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A REVIEW OF THE ORIGINS OF MICROPLASTICS ARRIVING AT WASTEWATER TREATMENT PLANTS

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ABSTRACT

Concerns regarding the impacts of microplastics in the global environment have brought into focus the need to understand better their origins, transport, and fate. Wastewaters (WW) are important in this regard: discharges from households, commercial and industrial premises, and surface run-off deliver microplastics to wastewater treatment plants (WWTPs) via sewerage systems, through which they are removed along with sewage sludge or destined for release into the environment in treated effluent. This review provides a contemporary and critical analysis of factors influencing the quantities and composition of microplastics (MPs) reaching wastewater treatment plants, including both primary and secondary sources. Three specific areas of concern were highlighted. First, current legislation, where present, needs to address regulation of microplastics in personal care and cosmetic products that cross international borders. Secondly, accurate estimation of microplastics arising from some sources and activities (e.g., mis-managed waste and hand washing of textiles) is challenging and estimated contributions of associated microplastics remain unsatisfactory as a basis for management decisions. Thirdly, information relating to microplastics in personal care and cosmetic products used by male consumers is lacking and contributions of such products to wastewater remain uncertain. We recommend that (1) voluntary practices and programmes should be replaced with formal regulation to achieve compliance, and (2) the role of consumers' behaviour in generating microplastics that are destined for wastewater treatment plants remains largely unknown and that more research in this domain is needed.

1. INTRODUCTION

Plastics have traced an era of development and have had a great impact on society, as they exhibit a great variety of properties and functions (Zalasiewicz et al., 2016). Since 1950, global plastic production has increased from 1.5 to 367 million metric tons in 2020 (PlasticsEurope and EPRO, 2021). Polypropylene (PP) and low- and high-density polyethylene (LD-PE and HD-PE) are the most widely-distributed plastics, whilst packaging, buildings and construction, transportation and textiles are the sectors with the highest demand for these materials (Ellen MacArthur Foundation, 2017; Ryberg et al., 2018; PlasticsEurope and EPRO, 2021). Polyethylene terephthalate (PET), polyvinyl chloride (PVC) and polystyrene (PS), are also amongst the most common type of polymers. However, concerns regarding the increased use of plastics and their presence in the environment are widespread. One of the issues is the generation and presence of small plastic pieces, commonly referred as microplastics.

These particles present a potentially significant threat to marine and terrestrial ecosystems. For example, ingestion of microplastics (MPs) by fish can result in reduced feeding and starvation, choking and internal damages, leading to mortality (Horton and Clark, 2018; Gola et al., 2021). Possible effects on humans have been reported, such as respiratory lesions and inflammatory responses due to the inhalation of fibrous material (Prata et al., 2020). Furthermore, Zaheer et al., (2022) by studying mice as model organisms exposed to polyethylene (PE) microplastics in an early stage of life, concluded that MPs might become a potential risk factor to develop autism spectrum disorder.

The origins and fate of microplastics have been investigated and numerous sources and destinations have

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been identified. Marine ecosystems have been detected as the ultimate fate for much microplastic waste, and even the treated effluent from wastewater treatment plants (WWTPs) may be a potentially important contributor of these particles to this environment (Ziajahromi et al., 2016). However, there is a lack of knowledge and deep understanding regarding the emissions from sources until they arrive at WWTPs. Some routine human activities and plastic objects that unintentionally release microplastics to the environment are not always taken in account, and are thus overlooked and underestimated (e.g. cleaning activities at home vs. mismanaged waste in the streets).

In this review, activities and social factors that may be linked to the delivery of microplastics to sewerage systems, and eventually to WWTPs, are identified and critically reviewed. The characteristics of the microplastics involved are described according to their origin categories: primary and secondary sources. Recommendations for further research are provided.

2. SEWERAGE NETWORK SYSTEMS

The European Union Urban Waste Water Treatment Directive (91/271/EEC) describes urban wastewater (WW) as a mix of WW from households, commercial, industrial premises, and run-off from streets and other surfaces (The Council of the European Communities, 1991). WW may have different fates: 1) collected by a sewerage network that is connected to a treatment facility, where ultimately it is treated; 2) collected by a sewerage network and emitted directly to the environment without any clean-up; 3) discharged directly to the environment; or 4) treated individually (e.g. collected in septic tanks) (Ryberg et al., 2018).

A sewerage network is the infrastructure that collects the wastewater from a community, and it can be a combined or separate sewer system. A combined sewerage system is designed to collect domestic sewage, industrial and runoff from streets (U.S. EPA., 2022). Globally, only 20-48% of WW is collected and treated, whereas the rest is returned to the environment without any treatment due to a lack of technical capacity, infrastructure, or financial support (UN-Water, 2019; Jones et al., 2021). Radcliffe (2019) observed that cities within developing countries have less developed water treatment systems. For example, in the capital of Cambodia, Phnom Penh, the crude sewage from a combined sewer system is often discharged into open channels, as there are no treatment plants to sanitise the wastewater (Radcliffe, 2019).

Furthermore, the capacity of the pipes from combined systems occasionally can be overloaded during heavy periods of rainfall or snowmelt, and consequently, the collected WW may be discharged into nearby water bodies without treatment (Chaplin, 2020; U.S. EPA., 2022). Increased runoff from urbanization, growing urban populations and the accumulation of solid waste in the streets are factors that affect these systems (Zambrano et al., 2018; Hutson and Moscovitz, 2019; Radcliffe, 2019).

It is important to recognise that the WW generated is not always directed to a treatment process, but can be directly discharged to aquatic ecosystems, causing pollution to the environment. Microplastics are just one possible type of pollutant found in WW.

3. MICROPLASTICS

3.1 Definition

In 1971, small pieces of plastic in the environment were first detected in the Sargasso Sea (Carpenter and Smith, 1972). The term "microplastic" was first applied to a study made on marine sediments (Thompson, 2004). In 2008, in the first microplastics marine research workshop hosted by the NOAA agency, a consensus was reached to define microplastic as a plastic particle smaller than 5 mm (Arthur et al., 2008); this definition has since been commonly used. Newer proposed definitions consider microplastics to be sized from 1 µm to 1 mm (Hartmann et al., 2019). For the purposes of the present review, particles from 1 µm to 5 mm in size are considered MPs, in accordance with the definition by Frias and Nash (2019). However, it is important to mention that there are other categories that describe smaller particles: submicron- (1 µm to 100 nm) and nanoplastics (< 100 nm), showing different properties and characteristics, but likely to have the same fate as MPs (Caldwell et al., 2022).

According to Frias and Nash (2019) and Hartmann et al., (2019), apart from size, a plastic microparticle should be a solid synthetic polymer with a regular or irregular shape (e.g. fibres, fragments, films and microbeads), and be insoluble in water at 20°C. However, modified natural polymers such as cellophane and rayon, might be included as well under the definition of plastics as these polymers have gone through a chemical transformation (Hartmann et al., 2019).

