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WHERE DO THEY GO? A REVIEW OF THE WASTEWATER TREATMENT PROCESS AND ITS IMPACT ON THE FATE OF MICROPLASTICS

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ABSTRACT

Wastewater treatment plants (WWTPs) are facilities designed to sanitise wastewater (WW) from different sources. A WW treatment process combines physical, chemical and biological reactions divided into 3 or 4 stages, according to the effluent standards of each site. WWTPs are intended to remove certain pollutants from the water stream, but not microplastics (MPs), plastic particles ranging from 5 mm to 1 µm. WWTPs work as source and destination for MPs. This review provides an insight into the stages included in WW treatment processes according to different technologies employed, alongside a critical evaluation of their influence on MPs. Relevant information and knowledge gaps have been identified. The removal of MPs depends on a variety of factors than just the type of incoming water or WWTP location. The most significant removal of MPs from WW occurs during the preliminary and primary stages. MPs removal efficiency fluctuates depending on the systems operating during the secondary and tertiary phases. As the comparison of information between studies is complicated due to the units and terms used to report and label the collected WW, we recommend that (1) kg/day units are applied if mass balance is of interest, and number of particles per L/day, considering the flow rate data from the sampling dates or annual average flow; (2) to further explain the selection criteria to classify MPs based on their size; and (3) to reach a consensus to name and identify the sampling points, such as the raw WW and effluent from the next stages.

1. INTRODUCTION

WWTPs are facilities that uses physical, chemical, and biological methods to treat wastewater (both sanitary wastewater and urban runoff) step by step, making it substantially cleaner, odourless, and compliant with regulatory criteria for safe release into water bodies (The Council of the European Communities, 1991; Vesilind, 2003). WWTP receives, treats, and stores wastewater from domestic dwellings, commercial and industrial premises, run-off from streets and other surfaces. These treatment works are designed to remove undesirable constituents in a systematic and stepwise fashion; such constituents might include large floating solid items (e.g. litter, vegetation, nappies); fats, oils and grease (FOG); organic matter; nutrients; pathogens (e.g. bacteria and viruses); toxic compounds, etc, and might be in form of suspended or dissolved solids. Some emerging contaminants may not be removed from water stream, such as MPs (Talvitie et al., 2017).

Microplastics are commonly (but not universally) defined as plastic pieces ranging from 5 mm to 1 µm, encompassing a variety of types, colours and shapes (Crawford and Quinn, 2017a; Frias & Nash, 2019). This definition is used for this study. Particles smaller than 1 µm are catalogued as submicron and nanoplastics (Caldwell et al., 2022). There are two categories according to the source of MPs (Cole et al., 2011). Primary MPs are deliberately created to meet specific tasks and products, whilst secondary MPs originate from further fragmentation of larger plastic items, due to mechanical abrasion, UV radiation and/or biological degradation (Crawford and Quinn, 2017a).

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The origins and fate of MPs have been investigated and numerous sources and destinations have been identified (Hasan Anik et al., 2021). As WWTPs receive many waste products from different sources, including large and small plastic particles, these treatment systems could work as an ultimate fate of MPs (Landeros Gonzalez et al., 2022). However, WWTPs are known to be sources of microplastics to water and soil, via the discharge of aqueous effluent into waterbodies and the use of sludge as an agricultural fertiliser and soil conditioner (Weithmann et al., 2018; Cowger et al., 2019).

The route from the raw wastewater (i.e. influent or crude sewage) to the treated water (i.e. final effluent) during WW treatment is a multifaceted procedure. Therefore, it is crucial to identify the different stages included in the WW treatment process and understand the working mechanism behind each step. Likewise, MPs throughout the WW treatment process is complex as there are many factors involved that might change their composition and external features in the final effluent and sewage sludge. However, these factors in some cases are not taken into account when it comes to study the presence of MPs in WWTPs. In this comprehensive review, we describe each stage in the wastewater treatment process according to different technologies applied, alongside a contemporary, critical evaluation of the state of knowledge of the impact of the treatment systems on MPs.

