browne et al. (2011) were the first to suggest that the MFs' contamination is related to the laundering of textile articles. In their publication, they noted that receiving water points of sewage effluents contained a higher abundance of MPs when compared to other reference sites. Besides, they found that the proportion of polymeric materials found in the sewage effluents resembled the MPs contaminating shore sediments and disposal sites. Subsequent investigations verified and quantified the detachment of MFs from laundering processes, which happens to be in the magnitude of millions of MFs per cycle. In a first estimation, it was calculated that about 0.5 million tons of MFs reach the oceans every year (Boucher & Friot, 2017). However, it was posteriorly noticed that the estimation used a parameter that was 100 to 300 times larger than a more proper value. In particular, the linear weight of the MFs was considered at 300 dtex (1 dtex = 1 gram of fiber per 10000 meters), when the MFs will have a linear weight between 1 to 3 dtex. In this sense, Belzagui et al. (2020) re-estimated the MFs' flow by applying a new methodology with new parameters and it was obtained an annual MFs' stream to aquatic environments of 0.3 million tons. However, if the ECHA definition proposal for MFs is accepted (length 0.003 mm to 15 mm and a length/diameter ratio > 3), only Pirc et al. (2016) have evaluated MFs sized above 5 mm. In this case, and adding that rayon MFs can also be considered as MFs, the amount of MFs detached per laundering reported by most of the published articles will be probably underestimated.

Common clothes and housing linens MFs' emissions were studied by Galvão et al. (2020). They assessed the detachment from a mix of articles and fabrics making a total of 205 pieces of daily used textiles, i.e., articles used by themselves. They found a MFs release of approximately 18 million MFs per 6 kg of washed garments, making 3 million MFs per kg of washed article. This is in line with all previous studies. Besides, they reported that more than 90% of the MFs have a length < 500 μm, with half of them being < 100 μm. The washing temperature and time of the cycles were assessed by Cotton et al. (2020). They compared a "normal" washing cycle (40ºC and 85 min) with a "cold-quick" program (25ºC and 30 min). As expected, they found that higher temperatures and times significantly increased the MFs' detachment, reduced the garment longevity, and accelerated the color loss, which is in line with the work of De Falco et al. (2018b). However, an interesting finding was that when the washing time is increased to a certain point, the fabric abrasion (and the MFs' release) become stable. The estimated time stabilizing point was found at 35 minutes. However, no mechanism explanation was found to this behavior (Bao et al., 2017).

Furtherly, Dalla et al. (2020) evaluated the MFs' detachment from 100% polyester knitted fabrics with different operational washing conditions. They reported that the operative conditions (program time, temperature, speed of centrifugation, number of inversions of drums, etc.) have a direct impact on the "stressing" or friction action on the garments. In this sense, the minor stress applied to fabrics was seen during "delicate" cycles, which were found to decrease the MFs' release by 16% when compared to the "cotton" program. However, it must be furtherly tested to understand if this is a generalized situation or a particular observation achieved in the washer used. If it happens to be a generalized condition, the appliance industry could apply these data to improve their programs to achieve the reduction of the generation of MFs.

Regarding the type of washers, Hartline et al. (2016) reported that top-loading washing machines detach more MFs than front-loading ones. As hypothesized by the authors, this may be a consequence of the central agitator of the top-loading models. In this sense, highly populated regions as Asia and North America have a high percentage of top-loading washers (90%). Hence, it could partially explain the usually big concentration of MFs found in the environment. To estimate what could happen if those countries have more front- than top-loading washers, Belzagui et al. (2020) inverted the situation by considering the opposite condition (10% of top-loading). This modification resulted in a global MFs' reduction of approximately 30%. This change reduces the generation of MFs, hence, it must be noticed that it could decrease not only MFs from reaching aquatic systems but to the whole environment too. However, as mentioned before, further investigation is needed to determine if this is a generalized situation or it only applies for the washers used in Hartline et al. experiments (Hartline et al., 2016). In this sense, improving the designs of washers to cause less friction to the garments or the inclusion of built-in MFs' filters can be also considered as feasible alternatives. By doing so, white goods manufacturers can include in their catalogues a qualification class regarding MFs' emissions of their machines. In this way, consumers will be able to consider this factor when acquiring a new washer.

Concerning the washing additives, there is still big uncertainty about their relevance in the detachment of MFs. For instance, Zambrano et al. (2019) encountered that the usage of detergent and higher temperature conditions showed a significant increase in the detachment rate for cotton. In this line, De Falco et al. (2018b) reported that liquid and powder detergents increase the MFs' release. They also found that powder detergents might cause a higher release of MFs due to their inorganic and water-insoluble compounds (like zeolite) that can cause friction to the textiles, and also due to the higher pH of these detergents. However, Napper and Thompson (2016) reported that no clear trend was found with the presence of detergent and conditioner. Although, fewer MFs were "occasionally" found when no- or bio-detergent were applied. Moreover, Salvador et al. (2020) reported that the use of liquid detergent reduced the MFs' detachment when compared with no detergent for synthetic fiber garments (polyester, acrylic, and polyamide) but not for cotton ones. In this sense, making an extended assessment of the effects of the additives will benefit on the formulation of new cleaning agents that could provide protection to the garments and reduce their MFs' detachment. In Table 5.2 it can be found a summary of the findings reported by these articles.

Table 5.2. Published works regarding the MFs' detachment in industrial and household laundering.

Work	Analytical method / Comments	Results
Browne et al., 2011	- *No clear information on the methodology used.* - *Conservative estimations reported.*	130 – 280 MFs/L per garment > 1 900 MFs/garment
Napper and Thompson, 2016	- *Indirect method* [(a)]. - *Fibers with a mean length > 5 mm considered* [(b)].	140 000 – 730 000 MFs/6 kg of washed garments 500 000 MFs/mg
Pirc et al., 2016	- *Indirect method* [(a)]. - *Filters of 200 µm.* - *Mean length considered > 5 mm.*	135 000 MFs/6 kg of washed garments
Hartline et al., 2016	- *Indirect method* [(a)].	29 – 431 mg of MFs/garment-washed (front-load) 1 471 – 2 121 mg of MFs/garment washed (top-load)
Carney Almroth et al., 2018	- *Gyrowash one bath 815/8 according to modified SS-EN ISO 105-C06.* - *Commercial liquid detergent.*	7 360 MFs/(m² L) for PES fleece 87 MFs/(m² L) for knitted PES
Salvador, 2017	- *Used Napper & Thompson (2016) and Pirc et al. (2016) methods.*	184 000 – 250 000 MFs/garment
De Falco et al., 2018b	- *Linitest apparatus used to simulate washing machine.* - *Direct quantification.*	6 000 000 – 17 700 000 MFs/5 kg of washed garments 0.43 – 1.27 g of MFs
Belzagui et al., 2019	- *Commercial front-load washer.* - *Quantification of the MFs by visual counting.* - *Polyamide 20 µm filter.* - *Indirect method to estimate the weight of the MFs (applying the diameter and density).*	1 000 000 – 6 500 000 MFs for 6 kg of washed garments 30 000 – 230 000 MFs/garment 2 – 29 mg of MFs/garment washed 1 200 – 33 000 MFs/(m² L)
Zambrano et al., 2019	- *AATCC standard SDL Atlas Launder-Ometer.* - *Whatman glass 1.2 µm filter.* - *HiRes Fiber Quality Analyzer (FQA) for MFs' quantification.* - *25°C and 44°C.* - *AATCC 135-2015 with Washing machine* - *Nylon 20 µm and then Whatman glass 1.2 µm filter.*	*Mass lost during accelerated laundering:* 0.2 – 4.0 mg MFs/g (cellulose-based fabrics) 0.1 – 1.0 mg MFs/g (polyester fabric)

Salvador et al., 2020	- *Commercial top washer used in the experiments.* - *Subsequent steel sieves 500 μm and 65 μm.* - *Indirect method to estimate the weight of the MFs (applying the diameter, linear weight, and density).*	*% of lost garment weight from 10 cycles*: 0.16 to 0.20 (100% cotton) 0.07 to 0.13 (100% acrylic) 0.03 to 0.06 (100% polyester) 0.04 to 0.10 (100% polyamide)
Galvão et al., 2020	- *Clothing and housing linens from a household of 4 people.* - *Commercial front-loading washer.* - *Aliquots of 20 mL were filtered through 12 μm nitrocellulose membrane filters* - *MFs were visually counted in one quadrant of the filter and extrapolated.*	18 000 000 MFs for 6 kg of washed garments
Dalla et al., 2020	- *100 % PES knitted fabrics.* - *Pre-washing before experiments.* - *Commercial washer, detergent, and softener used.* - *Effluent was filtered through 40 μm pore.* - *Different programs tested.*	36.60 mg/kg (cotton program) 32.51 mg/kg (delicate/silk program with liquid stain remover) 33.86 mg/kg (delicate/silk program.)

(d) Indirect method: the quantification is estimated from the weight, length, and/or density of the MFs.
(e) According to the ECHA, fibers with a length < 15 mm and a maximum diameter of 5 mm should be considered as MFs.

Although most methods shown in Table 5.2 exhibit similar results, there is an urgent need for a standardized analytical method (Henry et al., 2019). In this sense, some textile and interdisciplinary coalitions are working towards a unique analytical method. Indeed, in a recent publication from Tiffin et al. (2021), they proposed and published the validation of a method applying a *Gyrowash* machine. Besides, an inter-laboratory study was executed and they found that the method presented good replicability. However, it is important to mention that Zambrano et al. (2019) found that accelerated laboratory launderings, like those made in a Gyrowash, might release more MFs than household ones. This might indicate that these types of tests might only be applicable when relative detachment rates are the objective of the assessment. In other words, the absolute outcomes of MFs' detachment might be evaluated it in real conditions (conventional washers, textile articles and additives).

It should also be considered that each methodology has limitations or disadvantages. For instance, Belzagui et al.'s (2019) method and similar procedures are time-demanding. Besides, visual counting might be preferably used when small amounts of garments with strong colors are tested. On the other hand, methods such as those proposed by Salvador et al. (2020) might not be able to discern between MFs and longer fibers, or could erroneously include the mass of the detergent or other impurities in the weighting process of the MFs. To that effect, a method that considers different situations, types of samples or requirements will probably be needed, for example, one for relative and the other for absolute amount of released MFs.

On the subject of dryers, O'Brien et al. (2020) studied the airborne emission of MFs from a domestic vented dryer. They found that the lint emission corresponded to approximately 0.012% of the garment mass dried. Kapp and Miller (2020) also studied the MF emission from dryers. They collected the MFs vented to the surroundings of the experiment site and provided the results in different units for further comparability. In this sense, they reported averages from 35 mg to 70 mg of lint from three consecutive dry cycles. As can be seen, both studies confirmed that MFs can escape the lint trap of the dryers and therefore should be considered as a source of MPs to the environment. However, further investigation is needed to assess the detachment rate from these machines. In this sense, clothes tumble dryers can also be modified to be more efficient regarding the MFs' generation. For instance, TeGrotenhuis et al. (2017) proposed a hybrid heat pump dryer that could achieve savings in both energy and drying cycles, providing an indirect reduction of the MFs generated.

Devices to reduce the MFs in the effluents of the laundering equipment

There are commercially available devices intended to reduce the release of MFs from washers. These might work by reducing the MFs either inside the washing machine as in-drum devices [e.g., GuppyFriend (2021) or Cora Ball (2021)], or at the effluent as external filters [e.g., Lint LUV-R (2021) or PlanetCare (2021)]. These devices work differently when reducing the concentration of MFs in the washers' effluent. *GuppyFriend* and analogous systems work basically to protect the garments from the mechanical stress generated in the washing process. Hence, it is expected that they will reduce the generation of MFs. In contrast, external filters and the *Cora Ball* act by retaining the already generated MFs. It must be noticed that these devices might be complementary between them in to goal to get a higher reduction of the MFs.

According to McIlwraith et al. (2019), these technologies can accomplish a MFs' reduction in the washer effluent between 26% for the *Cora Ball* to 87% for the *Lint LUV-R* filter. Additionally, they reported no significant difference for the MF length when using the *Cora Ball*, suggesting that this device captures MFs in a wide range of sizes. Meanwhile, Napper et al. (2020) reported that the external device XFiltra (2021) was the most successful in retaining MFs, followed by the in-drum *GuppyFriend*. For the *XFiltra*, two main explanations were given, a finest mesh pore in contrast with other similar devices (60 µm vs > 175 µm), and the use of an integrated electrical pump to facilitate the flow through the mesh. On the other hand, *PlanetCare* filter sustains a > 90% of MFs' retention efficiency for their product, which contradicts the findings of Napper et al. (2020). The results of these studies are summarized in Figure 5.2.

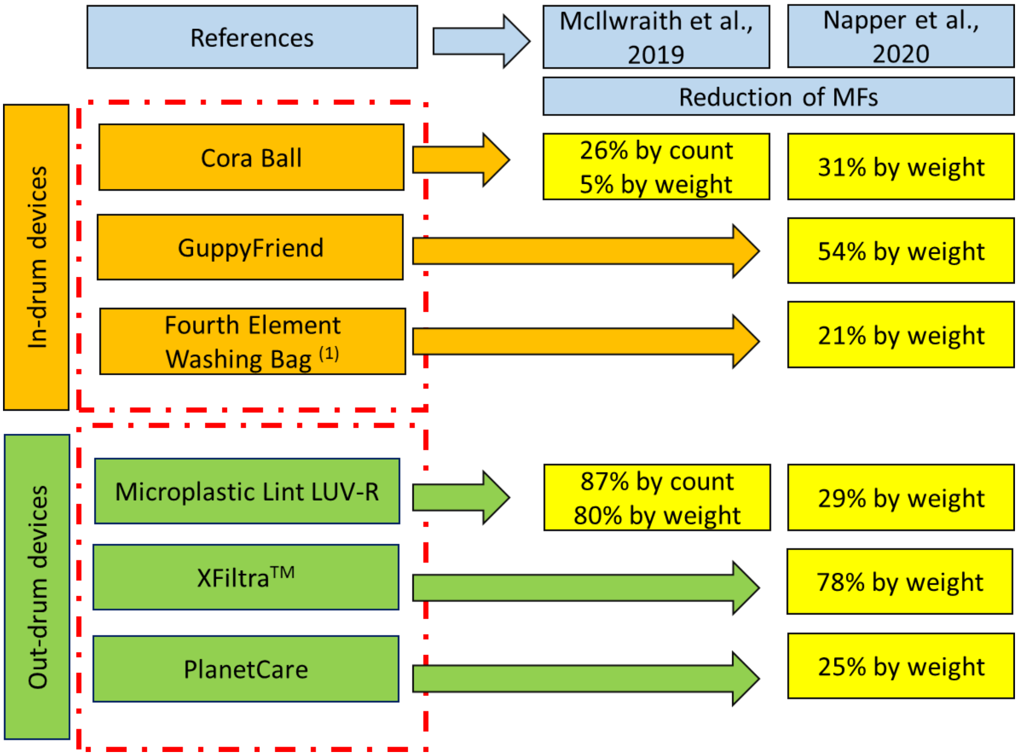

Figure 5.2. Microfibers' reduction at the effluents of the washing machines. (1) Fourth element washing bag only appears in Napper et al., (2020)'s work.

As can be seen in Figure 5.2, distinct results are reported for the same devices; hence, further experimental inter-laboratory replications are required not only for an accurate understanding of the devices but for their enhancement too. These options seem feasible for the short- to medium-term period as can be easily introduced into the market. Nevertheless, the final disposal of the retained MFs is an urgent concern yet to be addressed. For instance, as commented by Napper et al. (2020), once the filters are cleaned by collecting the MFs, these can be "*thrown into the everyday household waste*". In this sense, depending on the final disposition given to that specific waste, the MFs could also finish in water environments, turning the devices just into a mere MFs' "by-pass". Hence, this is very far from being a practical solution.

The usage or final disposition of the MFs has been poorly studied. For instance, Yousef et al. (2021) have proposed the use of lint from household dryers as a source of renewable energy through pyrolysis. They found that lint can produce an activation energy higher than that of textile waste as a result of its high purity, chemical composition, and uniform size, which facilitates the conversion process. However, it must be noticed that many of the lint materials, like polyester, will not have a renewable origin. In this sense, this fossil-based waste should not be considered as a renewable energy source. Besides, there is a logistic issue that needs further consideration to transport this waste. A good idea would be to persuade the users who have a MFs' retention system to take them to a MFs "disposal" site, from where these could be collected and submitted to a future final disposal treatment facility. In this sense, we can make an analogy with gases' emissions. There are diffuse emissions (cars – household laundering), which are harder to treat, and point sources which are easier (fabrics – textile industries). In this line, a good alternative could be to reuse them when these are collected in industries (point sources) and to immobilize them in treating facilities when these are retained in household launderings (diffuse sources).

Currently, it has been determined that textile articles are washed not because they are dirty, which implicates a futile use of water and energy resources (Stawreberg & Wikström, 2011). Hence, an effective and obvious method to reduce the MFs' shedding is to wash them only when it's required. For instance, in a hypothetical situation simulated in the work of Belzagui et al. (2020), a reduction of 30% on the generation of MFs can be achieved by reducing the water consumption for the operation of washers in the regions with a higher consumption rate (which is 19 m^3/washer per year). In other words, doing shorter washing cycles or washing less generates fewer MFs. This measure can be accomplished by instructing consumers to use quicker but adequate laundry programs and/or more efficient washers. This strategy can be implemented in the short- to medium-term to reduce the MFs' generation, which consequently will decrease the emission of MFs to the whole environment, affecting especially to the usage and contamination of water ecosystems.

It must be noticed that "washing less" is very subjective, yet, there are some guides to provide sufficient knowledge to consumers about when and how to wash their clothes. For instance, reported "better practices" are: filling up the washing machine, using liquid detergents, selecting colder and quicker laundry settings, among others (Cotton et al., 2020; De Falco, Gullo, et al., 2018; Mermaids, 2017; Plastic Pollution Coalition, 2017). This could be strengthened with textile campaigns indicating how many "normal" uses a determined garment can be subjected to before washing. In this sense, the population insight about MFs and their means to reduce the generation must be continuously reinforced. Social networks are strong tools that are constantly making publications on this topic (GESAMP, 2015b; SAPEA, 2019; Wagner & Lambert, 2018); however, environmental education should be guaranteed from the early stages of our education, and MFs should be a part of this education.

Regarding the mass or quantity of garments washed per cycle, in a quick experiment made in our laboratory to measure the MFs' detachment, polyester fabrics were independently submitted to one washing cycle at equal conditions (Fagor Innovation F-2180 washer, 40ºC, 1000 RPM, 57 minutes, and 59 liters of effluent). The textiles used were identical fabrics but differing in their weight and dimension; two weighted 0.13 kg, while the other pair weighted 3.00 kg (4 samples in total). Each sample was washed with liquid detergent. In this case, the contrast was from a washer occupation of 25% to 70% in volume and from 20% to 50% in weight. The relation between the mass of garments is about 23 (3.00 kg / 0.13 kg), while the relation between the mass of the MFs detached was seen to be approximately 5 (0.025 g / 0.005 g). The reduction of the MFs could be a consequence of a decrease in the friction between the textile articles and the washer drum. Besides, this quick experiment verified what is said in the "Mermaids good practice guide" (Mermaids, 2017): filling up the washing machine considerably reduces the MF detachment.

5.4 Municipal, Industrial and Drinking Water Treatment Plants

This section discusses the MFs and MPs found in water treatment plants (WTPs) and water products. The addressed perspective is to inform about the concentrations typically encountered in the effluents, the effectiveness of the different technologies for the MPs' removal, and the effects of the MPs on the treatments.

Drinking water treatment plants

It has been published that the consumption of bottled and tap water is a source of ingestion of MFs (Ossmann, 2021). The average concentrations reported were 94 MPs/L for bottles and 32 MPs/L for tap water (Eerkes-Medrano et al., 2019; Koelmans et al., 2019; Mintenig et al., 2019; Schymanski et al., 2018). Cox et al. (2019) estimated that the annual MPs' intake could be 52x10^3 items. However, it may raise to 90x10^3 MPs if only bottled water is consumed. Kosuth et al. (2018) found that most of the particles were MFs (98%). Regarding the size, Ossmann et al. (2018) reported that over 90% of the MPs found in bottle water corresponded to particles smaller than 5 µm, which are smaller than the mean 960 µm reported by Kosuth et al. (2018) for tap water. Hence, drinking WTPs are not completely effective in eliminating MPs, whether as for uncompleted retention or as for an "in-situ" generation.

In this line, Wang et al. (2020) reported that the overall MPs' removal from different technologies in drinking WTPs was from 82% to 89%. On the other hand, Y. Zhang et al. (2020) found a low retention of micro- and nanoparticles (< 2%), with an increase of 16% when applying a coagulant aid. Works from Ma et al. (2019a, 2019b) also reported low retention efficiencies (from 1% to 8%) at coagulants' conventional dosages. Hence, there is a need to mend these contradictions. Regarding the ultrafiltration process, it has been reported that it could be the most effective process for MFs' removal (Ma, Xue, Ding, et al., 2019; Y. Zhang et al., 2020). Sand filtration was also tested but it is not considered to play a primary role in removing MPs. A key finding was that drinking WTPs can also act as a source of MPs. In this specific case, greater concentrations of polyacrylamide (PAM) were detected in the effluent compared to the raw water. PAM is used as a component of the coagulant used in the process. In this sense, more investigation is needed to evaluate if the presence of MPs and MFs in drinking water is due to the physical degradation of the plastic bottles or the water treatment itself (Ossmann, 2021).