Cole et al., (2011) identified two categories of MPs defined according to their origin: "primary" (section 2.2) and "secondary" (section 2.3). Primary microplastics are intentionally created for a variety of purposes and are found in a diverse range of products. Secondary microplastics are irregular particles originating from the fragmentation of larger plastic items (Crawford and Quinn, 2017b; Weithmann et al., 2018).

The following overview focuses on MPs that are likely to reach WWTPs via the sewerage network (Table 1).

3.2 Primary microplastics

3.2.1 Personal Care and Cosmetics Products (PCCPs)

The application of synthetic plastics in dermal care products emerged in the 1970s, initially as an exfoliant skin cleanser, and since then, their functions and benefits expanded rapidly to other products (Beach, 1972).

Two types of cosmetics products exist according to their duration of use on the human body: rinse-off and leave-on products. Rinse-off products include those intended to stay a short period on the skin, hair or mucous membranes to achieve their purpose. Examples are shampoos, toothpaste, liquid soap, shaving foam and bath/shower additives. Leave-on products, such as skin care, make-up, and nail varnish are designed to stay in prolonged contact with the body. There are individual, specific reasons for adding microplastics as to products such as dermal exfoliator

TABLE 1: Microplastic categories according to their source.

Source	Examples
Primary	
Personal Care Products and Cosmetics	Rinse-off and leave-on products
Medical Applications	Encapsulating agents, teeth polishing,
Paints and Inks	Surface coating, polishing agents
Clothing and Textiles	Decorative objects
Industrial abrasives	Abrasive media
Accidental spills	Release of pre-production pellets
Wastewater Treatment Plants	Flocculant agents, 'bio-bead' filtration media
Secondary	
Mismanaged plastic waste	Littering, illegal dumping, leakage from landfill
Synthetic textiles / clothing	Abrasion from washing machines and tumble dryers, natural weathering
City pollution	Car/bike tyres and car brake abrasion, city dust
Accidental spills	Household items to the sink / toilet

agents, skin conditioners, binding and viscosity regulators and aesthetic agents (Leslie, 2014; Amec Foster Wheeler Environment and Infrastructure UK Limited, 2017; Crawford and Quinn, 2017c) (Table 2).

Primary MPs in Personal Care and Cosmetics Products (PCCPs) are typically found in spherical and regular shapes. Sizes can be variable as some can be large enough to be seen without the need for a magnification, and others less easily seen with the naked eye (Gouin et al., 2015). Microbeads bigger than 450 µm account for 70 % of the particles used in cosmetics; beads of this size produce more friction in cleansers and, therefore, a better scrubbing performance (Gouin et al., 2015; Sun et. al. 2020). More than 90% of total microplastics in PCCPs are PE beads designed for use as an exfoliant (Gouin et al., 2015). Facial cleansers have the highest abundance of plastic particles, whilst the particles found in shower gels are larger than in other PCCPs (Duis and Coors, 2016; Sun et. al., 2020). Plastic content sometimes can represent more than 90% of the ingredients (Amec Foster Wheeler Environment and Infrastructure UK Limited, 2017).

Microbeads from PCCPs are suspected to be the most common primary MPs in the influents to WWTPs. Hidayaturrahman and Lee (2019) found high concentrations of microbeads in both influent and effluent streams, higher than any other type of particles. They reported that these beads had similar characteristics to the ones contained in PCCPs. Likewise, in a study in the U.S., it was observed that MPs were probably derived from personal care and cosmetics products (Carr et al., 2016).

Napper et al. (2015) examined six different brands of facial scrubs to determine the characteristics of their microplastics content. On average, each 150 ml bottle contained ~1.4 million particles, ranging from 8 µm up to 2 mm in size (Napper et al., 2015); it was concluded that in a single application about 50,000 microparticles could be discharged to the WW system. However, to obtain a robust and reliable estimate of the number of MPs released to the environment, other factors such as sales data, the number of people using facial scrubs, the frequency of consumption and the characteristics of the product, must be taken in account. Furthermore, PCCPs consumption statistics mostly focus on the female population, rarely involving male consumers on market reports.

In 2019, the European Chemicals Agency proposed a ban on the addition of microbeads to "rinse-off" PCCPs in the EU, anticipating a reduction of ~500,000 tonnes of microbeads released to the environment over 20 years (European Chemicals Agency, 2019). According to the trade association Cosmetics Europe, due to a voluntary initiative by the cosmetics industry encouraged by the aforementioned association, there was a decrease of ~97% in the use of microbeads for exfoliating and cleansing purposes between 2012 and 2017 (Cosmetics Europe, 2018). However, apart from Italy, Sweden, France, Canada, South Ko-

TABLE 2: Plastics contained in Personal Care and Cosmetics Products (PCCPs).

Polymer	Function	Size range	Reference
PAC	Viscosity control	2 - 10 µm	Leslie, 2014 Bintein, 2017
PE	Film formation (e.g. sunscreen), binding agent for powders, exfoliant peeling agent	200 µm - 1.25 mm	Leslie, 2014; Bintein, 2017
PET	Aesthetic agent (e.g. glitter in make-up), film formation, viscosity control	200 - 320 µm	Leslie, 2014; Bintein, 2017
PLA	Exfoliant peeling and texturing agent	< 315 µm	Bintein, 2017; SpecialChem, 2020
PMMA	Sorbent material for delivery of active ingredients	200 - 320 µm	Bintein, 2017
PP	Viscosity increasing agent	< 1 mm	Leslie, 2014; Bintein, 2017
PS	Film formation		Leslie, 2014
PTFE (Teflon™)	Binding, bulking agent and slip agent	5 - 15 µm	Leslie, 2014; Bintein, 2017
PUR	Film formation	200 µm - 1.25 mm	Leslie, 2014: Bintein, 2017
Styrene acrylates copolymer	Coloured microspheres as aesthetic agents	2 - 10 µm	Leslie, 2014

PAC: Polyacrylate; PE: Polyethylene; PET: Polyethylene Terephthalate; PLA: Polylactic Acid; PMMA: Polymethyl Methacrylate; PP: Polypropylene; PS: Polystyrene; PTFE: Polytetrafluoroethylene; PUR: Polyurethane rea, New Zealand, Netherlands, Ireland, USA, UK, China, recently Portugal, and the first country in Latin America, Argentina, regulations and restrictions regarding the presence of microbeads in PCCPs are not yet agreed on a global basis (New Zealand Government, 2017; ChemicalWatch, 2018, 2021; DEFRA, 2018; Daliday, 2019; Oireachtas Library & Research Service, 2019; Watkins et al., 2019; Argentina Presidencia, 2020).