2. METHODS

This paper reviews the treatment process that takes place in wastewater treatment plants, evaluating the technologies implemented at each stage. The first section presents the stages of the treatment activity (preliminary, primary, secondary and tertiary) in relation to their general function and effects. Furthermore, in the second chapter of this study we analyse the impact of the treatment process on the microplastics in wastewater. The literature cited was obtained using “wastewater treatment plants” and “microplastics” as search terms. Published works referred to in the present study focused on: the occurrence and characteristics of microplastics in wastewater and sludge, and how the treatment equipment and other factors influence microplastics. Information such as location, sites for study, population served, average inflow, type of samples taken (WW and/or sludge), sampling collection period, sampling duration, sampling filter pore size, type of polymers, shape and removal efficiency from WW, were essential for the purposes of this review. The research for scientific articles relating to microplastics and WWTPs was completed in April 2022.

3. STAGES OF A WASTEWATER TREATMENT PLANT

3.1 Preliminary stage

The preliminary treatment stage typically consists of the physical exclusion of untreatable material (e.g. large plastic items) from the raw WW by screening and grit/grease removal (Butler et al.,1995; Oakley, 2019). Screening may include coarse and fine bar screens, usually from 25 mm to 6 mm (Butler et al., 1995; Oakley, 2019). The screened WW then continues to aerated grit chambers that facilitate the sinking of heavier solids - particles such as grit, gravel and sand (density of ~2500 kg/m3) (Stuetz and Stephenson, 2009). Fats, oil, grease and other floating materials are skimmed from the tanks (Butler et al., 1995; U.S. EPA., 1999b). Dissolved air flotation (DAF) is a common technique designed to remove insoluble substances such as oils, greases and suspended solids (Englande et al., 2015). During this process, small bubbles are produced by introducing air into WW at high pressure. These bubbles cling to the suspended solids, and consequently these solids rise to the surface and are removed by skimming (Englande et al., 2015; Bui et al., 2020; Chaubey, 2021).

Bar screens are usually cleaned mechanically but also can be cleaned manually. Sludge is collected separately and the effluent is then discharged to the next treatment stage. This preliminary stage of the process is mainly to protect a WWTP’s equipment from any damage or loss of efficiency derived from excessive scum formation, debris, oil films or fat accumulations (Oakley, 2019; Jasim, 2020). The grit removed by the screening process can be further split according to its solid composition. The more fibrous fraction, such as from sanitary products, might be taken out after screening and incinerated, whilst the solid fraction (e.g. grit) goes to landfill or other disposal facility (U.S. EPA., 2003; Michielssen et al., 2016; WWTPs staff, personal communication, October 18, 2021). Figure 1 shows the material discarded from this first stage.

3.2 Primary stage

After preliminary treatment, WW is introduced into a sedimentation tank or clarifier. In the primary clarifiers, the settleable solids and buoyant organic material are removed, which reduces the amount of suspended solids (Oakley, 2019). Chemical coagulants such as iron salts, ferric chloride and aluminium sulphate, are then used to produce dense and rapid-settling flocs (loosely clumped masses of fine particles) in order to increase the removal of suspended solids, phosphorus and heavy metals; and to reduce five-day biological oxygen demand (BOD_5), (Yargeau, 2012; Oakley, 2019; Jasim, 2020). The principal objective is to attain a homogeneous liquid that can subsequently be biologically treated.

The design of these sedimentation tanks falls into three classifications: a) horizontal flow, b) inclined surface and c) circular, radial-flow (Englande et al., 2015; Jasim, 2020) (Figure 2). The horizontal tank is a large rectangular tank where solid settle by gravity whilst the supernatant continues to the next wastewater treatment stage (Englande et al., 2015). The Lamella gravity settler system is an example of the inclined surface sedimentation tank. The inclined plates aid acceleration of deposition of the particles, as the particles agglomerate as soon as they reach the surface of the plate (Tarpagkou and Pantokratoras, 2014; Ma et al., 2019). In a circular tank, the solids that settle at the bottom (primary sludge) are removed continually by a rotary sludge scraper. The sludge is then pumped out and stored prior to treatment (Englande et al., 2015; Oakley,

FIGURE 1: Discarded waste from the preliminary stage. A) Fibrous waste; B) Grit removed.