Used Water treatment plants

In municipal or industrial used-WTPs, a proportion of the incoming MFs will be transferred into the sludge throughout the consecutive treatments. It is important to notice that WTPs should not be considered as a source of MPs and MFs but a pathway where they can be removed from the liquid stream. Globally, the percentage of municipal used-waters that are subjected to any kind of management is approximately 20% (ONU, 2017; Pham & Kuy, 2013). Populations connected to urban WTPs are markedly variated across the countries. For instance, South American and Asian countries treat around 20% of their municipal waters, while Central European countries have achieved a 97% of treatment coverage (EEA, 2017; Mara, 2004). WTPs are considered significant pathways for all types of MPs to aquatic and soil environments (Raju et al., 2020; Rolsky et al., 2020). Yet, as mentioned before, fiber-shaped ones coming from the laundering of textile articles are within the most encountered types in these streams.

Regarding the material, polyester MFs usually surpass other types of MPs (Browne et al., 2011; Lares et al., 2018; Magnusson et al., 2016; Murphy et al., 2016; Sun et al., 2019). Currently, polyester is the top synthetic material used by the textile industry. Besides, as pointed out by a series of publications, regardless of the material, each textile article detaches thousands to millions of MFs in every domestic washing cycle (Belzagui et al., 2019; De Falco, Gullo, et al., 2018; Folkö, 2015; Hartline et al., 2016; Pirc et al., 2016). For instance, Alavian Petroody et al. (2020) reported that most of the MPs found in a WTP were in the form of MFs, from which polyester was the most abundant, followed by polyamide and acrylic fibers. This makes sense, as an important part of the MPs entering a WTP will come from the household washers' effluents. Following the current data, it has been estimated that 0.48 million tons of MFs are globally generated in domestic washers, from where 0.20 million tons might be retained in WTPs' sludge and 0.28 million tons might reach aquatic environments (Belzagui et al., 2020).

In primary treatments, raw water usually passes through a set of screenings. If fine screens are applied (2.5 to 10 mm), MPs > 2.5 mm can be removed from the water but not completely because of the morphology of these particles (big length to diameter ratio). Next, grit removal, flotation, and primary settlement could also retain MPs with different densities than water (Sun et al., 2019). Primary treatments are effective to retain MFs as these might be adsorbed, aggregated, and entrapped in flocculating particles and separated by sedimentation (Sun et al., 2019; Wei, Zhang, et al., 2019). An important removal of the MFs can happen in this stage, in some cases, MPs' removal was found to be even greater than 90% (Lares et al., 2018). Secondary treatments, as activated sludge, are basically biological processes that degrade organic pollutants. Here, MPs can be retained by the extracellular polymer substances secreted by microorganisms and furtherly removed with the generated sludge; i.e., MPs are transferred to the sludge (Sun et al., 2019). In general, as reported in Bakaraki Turan et al. (2021), most of the MPs' and MFs' removal is made in the primary and secondary treatments. Finally, advanced treatments as coagulation and filtration, ultrafiltration, or membrane bioreactors can remove part of the remaining and lower-sized MPs. As can be seen in Figure 5.3, the retention and transfer rates might depend on the treatment applied. From different studies (Alavian Petroody et al., 2020; Blair et al., 2019; Carr et al., 2016; Franco et al., 2020; Gasperi et al., 2015; Gies et al., 2018; Gündoğdu et al., 2018; Lares et al., 2018; Leslie et al., 2017; Magni et al., 2019; Magnusson & Norén, 2014; Michielssen et al., 2016; Mintenig et al., 2017; Murphy et al., 2016; Talvitie et al., 2015; Talvitie, Mikola, Koistinen, et al., 2017; Talvitie, Mikola, Setala, et al., 2017; L. Zhang et al., 2021; Zhou et al., 2020), it can be stated that the MPs' and MFs' transfer efficiency will be between 76% to 98%.

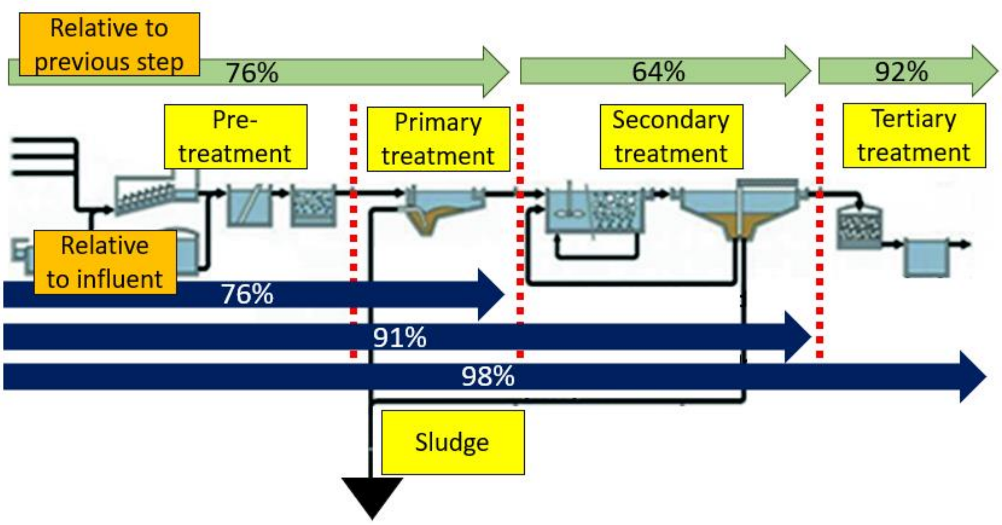

Figure 5.3. Percentage of retained microplastics and microfibers in a used-water treatment plant

For instance, Lares et al. (2018) reported that 99% of the MPs were removed in the primary treatment; afterward, a slight increase of the MPs' concentration (0.6 to 1 MPs/L) was found in the effluent of the activated sludge. This behavior might be a consequence of the sampling procedure, which didn't consider the different flows (turbulent and stable) that are found in each effluent. Hence, superficial sampling might retain more buoyant MPs when having a stable flux. MPs with a density higher than water will be mostly retained in primary and secondary treatments, whilst advanced treatments will eliminate floating particles from the final water effluent (Nizzetto et al., 2016). In this line, Bayo et al. (2020) found that particulate shapes were more prone (95%) to be retained than fiber ones (55%) in membrane bioreactors (MBR) and rapid sand filtration (RSF). It should be noticed that MFs might have an easier pass through RSFs as a consequence of their longitudinal shape and small diameters (~10 μm) (Hamidian et al., 2021). Edo et al. (2020) reported that the effluents of primary and secondary effluents were dominated by sizes between 25-104 μm, strengthening the conclusion that smaller and fiber-shaped MPs are more prone to be found in the effluents of WTPs (L. Li et al., 2020; Raju et al., 2020).

Despite having a relatively high retention efficiency, these facilities treat millions of liters every day, releasing high amounts of MFs (Sun et al., 2019). Also, as previously explained, the proportion of treated waters is still very low across the world, and it must be noticed that other sources of textile MFs (as garments' hand-washing) will be still left aside from these treatments. Equally important is that these particles can still enter the environment via the final disposal of the sludge, as conventional treatments don't remove sludge-based MPs (Z. Chen et al., 2020). The reported abundance of MPs in the sludge varies from 1.5x10^3 to 180x10^3 particles per kg of dry weight sludge (Edo et al., 2020; Lares et al., 2018; L. Li et al., 2020). Hence, MFs might still be dumped into the environment if the sludge is used as, e.g., an agricultural fertilizer (J. Bayo et al., 2016; Corradini et al., 2019; X. Li et al., 2018; Mahon et al., 2017).

The existence of WTPs was estimated to play a relevant role to transfer MFs from the liquid stream into the sludge. According to Belzagui et al., (2020), a global 30% reduction of MFs reaching aquatic environments can be achieved by increasing regions with a low percentage (<50%) of treated water to 60%. However, as pointed out in that study, further investigation is required to develop treatments for the MPs and MFs retained in the sludge. Yet, it must be noticed that Installing only primary treatments in places without WTPs could help to remove an important proportion of the incoming MPs and MFs from the liquid stream.

As previously indicated, the amount of MFs annually retained in the sludge of WTPs is about 0.20 million tons (Belzagui et al., 2020). Also, a publication estimated that the yearly amount of MPs entering agricultural lands from sludge might be between 63x10^3 to 430x10^3 and 44x10^3 to 300x10^3 tons in Europe and North America, respectively (Nizzetto et al., 2016). In this latter study, other sources of MPs, as particles collected by sewers, were also considered in the estimation. Partial removal of these fibers can be achieved by sieving and sifting procedures, however, a complete separation will not be possible (Weithmann et al., 2018). A recent promising strategy to reduce the MPs' concentration from the sludge is the hyperthermophilic composting technology (hTC), which was demonstrated to reduce 45% of the MPs after 45 days of treatment at a full-scale trial (Z. Chen et al., 2020).

Regarding passive treatments, Sarkar et al. (2021) studied the MPs' and MFs' pollution in freshwater wetland systems used for wastewater treatment. They estimated a MPs' removal of 50%, which is far lower than most conventional primary treatments. Besides, an important aspect to consider is that they identified variable amounts of heavy metals (As, Cd, Cr, Cu, Ni, Pb, Zn), ranging from 2.03 μg of arsenic per gram for MPs to 191.01 μg of zinc per gram of MPs. Fishes in the wetland ponds were also contaminated with MPs and heavy metals. In this sense, they concluded that natural wetlands are facing the risk of MPs pollution, where MPs have gotten to the trophic state hindering and stressing the wetland system. This article is in line with R. Li et al. (2020), who reported an abundance of 5.5×10^3 MPs per m^3 of water in a freshwater mangrove. However, as stated by Kumar et al. (2021), the number of MPs in wetlands will depend on various factors, such as the location, proximity to urban settlements, human interference, among others. The MPs' removal in natural wetlands could be very helpful for economically under-developed countries. In any event, it is evident that retaining the MFs before reaching WTPs will reduce their contamination to the environment, i.e., short- to medium-term alternatives as washers' filters can be a good alliance to reduce the flow of these pollutants either to water, soil or atmospheric ecosystems.

Effects of MPs and MFs on water treatment plants

It has been reported that MPs in biological treatment might reduce the abundance of the bacteria that is needed for the processes of nitrification, denitrification, among others. For instance, some MPs might inhibit the sludge anaerobic digestion in all its phases; hydrolysis, acidogenesis, acetogenesis, hydrogen, and methane production (Wei, Huang, Sun, Dai, et al., 2019; Wei, Huang, Sun, Wang, et al., 2019; X. Zhang et al., 2020; Z. Zhang & Chen, 2020). In some cases, depending on the polymer, the effects might have different origins. In this regard, polyethylene terephthalate (PET) and polyethylene (PE) induce the formation of reactive oxygen species (ROS, as OH* or H_2O_2). PET and Polyvinyl chloride (PVC) can release bisphenol-A (BPA) and di-n-butyl phthalate (DBP), respectively, both being toxic compounds (Wei, Huang, Sun, Dai, et al., 2019; Wei, Huang, Sun, Wang, et al., 2019; Wei, Zhang, et al., 2019). Minimum concentrations for the effect were established at 10, 20, and 100 MPs/g for PET, PVC, and PE, respectively, which are realistic concentrations found in sludge samples (X. Li et al., 2018; Wei, Zhang, et al., 2019). On the other hand, polystyrene nanoparticles affected the microbial community structures by reducing the cumulative methane production by 15% (Fu et al., 2018). Regarding polyester MFs, L. Li et al. (2020) found a reduction of methane production for several concentrations. Yet, the inhibition was lower than with other tested polymers. Also, Qin et al. (2020) tested with polyethersulfone MPs, and it was found that these particles slightly reduced the removal of ammonia nitrogen. In this sense, it is also important to have a better understanding of the effects that MPs and MFs will have on the biota of WTPs. For instance, if the potential damage is elevated, it will be necessary to remove them as much as possible before reaching WTPs.

5.5 Gaps, Criteria and Phases of the Solutions

As in every environmental issue, the solutions must fulfill a minimal set of conditions to be considered practical and to provide a positive net effect on the environment. Some of these criteria are mentioned hereafter:

1. An effective solution should avoid producing secondary issues when tackling the main concern. There are many ways to produce secondary problems, for instance, high energy and resources requirements, the usage of toxic or hazardous substances, among others. For example, the application of toxic compounds in the process could imply the subsequent event of environmental contamination (e.g., the use of methanol for producing textile coatings). Or, including washable coatings to textile articles could imply an increase of the BOD at the effluent discharging areas. In this sense, it could be of particular interest to perform a Life Cycle Assessment (LCA) for each MFs' processing or retaining system.
2. Alternatives should also seek the possibility of closing the gap of the MFs. In this line, some devices have shown great effectiveness in retaining MFs in washing machines. However, the interrogative of the posterior treatment must be equally and urgently solved.
3. The treatments or alternatives must be scalable. This is particularly complex in the manufacturing process of textile articles and the treatment of municipal waters. To better illustrate this, it must be noticed that the manufacturing of textiles is produced at uncountable points around the globe. In the same line, municipal waters are still poorly treated in terms of the worldwide proportion, being approximately 80% of the waters discharged to the environment without any treatment. On the other hand,

including devices in the washing machines could be easier in the short- and medium-terms.

4. In the case of devices intended for the users' application, these must be practical and easy to handle. The higher the device complexity the lower will be the implication of the users. This is furtherly explained in the work of Herweyers et al. (2020).
5. The economical parameter is also an important aspect to ponder. Unless policies or regulations (laws) to reduce this contamination are created, devices and/or processes should not exceed a "critical" economical point in order to promote the users' engagement.

In this line, Herweyers et al. (2020) assessed the consumers' perceptions and attitudes toward systems preventing MFs pollution. With this purpose, they determined the optimum requirement for these systems through a consumer survey. They found that the MFs' problem and peoples' washing behavior are underestimated. Besides, they constructed a minimal set of requirements from a user point of view, which are summarized next:

i. The solution must be effective and preferably visually experienced by the users so they can recognize their positive performance towards a cleaner environment.
ii. It must be durable and ensure long-term usage, i.e., disposable solutions should not be considered.
iii. The usage or installation of the product should be easy and user-friendly.
iv. People from all socio-economic levels should be able to acquire the product.
v. In case of a cleaning requirement of the product, it should be fast and user-friendly. The achievable amount of cleaning periods was found to be at every 15–17 washing cycles with a cleaning duration of 10 minutes.
vi. As also commented by them, further research is needed to investigate the possibilities to close the loop for the collected MFs. There are a lot of gaps still to be solved and to be investigated, as the MFs emitted to the air from the daily usage of the garments, or ways to immobilize or use the retained MFs. However, it must be noticed that there are environmental issues for all the life cycle of the garments. For instance, as reported by the Ellen MacArthur Foundation, less than 1% of the material used to manufacture garments is recycled into new garments (Ellen

MacArthur Foundation, 2017). In this sense, the MFs are one of the missing pieces to be solved in order to make the textile industry more sustainable, but further investigation is needed to develop new ideas or to improve already existing ones.

5.6 Conclusions

Many alternatives are available to reduce textile microfibers from reaching the environment. Some options are currently more viable in the short- and medium-term periods. The textile industry has the potential to drastically reduce the generation of microfibers by improving their processes or products. This could imply that downstream solutions might be dispensable or less severe. However, there are many small- and medium-sized textile industries around the globe, making this alternative feasible only in the long-term time. Also, there are currently some solutions for washers. These can reduce at least 30% of the microfibers' emissions from household laundry. Besides, new products as detergents or additives are being developed to reduce the generation of these particles.

On the other hand, water treatment plants can partially remove the microfibers from the liquid stream and retain them in the sludge. Depending on the technology applied, these facilities can remove up to 99% of the microfibers. Yet, the problem is still transferred to the sludge. In addition, installing these facilities is a long-term alternative. An important gap in every alternative is the final disposition or treatment of the microfibers. It is important to clarify that any solution must consider the whole process to certify that it is environmentally friendly and will not pollute more than the microfibers. Yet, it is very likely that the alternatives will be complementary between them, i.e., there will be no single solution for the microfiber pollution.

5.7 Acknowledgments

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Chapter 6:

Sustainable filtering system to retain textile microfibers during household laundering and a Novel treatment to immobilize and use them

6 Sustainable filtering system to retain textile microfibers during household laundering and a Novel treatment to immobilize and use them

The work presented is this chapter corresponds to the proposal of 4 different arrangements of a filter developed for the retention of released microfibers in domestic washing machines. No detailed explanations on the constitution of the 4 arrangements are given as this work is currently under patent process by the Universitat Politècnica de Catalunya. Spanish Patent register number P202130267:

"Dispositivo para retener microfibras textiles y método para obtener un producto hecho de un residuo generado por elementos textiles" F. Belzagui Elder, M.C. Gutiérrez Bouzán and M.M. Vilaseca Vallvé (OEPM Madrid, 26/03/2021).

In addition, a paper has been written to compare the developed filter arrangements with other marketed filtering options to retain MFs. It provides a novel way to retain and treat textile microfibers with a new filtering system. It is under revision for publication in a scientific journal:

F. Belzagui, C. Gutiérrez-Bouzán,V. López-Grimau,. Sustainable filtering system to retain textile microfibers during household laundering and a novel treatment to immobilize and use them.

Highlights

- 4 arrangements of a novel microfiber filtering system were tested in washers.
- All the arrangements showed a good microfibers' retention efficiency (> 90%).
- The replacement life interval for the cartridges is more than 30 washing cycles.
- The filters and the filtering media are made of recycled materials.
- The microfibers can be treated by including them in a polymeric matrix.

Abstract

According to the European Chemical Agency chemically treated and/or non-biodegradable textile microfibers (MFs) are a type of microplastics (MPs, size < 5 mm). These are considered one of the most environmentally threatening pollutants as they have a continuous and cumulative entrance to the environment. Once they reach natural ecosystems, they are technically very difficult to be removed. Currently, there are accessories to reduce this contamination, however, there are no alternatives to treat the MFs. This work tested a new and sustainable filtering system developed to retain the MFs

emitted from washing machines. The main characteristic of the system is that it uses recycled polymers for the filtering media and the filtering shell. Besides, it provides an alternative to furtherly treat and use the retained MFs by including them in the polymeric matrix used as the filtering media, adapting the filter to the circular economy philosophy. This treatment is explained in the next chapter of this thesis. In this sense, four filtering arrangements were evaluated in household washing machines. They presented a performance higher than 97% of MFs' removal from the washers' effluents. Also, all the tested arrangements showed a replacement time interval for the cartridge from 30 to more than 40 washing cycles, surpassing the durability expected by the users.

6.1 Introduction

Following the definition of the European Chemical Agency (ECHA), the microplastics (MPs) are fragments of chemically modified and/or non-biodegradable polymers with a length < 5 mm [6.1]. These particles have been widely encountered polluting every assessed ecosystem [6.2]. The estimations of the MPs concentration in the oceans are between 15 to 51 trillion buoyant MPs in and 14 million tons in the top 9 cm of sediments of the oceans [6.3–6.5]. Primary MPs are those emitted into the environment in a MPs size range; whilst secondary are those generated in the environment from larger plastic debris [6.6]. Regarding their impacts, it has been registered the ingestion across the trophic chain [6.7–6.10]. Besides, some effects of the MPs on organisms have been found, for instance, their retention and endocrine disruption, among others [6.11–6.14]. These particles can behave as vectors for organisms and hydrophobic toxic compounds [6.15, 6.16]. This contamination has been also found in products for human consumption and polluting the air [6.17–6.23], hence, there are many pathways for human exposure MPs [6.24]. Nevertheless, the potential risks for human health are still unknown [6.25–6.27].

The microfibers (MFs) are one type of MPs, these have a length to diameter ratio > 3 and a maximum length of 15 mm [1]. Textile MFs are among the most renowned as these have been widely found in the environment. These can be generated in the manufacturing, use, cleaning, and final disposal of a textile article [6.6]. This study is focused on those generated in the household laundering process, which can detach millions of MFs per washing cycle [6.28, 6.29]. Some solutions have been proposed for this contamination route. For instance, in-drum accessories to reduce the generation of MFs or out-drum filters to retain the already generated ones [6.30, 6.31]. However, in the "retaining" alternatives, none of the existing technologies has a final treatment for the MFs.

This article aims to evaluate the performance of a new MFs filtering technology. This system can be applied to retain MFs in equipment where these are emitted, like washing machines and dryers, among others. The technology and the subsequent treatment of the MFs are covered under the patent request P202130267. The principal novelty is that this system uses recycled thermoplastic pellets (low-density polyethylene) as the filtering media. Different arrangements of the technology were tested to know which one is more efficient for the catching MFs' purpose. Finally, the outcomes of this filter were compared with results reported in papers that tested other devices used with the same purpose.