In 2018, the sale of products containing "rinse-off" microplastics was banned in the UK: the number of microplastics from cosmetics released to the environment was consequently expected to decrease (DEFRA, 2018). As far as we are aware, currently this regulation does not apply to those PCCPs sold online coming from other countries where there microplastics are still used as ingredients.

3.2.2 Industrial Applications of primary microplastics

Microplastics are used as industrial abrasives and cleaning agents e.g. as blasting agents to remove paint, adhesives, dies from some material surfaces, or detergents (Essel et al., 2015; Hale et al., 2020). The plastic granules are usually made of polymethyl methacrylate (PMMA), polyester (PES), PE, melamine, and polycarbonate (PC), with an average size of 150 μm and 2.5 mm depending on their functions (Duis and Coors, 2016; Magnusson et al., 2016). Plastic media blasting is sometimes used instead of sand blasting as it offers lower damage to the surface and it does not remove a considerable amount of material (Gatto et al., 2019).

In Denmark, 5 to 25 tonnes per annum of MPs are estimated to be used as a substitute for sand in sandblasting purposes (Amec Foster Wheeler Environment and Infrastructure UK Limited, 2017). When water is used during blasting activity, wastewater should be pre-treated before being disposed to the sewage according to the local regulations, or be collected by a licensed waste carrier, to prevent pollution to waterbodies generated from the abrasive media and the waste from the blasted surfaces.

3.2.3 Accidental Spills of primary microplastics

Plastic pollution in the marine environment due to industrial plastic pellets was first described 50 years ago (Gregory, 1977). In industry, raw plastic materials in the form of powder, plastic resin pellets (or 'nurdles') or granulates are found in the plastic pre-production stage, as a feedstock to manufacture larger artefacts (Essel et al., 2015; Duis and Coors, 2016; Crawford and Quinn, 2017a). For their size (up to 8 mm), the pellets facilitate plastic processing and are easy to transport (Crawford and Quinn, 2017a). Problems arise when these pellets are released unmanaged from production facilities or distribution. There are several reasons for pellet releases, including improper packaging, infrequent or inadequate cleaning operations, lack of a containment system and human error. Residues from plastic manufacturing and regranulation during recycling are other sources of MPs, now from a secondary process (Duis and Coors, 2016; Crawford and Quinn, 2017a). These granules can reach drains and sewerage systems via surface run-off from interior and exterior sources.

At present, there are no reliable estimates of the pellets entering the environment or sewage networks from plastic production or from processing stages in an area. Some companies do not provide estimates of pellet production loss (Essel et al., 2015). In Europe, there is an estimated 55,000 to 550,000 million tonnes of MPs lost, assuming that the pellet loss is ~0.1 to 1.0% of the total European plastic production (Essel et al., 2015; PlasticsEurope and EPRO, 2021). Although a decline in loss of virgin pellets to the environment has been reported, some studies have found high concentrations of MPs close to plastic production facilities (Ryan et al., 2009; Crawford and Quinn, 2017a).

A study performed inside and outside three mechanical recycling facilities in Vietnam confirmed the release of small plastic particles due to improper wastewater management following the washing stage (Suzuki et al., 2022). In this case, recycling might help to reduce the amount of plastic waste destined for landfills. However, in circumstances where there is no further wastewater treatment from the recycling process, rather than being a solution it is just shifting the problem from one place to another. Corcoran et al., (2020) studied the beaches of the Laurentian Great Lakes of North America and discovered a direct and positive relationship between the number of plastic industries in the vicinity and the number of MPs. In the UK, a study determined that untreated WW spilled from industry is the main source of microbeads found in riverbeds, concluding that WWTPs are effective in preventing these MPs reaching the waterbodies (Woodward et al., 2021). This observation highlights that improper WW treatment from industry might potentially lead to large emissions of MPs to the environment.

There are initiatives and legislation aiming to reduce plastic loss from the industrial sector. In the USA, the California Water Code (Chapter 5.2) declares that to control and regulate the discharges from preproduction plastic points and non-point sources, a programme must be developed by the State Board and the Regional Boards (California Legislative Infomation, 2007). Internationally, The International Convection for the Prevention of Pollution from Ships (MARPOL) protocol created in 1973, has completely banned the disposal of all forms of plastics into the sea since 1988 by the addition of Annex V. (International Maritime Organization, 2019). In Europe, there are no laws specifically to measure and address plastic pollution due to industrial leakage. The Operation Clean Sweep (OCS) programme is a voluntary approach designed to prevent plastic resin loss, in any form, along the plastic supply chain from industry (PlasticsEurope, 2018). Nevertheless, there are no legal agreements that force industries to comply the objectives of the programme. It is just recently that parties are looking to develop an OCS certification system that will allow third parties to regularly audit the member companies, and hence to effectively quantify the achievements and failures of the programme. The EU Plastic Strategy (2018) highlights the need to implement cross-industry agreements to restrict the use of microplastics and tackle release of plastic to the environment (European Commission, 2018).

More efforts should be aimed towards the detection of accidental spillage of plastic pellets during the production, packing, transport and distribution, and in recycling facilities, to evaluate the magnitude of the problem and effectively develop management measures to tackle it. It is also necessary to implement regulatory figures supported by legislation instead of following voluntary programmes or agreements, jointly with international cooperation.

3.2.4 Primary Microplastics in Wastewater Treatment

Within a WW purification process, some polymers are used (Table 3). These polymers, also known as 'bio-beads', are used to provide a surface where micro-organisms can attach and grow (Cohen, 2001; Cockburn, 2022), but these can be lost from treatment systems and are regularly reported in coastal and marine environments (Turner et al. 2019).

The application of synthetic polymers during WW treatment has advantages over the use of natural polymers (e.g. alginate and carrageenan): synthetic polymers are less likely to dissolve in WW and are less vulnerable to biodegradation. Furthermore, these synthetic support materials are highly stable and their porosity can be controlled (Cohen, 2001). Flocculant agents are added to sludge in order to promote the aggregation of particles to larger solid particles (Murphy et al., 2016; Talvitie et al., 2017). To capture metals such as Pb, Cd and Zn from wastewater, ethyl acrylate is employed (Maleki et al. 2015).

It is important to be aware of these polymers at WWTPs. Even if their potential contribution to further contaminate the wastewater and sludge is low compared with raw wastewater, the synthetic polymers that aid to WW treatment process could cause further pollution problems in the environment. It was recently reported that sewage plants from the east coast of the UK released millions of black plastic bio-beads, causing great pollution not only at the North Sea but also to the Dutch coast (Cockburn, 2022). This is an example of the difficulty of containing microplastic material.

3.3 Secondary microplastics

Secondary plastics are irregular particles that originate from the breakdown of larger plastic pieces due to UV radiation, mechanical forces and/or biological degradation (Crawford and Quinn, 2017b; Weithmann et al., 2018). According to Horton and Clark (2018), these kind of particles are the most common and widespread microplastics in the environment.