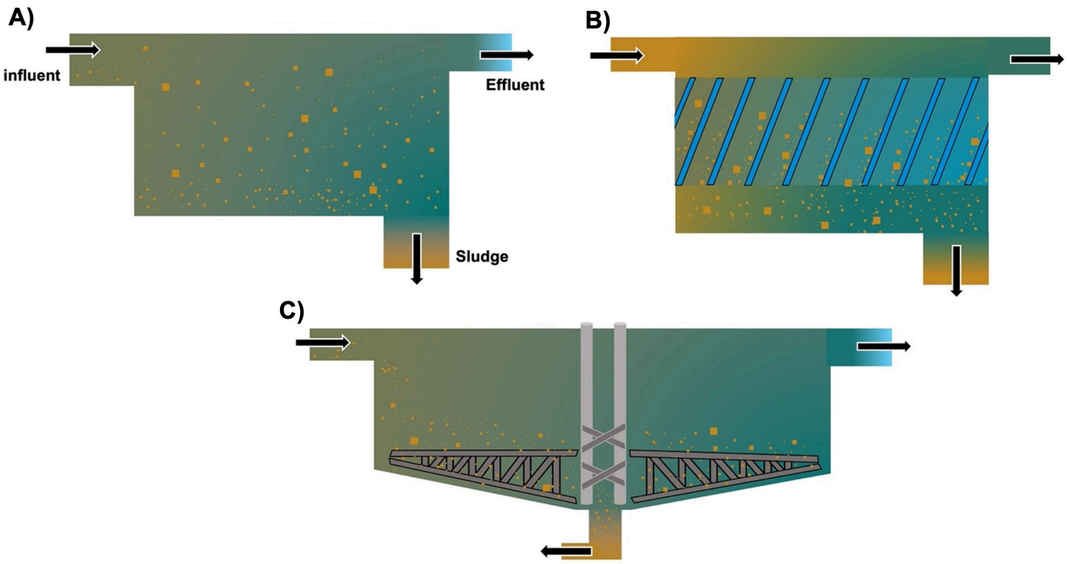

FIGURE 2: Types of Primary Sedimentation Tanks. A) Conventional (horizontal flow); B) Inclined surface (e.g. Lamella gravity settler system); and C) Circular.

2019). Across the preliminary and primary stages, a total of 50-70% of total suspended solids are removed (Qasim, 2017; Jasim, 2020).

3.3 Secondary stage

Secondary treatment applies biological processes to treat the WW (Englande et al., 2015). The purpose of secondary treatment is the removal of OM, suspended solids, reduction of biochemical oxygen demand, and removal of other compounds left from the previous stage (Vesilind, 2003; Jasim, 2020). There are two main categories of biological processes according to the microorganisms' performance mechanism: suspended- and attached-growth processes. In a suspended growth process, microorganisms are maintained in liquid suspension by mixing. In an attached growth (biofilm) process, microorganisms are attached to some type of medium, such as rock, gravel, plastics or other synthetic materials (Cohen, 2001; Metcalf & Eddy Inc., 2004). Both biological treatments sometimes occur in the same process (Metcalf & Eddy Inc., 2004).

Nitrification and denitrification are processed to remove nitrogen compounds from WW. Firstly, the nitrification process occurs when the ammoniacal nitrogen is converted to nitrate under an aerobic environment (Englande et al., 2015). Following this, the nitrate is converted to nitrogen gas, a process called denitrification (Englande et al., 2015; Frankel, 2020). During this process, bacteria degrades nitrogen products from which oxygen is released (Metcalf & Eddy Inc., 2004; Frankel, 2020). In the denitrification stage of treatment, the addition of oxygen is not required. Anaerobic treatment processes are used for treating WW with high concentrations of biodegradable organic materials. These microorganisms do not need oxygen to break down OM from the WW: the OM is decomposed to CH_4, CO_2 and other gases without oxygen (Englande et al., 2015; Ahammad and Sreekrishnan, 2016)

Under aerobic conditions, microorganisms use oxygen in the WW to decompose OM. Surface aerators and aeration diffusers are used to add oxygen to the system (Frankel, 2020). The surface aerators agitate the WW mechanically, so the surface mixes with oxygen from the atmosphere; aeration diffusers introduce air or pure oxygen in the form of bubbles into the WW from the bottom of the treatment vessel (Metcalf & Eddy Inc., 2004). There are many methods to achieve aerobic biological treatment processes, for example, activated sludge, trickling filtration, and fluidised and fixed-bed bioreactors (Yildiz, 2012; Englande et al., 2015; Frankel, 2020). In the following sections, these are explained/evaluated in detail.