One of the main advantages of this filter is that once exhausted, the retained MFs can be immobilized. For this, the patent mentions that the filtering media, consisting of thermoplastic pellets, can be merged, providing the MFs a matrix where these will be entrapped forming different types of composites. This point is explained in the next chapter of this thesis.

6.2 Materials and Methods

Tested filtering arrangements

The filtering system is covered by the patent request P202130267 (Spain), from which four different filtering arrangements were proven in this study. The arrangements were named as F1 (filter 1), F2 (filter 2), F3 (filter 3) and F4 (filter 4). Each of them is detailed afterward. As said in the introductory section, the novelty of this system is the application of recycled thermoplastic polymers as the filtering media. In this specific case, LDPE was employed in the form of pellets. These were usual commercial pellets, with a size of 3 to 5 mm on the bigger axis, and of 2 to 3 mm on the smaller axis.

The system has basically 3 main sections: (S1) a coupling sub-system to the washing machine, (S2) an empty section to facilitate the water flow and to provide space for the accumulation of MFs and dirt, and (S3) the filter cartridge filled with pellets. In this sense, the first three arrangements (F1, F2, and F3) were built following the simplest external filtering models shown in the patent. Whereas the F4 arrangement was structured to apply the filtration system in the existing washing machine filter. All the cartridges were structured to have an LDPE pellets' density of 0.5 to 0.6 g per cm^3. The external arrangements F1, F2, and F3 were made of translucent PVC to be able to observe their interior while conducting the experiments.

Materials and pre-treatments

Two types of black commercial polyester fabrics were selected. One was a woven fabric while the other was a fleece knitted fabric. For each experimentation, an equal number of pieces and weight (10 pieces, 280 g each piece) were distributed in two identical commercial washing machines (FAGOR Innovation F-2180, Spain), one for the control and the other to test each of the filtering arrangements. In this way, 70% (5.6 kg) of the maximum weight of these washers (8 kg) was introduced in each washing machine.

Before data compilation, two independent pre-treatments were made. On one hand, the washing machines were cleaned by doing 2 empty washing cycles. On the other hand, the fabrics were washed for five consecutive washing cycles prior to data collection. The latter pre-treatment was performed to achieve a constant detachment rate of MFs, which has been reported to be from the 5th washing cycle [6.28, 6.32].

Afterward, the MFs' filtering arrangements were independently tested versus a normal discharge of a washing cycle. The filtering arrangements were connected to one of the washers, while the other washer was kept unmodified to get comparable reference data of the concentration of MFs in the effluent. Then, the fabrics were washed with the "cold" program (30 min, 22 L of effluent, 1000 RPM, water grid temperature ~ 25ºC). A common detergent (Bosque Verde, Spain) was introduced to the washing trials with a volume of 75 mL per cycle.

The effluent of the washing cycle numbers 1, 5, 10, 15, and 20 were collected and evaluated by gravimetry. For this, from each effluent, a sample of 10 L was taken apart while continuously stirring. Then, 2 aliquots of 2 L were filtered through 20 μm polyamide filters. The polyamide filters were dried and weighed before and after the filtration of the discharged water (balance ± 0.12 mg). The difference between the weight of the filters was considered as the detached MFs. The retention efficiency of the filtering arrangements was calculated as the relation between the MFs found in the effluent of the filtered versus normal washing effluents (See Equation 6.1). The results were expressed in percentage of retained MFs.

$$R = \left[1 - \frac{(D_2 - D_1)}{(N_2 - N_1)}\right] \cdot 100\% \qquad Eq. 6.1$$

Where,

- R Retention efficiency (%)
- $D_2 - D_1$ Difference between the mean values of the weight of the filters when a filtering arrangement was applied (mg)
- $N_2 - N_1$ Difference between the mean values of the weight of the filters when no filtering arrangement was applied (mg)

The proven characteristics are listed hereunder:

Proven characteristic 1 – Different sized filters, F1 and F2: The external filter F1 was composed with a filtering diameter of 4 cm and a total high of 30 cm. The second external filter F2 had a filtering diameter of 6.3 cm and a total high of 41 cm. These were tested with the effluent of the washing machines flowing from top to bottom, with the filter cartridge at the bottom.

Proven characteristic 2 – Flow direction, F3: The F1-sized filter was also proven but turning the flow upside-down, i.e., the effluent flowed from the bottom to the top, with the filter cartridge at the top. It should be noticed that the filter arrangement F3 is the same as the F1.

Proven characteristic 3 – System inside the existing washer filter, F4: The same filtering criteria were applied in the already existing washer's filter. In this sense, the filtering media was the same as before (LDPE), and these were confined by using the same fiberglass. In this specific case, the volume of the existing washer filter was approximately 350 cm^3. Section S1 was an adapter to receive the water from the washing machine, section S2 was a hose with holes, in which the outside contained the pellets (S3).

Besides, in order to find the replacement time intervals of the cartridges, after the 20th washing cycle, the filtering arrangements were operated until they were clogged. In these trials, the efficiency was not measured.

Permeability coefficient and Porosity

The permeability coefficient was evaluated by applying Darcy's Law. The purpose was to determine the resistance to the flow of the devices when using LDPE as the filtering media with the conditions explained in the "filtering arrangements" section, specifically, the density of the pellets. Darcy's Law can be expressed by Equation 6.2.

$$Q = K\frac{\Delta h}{L}A \qquad Eq. 6.2$$

Where:

- Q Flow of the fluid (cm^3/s)
- K Permeability coefficient (cm/s)
- Δh Difference between the heights of the fluid at the influent and effluent of the filter (cm)
- L Height of the filtering section (cm)
- A Area of the filtering section (cm^2)

On the other hand, the porosity was calculated by introducing LDPE pellets with a density of 0.5 to 0.6 g per cm^3 in a recipient of 100 mL. Then, water was poured into the recipient and its volume was measured. The porosity was calculated with Equation 6.3.

$$\emptyset = \frac{V_W}{V_T} \qquad Eq. 6.3$$

From where $\emptyset$ is the porosity (dimensionless), V_W is the volume of water poured into the V_T 100 mL recipient filled with pellets.

6.3 Results and Discussions

Efficiency of the filtering arrangements

The four filtering arrangements described in the methodology section have been tested to evaluate their MFs retention performance. All the arrangements have shown a statistically significant difference between using or not the filtering system ($p < 0.05$ for all the cases, ANOVA). Regarding the tested models, as can be seen in Figure 6.4, the F1 arrangement has started retaining more than 50% of the MFs. It has also shown a constant and positive efficiency growth. Hence, at the 10th washing cycle, the retention climbed to 66%, while in the 20th washing cycle its retention was greater than 80%. In Figure 6.4 it can be also seen that in the last tested washing cycles the mass of MFs found in the filtered effluent was lower than 1 mg.

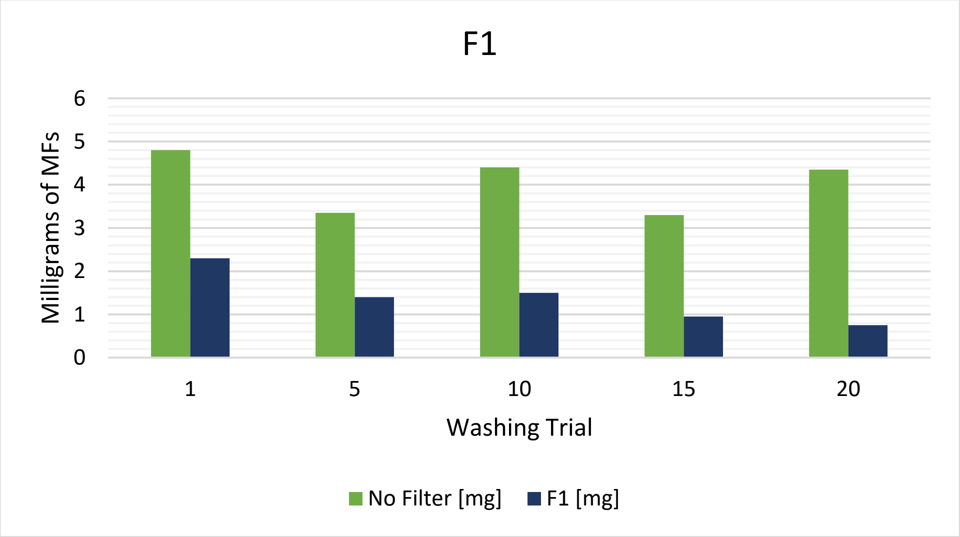

Figure 6.4. Retention of MFs with F1 arrangement (outside smaller filter).

Besides, the F1 arrangement was found to have a cartridge replacement time interval of approximately 30 washing cycles before clogging. On the other hand, the F2 arrangement showed a similar trend. As explained before, the F2 was larger and had more length and volume of filtering media (i.e., the LDPE pellets) than the F1. The F2 filtering model has shown a greater MF retention performance since the beginning (see Figure 6.5). In this line, the F2 model started with a MF retention greater than 55%, in the 10th washing cycle, it was already greater than 80%, reaching the performance that the F1 arrangement had at the 20th cycle. The F2 filter achieved a performance greater than 90% in the 20th washing cycle. This can be explained as a consequence of the larger length and volume of section S3, which gives more flow resistance and retention time to the MFs. Also, the volume of section S2 built to give space and time for the filtering process to occur was greater, meaning that the filtering can be done smoothly.

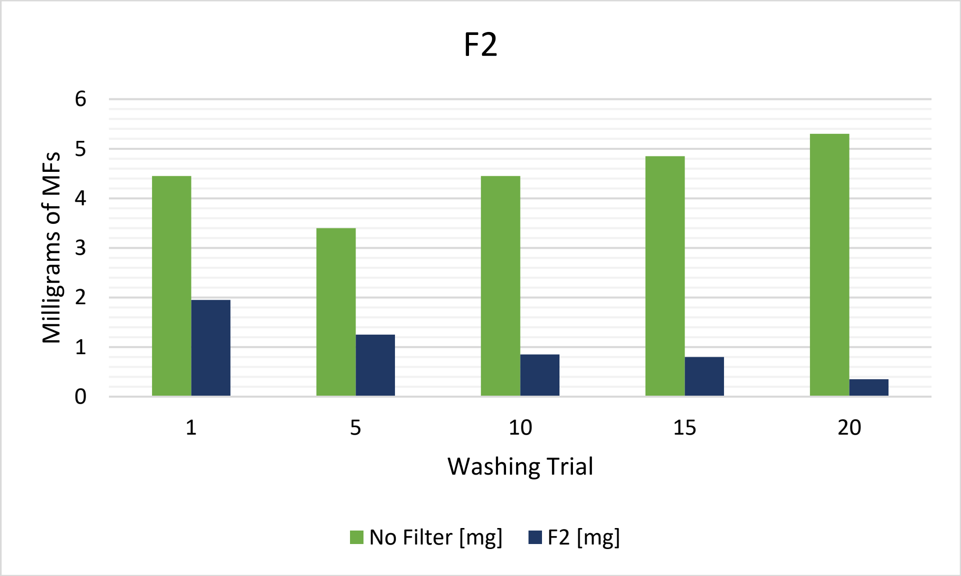

Figure 6.5. Retention of MFs with F2 arrangement (outside larger filter).

As can be seen in Figure 6.5, in the last washing cycles, the MFs that were found in the effluent approached zero. Besides, an extra advantage of using a larger filter is the increase in the replacement time interval of the cartridges and the subsequent comfort for the users. In this sense, the F2 arrangement was able to handle approximately more than 40 washing cycles before clogging, at least 10 more than the F1 model. Nonetheless, in the F1 and F2 models, the replacement time intervals might still be extended and the filtering efficiency improved by making some modifications mentioned in the patent. For instance, designing the filters with 2 non-consecutive filtering sections (i.e., interlayer sections).

As explained in the methodology section, the F3 filter model was arranged by turning the F1 model flow upside down, i.e., the effluent flowed from bottom to top. As can be seen in Figure 6.6, the F3 arrangement started with a retention efficiency greater than 85%. This means that F3 has shown an initial higher performance in contrast with the other tested arrangements. Besides, from the 15th washing cycle henceforward, the retention efficiency was almost 100%, indicating that the MFs' mass in the effluent was almost completely eliminated. Besides, the replacement time interval of the F3 cartridge was also improved in contrast to the F1 model reaching approximately 40 washing cycles. As these were identical in size, the conceived reason for the improvement was that the MFs didn't create a blockage at the begging of the filtering section (S3). Hence, the MFs and the dirt were easily retained in the previous section S2 and the filtering process was smoother as well.

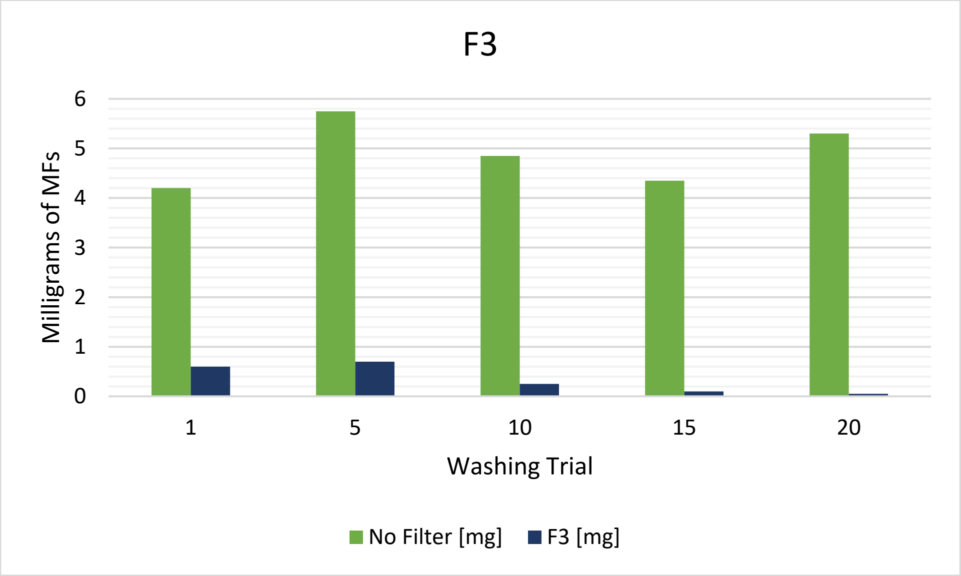

Figure 6.6. Retention of MFs with F3 arrangement (outside smaller filter with the bottom-up flow).

Finally, as mentioned before, the F4 arrangement was developed to be installed using the existing washing machine filter. This arrangement has shown a better starting performance than the F1 and F2 filters (see Figure 6.7). This might be explained as a consequence of the roughly radial filtering process that it ineluctably has; i.e., it has a concentric flow from the inside to the outside. The F4 started with 65% of efficiency and reached > 90% at the 20th washing cycle. As the F4 is constructed by using and surrounding the existing washer filter, the cartridge lifetime will depend on its size. Yet, in washers with small designed filters, a prolongation of the existing filter can be designed. In this way, the main advantage of this model is that it doesn't need the installation of external arrangements. Hence, the washers that have no space for external filters or that have the effluent hose welded to the discharge tube can still have an alternative to install a MFs catcher device. It should be mentioned that all the arrangements surpass the minimum expected by the users as an "optimal" replacement frequency (found at 17 washes by [6.33]).

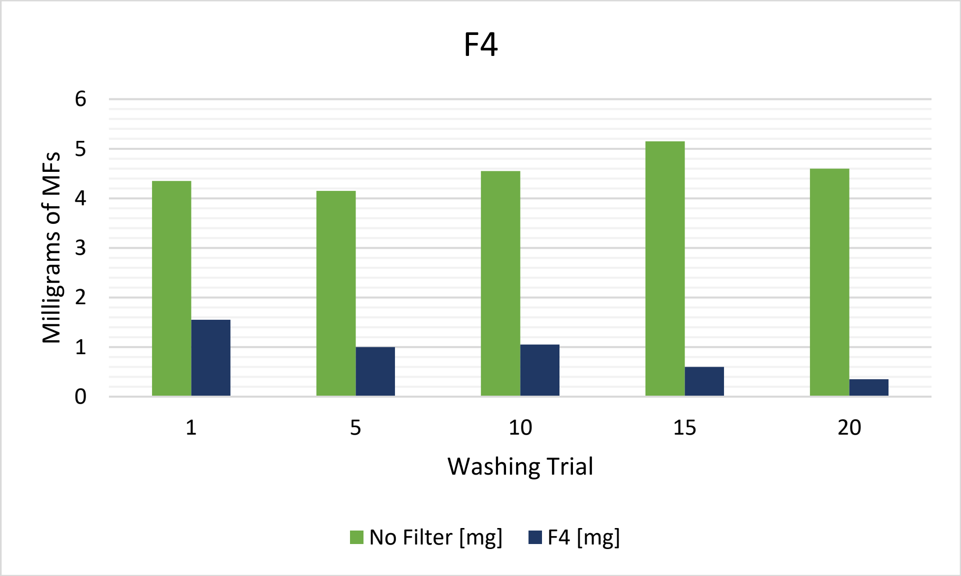

Figure 6.7. Retention of MFs with F4 arrangement: system introduced in the existing washer filter.

A new hypothesis arrived from these experiments, which was that the more MFs are presented in the effluent before reaching the filters the higher their retention efficiency. Hence, in a quick and separate experiment, a clean F2 model was subjected to a high concentration of MFs by introducing approximately 2 grams of prepared MFs into two consecutive washing trials (in addition to the textiles that were also washed). As expected, it was seen that the filtering arrangement was able to retain > 97% of the MFs. Hence, the more MFs are presented in the effluent the more MFs these arrangements will retain. In this sense, it can be deduced that a low percentage of retained MFs doesn't imply a poor performance but can be explained as a consequence of a low detachment of MFs. However, the more MFs are detached the lesser the replacement time interval for the filter cartridges.

Theoretically, in all the cases the filtering capacity of this system is determined by two main aspects. First, the lint that is formed between the MFs on and through the LDPE pellets. This latter feature provides the filter with an initial bump to start what can be referred to as a "chain reaction". In other words, the already retained MFs strengthen the filtering capacity by forming a new lint layer that catches more MFs. This can be seen in Figure 6.8, where the efficiencies of the filtering arrangements were found to be variable and positive (a linear trend with $R^2 > 0.9$) throughout the trials. This was also empirically seen through the translucent PVC filters, where the lint formed by the MFs was easily observed. In this sense, it was also appreciated that the lint formed "hot spots" where the accumulation was greater.

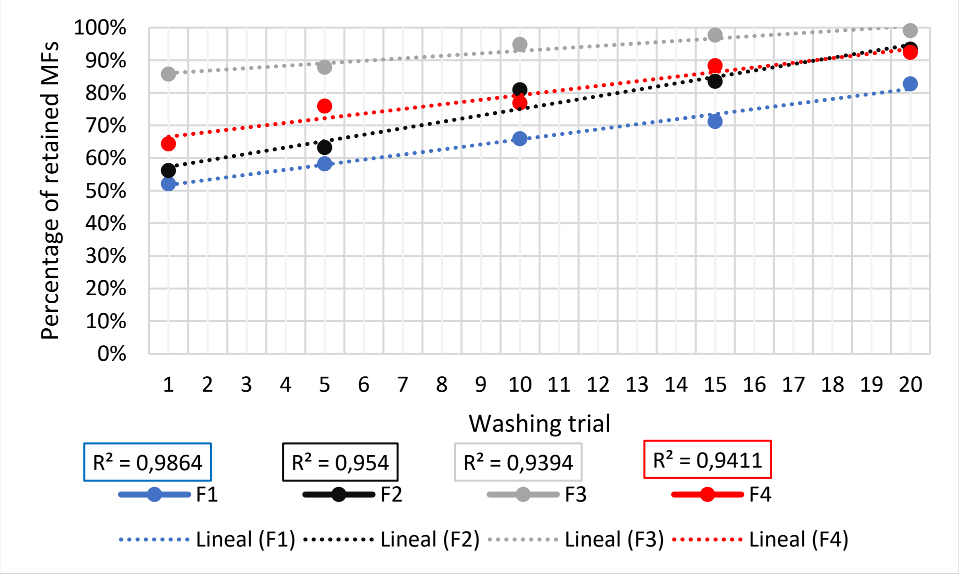

Figure 6.8. Comparison of MFs' retention efficiency for all the filtering arrangements.

Concerning the permeability coefficient, it was estimated in the order of 10^{-3} m/s (1.3 to 1.5 cm/s). This parameter is related to the structure of the filtering media and determines the resistance to the flow that the device will have when using 0.5 to 0.6 g per cm^3 of LDPE as the filtering media. As it can be seen, the encountered value can be considered as "very high" or "rapid" when compared to other similar filtering media concerning the filtration mode (e.g., sand) [6.34]. In this sense, the effluent of the washers can flow through the filters without any inconvenience. This latter feature was confirmed in the experiments made to assess the filter performance. Regarding the porosity, it was found at approximately 0.35, which was seen to give sufficient empty voids to let the water flow but to retain the MFs. It should be mentioned that the filter is a dynamic system, meaning that it will change most of the main parameters over functional time. However, as was seen throughout the experiments, these alterations affect the system by increasing the filtering efficiency.