3.3.1 Urban Pollution

Road dust is defined as earthen material such as gravel, soil, sand, and other materials (United States Environmental Protection Agency, 2010). It consists of a mixture of naturally-occurring and man-made particles. For the purposes of this review, the particles from synthetic objects are included within this definition. For example, the particles generated by plastic litter, traffic-related infrastructure and vehicle use, being the last one an area of concern in recent years.

In an urban environment, sources of plastic microparticles include mismanaged waste, road markings, vehicle/tyres (section 2.3.3), construction/roadwork activities (section 2.3.2), atmospheric debris, exterior paints, and items of personal footwear and clothing (Dehghani et al., 2017; Ryberg et al., 2018). In these cases, the release of MPs is caused by abrasion, unintentional emissions, and natural wear of the materials. Building coatings, applied for ornamental purposes and to prevent fouling and corrosion, might be degraded as a result of continual exposure to UV-irradiation (Gaylarde et al., 2021). These MPs are distributed by the wind and vehicle-generated currents to the atmosphere (Vogelsang et al., 2019; Järlskog et al., 2020). However, they can enter WWTPs through combined sewerage systems; this process is directly connected to meteorological conditions and wet cleaning-sweeping. Unlike PCCPs, MPs from outdoors enter the drainage network through man-made holes, catchment basins and storm drains, mostly by run-off water (Yukioka et al., 2020).

3.3.2 Road wear

Road surface markings are a combination of pigments, reflective materials and resins such as epoxy, acrylic, PE, PA and PMMA (Migletz et al., 2001; Sundt et al., 2014; Road Marking Services LTD, 2021). The function of these synthetic resins is to provide visible, durable and long-lasting marks on the surface. In 2014 in Sweden, it was calculated that ~500 tonnes of polymers were added to road paint and markings, slightly more than in Norway (Magnusson et al., 2016; Vogelsang et al., 2019). It was reported by a modelling study from the U.S. that these road markings could last from a few months to up to 4 years, depending on various factors: the roadway type, specifications, quality control, manufacturers and weather conditions (Migletz et al., 2001). Road surface markings can be damaged by vehicle traffic and the other factors mentioned above, and also by pedestrian traffic (Kitahara and Nakata, 2020).

Kitahara and Nakata (2020) investigated microplastic abundance in road dust from rural and urban areas in Ja-

TABLE 3: Synthetic Polymers used to aid Wastewater Treatment Processes.

Synthetic Polymer	Function	Reference
Ethyl acrylate	To capture heavy metals	Maleki et al., 2015
Polyacrylamide	Flocculation agent	Bintein, 2017; Talvitie et al., 2017
Polyethylene-based biobeads	Filtering media	Turner et al. 2019
Polypropylene glycol	Carrier agent	Cohen, 2001
Polystyrene	Carrier agent	Talvitie et al., 2017
Polyvinyl alcohol	Carrier agent	Cohen, 2001

pan; they also evaluated relationships between road markings and the detected MPs. Over 60% of microplastics were found to be PES, PVC, and PMMA, and some chemicals such as organophosphate flame retardants and UV stabilisers were detected in both MPs and the fragments samples from the road markings, suggesting that the surface markings are a potential source of MPs (Kitahara and Nakata, 2020). Furthermore, they found a positive correlation between total MPs abundance and the daily traffic density. Road surface markings may be an important source of MPs to the environment, as these synthetic paints are present on the surface of many roads around the world.

Other traffic-related infrastructure of concern includes traffic cones, drums, and speed bumps; some speed bumps are produced using recycled PVC from bottles (Dehghani et al., 2017). These plastic items found in the street might be further degraded, for example, via UV light from sunlight to which they are usually exposed and might eventually produce plastic particles; this issue merits further attention.

3.3.3 Vehicle Tyre and Brake wear

Vehicle tyres have been suggested as one of the most important sources of MPs in the environment and a significant pollutant in street dust (Kole et al., 2017). The contact between the road surface and tyres causes abrasion and heat in the tyre, and results in the release of small particles (Grigoratos and Martini, 2014; Kole et al., 2017). Figure 1 illustrates the general composition of the tyres, plus the aspects that can impact on a tyre's wear (Vogelsang et al., 2019). The average tyre's tread wear ranges from 0.006 to 0.1 g/km for small cars, to 1.0 g/km for heavy goods vehicles (Aatmeeyata et al., 2009; Pant and Harrison, 2013; Kole et al., 2017). It is estimated that a standard passenger car tyre can last from 20,000 to 50,000 km, assuming the tyres in front and back last differently, and can lose from 10 to 30% of the mass during their lifetime (Pant and Harrison, 2013; Kim and Lee, 2018). Examples of the estimated annual amount wear per country based on passenger car traffic are provided in Table 4. According to a simulation experiment to test tyre wear under specific driving conditions, braking and slippery events increase the number of particles emitted (Kim and Lee, 2018).

TABLE 4: Estimated annual amount wear per country based on passenger car traffic.

Country	Wear and Tear tonnes	Reference
Australia	20,000	a
UK	44,897	*
India	51,998	b
Mexico	91,932	**
Brazil	143,023	b
China	352,000	b
USA	586,800	b

** Calculated on the total vehicle registered in 2017, average milage per car and tyre tread wear emission factor of .1 g/km (Parry, 2017; Järlskog et al., 2020; National Travel Survey, 2020). ** Calculated on the total vehicle registered in 2019, average milage per car and tyre tread wear emission factor of .132 g/km (Office of Highway Policy Information, 2010; Kole et al., 2017; INEGI, 2019). a.- Kole et al., 2017; b.- Milani et al., 2004.*

It is estimated that 50% of the deposited tyre particles on the roads are mobilised via run-off water and can be directed either to the drainage or to surface waters, whilst the rest can accumulate on the roadside soil (Wagner et al., 2018; Unice et al., 2019; Järlskog et al., 2020). Rødland et al., (2022) analysed the number of MPs in roadside snow samples from different sites in Oslo, Norway, and concluded that high driving speed was the most important factor; as driving speed increases, road surface abrasion increases. In this study, the fact that melted roadside snow should receive treatment prior to release to the environment, since it might contain tyre plastic particles, is highlighted. Tyre wear can thus lead to water pollution directly to the surface waters (without any treatment) or through WWTPs via effluent if they are not removed from WW.

For vehicle-brake pads, it is challenging to define their material components as there are many variations in their structure. Usually, they consist of a rigid black laminate onto which a friction material is attached. Friction materials can be classified as non-metallic, low metallic, semi-metallic, ceramic and non-asbestos organic (Lyu et al., 2020). Brake pads, initially made from asbestos, are currently made of a mixture of materials such as rubber, silica, glass or fiberglass, and Kevlar (synthetic fibre trademark) bound together with resin. Semi-metallic brake pads are made of a mixture of organic materials and metal, such as iron, copper, and steel, held together by synthetic resin (Borawski, 2020; Bridgestone, 2021). Additionally, brake pads contain four groups of other components: fibres, fillers, binders, and friction additives (e.g. flame resistant oxides and lubricants) (Oluwafemi et al., 2019; Borawski, 2020). The binders are routinely made of phenolic and epoxy resin (Borawski, 2020).