3.3.1 Activated sludge

The activated sludge process (ASP) is a biological treatment in the secondary stage that usually includes aeration and clarification tanks (Yildiz, 2012) (Figure 3).This process is a suspended growth biological treatment, thoroughly mixing living microorganisms with organic compounds, using OM as a source of nutrients (Yildiz, 2012; Pennsylvania Department of Environmental Protection, 2014; Jasim, 2020).

As microorganisms are growing whilst breaking down OM, they clump together to form flocs ("flocculation"). Then flocs settle to the bottom of the tank and create an active mass known as activated sludge (Yildiz, 2012; Frankel, 2020). The mixture of activated sludge and WW in the aeration tanks (mixed liquor) flows to the final clarifier tanks where the sludge is removed. The remaining liquid is discharged as effluent. The sludge from this stage can be disposed to be separately treated or reused in the aeration tank (return activated sludge) (Chaubey, 2021). The return of activated sludge helps to maintain an adequate population of microorganisms (Pennsylvania Department of Environmental Protection, 2014; Frankel, 2020).

3.3.2 Trickling filtration

A trickling filter is an attached growth biological treatment process where the organisms are attached to a fixed bed instead of being suspended (Yildiz, 2012) (Figure 4). The effluent from the previous stage flows over a surface to which the microorganisms adhere and grow, creating a biofilm on the surface of the filter medium (Englande et al., 2015). As it is an 'open-air' system, the oxygen can be supplied from the atmosphere, or can be artificially pumped into the system (Englande et al., 2015). One characteristic is the need for continuous recirculation of the treated effluent into the influent, providing a wetting rate sufficient to keep the microorganisms active (Yildiz, 2012).

The filter medium is a bed of inert porous material inside a settling tank usually comprising stone, but it can comprise synthetic material such as glass, PVC and polyurethane foam that may offer a greater surface area and depth for microbial growth (Englande et al., 2015; Ahammad and Sreekrishnan, 2016).

3.3.3 Fluidised and Fixed Bed Bioreactors

Fixed-bed bioreactors consist of a packed bed medium wherein surface microorganisms can settle and grow, whilst the medium components remain immobile (Frankel, 2020). The influent is introduced from the bottom of the tank. For aerobic applications, air is mechanically injected into the chamber or spread across the reactor floor (Yildiz, 2012). In anoxic conditions, these reactors are used for denitrification purposes (Frankel, 2020).

Fluidised bed bioreactors use biocarriers that offer a growing surface to the microorganisms in an aerated environment. They are meant to provide a medium on which the microorganisms can grow and degrade OM (Englande et al., 2015). Biocarriers can be made of polyethylene (PE), polyurethane (PUR), polypropylene (PP), polyvinyl chloride (PVC), and high-density polyethylene (HD-PE), and their sizes range from 7 to 48 mm (di Biase et al., 2019). The hydrophobicity of these polymer carriers increases the microorganism's attachment rates (di Biase et al., 2019).

3.3.4 Membrane Bioreactors

A membrane bioreactor (MBR) is a biological reactor that uses ultra- and micro-filtration membranes to remove high levels of suspended solids (Englande et al., 2015; Frankel, 2020) (Figure 5). This process combines the activated sludge process with membrane filtration. The MBR technology enables a more efficient removal of pollutants, as it involves a relatively long retention time (Englande et al., 2015). The membrane pore size may be as small as 0.1 mm, guaranteeing a high-quality clarified effluent (Wen et al., 2010).

A MBR can be divided into three types according to their configuration: A) Submerged/immersed membrane bioreactor; B) Cross-flow membrane bioreactor; C) Hybrid membrane bioreactor (Figure 5). BOD and suspended solids removal efficiency using MBR technology is typically >90% (Wen et al., 2010).

3.4 Tertiary stage

Some WWTPs include an additional, final, stage of treatment. Tertiary treatment uses advanced techniques to improve the quality of the final effluent. The reasons for additional treatment are to protect water ecosystems and meet the required quality standards of the receiving water (DEFRA, 2012). These techniques include treatment by disinfection (chlorination, UV light, ozonation) and advanced filtration processes (Dhodapkar and Gandhi, 2019; Chaubey, 2021).