On the other hand, the statistic of the method was measured by estimating its standard deviation. As seen through Figure 6.4, Figure 6.5, Figure 6.6, and Figure 6.7, the value was found at ± 0.5 mg of MFs. As explained in the methodology section, the method was tested by using small and large amounts of textile fabrics. The standard deviation was seen to be practically the same for both quantities (see supplementary information). In this sense, the most plausible explanation is that the error generated when weighing the MFs gets proportionally smaller when larger amounts of fabrics are tested. This data strengthens the hypothesis that the more quantity of textiles is washed together the fewer MFs are detached as a consequence of the friction reduction between the textiles and the washer [6.35].

Once the filters are exhausted the thermoplastic pellets and the MFs can be enclosed in a polymeric matrix that can be furtherly used for different purposes. We are currently working on this topic, which will be furtherly published.

Comparison with other MFs' filtering devices

Concerning other filters, F1, F2, F3, and F4 have presented a higher performance than most of the devices found in the market (see Table 6.3). Particularly, the F3 arrangement has shown the best retention efficiency. Two studies have evaluated the MFs retention efficiency of commercial devices. McIlwraith et al. (2019) [6.36] tested the *Cora Ball* and the *Lint LUV-R*. They used 545 g of a 100% polyester fleece blanket and applied a cold temperature, a total washing time of 30 min, 26.5 L of effluent, and a spin speed of 660 RPM. Excluding the mass of textiles used, the conditions are similar to the ones used in this study. Besides, they only evaluated the MFs retention efficiency at the 1st washing cycle. The filtering procedure to assess the MFs retention was done through a 10 μm pore size filter. They reported that these technologies can achieve a MFs drop from 26% (*Cora Ball*) to 87% (*Lint LUV-R*).

On the other hand, Napper et al. (2020) [6.37] also tested some technologies. They used three synthetic fabrics (jumpers) made of 100% polyester, 100% acrylic, and 60%

polyester/40% cotton. Each washing cycle was filled with 1300 g of textiles. To avoid the initial peak that new garments have on the MFs' detachment, they also washed the clothes 4 times before data collection. The front-loading washers were set at 30ºC, 1000 RPM, and 45 min. The reported outcomes corresponded to the average of the data collected after the 1st, 5th, and 10th wash. The filter pore size used was of 1 µm. They have reported that the external device *XFiltra* [6.38] was the best device with an efficiency of 78%. According to their study, two key features can explain the higher efficiency of this filter: a finest diameter pore in contrast with other external devices (60 µm vs > 175 µm), and the usage of an integrated pump, which might need extra energy supply. On the other hand, *PlanetCare* [6.39] affirms that their device has a MF retention efficiency of > 90%, which contradicts the findings of Napper et al. (2020) [6.37]. The results of these studies and ours are summarized in Table 6.3.

Table 6.3. Results reported by two different publications regarding MFs' reduction devices and comparison with the present study.

Device	**McIlwraith et al., 2019** [36] (1)		**Napper et al., 2020** [37] (2)	**Our work**				
	Retention Efficiency (%)			**Retention Efficiency by weight (%)**				
	By count	**By weight**	**By weight**					
In-Drum								
				Washing	**1st**	**5th**	**10th**	**20th**
Cora Ball (No mesh)	26	5	31	**F4**	64	76	77	92
GuppyFriend (50 µm)	-	-	54					
4th element (50 µm) (3)	-	-	21					
External Filters								
Lint LUV-R (150 µm)	87	80	29	**F1**	52	58	66	83
XFiltra (60 µm)	-	-	78	**F2**	56	63	81	93
PlanetCare (200 µm)	-	-	25	**F3**	86	88	95	99
(1) Outcomes are for the MFs retention average from 1st washing cycle (2) Outcomes are for the average of the 1st, 5th, and 10th washing cycles' data. (3) Napper et al. [6.37] for the reference of the 4th element washing bag.								

As can be seen in Table 6.3, different outcomes were reported for the same devices; hence, further interlaboratory trials are required. In addition, to effectuate a more accurate comparison, the results of McIlwraith et al. (2019) must be compared with our 1st washing cycles' outcomes. Whilst, the outcomes of Napper et al. (2020) must be compared with the average of the outcomes obtained from the 1st, 5th and 10th washing cycles. These averages are 59% for F1, 67% for F2, 90% for F3, and 72% for F4. It has to be mentioned that the final disposal of the retained MFs is a crucial concern still to be addressed. Napper et al. (2020) [6.37] commented, for example, that once the filters are cleaned by collecting the MFs, these can be "*thrown into the everyday household waste*". Hence, the MFs could also finish in the environment. In this line, besides the fact that this filter is made and uses recycled materials, it has the advantage that the MFs can be furtherly treated with the filtering media.

Another parameter that must be considered is the time intervals to clean or replace the filters. The *Lint LUV-R* needs to be cleaned every 2 to 3 loads of laundry according to the manufacturers. *Planet Care* can function for 20 washing cycles. *XFiltra* can handle 30 washing loads [6.37]. In our case, the smaller arrangements F1 and F3 were found to hold at least 30 washing cycles, whilst the F2 arrangement can withstand at least 40 loads of washing. On the other hand, the in-drum F4 arrangement replacement interval will depend on the size of the existing washing machine filter. Regarding the commercial in-drum devices need, they need to be cleaned once every load of laundry. According to [6.33], the minimum cleaning time interval for the users is between 15 to 17 washing cycles. Hence, not all devices meet this requirement. In the case of the system assessed in this study, the replacement time interval for the cartridges surpasses the users' expectations.

6.4 Conclusions

Different models of a new textile microfiber retaining system were evaluated. It was found that all 4 assessed arrangements have a good performance for retaining microfibers from the washers' effluents. Depending on the model, the microfiber retention efficiency was estimated between 52% to 86% in the 1st washing cycle and up to 83% to 99% in the 20th. The best performance was encountered when the flow of the washers' effluent went from bottom to top, being the filtering media at the top. Besides, all the arrangements showed a sufficient replacement time interval for the cartridges, as these were capable of handling more than 30 washing cycles. It is important to mention that one of the arrangements didn't need an external artifact as it was applied by surrounding the existing washing machine filter.

In addition to the good performance of these filters, it should be highlighted that they hold two relevant features. First, the usage of thermoplastic recycled waste for the filtering media and the shell, which strengthens the circular economy philosophy and produces a "greener" product. And, that the retained microfibers can be further and easily immobilized in a polymeric matrix by merging the filtering media with the microfibers inside. This latter feature can be harnessed to develop different types of products, tackling one of the main issues of the existing alternatives to reduce the microfibers, which is the subsequent treatment of these pollutants.

6.5 Acknowledgments

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Chapter 7:

Novel treatment to immobilize and use textiles microfibers retained in polymeric filters through their incorporation in composite materials

7 Novel treatment to immobilize and use textiles microfibers retained in polymeric filters through their incorporation in composite materials

This chapter provides a novel and sustainable route to treat textile microfibers by immobilizing them in a polymeric matrix and turning them into new composite materials. The properties of carriers obtained by immobilization of Mfs are also studied.

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Additional information about this topic cap be found in the annexes of this thesis:

Annex B: Process for the immobilization of microfibers to obtain carriers for the treatment of used water in MBBR plants (mobile bed bioreactors). This study corresponds to a part of the Project "MBBR carriers" founded by UPC (R-02058 – Ajut als instituts 2021).

Annex C: F.Belzagui, B.Amante, C. Gutiérrez-Bouzán. Development of a Self-Sustaining Floating Water Treatment System with Renewable Energy Supply ETAF. This work corresponds to an application of carriers to the treatment of surface waters. It has been presented in the II International Congress on Water and Sustainability (Terrassa, Spain, 24-26 March 2021) and it is published in "Book of Abstract: II International Conference on Water and Sustainability (OmniaScience, Editors: B. Amante, F. Belzagui, V. Buscio a L.Canals). DOI: 10.3926/icws2021. ISBN: 978-84-123480-0-2

Highlights

- Textile microfibers have retaining alternatives but no further treatment has been proposed.
- Retained microfibers can be treated by immobilizing them in a polymeric matrix.
- Microfibers act as reinforcement of polymer composites improving mechanical properties.
- These composites can be furtherly used in different applications.
- Half of the European production of recycled plastics is sufficient to treat all microfibers.

Abstract

Microplastics (MPs, size < 5 mm) are within one of the most environmentally challenging pollutants. Their continuous and cumulative inflow to or generation in the environment is what makes them drastically problematic. These pollutants can come from a wide variety of sources; hence, they are potential vectors to carry environmental and human health extensive risks. Microfibers (MFs) are one type of MPs. One of the most renowned sources is the MFs detached from textile articles from household laundering or industrial processes. Currently, there are many alternatives to retain the MFs detached from textile articles. However, as far we know, there are no alternatives to valorize the retained MFs. In this sense, we are proposing a novel and sustainable treatment to immobilize the MFs in a polymeric matrix, turning them into a composite. To determine the mechanical properties of the expected composites, different proportions of polyester MFs were mixed with low-density polyethylene which is the material proposed for immobilization of MFs. The results showed that the optimum manufacturing composition was the use of 10% (vol/vol) of polyester MFs in the polymeric matrix. This composition improved some of the tensile mechanical properties of the polymeric matrix. Once the composites are obtained, these can be used for different purposes.

Keywords: Microplastic; Microfiber; Treatment; Textile; Contamination; composites.

7.1 Introduction

Considering the definition of the European Chemical Agency (ECHA), the microplastics (MPs) are fragments of chemically modified and/or non-biodegradable polymers with a length < 5 mm [7.1]. These pollutants have been encountered in every assessed ecosystem, hence, they are referred to as ubiquitous [7.2]. Every plastic product is a potential source of MPs. In this sense, sources have been divided into two main groups. Primary MPs are those emitted into the environment in a MPs size range; whilst secondary are those generated in the environment from the fragmentation of larger plastic debris [7.3]. The microfibers (MFs) are one type of MPs, these have a length to diameter ratio > 3 and a maximum length of 15 mm [7.1]. Textile MFs are among the most renowned as these have been widely found in the environment. These can be generated in the manufacturing, use, cleaning, and final disposal of a textile article [7.3]. Household laundering and textile industrial processes detach millions of MFs per year [7.4–7.6].

The impacts from MPs have been widely studied; for instance, the ingestion across the trophic chain is currently a widely known fact [7.7–7.10]. On the other hand, some effects of the MPs on organisms have been found, like their retention in the intestinal tract and endocrine disruption [7.11–7.14]. However, most of those articles might lack realism as the concentrations used are not usually found in the environment. Besides, the MPs used for laboratory trials are not the ones that are mostly encountered, which are MFs [7.15]. MPs can also behave as vectors for organisms and hydrophobic toxic compounds [7.16, 7.17]. This contamination has also reached products for human consumption and has polluted the air [7.18–7.24]; hence, there are several paths for human exposure to MPs [7.25]. Nevertheless, the potential risks for human health are still unknown [7.26–7.28].

Some solutions have been proposed to retain the MFs detached from washing machines. There are in-drum accessories to reduce the generation of MFs or out-drum filters to retain the already generated ones [7.29, 7.30]. However, as far as we know, there are no existing alternatives to finally treat the retained MFs. Napper et al. (2020) commented, for example, that once the filters are cleaned by collecting the MFs, these can be "*thrown into the everyday household waste*" [7.31]. Hence, the MFs could also finish in the environment. This article aims to present a novel, practical, and sustainable treatment for the valorization of the retained MFs. The treatment of the MFs is covered under a pendent Spanish patent. The method consists in making a composite with the MFs, and a thermoplastic polymeric matrix; in this case, polyester (PES) and low-density polyethylene (LDPE). PES was selected as it is the fiber mostly used by the textile industry [7.32, 7.33]. On the other hand, LDPE was selected because of its relatively low fusion temperature. The composites were made by mixing different proportions of PES and LDPE. The composites were subjected to tensile tests to evaluate mechanical properties and the morphology of the fracture's surface was analyzed by Scanning Electron Microscopy (SEM). The materials showed very good compatibility since some of the tensile mechanical properties of the LDPE were improved because polyester MFs act as reinforcement of the polymer matrix. In this line, we are proposing a novel and practical method for the valorization of textile MFs pollutants.

7.2 Materials and Methods

Composites Manufacturing

The composites were prepared by mixing different proportions of PES-MFs with LDPE. The mixing was carried out in a CollinW100T two-roll mixer (Dr. Collin, GmbH, Germany). The temperature in both cylinders was set at 130°C. Once the LDPE was melted the MFs were introduced and mixed for about 10 minutes to achieve a sufficient mixing between the polymers. The tested compositions were 5%, 10%, and 15% (vol/vol) of MFs in the polymeric matrix (LDPE). Higher proportions did not allow to obtain compact composites since more MFs did not have enough polymeric matrix to coat all the MFs. Once the mixing was completed, the blend was then consolidated at 100 kN and 140°C for 5 min, in a Collin Mod. P 200E hot plates press machine (Dr. Collin GmbH, Germany, forming square plates (100x100x2.5mm^3). The cooling process is carried out using cool water for refrigeration. Test samples were properly shaped according to the ASTM-D-412-98 specifications to be used in the tensile test. Plain LDPE without MFs was also treated in the same way as the filling materials to obtain suitable reference samples.

Tensile Test

The tensile strength tests were carried in an Instrom 3366 (*Instrom, UK*) universal machine by following the standard ASTM-D-638-14 [7.34]. The tensile test speed was set at 1 mm per minute. Young's modulus, tensile strength, and elongation at maximum load were calculated using Bluehill version 2 software. Five test specimens per composite were tested and compared with pure LDPE. The mean and standard deviation were calculated and used to understand the mechanical behavior of the composites.

SEM Images

Scanning Electron Microscopy (SEM) was used to qualitatively examine the fracture surface of the broken samples from mechanical testing. By observing the environment of the MFs it was possible to analyze the adhesion of the fibers to the matrix. Several images of every composite sample were studied. The microscope used was a Phenom G2 Pro device, (FEI company, USA) at an accelerating voltage of 15 kV. Previously to the observations, one sample of each of the composites was prepared by coating them with a fine layer of gold-palladium to increase their conductivity.

7.3 Results and Discussions

A picture of composites with 10% of MFs can be seen in Figure 7.9. However, as explained before, different proportions were proven (5%, 10%, and 15%). Visually speaking, no variation was observed between the different composites. Besides, no MFs were seen at the composites' surface, meaning that these were totally trapped inside the polymeric matrix. This observation was considered a good outcome, as composites with MFs' "limbs" did not appear on the surface. It has to be noticed that these composites were composed of discontinuous MFs randomly oriented in a polymer matrix. Hence, the aim of the work is not to improve the properties of a polymeric material but to get rid of the MFs that are detached from textile articles. In other words, to give a final treatment to a waste that is currently concerning the scientific community.

Figure 7.9. Composites made with 10% vol. of PES-MFs in a LDPE matrix. The lighter section of the photo is a visual effect of the incident light.

In Figure 7.9 a squared shape of a composite is shown, however, depending on the mold used in the hot plates press machine, other shapes can be formed. The shape will only depend on the mold used in the hot plates press machine. Hence, a wide variety of products can be made, as it will only need a mold to make replications. Besides, no novel or unconventional equipment is needed to make the composites. The development of this treatment was made considering the equipment commonly used in polymer recycling plants. In this sense, depending on the product that would be made, some recycling plants can introduce the MFs to their pre-products without making any significant changes to their processes.

Tensile Tests Outcomes

Once again, it has to be emphasized that we are not dealing with fibers that could be arranged and prepared to provide a series of better properties to a given polymer, i.e., a grid to insert inside the polymer. In this case, we are working with “garbage”, MFs obtained from textile articles that are randomly inserted into the polymer matrix. Nevertheless, as shown in Figure 7.10, the tensile strength, as well as the Young’s Modulus, were improved when inserting the MFs into the polymeric matrix. Hence, the term “garbage” could be transformed into “raw material”. In other words, we are transforming MFs’ pollutants into feedstock to improve the tensile stress of the LDPE. As can be seen in Figure 7.10, under the criteria of homogeneity of the properties, the outcomes showed that the most preferable proportion was when 10% of MFs were included in the polymeric matrix. This was elucidated from their standard deviation regarding the Young’s Modulus and the strain: 11 for 5%, 12 for 10%, and 28 for 15% (standard deviation values for Young’s Modulus). In this line, at a certain point between 10% to 15% of MFs, the standard deviation becomes larger, which could indicate an inflection point from where the composite begins to lose some of its homogeneity. However, depending on the application that these composites could have, i.e. for non-structural applications, the 15% MFs’ proportion composites can be sufficiently homogeneous if the objective product won’t be subjected to tensile stress greater than 8 MPa.

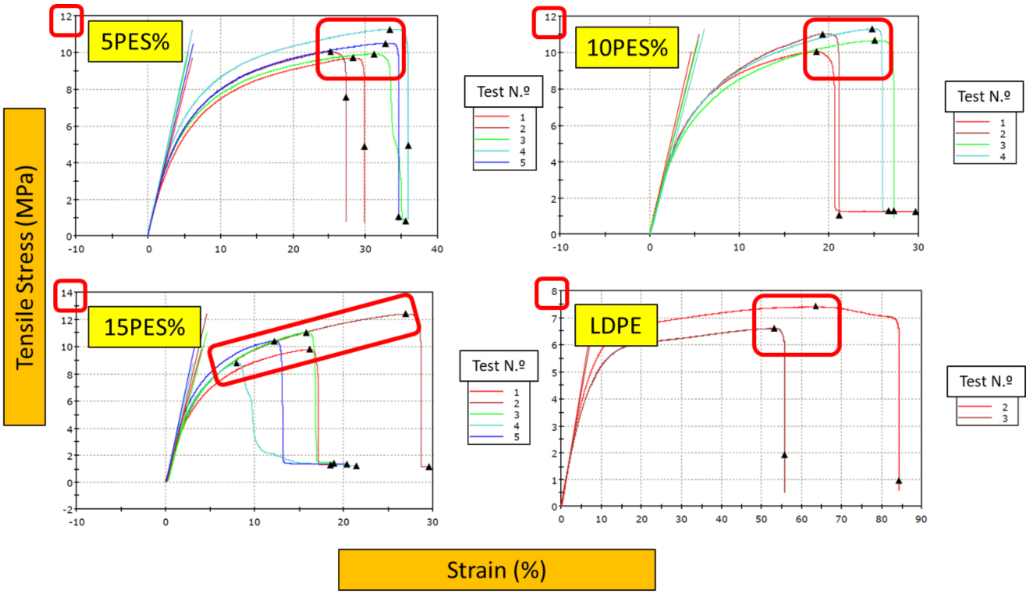

Figure 7.10. Strain (%) versus tensile stress (MPa) of composites with different compositions of polyester microfibers (PES-MFs) in the low-density polyethylene matrix (LDPE). Please, be aware that the "y-axes" do not present the same scale.

It has to be mentioned that what is gained in tensile stress is lost in elasticity. In other words, the composite gets more prone to be ruptured without a great deformation. As can be seen in Figure 7.10, in all the cases Young's modulus increased (see Table 7.4). The most plausible explanation for this outcome is that Young's modulus of the polyester fibers is much higher than that of LDPE [7.35]. From our experimental data, the LDPE Young's modulus was 9 MPa whereas for PES the values are between 0,92 and 10 GPa [7.37-7.39]. Hence, the contribution of PES microfibers causes an increase of tensile strength at maximum load compared with pure LDPE.

Table 7.4. Mean Young's modulus of each composite.

Composite	5% PES	10% PES	15% PES	100% LDPE
Mean Young's modulus (MPa)	172,85	200,03	276,92	89,24

In addition, tensile strength at maximum load of LDPE was lower than for composites but can suffer a longer deformation. With the obtained data, we can apply the rule of mixtures to estimate the parameter "K" [36] of Equation 7.1, which is a fiber efficiency parameter that gives an indication of the contribution of the MFs' properties to the composites.

$$E_C = K E_F V_F + E_M V_M \qquad Eq. 7.1$$

From where:

- E_C Young's Modulus of the composite
- E_F Young's Modulus of the polyester MFs (mean between 0.92 to 10 GPa) [37][38][39]
- V_F Volume of MFs included in the matrix
- E_M Young's Modulus of the LDPE (0.09 GPa, experimentally obtained)
- V_M Volume of LDPE included in the matrix

Hence, we can estimate the "K" for every measured point:

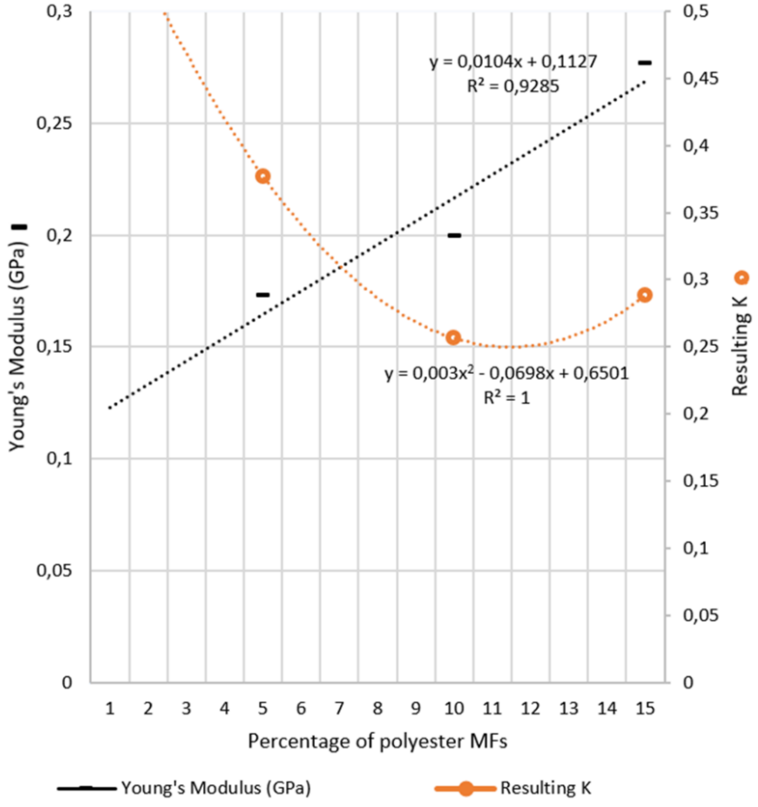

Figure 7.11. Young's modulus and the resulting "K" for each of the composites made.