Vehicle brake-related wear particles are released when brake components are forced against the inner surface of a rotating cylinder while undergoing deceleration (DEFRA, 2001; Grigoratos and Martini, 2014). Some of the materials worn during braking are deposited on roadways or roadsides and can be easily transported in the environment. In a study by Hagino et al., (2016) by creating a laboratory-brake dynamometer system, concluded that particles down to 10 µm are released at rates from 0.04 to 1.2 mg per km per vehicle. The quantity increases for heavy duty vehicles (DEFRA, 2001). According to Vogelsang et al., (2019), the road-related load of MPs to Norwegian WWTPs per year ranges from 7 tonnes for polymer-modified bitumen, to ~49 tonnes for road markings and up to ~1,185 tonnes for tread rubber from tyres. However, the authors recommend caution with these results, as they are highly speculative. WWTPs are expected to be an important sink for tyre wear particles via the surface runoff water, occurring during rain and meltwater events, also in the sweeping and cleaning activities of the ground surface if litter is deposited into the drainage system.

Nowadays particulate matter released from vehicle tyres and brakes is of specific concern (e.g. human health-related issues) due to the presence of plastic particles, and there are lots of factors to be further discussed.

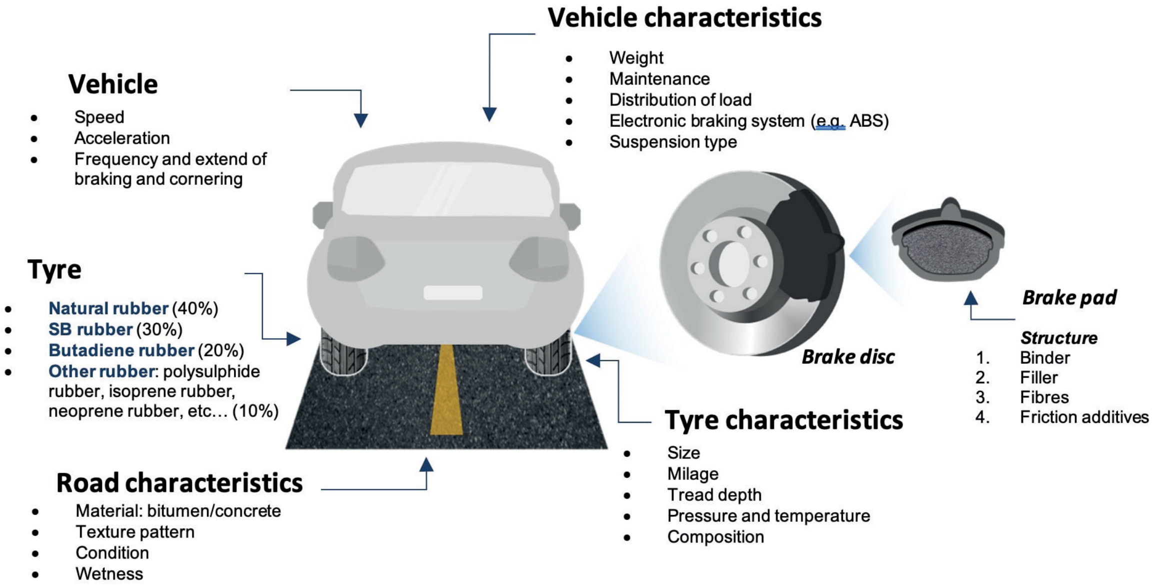

FIGURE 1: Tyre composition and factors affecting tyre wear. Also, the structure of the brake pad within the brake disc.

These factors are relevant if it is desired to estimate more precisely the emissions of MPs from vehicle and tyre brakes. For example, the material composition and properties from both tyres and brakes are not always consistent and vary according to their geographical origin; the structure of the road surface and its operation conditions; average daily traffic and types of vehicles transiting through it, amongst other factors.

3.3.4 Mismanaged Plastic Waste

Environmental contamination due to waste mismanagement is a worldwide problem. Municipal solid waste (MSW) is defined as the discarded residues from domestic and commercial premises, institutions and small-scale industries (Hoornweg and Bhada-Tata, 2012). Here, we add the waste produced in the streets, not involving an enclosed area (e.g. dog faeces bags, waste from picnics) According to Kaza et al., (2018), it is estimated that the global human population generates ~2.0 billion tonnes of MSW every year, projected to rise up to 3.0 billion tonnes by 2025. In general, the level of urbanisation, economic development and industrial growing, determine waste generation (Hoornweg and Bhada-Tata, 2012; Tsakona and Rucevska, 2020).

From the total waste produced, ~12% is attributed to the plastic sector (Kaza et al., 2018; Tsakona and Rucevska, 2020). In 2015, according to Geyer et al., (2020), LD-PE, PP, PES (plus acrylic fibres and polyamide) and HD-PE, represented the largest volume of polymers within the plastic waste generated worldwide, with 57 million tonnes (24%), 55 million tonnes (23%), 42 million tonnes (17%) and 40 million tonnes (16%) respectively. Most of the plastic waste comes from the post-consumer market, i.e. waste from during or after the product use (Geyer et al., 2020). The sectors that are mainly responsible for the waste due to plastics are packaging, followed by textiles and consumer goods, contributing 46%, 15% and 12%, respectively, to overall plastic waste generation (Tsakona and Rucevska, 2020).

According to Geyer et al., (2017), ~6 billion tonnes of plastic have been produced since 1950, and 79% has been disposed in landfills or lost to the environment. Plastic waste can be recycled into a secondary material, incinerated for energy recovery, or eventually disposed of. The two most probable scenarios are when the waste is discarded on land: it is either contained in managed systems such as sanitary landfills, or left in unmanaged systems in the natural environment (Geyer et al., 2017).

Mismanaged plastic waste is littered or inadequately disposed in dump sites, or in open and uncontrolled landfills (Jambeck et al., 2015). Secondary MPs can reach WWTPs by weathering of larger plastic items dumped in the environment via run-off water (Bui et al., 2020). Leachate, a liquid produced by the contact of water and solid waste, occurs in all types of landfills and needs to be managed. This liquid can be polluted by a number of suspended materials (Rao et al., 2017), within which MPs could be present. Not only these MPs but also their leaching additives from their surfaces can reach sewerage systems by run-off water during rainfall if the dump site is not covered to reduce leachate formation (Magnusson et al., 2016). Figure 2 shows an example of the liquid by-product from a municipal solid waste located in Mexico and the pieces of plastics present on the floor site (currently the site is no longer in operation).