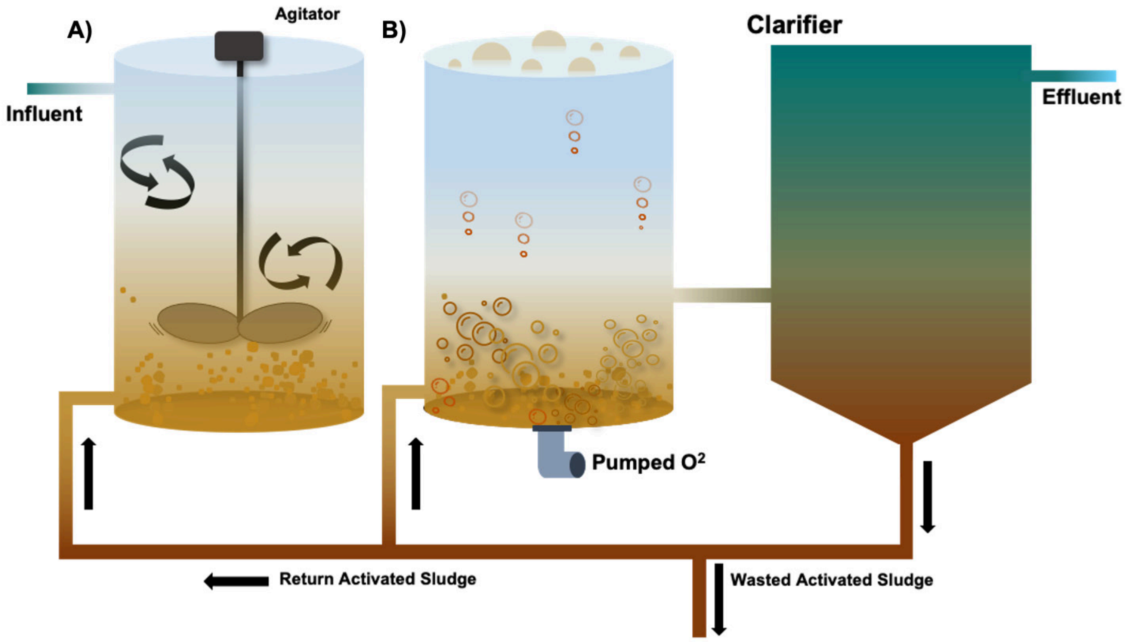

FIGURE 3: Activated sludge process scheme. A) Air mechanically added (surface aerators); B) Pure oxygen pumped into the system (aeration diffusers).

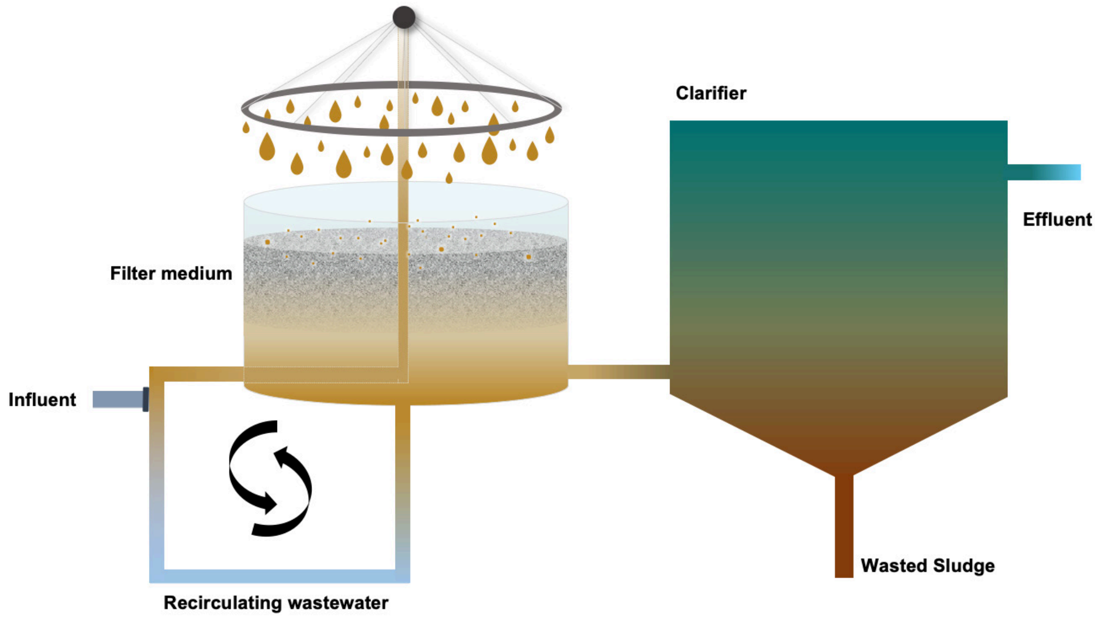

FIGURE 4: Schematic diagram of the trickling filtration process. This system consists of a settling tank, a filter medium, an influent wastewater distribution system, a clarifier and a recirculation pipe.