From Figure 7.11, we can see that initially, the "K" parameter decreases from 0,38 to 0,26 when the MFs' concentration changes from 5% to 10%. However, when the MFs increased to 15%, the "K" parameter also tends to increase. These changes observed in the "K" parameter may probably be caused by differences in the orientation and distribution of the MFs. Hence, when the concentration of MFs is low the fibers could be preferentially randomly and uniformly distributed within a specific plane expecting values of "K" of 0,375 (as mentioned in William and Rethwisch (2018) [7.36]). On the other hand, the increase of the MFs concentration up to 10% vol. may probably induce a random and uniform distribution of the fibers within the three dimensions in the space causing the reduction of the "K" parameter. Continuing on the line, between the 10% to 15% an increase of the "K" was noticed, revealing a change in the composites` behavior. Hence, with the obtained equations we can determine the "inflection" point. This could be, as explained before, the point at which the standard deviation of the composites' tensile stress begins to increase corroborating the results of the tensile tests shown previously. Setting the derivative of Equation 7.2 equal to zero, the MFs composition corresponding to the minimum "K" can be found:

$$y = 0.003x^2 - 0.0698x + 0.6501 \qquad Eq. 7.2$$

$$\frac{dy}{dx} = 0 = 0.006x - 0.0698$$

$$x = 11.6\%$$

Where "x" represents the percentage of MFs and "y" the fiber efficiency parameter "K".

In other words, at approximately 12% of MFs composition, the composite starts to be less homogeneous. This might be because the MFs fill a significant space of the composites, lowering the homogeneity. However, more data and experimental observations should be done to make a more precise conclusion. On the other hand, as can be seen in the SEM images (Figure 7.12), the adhesion of the MFs to the matrix could be improved by introducing other types of MFs with more asperity (e.g., cotton). In real conditions, cotton and other MFs will be present, as the filters used to retain the MFs do not discriminate between synthetic or natural fibers.

SEM Images

The fiber-matrix adhesion achieved in this case can be clearly seen in Figure 7.12, where a type of "tunnel" is formed between the PES-MFs and the LDPE-matrix. These tunnels might be formed when the LDPE contracts as a consequence of its cooling and hardening. The images revealed a lack of fiber-matrix compatibility mainly because the PES microfibers are smooth with negligible roughness [7.40]. However, as said before, when working with real mixed materials that are outflowed from a washing machine, other types of MFs will also be included in the composites. In this sense, materials with more roughness will appear, hence, a mechanical adhesion is expected, increasing the adhesion between the MFs and the polymeric matrix. Nonetheless, despite the low compatibility, this does not mean that the composite has lower tensile strength, because, as stated in the previous section, the tensile strength increases when the MFs are included. Hence, to achieve a composite with improved mechanical properties is imperative to make a good mixing before doing the final products. An important aspect to consider is that this treatment can include the MFs independently on the retaining method used (e.g., XFiltra, PlanetCare, Microplastics LINT LUV-R [7.31]).

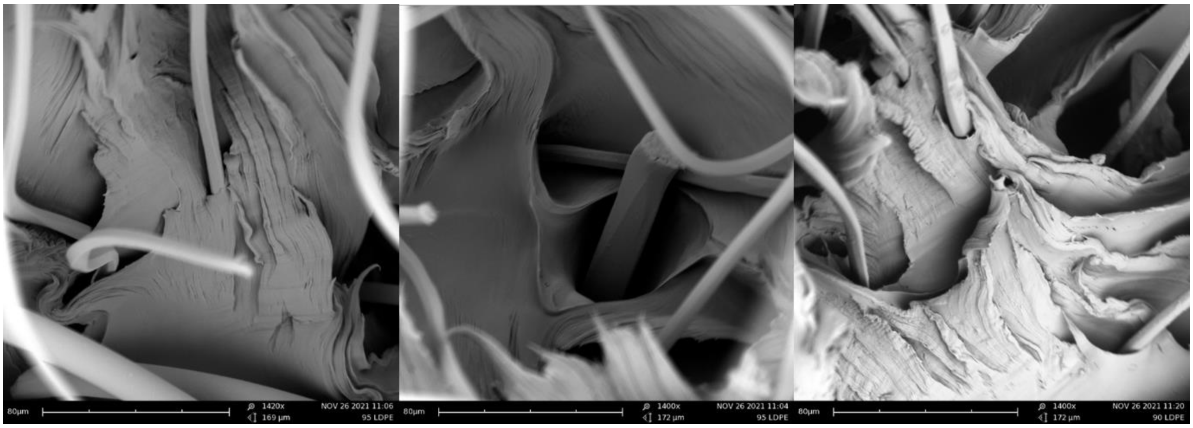

Figure 7.12. SEM images of the composites at x1400. The first two images are for 5% PES and the last one for 10% PES.

Practical solution?

Currently, only in Europe, there are 10 million tons of LDPE that are produced every year. If we only care about LDPE it occupies about 20% of the market, principally in reusable bags, trays, food packaging, among others [7.41]. Nowadays, in Europe, about 40% of the packaging waste, which commonly comes from a type of PE or polypropylene, is recycled [7.42]. On the other hand, globally, there are 0.5 million tons of MFs that are annually generated from domestic laundering [7.43]. This means that recycling 5 million tons, half of the European production, of LDPE will be enough to give the MFs a sustainable and practical treatment. In our facilities, we are currently working with 10% (vol/vol) MFs composites by using them in biological wastewater treatment plants as moving bed bio-reactor carriers. The results will be published once sufficient data are gathered.

7.4 Conclusions

This study presents a novel and sustainable treatment for microfibers detached from textile garments. The treatment is very simple and doesn't need sophisticated equipment or high energy or resources demand. In this sense, to treat the microfibers we are proposing to immobilize them into a polymeric recycled matrix from where they cannot escape. Once inside, MFs based polymeric composites can be obtained and different applications can be given to these composites. In this work, three different proportions of polyester microfibers (5%, 10%, and 15%, vol/vol) were introduced into recycled low-density polyethylene matrix. The purpose was to have an approximated knowledge of the higher concentration of microfibers that can be included and still be able to produce a high-quality product.

In this sense, it was seen that the composites having up to 10% of microfibers behaved homogeneously. A lower concentration of microfibers also worked fine, but our objective is to treat the currently "fibers' microplastic pollution from laundering". Hence, the more microfibers that can be included, the better. Nonetheless, it was seen that when including 10% of microfibers in the thermoplastic polymer matrix, some mechanical properties, as the tensile strength or Young's modulus improved at the expense of reducing the maximum deformation achievable. Besides, no microfibers were detached from the final composite, meaning that these were totally inserted in the matrix. On the other hand, SEM images showed a low fiber-matrix compatibility due to the non-existent roughness of the PES microfibers. However, in real conditions, other microfibers with more roughness than that of polyester will be included, increasing even more the gripping between the pollutants and the recycled polymer by promoting mechanical adhesion.

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Chapter 8:

Cigarette Butts as a Microfiber Source with a Microplastic Level of Concern

8 Cigarette Butts as a Microfiber Source with a Microplastic Level of Concern

This chapter evaluates the detachment rate, toxicity, and degradability of the microfibers detached from the cigarette butts. It was published in Science of the Total Environment (STOTEN, Elsevier, IF = 7.963. Q1, JCR-WoS):

F. Belzagui, V. Buscio, C. Gutiérrez-Bouzán, M. Vilaseca. *Cigarrette Butts as a Microfiber Source wiht a Microplastic Level of Concern*, 2021, 25; 762. .doi:10.1016/j.scitotenv.2020.144165.

Abstract

Microplastic pollution is a growing environmental concern among the scientific community. These small particles (<5 mm) might come from the fragmentation or direct emission of artificial and synthetic polymers. Among them, the microfibers (MF) are one of the most common types of microplastics identified in the environment. On the other hand, the most encountered type of garbage found in clean-up campaigns is the cigarette butts, which contains the smoked filters (SF) and unsmoked rests of tobacco. SFs are hazardous debris but are usually not properly disposed as such, and are composed of >15'000 strands that can be detached as MFs. This study aims to evaluate the detachment rate, acute aquatic toxicity, and the aquatic-, thermooxidative-, and photo-degradability of the MFs generated from SFs. In this way, it was found that SFs detach approximately 100 small MFs (< 0.2 mm) per day. In a rough estimation, about 0.3 million tons of potential MFs might be annually reaching aquatic environments from this source. Concerning the eco-toxicity, a statistically significant difference was found when MFs are present in the leachate generated by the SFs, where the *Daphnia magna* EC_{100} and EC_{50} were of 0.620 SF/L and 0.017 SF/L, respectively. Finally, the degradability of the SFs was evaluated by applying two methods (ATR-FTIR analysis and gravimetry). In both of them, a low degradability rate was observed. Thus, it may be concluded that MFs from SFs constitute an important source of microplastics, which might partially explain the high concentration of artificial polymers that have been found in the deep-sea sediments. Yet, the correct management of the SFs is an unsolved issue that should receive urgent attention.

Keywords: Microplastic; Microfiber; Pollution; Cigarette Filter, Cigarette Butts.

8.1 Introduction

The microplastics as pollutants are small fragments (length < 5mm) of non-chemically modified and non-biodegradable polymers that have been found contaminating every ecosystem. Within the types of microplastics, microfibers (MF) are those with a length to diameter ratio >3; i.e., a maximum length of 15 mm [8.1]. A possible but usually not considered source are the cigarette butts, or smoked cigarette filters (SFs). In this sense, several clean-up campaigns have reported that SFs are the most common litter in number of items across the world [8.2, 8.3]. In 2017, volunteers around the globe collected more than 2 million SFs, outnumbering other usual plastic wastes as straws, plastics bags, bottle caps, and food wrappers [8.4]. In this sense, Roder, Putschew, and Nehls 2014 reported a mean and a maximum of 2.7 and 48 cigarette butts per m^2, respectively. The current global consumption of tobacco is 6 trillion cigarettes per year [8.6–8.8], and it has been estimated that three-quarters are not correctly littered. Hence, about 4.5 trillion SFs per year are carelessly discarded into the environment [8.9–8.12]. Despite that some ideas have proposed to make use of this litter [8.13], these residues continue to constitute a severe global issue of incorrect toxic litter disposal, yet, deficient attention has been placed onto them [8.14].

The tobacco cigarette filters are generally composed of more than 15’000 fibers strands made of cellulose acetate with plasticized additives. This material can last up to 30 years to degrade in certain conditions [8.15–8.17]. When incorrectly disposed, SFs might suffer a quick release of the strands, which can be detached as a MF, or get fragmented over time. Large concentrations of “rayon” fibers have been identified polluting the deep ocean sediments, which can be partially attributed to the strands detached from littered SFs [8.18, 8.19]. On the other hand, tobacco cigarettes carry more than 4’800 chemical compounds, from where more than 70 are carcinogens and over 200 are toxics [8.20]. When cigarettes are smoked, a proportion of these compounds are adsorbed by the filters. Afterward, some can be rapidly leached into the environment, while others remain adsorbed in the strands for an indefinite period [8.12, 8.21, 8.22]. Hence, MFs from SFs can act as vectors for many hazardous chemicals and could even behave as a source for prolonged radionuclide contamination [8.23].

This study aims to evaluate the releasing rate, ecotoxicity, and the aquatic-, thermooxidative-, and photo-degradability of the MFs detached from SFs, and to approximate its relevance to the overall MFs' pollution. To do so, the releasing rate of MFs from these filters was measured in synthetic seawater. A rough estimation of the potential amount of these MFs that reach aquatic environments was done by considering the smoking trends of all the countries and the worldwide municipal water treatment rate. Unsmoked cigarette filters (UFs) and SFs were tested to measure the degradation of the cellulose acetate and to determine the possible influence of the smoking process in this degradability. Also, the toxicity of the MFs was assessed by means of an acute aquatic toxicity test on *Daphnia magna*.

8.2 Materials and Methods

8.2.1 Smoking process of the cigarettes

Four different international and widely consumed commercial brands of tobacco cigarettes were purchased from a common store in Terrassa, Spain. The smoking process of the cigarettes was carried out by using a self-prepared gadget that consisted of an air bomb connected to a valve. The system was regulated to obtain a smoking velocity of approximately 1 cigarette per 2 minutes. The smoking velocity selection was a consequence of previous attempts, in which an overheating of the cigarette filters was seen when higher smoking velocities were applied. This could have derived on the filter's physical degradation. Besides, it does not correspond to the normal smoking procedure.

8.2.2 Smoked cigarette filters as a source of microfibers

Ten SFs were carefully separated from their wrapping papers to facilitate the final visual identification of the MFs. The wrapping paper is rapidly detached when coming in contact with water, hence, its removal doesn't influence the release of MFs. Then, the SFs were placed in 10 L of distilled water to obtain a sample with a known concentration of 1 SF/L. Afterward, the sample was submitted to a slow but continuous mechanical agitation for two weeks. The agitator consisted of a bowl with a blade that was assembled to generate a slow waving motion (frequency of 1 cycle per 2 seconds, maximum amplitude of the waves of approximately 2 cm). The purpose was to emulate the slow movement of natural water bodies. Two times per week, three aliquots of 50 mL were taken, filtered through 20 µm polyamide filters (*Merck Millipore Ltd. Nylon Net*, 47 mm of diameter), and dried for 24 h at 60ºC. The filtering process was done by using a vacuum pump. The counting of the MFs was executed by visually inspecting the whole area of the polyamide filters in a stereomicroscope (*Carton Stereo Zoom SC*).

8.2.3 Ecotoxicity assessment of the microfibers released from smoked cigarette filters

The evaluation of the toxicity of the SFs and the released MFs was executed by following the standard toxicity test "*OECD Test No. 202, Daphnia sp. Acute Immobilisation Test*" [8.24]. Because the leachate of the SFs can have a negative effect by themselves (i.e., it contains toxic substances desorbed from the cigarette filter), two settings were prepared to evaluate the toxicity that can be attributed to the MFs.

In the first one, 10 SFs were vigorously agitated in 1 L of distilled water for 24 h. The generated leachate was filtered through 20 µm polyamide filters to remove the SFs. In the second set, 10 SFs were cut into MFs of approximately 1 mm in length, put into 1 L of distilled water, and vigorously agitated for 24 h. Afterward, the *Daphnia magna* matrixes were prepared with concentrations of 10, 4.8, 1.0, 0.48, 0.10, 0.048, and 0.010 SF/L from both sets of samples.

Every matrix was prepared by following the standardized OECD test. In this way, for both sets and every concentration, 4 replicates were executed with 5 *Daphnia magna* each. Additionally, a control sample was prepared. The organisms were evaluated after a period of exposure of 48 h, where self-moving *Daphnia magna* were counted as living organisms and compared with the control sample.

8.2.4 Degradation of the UFs and SFs

The evaluation of the degradation rate of the cigarette filters was developed by following two methods: (1) chemical analysis through FITR after salty water and UV-light treatment to the filters, and (2) gravimetrical analysis after applying the filters to a microplastic separation method proposed by the *National Oceanic and Atmospheric Administration* (NOAA).

In the first method, UFs without wrapping papers were put in 2 glass containers (3 UFs in each) one with distilled water and the other with synthetic seawater (3.5% NaCl w/v). Then, these were submitted to a slow mechanical agitation (frequency of 1 cycle per two seconds, maximum wave amplitude of 2 cm) and continuous UV-light (365 nm, intensity of 7 mW/cm^2, *Vilber Lourmat lamps*) for one month. The same procedure was simultaneously conducted with the same number of SFs. Afterward, to measure the possible degradation of the acetate cellulose strands, the outermost layer of every filter was sampled with a clamp to evaluate its chemical structure employing an ATR-FTIR (Nicolet 6700 FTIR, *ThermoScientific*). The outermost layer was selected as it was the portion of the filter with the highest UV-radiation exposure. Besides, UFs and SFs non-submitted to the test were also analyzed for comparative purposes. On the other hand, MFs from the SFs were left in containers with freshwater for 18 months in a place where the daylight could reach them. Afterward, three samples were analyzed with the same FTIR equipment.

On the other hand, the microplastic identification method proposed in NOAA 2015 [8.25] was also applied. This method relies on a modified Fenton Reaction (Fe(II)/H_2O_2) to oxidize "non-synthetic" polymers into carbon dioxide. The procedure was established as follows: SFs (made of cellulose acetate) were weighted without the wrapping papers. To have a reference frame for evaluation, woven fabrics of 100% cotton (COT) and 100% polyester (PES) were cut into regular pieces of similar weight. Thus, a total of 6 samples of each material were dried for 24 h at 60 ºC before final weighting. Then, the initial solutions for the Fenton reaction were prepared, which consisted of a mix of (A) Fe (II) 0.5 M, and (B) H_2O_2 at 30%. Equal volumes (20 mL) of each solution were mixed in glass containers and heated up to 75 ºC in a water bath. Then, the samples (filters, PES, and COT) were independently immersed in the glass containers for 15 minutes while kept in continuous stirring at 75 ºC. After the first 15 minutes, three runs of experiments were executed, where two samples of each material were tested in each run.

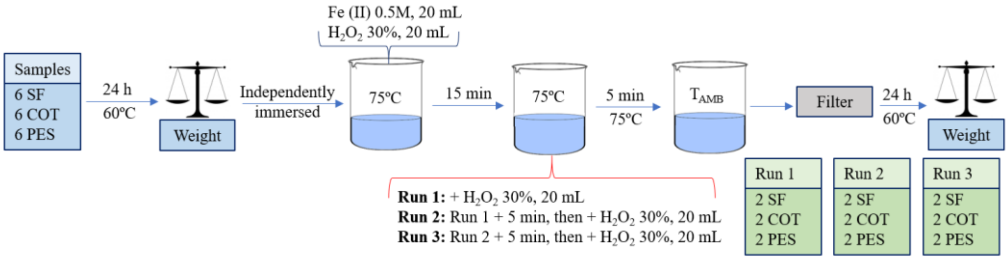

Figure 8.1. Schematic overview of the experimental set to evaluate the degradability of the filters (SF) compared with cotton (COT) and polyester (PES). Each sample was independently treated

In the first run, 20 mL of solution B was added after 15 minutes. In the second run, the same was done but adding 20 mL of solution B after 20 minutes. And, in the third run, one last volume of 20 mL of solution B was added after 25 minutes. For each time the solution B was added, the samples were maintained for 5 more minutes at 75 ºC, which is the optimal condition for the chemical reaction to occur. The three times interval of 5 minutes each were selected to quickly increase the reaction velocity. Afterward, the glass containers were covered with a plastic film until room temperature was reached, and then the solutions were filtered through 20 µm polyamide filters (*Merck Millipore Ltd. Nylon Net*, 47 mm of diameter), and oven-dried for 24 h at 60 ºC. The filtering process was done by using a vacuum pump. The polyamide filters were previously weighted after being dried for 24 h at 60 ºC. The difference in mass between the initial sample and that retained on the polyamide filters was taken as a direct measurement of the degradability (Figure 8.1).

8.3 Results and Discussions

8.3.1 Smoked cigarette filters as a source of microfibers

As explained in the methodology section, SFs were immersed in water and submitted to slow agitation. As expected, SFs released many MFs from the very beginning of the experiment. As can be seen in Figure 8.2, every SF detached approximately 100 MFs per day in the first two weeks. However, this detachment might accelerate over a long-time period as a consequence of the physical deterioration of the fibers that compose the filters [8.26, 8.27].