It is important to develop studies referring to the presence of MPs in the leachate from landfills, specially, in this case, if it is discharged directly to the sewer. For example, van Praagh and Liebmann (2021) studied the concentration of MPs in the leachate from 11 landfills, reporting that in some sites this fluid was discharged to the WWTPs without receiving any previous treatment. They reported an annual average load of 20 kg of MPs, based on the leachate volume production per year, with sizes ranging from

FIGURE 2: Leachate presence in an open dump site located in Mexico.

5 mm to 50 µm (due to size limitations). Within the type of plastics found, rubber and polymer-modified bitumen are included, whilst PE was the most dominant type of plastic. They reported that the kind of plastics detected might be from other sources, such as the different operations inside the facilities and atmospheric deposition (van Praagh and Liebmann, 2021). These examples of unexplored potential sources of MPs to the environment require a research focus on countries that rely on the disposal of plastic waste to landfills and further improvement of sampling techniques to detect lower sizes of MPs. Silva et al., (2021) calls for appropriate management and monitoring programmes alongside mitigation strategies, to control the pollution due to the leachate from landfills.

In a study undertaken in Teheran, Iran, by collecting dust from streets for two months, ~2600 MPs were detected, mostly fibres, and estimated from 83 to 605 particles per 30 g/dry dust (Dehghani et al., 2017). The authors mentioned that particles of plastic in the streets are likely because of the poor recycling rates and the lack of interest by citizens. Yukioka et al. (2020) carried out a study by collecting street dust in three different Asian countries. They discovered that in Japan, at the reference site, the highest numbers of MPs were found nearby commercial facilities, whilst in two other places, Vietnam and Nepal, were detected nearby restaurants and many dump points. The latter suggests that the existence of illegal dump wastes might increase the number of MPs on streets. PE, PP, from plastic bags and packaging; and SBS rubber, from vehicles and shoe soles, were the most frequently found polymers (Yukioka et al., 2020). Further research is needed regarding the deeper layers from unpaved roads, as MPs might get trapped by the continuous pressure of the vehicles against the ground. MPs trapped in soil might delay their transfer to the WWTPs.

During the COVID-19 pandemic, cases of mismanaged plastic waste have increased, especially personal protective equipment such as gloves and face masks (Akber Abbasi et al., 2020; Roberts et al., 2021). According to Prata et al., (2020), at the beginning and during the pandemic, globally every month ~130 billion and 65 billion face masks and gloves were used respectively. In cities, many of these items are found on the streets, and subsequently they can get to the sewerage systems, depending on the drainage design. Finding the proportions of these products leading to the treatment plants is a way to reveal the situation of waste thrown in the environment.

Although fly-tipping occurs almost everywhere, ~2 billion people lack basic waste collection services (OECD, 2018). These people are mostly from middle- and low-income countries, where they rely on informal waste collection methods. Informal recyclers collect plastic waste from streets and landfills, performing a critical function for the plastic waste management in those places (Ferronato and Torretta, 2019). Lebreton et al., (2017) estimated that ~1.8 million tonnes of plastic waste (average size of 50 cm to 300 µm) is transported from rivers into oceans each year, with Asia the largest contributor; nevertheless, the portion of MPs derived from mismanaged waste arriving at WWTPs is difficult to estimate.

3.3.5 Synthetic textiles

Microfibre pollutants of concern, mainly textile-related MPs, come from synthetic textiles (Salvador et al., 2017). PES, PA, PP and acrylate are the most common types of plastic that synthetic fibres are composed of, representing ~60% of global fibre consumption (Salvador et al., 2017). These fibres can be made in a continuous yarn form or twisted short yarns (Salvador et al., 2017). The production of fabric using these materials involves yarns, knitting, braiding, woven and nonwoven styles (Salvador et al., 2017; Félix-de-Castro et al., 2019). The fibres can include additives, such as dyes or chemicals, to improve colour fastness, which are used to modify appearance and improve a garment's performance (Darbra et al., 2011).

The release of fibres from textiles during laundry has been widely reported (Hartline et al., 2016; Napper and Thompson, 2016; Pirc et al., 2016; De Falco et al., 2018). The weathering of synthetic textiles in washing machines is determined by a combination of several factors, such as the fabrication parameters of the motor, characteristics of the garments, laundry products used and consumer behaviour (Hartline et al., 2016; Napper and Thompson, 2016; Salvador et al., 2017) (Figure 3). The age and quality of the materials are important. According to Carney et al., (2018), older garments release more fibres that newer ones. This is contrary to the results presented by Kelly et al. (2019) who found that on the first washing there is the greatest release of fibres from synthetic textiles, gradually decreasing over subsequent washing cycles. Browne et al. (2011) observed that >1900 microfibres could be released from a cloth per wash. Three types of fabrics treated at different conditions using washing machines released between ~138,000 to ~729,000 fibres per 6 kg load (Napper and Thompson, 2016). Another study discovered a range of 6-17 million fibres per 5 kg wash by using a laboratory simulator of a washing machine (De Falco et al., 2018). The variance in results can be explained by the different methodologies applied and the objectives to achieve. It is important to highlight that there are many factors involved apart from the textiles tested.

During laundry, textiles experience deformation, compression and expansion by the washing machine (Warmoeskerken et al., 2002). This may cause some detachment of fibres from the garments. However, the washing products might prevent damage to the materials as the foam produced might reduce the rubbing action among fibres and the mechanical abrasion (De Falco et al., 2018). There are contrasting opinions in relation to the use of fabric softener and the release of fibres. Pirc et al., (2016) and Lant et al., (2020) studied the production of fibres during machine washing and concluded that softener and detergent do not significantly influence the release of fibres. In fact, De Falco et al., (2018) observed that the use of softeners could reduce by ~35% the liberation of fibres. In contrast, Napper and Thompson (2016) found that textiles shed more fibres when fabric softeners are used. Temperatures >70°C can damage the structure of clothing (Laitala et al. 2011). Washing time can also induce the release of fibres from textiles (De Falco et al., 2018). Lant et al., (2020) confirmed that lower temperatures (e.g.15 °C) and shorter washing cycles could help to reduce fibres' detachment from synthetic fabrics. The make/model of the washing machine influences the behaviour of the fibres likely to spread. According to Hartline et al., (2016), front-loading washers release fewer fibres as they typically have a lower cycle duration and water consumption, compared with top-loading models. The aforementioned might be related to the results from Kelly et al., (2019) and Lant et al., (2020), which concluded that a low water-volume-to-fabric ratio reduces the fibres release.

The different washing procedures and equipment should be taken into account when the relationship between washing activity and the release of fibres from clothes is studied. For example, testing under the European and the North American washing conditions will differ from each other on the style and brand of the washing machine, loading capacity, laundry products, and the setting programmes which include water temperature, cycle duration and spin speed (Kelly et al., 2019; Lant et al., 2020).