3.4.1 Disinfection

Chlorination, ozonation and UV light exposure are the basic mechanisms used to further disinfect WW. Chlorine is widely used for its capacity to eliminate many pathogenic microorganisms. Chlorination agents include chlorine gas, chlorine dioxide, calcium hypochlorite ($Ca(OCl)_2$) and sodium hypochlorite (NaOCl) (Stuetz and Stephenson, 2009; Collivignarelli et al., 2017). Their performance is determined by the time of exposure, dosage, and the pH and temperature of the WW (Collivignarelli et al., 2017; Chaubey, 2021). Chlorine residuals need to be removed from the final effluent as they are toxic to the aquatic environment (U.S. EPA., 1999a).

Another alternative for WW disinfection is the application of ozone (O_3). O_3 generation starts by applying a high electrical discharge in a molecule of oxygen (O_2), taken from air or high-purity oxygen, to be dissociated into oxygen atoms. Subsequently, these atoms collide with an oxygen molecule to form this unstable gas (Metcalf & Eddy Inc., 2004; Collivignarelli et al., 2017). Ozone is a strong oxidant capable of disintegrating the cell wall and membrane

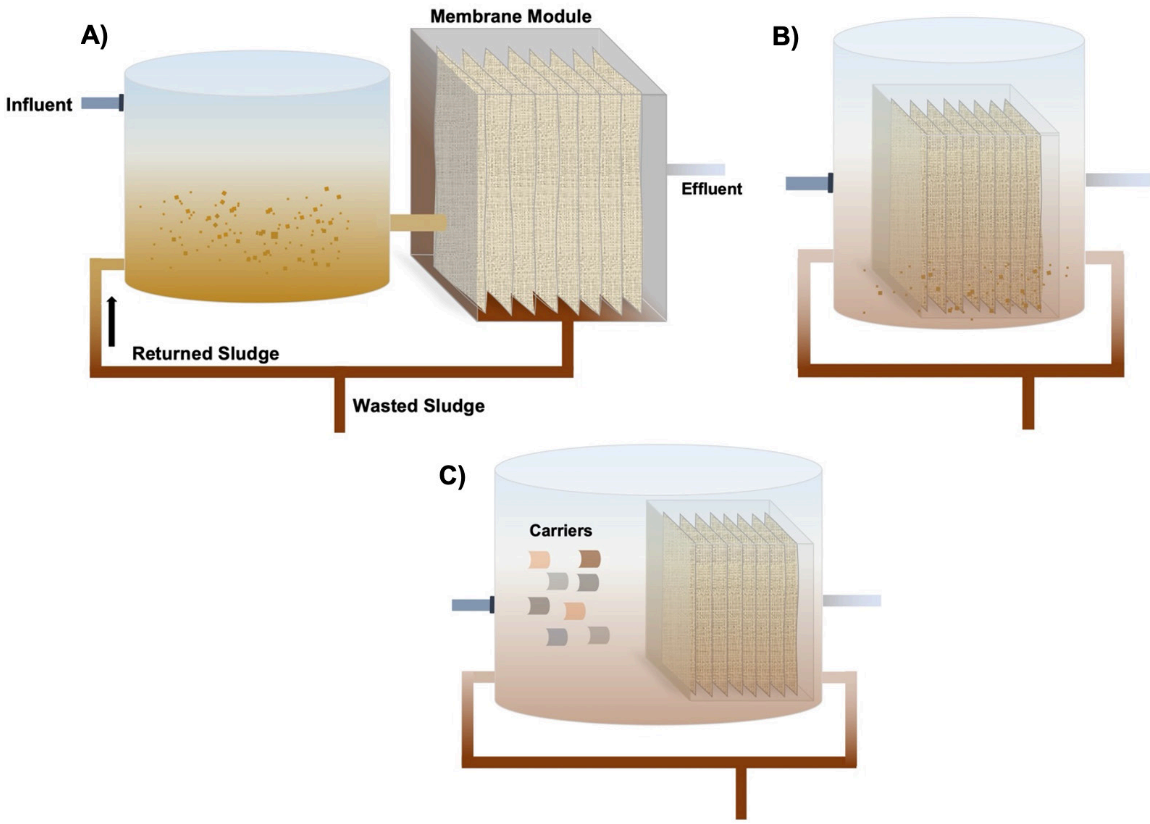

FIGURE 5: Schematic diagrams of the 3 main types of membrane bioreactors. A) Submerged membrane bioreactor; B) Cross flow membrane bioreactor; C) Hybrid membrane bioreactor. The biocarriers are polymers used in the fluidised bed reactors.

of microorganisms. However, it is not suitable for WW containing high levels of BOD and suspended solids due to its high cost (U.S. EPA., 1999b).

Ultraviolet (UV) radiation is a form of electromagnetic energy covering the wavelength range of 100–400 nm, with the optimum germicidal fraction between 220 and 320 nm (USEPA., 1999c; Collivignarelli et al., 2017). Disinfection by UV radiation is a physical process effective against most viruses, cysts and spores as it destroys the genetic material of microorganisms, preventing further reproduction. UV radiation has little effect on nitrogen compounds or ammonia, and some factors such as turbidity and suspended solids can decrease the performance of UV disinfection (USEPA., 1999d).

3.4.2 Advanced filtration systems

Advanced filtration is a process where WW flows through a permeable layer. The materials include a filter paper, a membrane, a sieve or other suitable porous medium (Beverly, 2011). Filtration removes substantial quantities of solids from wastewater, promoting effective disinfection.