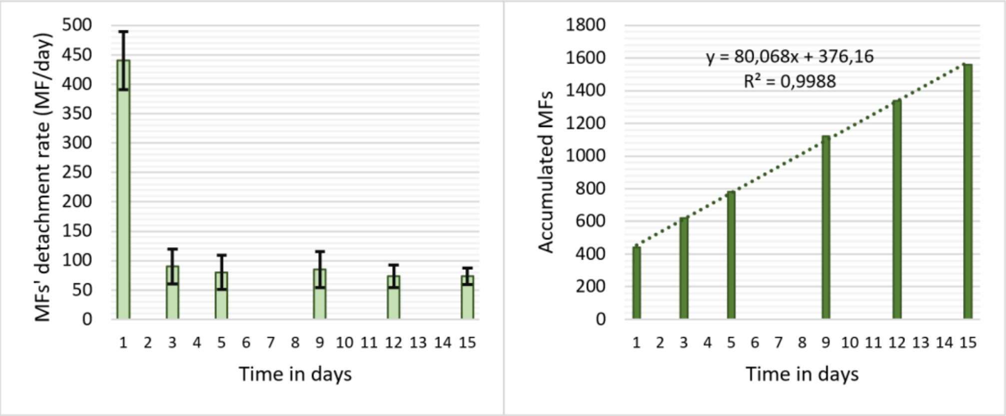

Figure 8.2. Microfibers (MFs) released per smoked cigarette filter per day in the left image, standard deviation is included. Accumulated MFs per smoked cigarette in the right image, the fitted model is included.

On the other hand, on the first day of soaking, the detachment was observed to be relevantly superior (450 MFs). In this line, Mepani et al. 2002 reported that some strands might present defective, smaller, and poorly attached fibers from the manufacturing process of the filters. These fibers might explain the rapid first release of MFs when the filters are immersed in water. On the other hand, it is not clear that if the usual process of extinguishing the cigarette ember onto a hard surface will accelerate the detachment of the MFs by the physical deterioration of the SF, or to lower it as a consequence of the tar that might bind the MFs together. Regarding the size, most encountered lengths were <0.2 mm, with many MFs being too small to be visually measured. It must also be emphasized that, once SFs reach an aquatic environment, the MFs are released straight into it, since the unique intermediary between these pollutants and the ambient is the wrapping paper. It would be interesting if future studies evaluate the role of the adsorbed chemicals to the MFs' detachment rate.

Concerning the worldwide situation, by applying the adjustment of the daily amount of MFs detached per SF for the last 10 years, it can be estimated a conservative value of $4 \cdot 10^{18}$ particles of MFs being annually released. On the other hand, by considering the global production of cellulose acetate destined for cigarette filters (640'000 tons) [8.29, 8.30], and by assuming that 60% of the SFs reach aquatic environments, about 0.3 million tons of potential MFs are being directly and constantly introduced from this source. Comparing this value with a known and relevant source of MFs, the domestic laundry of textile garments, which release was estimated at 0.28 million tons MFs per year to aquatic environments [8.31], it can be concluded that MFs detached from SFs might be almost in the same order of magnitude than those released from domestic laundry. Surprisingly, very low attention has been put onto this source, whether from the tobacco producers or consumers. Yet, further investigation is needed to make an accurate evaluation regarding the MFs generated from discarded cigarette butts.

Also, a rough estimation was executed to spot the most probable contaminating countries regarding MFs from SFs. As seen in Torkashvand et al. 2020 review, it can be assumed that every country behaves similarly concerning the incorrect littering of SFs. Besides, the waste generation is mostly a function of the population size [8.32], which in this case must be correspondingly connected to the cigarettes consumed per country. Also, it can be assumed that the littered SFs will be proportional to the amount of generated MFs. On the other hand, the prevalence of electronic cigarette smokers was considered but it was neglected at this value didn't suppose a relevant proportion of the total amount of smokers; for instance, 0.5% in China [8.33]. In this sense, for each country, the data compiled for this purpose were the cigarettes smoked per day per smoker, the percentage of smoker population, and the total population. Hence, as seen in Figure 8.3, only 17 countries might generate almost 80% of these pollutants, with China producing about 35% of it.

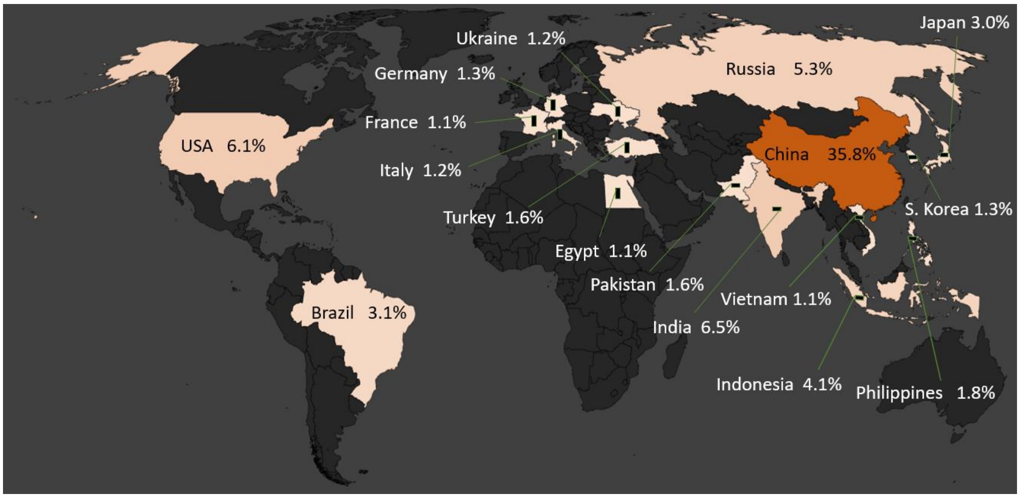

Figure 8.3. Percentage of SFs littered in countries with a contribution of >1%.

The reduction of the littered SFs could be attained by decreasing the tobacco consumption, increasing the consumers' awareness regarding the environmental issues of the littered SFs, proper disposal management, banning single-use filters, or replacing them with more durable but easily disposable options, among others. Regarding a second life for this waste, Marinello et al. 2020 made an excellent contribution by reviewing the published possible solutions. In that work, it can be found that depending on the treatment applied, this waste can be recycled into insecticides, buildings' fills, etc. For instance, Mohajerani, Abdul, and Larobina 2016 estimated that, if 2.5% of the world's annual brick production would incorporate 1% of cigarette smoked filters (by weight), it will completely offset the yearly cigarette production. On the other hand, a declining trend in the global consumption of tobacco has been achieved in the last years [8.35]. However, many low- and middle-income countries are expected to increase their tobacco prevalence [8.36], which could lead to a net increase of the MFs generated from SFs as a consequence of their deficient waste management systems. As can be seen, there is a broad set of solutions for this specific potential contamination, yet, it does not generate the same concern throughout the scientific and/or social communities as other types of MPs.

8.3.2 Ecotoxicity assessment of the microfibers released from smoked cigarette filters

To evaluate the acute aquatic toxicity due to the MFs released from SFs, the acute immobilization test with freshwater crustaceans *Daphnia magna* was carried out. At lower concentration values (<0.1 SF/L), a statistically significant difference in the effect on D. magna was obtained when the MFs were aggregated (Shapiro-Wilk for normality, one-way ANOVA, n = 30, p-value = 0.038, no changes in the control sample). The obtained data are summarized in the boxplots of Figure 8.4, where each trial and concentration are represented against the percentage of immobilized daphnids. From these results, the 100% lethal concentration was estimated at 0.620 SF/L for MFs with leachate and 0.888 SF/L for leachate only. This value is much higher than the reported by Kathleen M. 2013 which found a 100% effect at a concentration of 0.125 SF/L (only leachate), the difference might be attributed to the use of different tobacco brands and/or the application of different smoking methods.

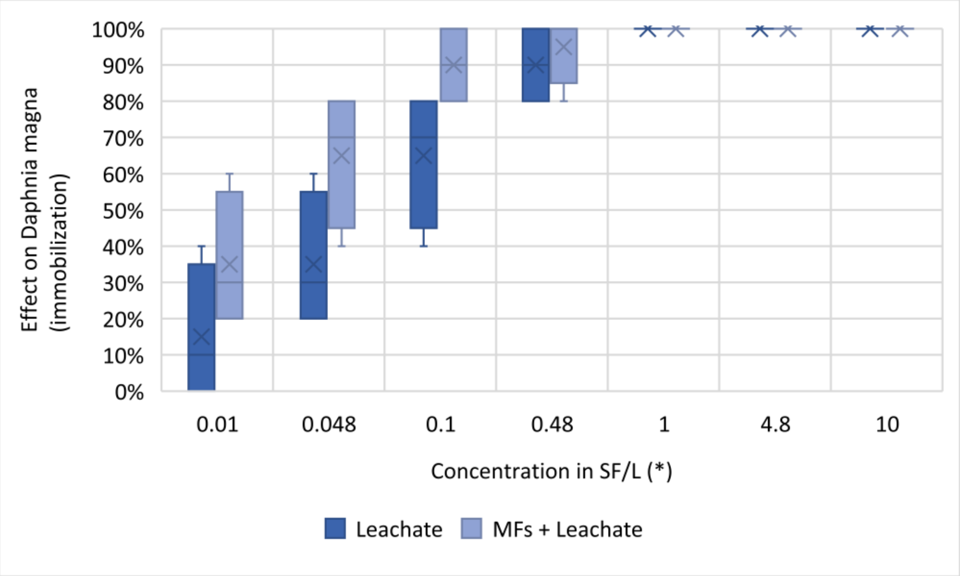

Figure 8.4. Effect (immobilization) on *Daphnia magna* after 48-h exposure to different concentrations of smoked filters (SF): only leachate in dark blue, MFs and leachate in light blue. (*) It should be noted that the "x" axis does not present continuous or lineal space between the values and that each boxplot corresponds to the concentration shown below.

As can be seen in Figure 8.4, as the concentration of SFs decreased, the effect of the presence of MFs became more notorious. In this way, the 50% effect (EC_{50}) is 0.017 SF/L for leachate with MFs, whereas leachate only shows an EC_{50} value of 0.067 SF/L, which indicates that in this case, the microfibers have increased the toxicity by 4 times. At lower concentrations, this ratio was even increased from 5 to 10 times. The reason for this difference might be that at higher concentrations the toxic effect of the lixiviate has a strong and quick influence, while only at lower concentrations the presence of MFs is discernible. Possible explanations are the ingestion of the MFs with their adsorbed toxic substances, and the continuous desorption of lixiviate, among others.

In this way, despite being many variables in the production of tobacco and cigarettes [8.38], all the studies that have conducted cigarette waste toxicity tests on living species have reported negative impacts (e.g., Novotny and Slaughter 2014; Slaughter et al. 2011; Wright et al. 2015). Besides, it has been published that a single SF can leach enough substances to contaminate 1'000 liters of water [8.5]. This can be explained because SFs and their MFs adsorb and carry many compounds that are included in the "*Toxic Release Inventory Program*" of the US-EPA, like pesticides, heavy metals, and organic chemicals, among others [8.10].

This program covers toxic chemicals that can cause *"cancer or other chronic human health effects, and significant adverse acute human health and environmental effects"* [8.40].

After the SFs are soaked both in natural or salty water, the elution of the toxic compounds can last from a few minutes to months [8.5, 8.21, 8.39]. For instance, it was reported that nicotine, one of the most studied compounds of the cigarette butts, rapidly elute in contact with water, with almost 50% eluted in the first of 15 rainfall experiments [8.5]. On the other hand, Moerman and Potts 2011 studied the leachate of metals from smoked cigarette filters. They found that differences in the pH within typical values of rainfall precipitation (4 to 6) didn't have any effects on the heavy metal leaching. They also reported that the metals have different leaching behaviors over time, suggesting that the longer a smoke cigarette filter remains in the environment, the greater will be the contamination. In this sense, the MFs released from the SFs can still transport toxic substances for an indefinite time, which can be furtherly ingested by a wide range of organisms [8.20]. This ingestion has been reported, for instance, by Jemec et al. 2016, who found that even long MFs (1.4 mm) can be consumed by small daphnids.

Along our experiments to obtain the toxicity values, a particular behavior of *Daphnia magna* was observed. One specific physical harm derived from the morphology of these pollutants was identified. During the trials, it was seen that some *Daphnia magna* suffered an external entanglement with the MFs, drastically reducing their capability of movement, leading to a certain and quicker decease. This was mainly seen in the lower range of the tested concentrations (<0.1 SF/L) because there were more living daphnids on which to verify this. It must be also noticed that lower concentrations are more likely to be found in the environment. This specific impact is expected to occur with other fiber-shaped pollutants on organisms similar in size and movement to *Daphnia magna* and would be of special interest to be furtherly studied.

Hence, MFs from SFs should be considered as a potential risk for the aquatic environment for several reasons: the possible hazardous linked to their size, morphology, and capacity to adsorb, transport, and release toxic substances; and the potential exposure to biota linked to the large quantity of SFs dumped into the environment. In any case, aquatic ecosystems are especially vulnerable to SFs, as they are a sink with favorable conditions for the detachment of MFs and the transport of the adsorbed toxic compounds. In this way, as pointed out in Novotny et al. 2009, SFs should be treated as hazardous waste even under correct disposal

8.3.3 Degradation of the UFs and SFs

As explained before, UFs and SFs were submitted to distilled and salty water with UV-light for one month to emulate marine conditions. Subsequently, they were analyzed through FTIR technique to compare and evaluate possible changes in their chemical structures. Figure 8.5 shows the FTIR spectra for the tested samples. In this way, the samples' FTIR resemblance to CA was approximately 95% in all the tested settings (*Specta Polymers and Plasticizers by ATR - corrected* library), indicating that they remained chemically unaltered.

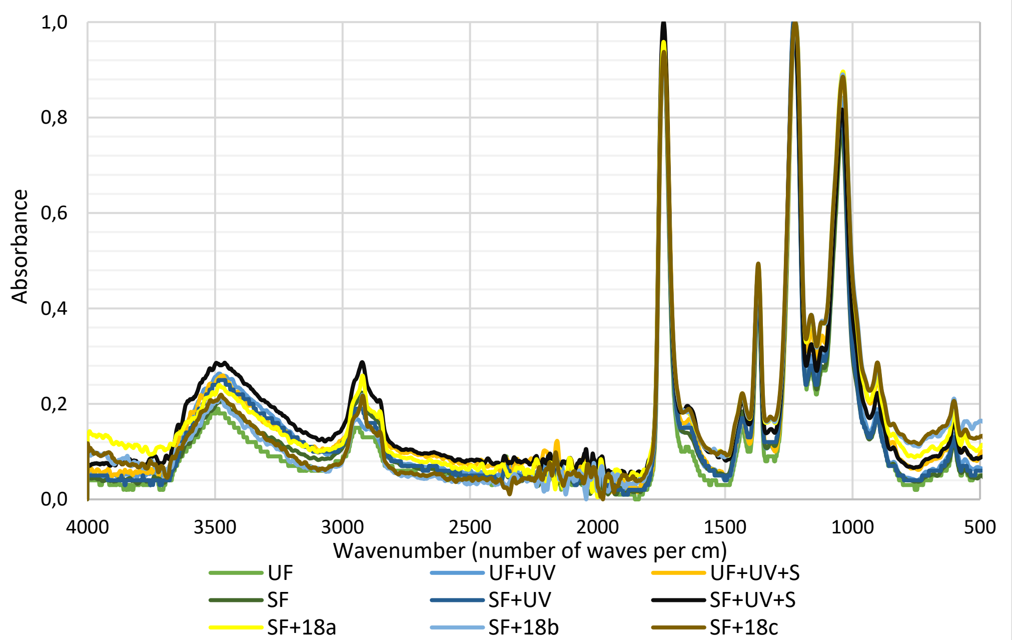

Figure 8.5. FTIR spectra of unsmoked (UF) and smoked filters (SF). "UF" and "SF" (filters) were not submitted to any trial. Filters "+UV" were submitted to a month of UV-light in distilled water. Filters "+UV+S" were submitted to a month of UV-light in salty water. "SF+18a, b, and c" are the samples analyzed after 18 months.

In Figure 8.5, the cellulose acetate (CA) composition of the cigarette filters can be identified by the acetyl groups characteristic bands at 1'240, 1'370, and 1'750 cm^{-1} wavenumbers. Hence, it can be observed that the FTIR spectra of the UFs and SFs treated samples versus the untreated ones are practically identical. Besides, no difference was found between UFs and SFs, which indicates that the smoking process does not change the chemical structure of the filter or turns it more photodegradable. Also, the SFs that were left in freshwater and natural sunlight for 18 months didn't show any chemical changes. For these reasons, it can be concluded that the CA has a very low degradability under aquatic environments.

On the other hand, the low degradability behavior was also noted with the NOAA microplastic separation methodology. This process was carried out to compare the degradability of different fibers' materials under the same test (Figure 8.6). As can be seen, the degradation of the CA cigarette filters (SF) appeared to reach a plateau when less than 10% of the mass was degraded. This can be explained as a consequence of the rapid degradation of the external cellulose layer [8.42]. In contrast, cotton (COT) samples showed a continuous degradation trend, while polyester (PES) samples did not respond to the Fenton reaction.

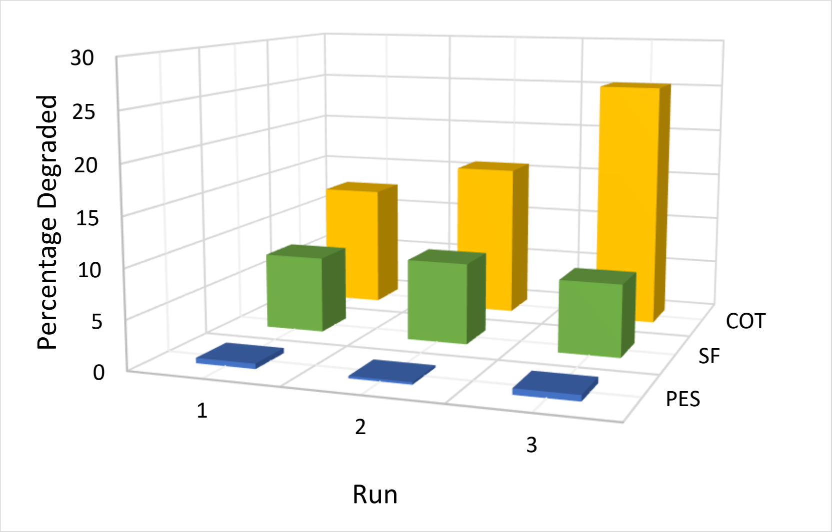

Figure 8.6. Degradation of the smoked filters (SF), and woven fabrics of cotton (COT) and polyester (PES) with a modified Fenton reaction.

Also, to evaluate the longer-term degradation of the affected fibers, one more experimental test was conducted on cotton and filters. In this last trial, 20 mL of the H_2O_2 solution was added in the 3rd run and left overnight covered with a glass dish. Under this circumstance, the cotton sample was fully degraded, while the filters did not surpass the 10% plateau. As explained before, the test proposed by the NOAA is intended to separate microplastics from complex samples [8.25]. Therefore, under the premises of this analysis, the MFs detached from cigarette filters can be considered as microplastics and consequently could remain in the environment for a long time.

The low degradability of the filters can be explained for a series of reasons. First of all, the properties of CA depend on the degree of substitution (DS), which is the average number of acetyl groups per monomer [8.43, 8.44]. In this sense, biological degradation can occur in specific environmental conditions, but the higher the DS the lesser the extent of biodegradation [8.12, 8.42]. In the tested cases, by observing the relation between the length of the characteristic bands of the acetyl groups in Figure 8.5, it can be inferred that the samples have a high DS of approximately 2.6 to 2.8. This high DS makes cigarette filters difficult to degrade by microorganisms or enzymes, i.e., to biodegrade [8.43–8.46].

On the other hand, although the ketonic carbonyl groups of CA could be photodegraded at UV-radiation < 280 nm [8.27, 8.46], little or no UV-radiation <290 nm reaches the Earth's surface as it's absorbed in the atmosphere [8.47, 8.48]. Furthermore, in aquatic environments, the thermal loading is inhibited and the UV-radiation exposure is limited, reducing, even more, the photodegradation process [8.27]. These results are consistent with other studies, for instance, Hosono et al. 2007 reported that CA did not suffer any chemical changes after 28 days of being in a xenon fadeometer. Moreover, whilst Bonanomi et al. 2020 reported that in optimal land conditions an 80% of degradation can be reached in 5 years, they also pointed out that, in the absence of soil (typical urban environment), the filters remained unaltered after 5 years. In this sense, it has been estimated that the degradation of SFs might last up to 30 years in certain conditions [8.16, 8.27]. Nonetheless, these are only estimations that applied kinetic models and there is no reliable information about its full-degradation time scale [8.17].

The SFs as a source of MFs might seem obvious, yet, in most publications, almost no attention is fixed on these or other artificial microplastics. For instance, in the SAPEA 2019 report, only synthetic materials as tires and textiles garments are related to relevant sources of microplastics. On the other hand, cigarette butts are only mentioned as an example of bad littering behavior. It is important to underline that Woodall et al. 2014 reported enormous amounts of "rayon" MFs on the seabed around the globe, surpassing the concentrations of synthetic polymer microplastics. However, in our opinion, this work might have identified a group of generic artificial cellulosic fibers (such as cellulose acetate) as rayon. Technically, the material characterized as "rayon/viscose" is related to a textile fiber that has a global low relative production [8.50]. Hence, the probability of its presence as a microfiber in the sediments of the oceans is very low. For this reason, the cellulosic fibers identified by Woodall et al. must have other origins, in which the MFs from SFs might constitute an important proportion of those pollutants. Therefore, given the low degradability of the SFs and their constant and incorrect disposal, it can be stated that these items are an increasing source of long-lasting MFs that are not receiving adequate consideration.