The design of textiles can influence microfibre shedding (De Falco et al., 2018). For example, shorter staple fibres might easily detach from yarn (Félix-de-Castro et al., 2019), and also, tightly constructed yarns in clothes are preferred as they reduce fibre shedding (Carney et al., 2018). PES is often a preferred material as it brings durability and strength to clothes (Napper and Thompson, 2016; Gündoğdu et al., 2018). An Italian study found that 83% of fibres in the influent from a single WWTP were PES (Magni et al., 2019). According to Browne et al. (2011), fleece fabric can spread ~180% compared with other types of fabrics. The type of textile material and the design are factors that must be taken in account together to analyse the detachment of fibres from garments.

The types of clothing used are dependent on the time of year and weather conditions. During wintertime, the usage of washing machines increases by ~700% as the public usually wear more clothing (to stay warm), which is expected to lead to more fibres entering the WWTPs (Browne et al., 2011). This was confirmed by Ben-David et al., (2021) as in winter they noticed a doubling of MPs compared with the other seasons. Conversely, in Thailand, Kittipongvises et al., (2022) detected higher MPs abundance in the WWTPs during the dry season in two years, assuming that the mobility and dilution of the MPs, road run-off or other transport-related emission sources plus combined sewerage systems, were the determining factors on their findings.

Plastic particles used for ornamental purposes, such as glitter or plastic pearls, are catalogued as primary MPs as they were purposely manufactured to be small. However, they can accidentally detach from clothes during laundry (Crawford and Quinn, 2017d). Figure 3 summarises the factors involved in the fibres shedding from clothing.

It is estimated that ~95% of households in developed countries have washing machines (Salvador et al., 2017). The residents of some developing countries do not have full access to electricity (WorldBank, 2018), and therefore do not have access to electrically-powered washing machines. Some rely on washing their clothes in rivers, or discharge laundry effluents directly to water bodies, representing a source of MPs to water surfaces in addition to treated effluent from WWTPs. Currently there is a lack of data regarding the emission of fibres during handwashing, creating a limitation on estimating a more accurate number of fibres released from laundry activities. Some factors such as the type of detergent and softener, type of the synthetic material, water temperature, and washing tools and techniques (e.g. use of a clothes' stone, wash basin or brush), need to be taking in account to test the detachment of microfibres from handwashing to simulate a real situation.

The concerns related to tumble dryers mainly focus on microfibre pollution in air and terrestrial environments, and the consequent human exposure to these airborne microplastics. The drying of clothes is typically done in three ways: 1) Outdoors, by hanging the textiles on a clothesline, 2) Indoors, without any mechanical intervention, using for example drying racks or, 3) Indoors, by using a tumble dryer (Lant et al., 2022). There are three types of tumble dryers: vented tumble dryers, condenser dryers and those with combined washing and drying functions. Vented tumble dryers expel warm damp air through a hose to the exterior, using a lint filter inside the appliance to collect the fibres detached from textiles. This filter is later cleaned by the consumer and the debris is usually deposited in the waste bin. The second type, the condenser dryer, is a sealed system that condenses the moisture from clothes and the water collected is emptied by the consumer or automatically drained away. Some combined washer/dryer appliances, however, do not have a lint filter, meaning that the accumulated fibres are released to the drainage pipe (Lant et al., 2022). The latter two types of tumble dryers could be a potential source for microplastic pollution to wastewater, as they rely on the disposal of collected debris to the drainage system.

Tao et al., (2022) evaluated the microfibre release of 22 shirts made separately from cotton and PES materials, using a vented tumble dryer in different time settings. They detected that 1000 g of cotton textiles can release an average of 42,000 fibres, whereas the PES ones up to 55,330 fibres during 15 min of use, concluding that the PES textiles can produce more microfibres than the cotton ones. In contrast, the results presented by Lant et al., (2022) found that the fibres collected in both lint filter and the mesh adhered to the dryer exhaust were mostly cotton. They highlighted the importance of the pore size of the lint filter, as they discovered that changing from 100 µm to 40 µm reduces up to 35% the release of fibres through the exhaust. Other factors such as the previous treatment during the washing, the dryer products and the fabric are also important in this regard. In the context of the present review, the risk arises when these particles are deposited on the land surface and travel to the drainage system via the flow of water. For example, a study in the US analysed snow samples collected nearby dryer exhaust and confirmed the presence of microfibres up to 9 m away from the exhaust vent itself (Kapp and Miller, 2020). Once the snow melts, those trapped particles will redistribute in the environment. Also, drying textiles indoors involves the presence of airborne fibres in the environment, that can later settle down and contribute to indoor dust. This issue is further discussed in the next section 2.3.5.

Recommendations have been made in order to stop the release of synthetic textiles from washing machines and dryers. As an example, the use of more environmentally friendly textiles or cotton clothing has been suggested (Tao et al., 2022). However, it is important to take in account the water consumption behind washing a single shirt: to reach the amount of water necessary for the growing and processing of cotton, the garment needs to be washed and reused an average of 630 times (Pakula and Stamminger, 2015). Additionally, apart from the agrochemicals use in crops, some additives and dyes of synthetic origin are added to preserve the fabric longer and the fibres released might take longer to degrade. Therefore, the concept of so-called environmentally friendly textiles or stopping the use of synthetic textiles is more complex than it initially appears.

There is thus opportunity and need for research in this field. For instance, it would be of merit to test the different types of tumble dryers with particular emphasis on those discharging loose fibres and other particles. Investigating if the temperature applied, clothing load or the spin-dry rate of the dryer are factors contributing to the shedding of fibres, regardless of the laundry products used, would also be of merit. Apart from testing PES and cotton textiles, more experiments involving mixed laundry loads of clothes (Lant et al., 2022) or with different percentage of fabrics should be undertaken. As laundry activities are very common, it is crucial to understand consumer behaviours and habits behind laundry activities. Around the world, many people cannot afford to use tumble dryers or washing machines or do not have access to an electricity supply, so they rely on other methods to wash and dry their clothes.