8.4 Conclusions

Smoked cigarette filters (SFs) are the most encountered type of litter around the world. Surprisingly, a still not resonated but significant issue is the possible impact generated from the releasing of the more than 15'000 strands that compose every SF, which can be detached as a microfiber (MF) or eventually get fragmented into lower sizes. This study evaluated the releasing rate, toxicity, and degradability of the MFs detached from SFs.

Regarding the releasing rate of MFs in water, every SF detaches around 100 MFs per day, with most MFs begin smaller than 0.2 mm. Concerning the ecotoxicity of these MFs, the EC_{50} on *Daphnia magna* organisms was observed at 0.017 SF/L. Furthermore, it was also detected that some daphnids got externally entangled with the MFs, significantly reducing their capability of movement and accelerating their decease. Concerning the degradability, the cellulose acetate that composes SFs showed no chemical alterations after a month under synthetic seawater conditions (UV-light and salty water) and after 18 months in freshwater and daily sunlight. Besides, the *National Oceanic and Atmospheric Administration* microplastic separation method showed that the cellulose acetate reached a plateau of degraded mass at 10%, from which no more changes were observed.

As can be seen, MFs from SFs are unnatural and hazardous particles that have a low degradation rate meaning a potentially high exposure and a risk for the aquatic environment. In particular, these small pollutants and their adsorbed toxic compounds can be introduced to the trophic chain, as can be easily ingested by a wide range of organisms. In a rough estimation, it was approximated that about 0.3 million tons of MFs per year might be introduced to aquatic environments from this source, with almost 80% generated by 17 countries. In this way, the MFs detached from SFs are an important source of MFs, which might partially explain the ubiquitous concentrations of modified cellulose MFs that have been reported as "rayon" polluting the deep-sea sediments. Therefore, in terms of the efforts put on developing solutions to reduce this contamination, these potential pollutants should also be treated by the scientific and social communities as an important microplastic source.

8.5 References

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Chapter 9:

Conclusions

9 Conclusions

This chapter should correspond to the general conclusions of the thesis. But Francisco could not write it. To maintain his original work, the conclusions of each chapter have been collected and copied here after:

Quantification of detached MFs

A direct and highly reliable method to quantify the detachment of textile microfibers from whole finished garments was developed and applied. In order to normalize the microfiber detachment rates results, comprehensive and comparable results are needed. In this way, we recommend a set of units that give fundamental conclusions of the microfiber detachment with respect to the textile article. In addition, a methodology to estimate the relation between the number of MFs and their mass was developed.

From consecutive washing trials, it was found that the microfiber detachment rate (MFDR) decreases until stabilization is reached in the 5th washing cycle. The MFDR in that point is between 175 to 560 MF/g or 30'000 to 465'000 MF/m^2 of garment. It was also found a high and positive relation ($R^2 = 0.71$ to 0.89) between the MFDR and the superficial density (g/cm^2) of the garment. Transforming the results into units of mass, we estimated a MF loss between 23 to 73 mg/kg of garment or 4 to 61 mg/m^2 of garment.

Moreover, the morphology of the microfibers was analyzed, and two different shapes were found: one group that comes from microfibers that were already loosely entangled with the fibers' grid of the garments, while the other corresponds to microfibers that were ripped-off from the fiber grid as a consequence of the mechanical stress suffered in the launderings. This latter case could be perpetuated by the garment use and its UV degradation. With respect to the microfiber length, it was found that it decreases from the 1st to the 3rd washing cycle. Both findings are helpful to evaluate the applicability of new microfibers' reduction solutions in different steps of the garment life cycle.

Finally, our results were used to re-estimate the mass flow of microfibers to the oceans, which was found to be overestimated by other authors. However, according to our results, the amount of MFs reaching the oceans is $1.4 \cdot 10^{17}$ MFs/year, which is higher than the value obtained when our calculation methodology is applied to the previously published data. This implies that a higher quantity of smaller and more easily ingestible microfibers is heading towards the oceans.

Estimation of MFs reaching aquatic environments

An estimation of the mass flow of microfibers (MFs) to aquatic environments was accomplished by developing a new calculation methodology. The method applies a set of known-parameters that are linked to the MFs' pollution, which are: (1) MFs detachment rate from different textile garments; (2) volumes of laundry effluents; (3) percentage of municipal used-water treated per world region; (4) type of water treatment applied, and (5) proportion of front- versus top-loading washing machines. In this way, different scenarios were studied and a central value of 0,28 million tons per year of MFs was obtained, which is approximately 50% lower than previously published.

On a regional basis, 65% of all the MFs that reach aquatic environments come from Asia. The explanation for this major influence is a combination of the high proportion of top-loading washing machines, an inefficient water-usage in the washing cycles, a low rate of municipal water treated, and a high population density. In contrast, other regions such as Europe have a relatively low contribution to the MFs' pollution, basically, as a consequence of the opposite conditions. On the other hand, when estimating the overall mass of generated MFs in the laundering process, North America gets situated in the first place with 18% of the global MF generation, from where a high proportion of these MFs is retained in municipal water treatment plants.

In addition, three hypothetical situations were analyzed with the attempt to quantify the impacts on the MFs release and to make positive proposals able to be applied at government, industries, and consumer levels. Concerning the washing machine types, the current proportion of front- versus top-loading washers in the Asian region was inverted. In this way, a global MFs release reduction of 29% was accomplished. Regarding the consumers, regions with high consumption of water per laundry were matched to the worldwide average. Thus, the attained MF reduction was of 29%, meaning that it is an efficient and sizeable MF reduction strategy. Additionally, at a governmental level, the evaluation was done by increasing the percentage of treated water in regions with a low used-water treatment rate. By doing so, a global MFs' reduction of 31% MFs was achieved. Finally, if all strategies were combined, a MF reduction of 65% could be achieved. However, it must be noticed that while all measurements decrease MFs from reaching aquatic environments, only modifications in the washer type and washing behaviors (e.g., lower but sufficient washing time) could efficiently reduce the detachment of MFs. Henceforth, major importance should be applied in those strategies that tackle the generation of MFs.

Solutions to mitigate the presence of textile MFs

Many alternatives are available to reduce textile microfibers from reaching the environment. Some options are currently more viable in the short- and medium-term periods. The textile industry has the potential to drastically reduce the generation of microfibers by improving their processes or products. This could imply that downstream solutions might be dispensable or less severe. However, there are many small- and medium-sized textile industries around the globe, making this alternative feasible only in the long-term time. Also, there are currently some solutions for washers. These can reduce at least 30% of the microfibers' emissions from household laundry. Besides, new products as detergents or additives are being developed to reduce the generation of these particles.

On the other hand, water treatment plants can partially remove the microfibers from the liquid stream and retain them in the sludge. Depending on the technology applied, these facilities can remove up to 99% of the microfibers. Yet, the problem is still transferred to the sludge. In addition, installing these facilities is a long-term alternative. An important gap in every alternative is the final disposition or treatment of the microfibers. It is important to clarify that any solution must consider the whole process to certify that it is environmentally friendly and will not pollute more than the microfibers. Yet, it is very likely that the alternatives will be complementary between them, i.e., there will be no single solution for the microfiber pollution.

Development of a filter to retain the MFs

Different models of a new textile microfiber retaining system were evaluated. It was found that all 4 assessed arrangements have a good performance for retaining microfibers from the washers' effluents. Depending on the model, the microfiber retention efficiency was estimated between 52% to 86% in the 1st washing cycle and up to 83% to 99% in the 20th. The best performance was encountered when the flow of the washers' effluent went from bottom to top, being the filtering media at the top. Besides, all the arrangements showed a sufficient replacement time interval for the cartridges, as these were capable of handling more than 30 washing cycles. It is important to mention that one of the arrangements didn't need an external artifact as it was applied by surrounding the existing washing machine filter.

In addition to the good performance of these filters, it should be highlighted that they hold two relevant features. First, the usage of thermoplastic recycled waste for the filtering media and the shell, which strengthens the circular economy philosophy and produces a "greener" product. And, that the retained microfibers can be further and easily immobilized in a polymeric matrix by merging the filtering media with the microfibers inside. This latter feature can be harnessed to develop different types of products, tackling one of the main issues of the existing alternatives to reduce the microfibers, which is the subsequent treatment of these pollutants.

Immobilization of the collected MFs

This study presents a novel and sustainable treatment for microfibers detached from textile garments. The treatment is very simple and doesn't need sophisticated equipment or high energy or resources demand. In this sense, to treat the microfibers we are proposing to immobilize them into a polymeric recycled matrix from where they cannot escape. Once inside, MFs based polymeric composites can be obtained and different applications can be given to these composites. In this work, three different proportions of polyester microfibers (5%, 10%, and 15%, vol/vol) were introduced into recycled low-density polyethylene matrix. The purpose was to have an approximated knowledge of the higher concentration of microfibers that can be included and still be able to produce a high-quality product.

In this sense, it was seen that the composites having up to 10% of microfibers behaved homogeneously. A lower concentration of microfibers also worked fine, but our objective is to treat the currently "fibers' microplastic pollution from laundering". Hence, the more microfibers that can be included, the better. Nonetheless, it was seen that when including 10% of microfibers in the thermoplastic polymer matrix, some mechanical properties, as the tensile strength or Young's modulus improved at the expense of reducing the maximum deformation achievable. Besides, no microfibers were detached from the final composite, meaning that these were totally inserted in the matrix. On the other hand, SEM images showed the most plausible interaction between the polyester and the low-density polyethylene is a wettability adhesion. However, in real conditions, other microfibers with more roughness than that of polyester will be included, increasing even more the gripping between the pollutants and the recycled polymer by promoting mechanical adhesion.

Other type of MFs: cigarette butts

Smoked cigarette filters (SFs) are the most encountered type of litter around the world. Surprisingly, a still not resonated but significant issue is the possible impact generated from the releasing of the more than 15'000 strands that compose every SF, which can be detached as a microfiber (MF) or eventually get fragmented into lower sizes. This study evaluated the releasing rate, toxicity, and degradability of the MFs detached from SFs.

Regarding the releasing rate of MFs in water, every SF detaches around 100 MFs per day, with most MFs begin smaller than 0.2 mm. Concerning the ecotoxicity of these MFs, the EC50 on Daphnia magna organisms was observed at 0.017 SF/L. Furthermore, it was also detected that some daphnids got externally entangled with the MFs, significantly reducing their capability of movement and accelerating their decease. Concerning the degradability, the cellulose acetate that composes SFs showed no chemical alterations after a month under synthetic seawater conditions (UV-light and salty water) and after 18 months in freshwater and daily sunlight. Besides, the *National Oceanic and Atmospheric Administration* microplastic separation method showed that the cellulose acetate reached a plateau of degraded mass at 10%, from which no more changes were observed.

As can be seen, MFs from SFs are unnatural and hazardous particles that have a low degradation rate meaning a potentially high exposure and a risk for the aquatic environment. In particular, these small pollutants and their adsorbed toxic compounds can be introduced to the trophic chain, as can be easily ingested by a wide range of organisms. In a rough estimation, it was approximated that about 0.3 million tons of MFs per year might be introduced to aquatic environments from this source, with almost 80% generated by 17 countries. In this way, the MFs detached from SFs are an important source of MFs, which might partially explain the ubiquitous concentrations of modified cellulose MFs that have been reported as "rayon" polluting the deep-sea sediments. Therefore, in terms of the efforts put on developing solutions to reduce this contamination, these potential pollutants should also be treated by the scientific and social communities as an important microplastic source.

Chapter 10:

Annexes

10 Annexes

Annex A

Supplementary information of chapter 4: Textile Microfibers Reaching Aquatic Environments: A New Estimation Approach

- As there is no information regarding the washer type for some regions, **three different scenarios were established**. In all these scenarios, regions with information maintain their known proportions. However, to include the possible settings, regions without information were considered to have ratios of front versus top-loading (FL:TL) washers of 7:3, 5:5, and 3:7 in scenarios S1, S2, and S3, respectively. See complete data values for P_F and P_T in the following Tables:

Table 10.1. MFs release from household laundering to aquatic environments, scenario S0: 100% Frontal-loading washing machines in regions without information.

	Scenario S0 (100% Frontal-loading washing machines)			
	MF/year		Ton/year	
	Optimistic	Pessimistic	Optimistic	Pessimistic
North America	2.16E+16	1.68E+17	2.85E+03	2.22E+04
Latin America and the Caribbean	4.06E+16	3.16E+17	5.35E+03	4.17E+04
Europe	1.20E+16	9.32E+16	1.58E+03	1.23E+04
NIS	8.32E+15	6.48E+16	1.10E+03	8.54E+03
Central Asia + China	5.26E+16	4.09E+17	6.93E+03	5.40E+04
South Asia	5.23E+16	4.07E+17	6.89E+03	5.37E+04
Other Pacific Asia	2.24E+16	1.75E+17	2.95E+03	2.30E+04
Pacific OECD + South Korea	1.65E+16	1.29E+17	2.18E+03	1.70E+04
Middle East and North Africa	1.11E+16	8.65E+16	1.46E+03	1.14E+04
Sub-Saharan Africa	6.28E+14	4.89E+15	8.28E+01	6.45E+02
Worldwide	2.38E+17	1.85E+18	3.13E+04	2.44E+05
Central Value	**1.05E+18**		**1.38E+05**	

Table 10.2. MFs release from household laundering to aquatic environments, scenario S1: 70% frontal- and 30% top-loading washing machines (FL:TL) in regions without information.

	Scenario S1 (70% Frontal; 30% Top-loading washing machines)				
	FL:TL	MF/year		Ton/year	
		Optimistic	Pessimistic	Optimistic	Pessimistic
North America	0.1:0.9 *	4.64E+16	3.61E+17	6.11E+03	4.76E+04
Latin America and the Caribbean	0.7:0.3	6.78E+16	5.28E+17	8.93E+03	6.96E+04
Europe	0.98:0.02 *	1.28E+16	9.96E+16	1.69E+03	1.31E+04
NIS	0.7:0.3	1.39E+16	1.08E+17	1.83E+03	1.42E+04
Central Asia + China	0.1:0.9 *	1.13E+17	8.79E+17	1.49E+04	1.16E+05
South Asia	0.1:0.9 *	1.12E+17	8.74E+17	1.48E+04	1.15E+05
Other Pacific Asia	0.1:0.9 *	4.81E+16	3.75E+17	6.34E+03	4.94E+04
Pacific OECD + South Korea	0.1:0.9 *	3.55E+16	2.76E+17	4.67E+03	3.64E+04
Middle East and North Africa	0.7:0.3	1.85E+16	1.44E+17	2.44E+03	1.90E+04
Sub-Saharan Africa	0.7:0.3	1.05E+15	8.16E+15	1.38E+02	1.08E+03
Worldwide	-	4.69E+17	3.65E+18	6.18E+04	4.81E+05
Central Value	-	2.06E+18		2.72E+05	
*Fixed values.					

Table 10.3. MFs release from household laundering to aquatic environments, scenario S2: 50% frontal- and 50% top-loading washing machines (FL:TL) in regions without information.

	Scenario S2 (50% Frontal; 50% Top-loading washing machines)				
	FL:TL	MF/year		Ton/year	
		Optimistic	Pessimistic	Optimistic	Pessimistic
North America	0.1:0.9 *	4.64E+16	3.61E+17	6.11E+03	4.76E+04
Latin America and the Caribbean	0.5:0.5	7.68E+16	5.98E+17	1.01E+04	7.88E+04
Europe	0.98:0.02 *	1.28E+16	9.96E+16	1.69E+03	1.31E+04
NIS	0.5:0.5	1.57E+16	1.23E+17	2.07E+03	1.61E+04
Central Asia + China	0.1:0.9 *	1.13E+17	8.79E+17	1.49E+04	1.16E+05
South Asia	0.1:0.9 *	1.12E+17	8.74E+17	1.48E+04	1.15E+05
Other Pacific Asia	0.1:0.9 *	4.81E+16	3.75E+17	6.34E+03	4.94E+04
Pacific OECD + South Korea	0.1:0.9 *	3.55E+16	2.76E+17	4.67E+03	3.64E+04
Middle East and North Africa	0.5:0.5	2.10E+16	1.64E+17	2.77E+03	2.16E+04
Sub-Saharan Africa	0.5:0.5	1.19E+15	9.26E+15	1.57E+02	1.22E+03
Worldwide	-	4.82E+17	3.76E+18	6.36E+04	4.95E+05
Central Value	-	2.12E+18		2.79E+05	
*Fixed values.					

Table 10.4. MFs release from household laundering to aquatic environments, scenario S3: 30% frontal- and 70% top-loading washing machines (FL:TL) in regions without information

	Scenario S3 (30% Frontal; 70% Top-loading washing machines)				
	FL:TL	MF/year		Ton/year	
		Optimistic	Pessimistic	Optimistic	Pessimistic
North America	0.1:0.9 *	4.64E+16	3.61E+17	6.11E+03	4.76E+04
Latin America and the Caribbean	0.3:0.7	8.28E+16	6.45E+17	1.09E+04	8.50E+04
Europe	0.98:0.02 *	1.28E+16	9.96E+16	1.69E+03	1.31E+04
NIS	0.3:0.7	1.70E+16	1.32E+17	2.24E+03	1.74E+04
Central Asia + China	0.1:0.9 *	1.13E+17	8.79E+17	1.49E+04	1.16E+05
South Asia	0.1:0.9 *	1.12E+17	8.74E+17	1.48E+04	1.15E+05
Other Pacific Asia	0.1:0.9 *	4.81E+16	3.75E+17	6.34E+03	4.94E+04
Pacific OECD + South Korea	0.1:0.9 *	3.55E+16	2.76E+17	4.67E+03	3.64E+04
Middle East and North Africa	0.3:0.7	2.27E+16	1.76E+17	2.99E+03	2.33E+04
Sub-Saharan Africa	0.3:0.7	1.28E+15	9.98E+15	1.69E+02	1.32E+03
Worldwide	-	4.92E+17	3.83E+18	6.48E+04	5.04E+05
Central Value	-	2.16E+18		2.85E+05	
*Fixed values.					

Table 10.5. HA: MFs release from household laundering to aquatic environments: 90% frontal- and 90% top-loading washing machines in Asian regions. In light blue regions that were modified

	Central values for Scenario HA (90% frontal- and 90% top-loading washing machines in Asian regions)					
	HA1		HA2		HA3	
	MF/year	Ton/year	MF/year	Ton/year	MF/year	Ton/year
North America	1.23E+17	1.62E+04	1.23E+17	1.62E+04	1.23E+17	1.62E+04
Latin America and the Caribbean	2.98E+17	3.92E+04	3.37E+17	4.45E+04	3.64E+17	4.80E+04
Europe	5.62E+16	7.41E+03	5.62E+16	7.41E+03	5.62E+16	7.41E+03
NIS	6.10E+16	8.04E+03	6.91E+16	9.11E+03	7.46E+16	9.83E+03
Central Asia + China	3.00E+17	3.95E+04	3.00E+17	3.95E+04	3.00E+17	3.95E+04
South Asia	2.98E+17	3.93E+04	2.98E+17	3.93E+04	2.98E+17	3.93E+04
Other Pacific Asia	1.28E+17	1.68E+04	1.28E+17	1.68E+04	1.28E+17	1.68E+04
Pacific OECD + South Korea	9.42E+16	1.24E+04	9.42E+16	1.24E+04	9.42E+16	1.24E+04
Middle East and North Africa	8.14E+16	1.07E+04	9.23E+16	1.22E+04	9.95E+16	1.31E+04
Sub-Saharan Africa	4.61E+15	6.07E+02	5.22E+15	6.88E+02	5.63E+15	7.42E+02
Worldwide	1.44E+18	1.90E+05	1.50E+18	1.98E+05	1.54E+18	2.03E+05

Table 10.6. HB: MFs release from household laundering to aquatic environments: Regions with a high consumption of water are matched to the average (19 m^3 per washing machine). In light blue regions that were modified.

	Central values for Scenario HB (Regions with a high consumption of water are matched to the average, 19 m^3 per washing machine)					
	HB1		HB2		HB3	
	MF/year	Ton/year	MF/year	Ton/year	MF/year	Ton/year
North America	1.38E+17	1.82E+04	1.38E+17	1.82E+04	1.38E+17	1.82E+04
Latin America and the Caribbean	1.98E+17	2.61E+04	2.24E+17	2.95E+04	2.42E+17	3.19E+04
Europe	5.62E+16	7.41E+03	5.62E+16	7.41E+03	5.62E+16	7.41E+03
NIS	6.10E+16	8.04E+03	6.91E+16	9.11E+03	7.46E+16	9.83E+03
Central Asia + China	4.96E+17	6.53E+04	4.96E+17	6.53E+04	4.96E+17	6.53E+04
South Asia	2.36E+17	3.11E+04	2.36E+17	3.11E+04	2.36E+17	3.11E+04
Other Pacific Asia	1.33E+17	1.75E+04	1.33E+17	1.75E+04	1.33E+17	1.75E+04
Pacific OECD + South Korea	6.70E+16	8.82E+03	6.70E+16	8.82E+03	6.70E+16	8.82E+03
Middle East and North Africa	7.89E+16	1.04E+04	8.95E+16	1.18E+04	9.65E+16	1.27E+04
Sub-Saharan Africa	4.61E+15	6.07E+02	5.22E+15	6.88E+02	5.63E+15	7.42E+02
Worldwide	1.47E+18	1.93E+05	1.51E+18	2.00E+05	1.54E+18	2.04E+05

Table 10.7. HC: MFs release from household laundering to aquatic environments: Percentage of used-water treated of 60% for regions with a current treatment extent < 50%. Government Entities and Used-Water Treatment Plants. In light blue regions that were modified.

	Central values for Scenario HC (Percentage of used-water treated of 60% for regions with a current treatment extent < 50%)					
	HC1		HC2		HC3	
	MF/year	Ton/year	MF/year	Ton/year	MF/year	Ton/year
North America	2.04E+17	2.69E+04	2.04E+17	2.69E+04	2.04E+17	2.69E+04
Latin America and the Caribbean	1.75E+17	2.30E+04	1.98E+17	2.61E+04	2.14E+17	2.81E+04
Europe	5.62E+16	7.41E+03	5.62E+16	7.41E+03	5.62E+16	7.41E+03
NIS	3.62E+16	4.78E+03	4.11E+16	5.41E+03	4.43E+16	5.84E+03
Central Asia + China	3.03E+17	3.99E+04	3.03E+17	3.99E+04	3.03E+17	3.99E+04
South Asia	3.03E+17	3.99E+04	3.03E+17	3.99E+04	3.03E+17	3.99E+04
Other Pacific Asia	1.30E+17	1.71E+04	1.30E+17	1.71E+04	1.30E+17	1.71E+04
Pacific OECD + South Korea	1.56E+17	2.05E+04	1.56E+17	2.05E+04	1.56E+17	2.05E+04
Middle East and North Africa	6.90E+16	9.10E+03	7.83E+16	1.03E+04	8.44E+16	1.11E+04
Sub-Saharan Africa	2.25E+15	2.97E+02	2.55E+15	3.37E+02	2.75E+15	3.63E+02
Worldwide	1.43E+18	1.89E+05	1.47E+18	1.94E+05	1.50E+18	1.97E+05

Table 10.8. HD: MFs release from household laundering to aquatic environments: combination of all previous scenarios. In light blue regions that were modified.

	Central values for Scenario HD					
	HD1		HD2		HD3	
	MF/year	Ton/year	MF/year	Ton/year	MF/year	Ton/year
North America	8.35E+16	1.10E+04	8.35E+16	1.10E+04	8.35E+16	1.10E+04
Latin America and the Caribbean	1.16E+17	1.53E+04	1.32E+17	1.73E+04	1.42E+17	1.87E+04
Europe	5.62E+16	7.41E+03	5.62E+16	7.41E+03	5.62E+16	7.41E+03
NIS	3.62E+16	4.78E+03	4.11E+16	5.41E+03	4.43E+16	5.84E+03
Central Asia + China	1.83E+17	2.41E+04	1.83E+17	2.41E+04	1.83E+17	2.41E+04
South Asia	8.75E+16	1.15E+04	8.75E+16	1.15E+04	8.75E+16	1.15E+04
Other Pacific Asia	4.93E+16	6.50E+03	4.93E+16	6.50E+03	4.93E+16	6.50E+03
Pacific OECD + South Korea	4.05E+16	5.33E+03	4.05E+16	5.33E+03	4.05E+16	5.33E+03
Middle East and North Africa	6.69E+16	8.82E+03	7.59E+16	1.00E+04	8.18E+16	1.08E+04
Sub-Saharan Africa	2.25E+15	2.97E+02	2.55E+15	3.37E+02	2.75E+15	3.63E+02
Worldwide	7.21E+17	9.51E+04	7.51E+17	9.90E+04	7.71E+17	1.02E+05

Table 10.9. HAB: MFs release from household laundering to aquatic environments: Combination of alternatives A and B. In light blue regions that were modified.

	Central values for Scenario HAB					
	HAB1		HAB2		HAB3	
	MF/year	Ton/year	MF/year	Ton/year	MF/year	Ton/year
North America	8.35E+16	1.10E+04	8.35E+16	1.10E+04	8.35E+16	1.10E+04
Latin America and the Caribbean	1.98E+17	2.61E+04	2.24E+17	2.95E+04	2.24E+17	2.95E+04
Europe	5.62E+16	7.41E+03	5.62E+16	7.41E+03	5.62E+16	7.41E+03
NIS	6.10E+16	8.04E+03	6.91E+16	9.11E+03	6.91E+16	9.11E+03
Central Asia + China	2.98E+17	3.93E+04	2.98E+17	3.93E+04	2.98E+17	3.93E+04
South Asia	1.43E+17	1.88E+04	1.43E+17	1.88E+04	1.43E+17	1.88E+04
Other Pacific Asia	8.03E+16	1.06E+04	8.03E+16	1.06E+04	8.03E+16	1.06E+04
Pacific OECD + South Korea	4.05E+16	5.33E+03	4.05E+16	5.33E+03	4.05E+16	5.33E+03
Middle East and North Africa	7.89E+16	1.04E+04	8.95E+16	1.18E+04	8.95E+16	1.18E+04
Sub-Saharan Africa	4.61E+15	6.07E+02	5.22E+15	6.88E+02	5.22E+15	6.88E+02
Worldwide	1.04E+18	1.38E+05	1.09E+18	1.44E+05	1.09E+18	1.44E+05

Annex B:

Moving Bed Bio-Reactors Carriers Manufacturing Trials and Process

Abstract

The manufacture of MBBR carriers was carried out by merging plastic (HDPE and LDPE pellets) with textile MFs. Both are the waste obtained when the filter designed for domestic washer is exhausted. The resulting mix of polymers are called "composites". The plates of composites were subsequently cut to increase the specific area of carriers. In this way, carriers with high specific surface and relatively homogeneous shapes were obtained.

Previous to the carrier manufacturing, some mechanical properties of composite polymers with different proportions of MFs and low-density propylene were tested (tensile test, ASTM-D-638-14). In spite that the microfibers (MFs) are not included in a homogeneous way, these properties were improved over the polymeric matrix used, which was Low-Density Polyethylene (LDPE). The optimal mixture was 10% MFs in the polymeric matrix. A detailed description of the carriers manufacturing is indicated below.

1. Manufacturing of the plates

The plates were manufactured as follow:

a) The LPDE pellets (which were recycled) were put in a two-cylinder rotating heating machine (Figure 10.13). The temperature used was of approximately 130ºC. Once the LPDE was merged, the MFs were gradually inserted in the machine. Afterward, approximately 10 minutes were waited to let a sufficient mixing between the polymers.

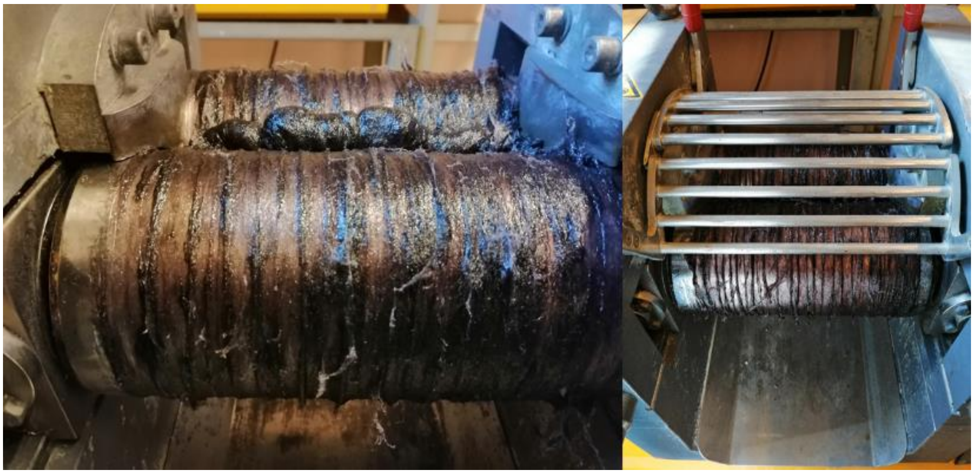

Figure 10.13. Rotating cylinders used to mix the MFs and the LPDE. The outcome was a composite.

b) The composites where then introduced in a pressuring machine to form the plates. This machine was heated up to 140 °C. The morphology of the plates depended on the plaques used. Figure 10.2 shows the machine used to compress and form the composites' plates.

Figure 10.2. The composite is being compressed by the pressuring machine.

c) As said, the plates where made with different morphologies depending on the plaques used. The plates used to make the MBBR carriers were the squared ones.

Figure 10.3 shows the formed plates. The last circular composite was used to make the carriers.

Figure 10.3. Plates formed with the pressuring machine. The final circular composites where obtained from the squared composites.

2. Manufacturing of the carriers

Once the circular composites were made, these were drilled with a drilling machine to increase the specific area and to give holes for the microorganisms to attach. In total, approximately 100 MBBR carriers were formed, enough to fill 1 liter, which was the volume needed to make the biological water treatment experiment. Figure 10.4 shows the final form of the MBBR carriers.

Figure 10.4. MBBR carriers with holes to increase the specific area and give supporting places for the microorganisms to attach.

3. Mechanical trials made to the plates

The percentage in volume for the manufacturing of the plates for the trials where of 5%, 10% and 15% of MFs in volume of MFs of polyester. More MFs didn't have enough polymeric matrix to mix them. Test pieces and the experiment were made accordingly to the ASTM-D-638-14 standard. The machine (Instron 3366) used is shown in Figure 10..

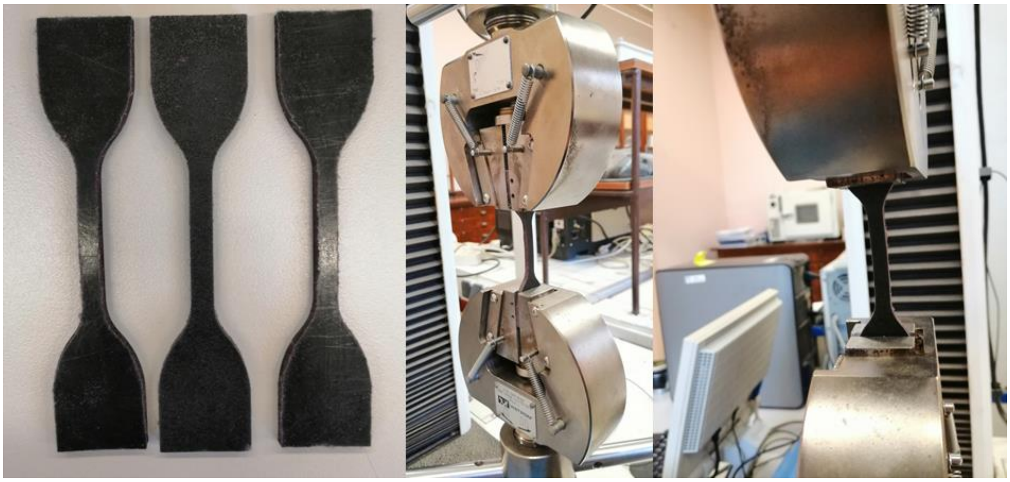

Figure 10.5. Tensile test machine used to test the composites. The image from the left corresponds to the test pieces formed to make the testing experiment according to the ASTM-D-638-14.

The results were shown in Figure 10.6, where it can be seen that the homogeneity of the composites is relatively good even at 10% of polyester in the composites. Even better, the tensile stress of the LPDE was improved from MPa to 12 or 14 MPa. This means that despite the MFs are not included in an organized structure, they are capable of forming a consistent composite.

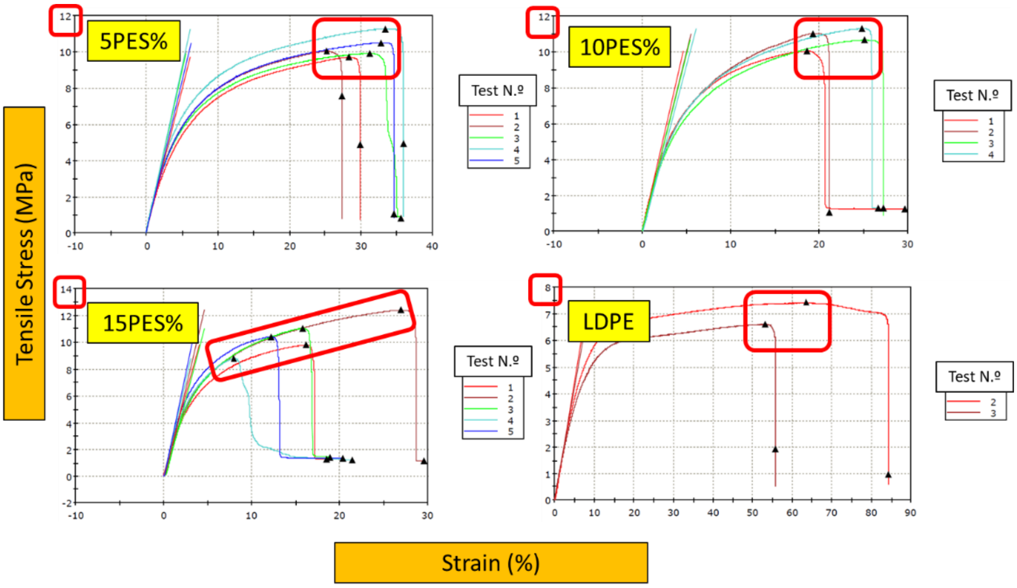

Figure 10.6. Strain (%) versus tensile stress (MPa) of composites with different compositions of polyester microfibers (PES-MFs) in the low-density polyethylene matrix (LDPE). Please, be aware that the "y-axes" do not present the same scale.

4. SEM images to the samples

SEM images were taken to see the adherence type between the polyester MFs and the LDPE matrix. In this sense, the broken edges of the test pieces in the tensile test were prepared to make the SEM process. The main conclusion was there is no complete adherence as the MFs form some kind of "tunnel" in the LDPE matrix. Figure 10.7 shows the "tunnels".

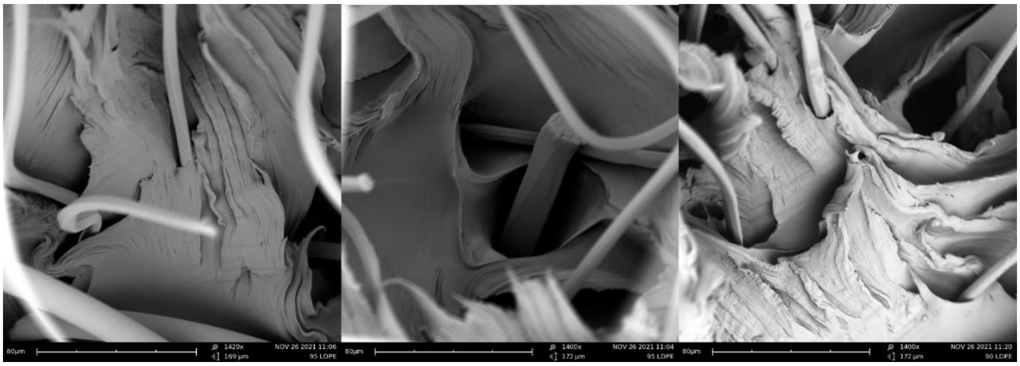

Figure 10.7. SEM images of the composites at x1400. The first two images are for 5% PES and the last one for 10% PES.

On the other hand, and as expected, the MFs didn't form any type of interlocked or reticulated formation, as these were put and mixed without any control of the location that the MFs can have. However, it is not possible to put them in order, that's why the mixing procedure lasted 10 minutes. Nonetheless, the MFs were homogeneously distributed across the entire composite. This can be seen in Figure 10.8.

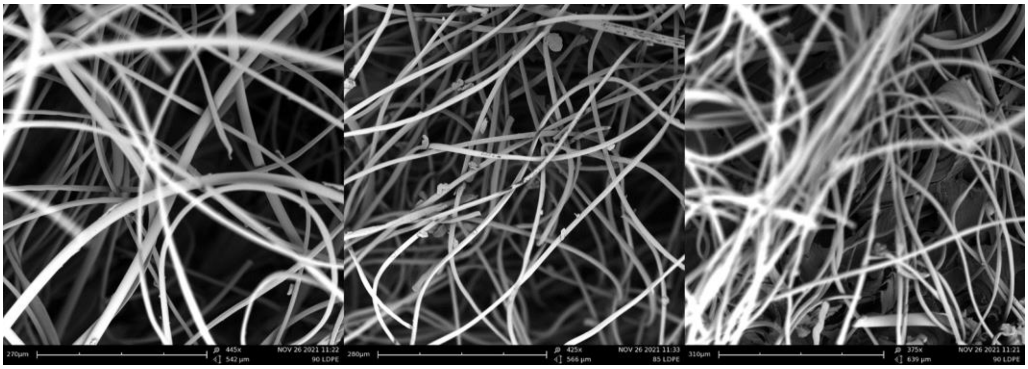

Figure 10.8. MFs shown above the composite matrix.

Annex C

Development of a Self-Sustaining Floating Water Treatment System with Renewable Energy Supply ETAF

This study corresponds to the use of carriers into a MBBR plant to treat superficial water. This work has been presented in the II International Congress on Water and Sustainability (Terrassa, Spain, 24-26 March 2021) and it is published in "Book of Abstract: II International Conference on Water and Sustainability (OmniaScience, Editors: B. Amante, F. Belzagui, V. Buscio a L.Canals). DOI: 10.3926/icws2021. ISBN: 978-84-123480-0-2

Abstract:

The scarcity of water is one of the most threatening global concerns, which will be even more exacerbated due to climate change and population growth. Water has a complex nexus with everything that surrounds us. It is the main bloodstream of the ecosystems and the key for food and energy security. Water is a limited resource but is not treated as such. In this sense, water pollution is a worldwide issue that needs urgent action. This pollution can be generated from point and diffuse sources, being the latter harder to control. In this sense, we are proposing an efficient floating water treatment system to treat both contamination sources. It will be deployed on surface reservoirs like lakes and ponds. The floating characteristic will permit the treatment of different sections of the reservoir by moving the ETAF. Besides, there will be no requirements for external chemical agents and only biological sludge will be generated. For this, the system will consider a synergetic combination between active and passive treatments.

Besides, the ETAF will be designed to withstand the inherent intermittence of typical renewable energy sources. On the other hand, common plastic wastes will be used to build it, supporting the circular economy philosophy. In this sense, a peculiarity will be the use of microplastics and microfibers to elaborate some of the components. This system is also intended to be efficient in terms of costs and energy for operation and maintenance; hence, it can be suitable for developed and developing countries.

Development of a Self-Sustaining Floating Water Treatment System with Renewable Energy Supply
ETAF

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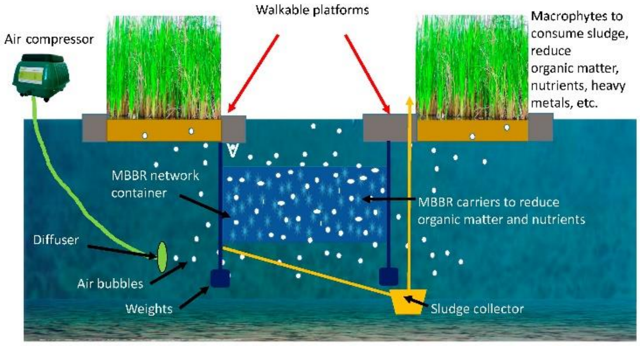
1. Introduction

Water has a complex nexus with everything that surrounds us and its scarcity is one of the most threatening global concerns. In this sense, water pollution is a worldwide issue that needs urgent action. This pollution can be generated from point and diffuse sources, being the latter harder to control. We are proposing an efficient floating water treatment system to treat both type of contamination sources.

2. The Proposed Treatment – **ETAF** – *"Estación de Tratamiento de Aguas Flotante"*

- Synergetic combination between active (**MBBR**, ***Moving Bed Bio-Reactor***) and passive (**Macrophytes**) treatments
- For surface water reservoirs like lakes and ponds
- Treat excess of organic matter and nutrients, among others
- The floating characteristic will permit to treat different sections of one reservoir
- No requirement for chemical agents; only biological sludge will be generated
- It's deployment and operation will have no negative impacts on the ecosystem to be treated
- It will withstand the inherent intermittence of renewable energy sources
- Common plastic wastes will be used to build and elaborate some of the components like the MBBR carriers
- Intended to be efficient in terms of costs and energy for operation and maintenance
- Suitable for all type of countries



3. Recycling Materials

Waste and microplastics can be used as a resource to:

- Make the MBBR carriers
- Build the "floating" components of the ETAF

https://inhabitat.com/bottles-up-fijis-first-plastic-bottle-boat/

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