Apart from textile fibres, nonwoven fibres from sanitary products such as diapers (also known as nappies), tampons, pads and face/surface wipes are commonly found in the influent to WWTPs (Le Hyaric et al., 2009), as they are inappropriately flushed down household lavatories. A British water company (Thames Water) announced that

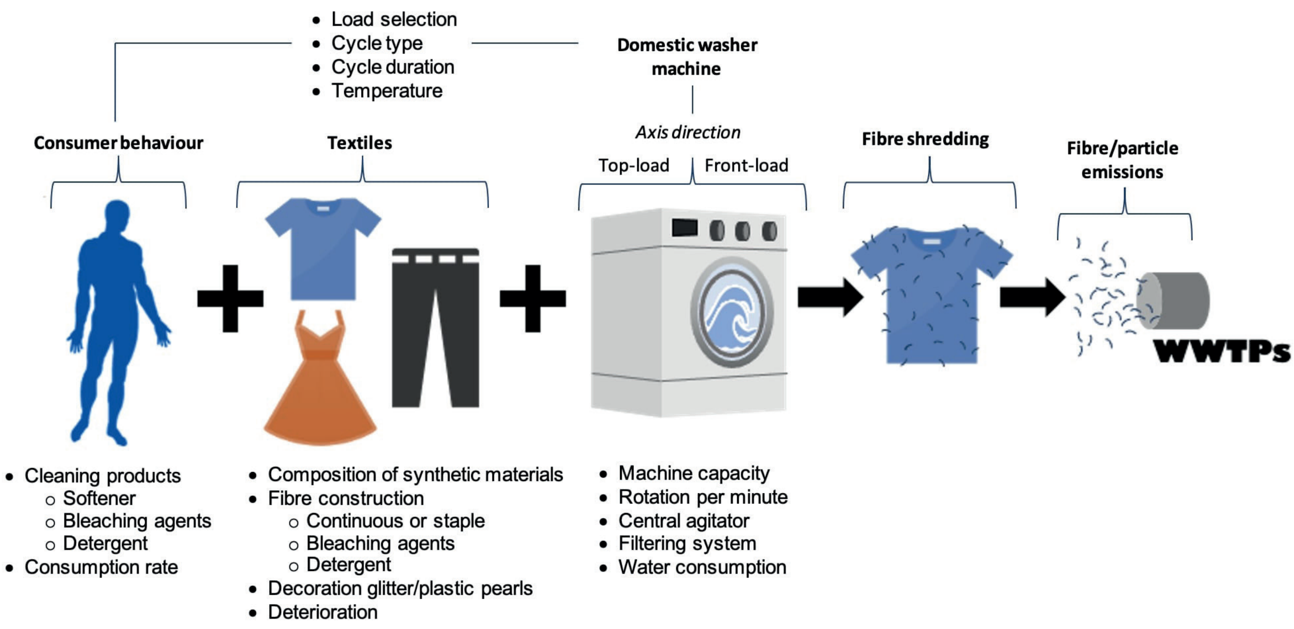

FIGURE 3: Factors influencing fibres emission from textiles during laundry. Adapted from Salvador et al., (2017).

due to the coronavirus outbreak in 2020, they registered a rise of 20% of sewer blockages due to wet wipes and other "unflushable" items (Thames Water, 2020). These "unflushable" materials cling to the fat and other debris, creating larger blockages in the sewage system, causing major plumbing problems and pipes to overflow the contained WW to the environment.

3.3.6 Indoor pollution

Human activities are often carried out indoors (Liu et al., 2020). Dris et al. (2017) estimated a value of ~5 fibres per m^3 and a mean deposition rate of 6,300 fibres/day/m^2 from three different sites: an office and two flats, showing one of the flats the highest concentration of fibres. This might be due to the different activities performed at each site and the inhabitant's lifestyle. They also detected a decrease in the number of fibres as their sizes increased, assuming that the larger the fibres, the faster they settle.

Zhang et al. (2020), identified that ~37% from all the particles they collected were MPs, mostly fibres, from three different locations inside a university. The same study found ~5 times more microplastics in a student dormitory compared with an office. MPs are directly linked to the number of occupants and activities taking place on-site (Dris et al., 2016). A study in Indonesia confirmed that the more crowded the room, the more plastic particles in the environment (Bahrina et al., 2020). In Denmark, by using a breathing human simulator connected to a filtering membrane, only 4% of all the particles detected were synthetic, with 87% fragment-shaped and the rest fibres (Vianello et al., 2019).

The most common polymers in indoor fibres are PES and PP (Dris et al., 2017; Bahrina et al., 2020; Zhang et al., 2020). PE, acrylic and PA fibres have also been detected (Dris et al., 2017; Zhang et al., 2020). Findings of PP microplastic particles are consistent with the objects contained in houses and offices, such as carpets, chairs and couches (Dris et al., 2017). Indoor paint, soft toys, plastic utensils, packaging and building materials are other potential sources of MPs (Vianello et al., 2019; Bahrina et al., 2020). However, contamination by MPs is mainly derived from clothing and textile products (Bahrina et al., 2020; Zhang et al., 2020).

The behaviour of MPs in indoor environments are determined by key factors e.g. mechanical ventilation inside and external airflow turbulence (Dris et al., 2017; Zhang et al., 2020). MPs could settle and be retained in carpets and rugs; resuspension of these particles might be more likely from hard floors and other hard surfaces in rooms (Dris et al., 2017). Humidity affects particle adhesion to surfaces and impedes the movement of particles (Mukai et al., 2009). Larger particles are detected in dustfall as they settle faster than smaller particles and so tend to accumulate (Dris et al., 2017; Gasperi et al., 2018).

Human activities and lifestyle contribute to MPs behaviour. Laundry habits, such as indoor drying, sorting and storage, can shed fibres from textiles (Sundt et al., 2014; Dris et al., 2017; Zhang et al., 2020). Another example is habitual behaviour when entering indoors; clothes and shoes might carry particles from outside to introduce and spread them inside. Cleaning activities, such as wiping furniture and surfaces, sweeping and mopping different material floors, and vacuuming rugs and carpets, disturb particles in the indoor environment. Dris et al. (2017) highlight that settled MPs on indoor surfaces are likely to end up in WWTPs if WW or cleaning products (e.g. wipes) are disposed via the sewerage system.

4. CONCLUSIONS

This review has established that a wide range of activities and processes contribute to the inflow of microplastics to wastewater treatment plants. Of these activities and processes, those that contribute primary microplastics are more readily estimated than those contributing secondary microplastics. Sources such as mismanaged waste, hand washing of textiles and leachate from waste disposal sites, for example, lead to the release of microplastics and all are primarily sources of secondary microplastics for which quantification is challenging. Likewise, microplastics from road surfaces are subject to a wide range of factors that influence their retention in or mobilisation from unpaved roads and are also challenging to quantify. For sources of primary microplastics such as pellets (nurdles), more robust monitoring could enable better management and control of spillage and loss. More stringent legal frameworks should lead to fuller compliance in this regard. Regulatory control should also consider the import of products from locations where less stringent controls apply, for example when purchases of personal care and cosmetics products are made via the internet.

The habits and behaviour of consumers are highlighted as important influencers of microplastic quantities and composition in wastewaters. Choices of textiles and fabrics have implications for secondary microplastics entering wastewater treatment plants, as do means and methods used for washing textile products. These outcomes emphasize that local actions and initiatives as well as broadscale measures are clearly needed if progress is to be made to remediate problems associated with microplastics in the environment.

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