Disc-filter technology uses fabric membranes instead of a granular medium to filter the effluent. WW enters thorough a channel and flows through the filter. This filter cloth is made of either polyester (PES) or stainless steel (Metcalf & Eddy Inc., 2004). This technique employs a rotary system that operates in an intermittent or a continuous-backwash mode. The particles retained are thereby removed from the screen (Metcalf & Eddy Inc., 2004).

Granular filtration WW filters WW through materials such as sand, anthracite grains or gravel (Bui et al., 2020). During the flow process, suspended solids are retained, and some of them are biochemically decomposed and removed later (Arndt and Eric, 2004; Beverly, 2005). There are two types of sand filtration: 1) slow (known as gravity) sand- and 2) rapid sand- filtration. Slow sand filtration uses gravity-fed flows, whilst rapid sand filtration uses either pumping or head pressure (Arndt and Eric, 2004).

In order to conduct projects related to WWTPs, it is crucial to acknowledge the design and function of these facilities and understand, in this case, the performance of MPs throughout the treatment system.

4. IMPACTS OF WASTEWATER TREATMENT STAGES ON MICROPLASTICS CONCENTRATIONS

4.1 Effects on microplastics throughout treatment process

The numbers of MPs in the influent and effluent fluctuate in each stage and system, leading to an overall decrease throughout the treatment process. It is during the preliminary and primary stages where most of the MPs are removed via particle skimming and sludge settling processes (Carr et al., 2016; Michielssen et al., 2016; Murphy et al., 2016; Qasim, 2017; Talvitie et al., 2017; Lares et al., 2018; Gies et al., 2018; Magni et al., 2019; Bui et al., 2020). Depending on the plastic size and density (Table 1), mate-

TABLE 1: Density range of some plastics.

Polymer	Density (g/cm^3)	Reference
PE-LD	0.94	
PE-HD	0.97	
PLA	1.24	
PS	0.01-1.50	
PUR	0.08-1.03	
CR	0.11-1.25	
PP	0.9-1.27	Crawford and Quinn, 2017a
ABS	1.00-1.21	
PMMA	1.10-1.25	
PA	1.12-1.38	
PVC	1.15-1.50	
PET	1.30-1.50	
PC	1.40-1.50	
Cellophane	1.40–1.53	Gago et al., 2018
Rayon	1.53	
PTFE (Teflon™)	2.10-2.30	Crawford and Quinn, 2017a

ABS: Acrylonitrile butadiene styrene; CR: Polychloroprene (neoprene) PA: Polyamide; PC: Polycarbonate; PE-LD: Low Density Polyethylene; PE-HD: High Density Polyethylene; PET: Polyethylene Terephthalate; PLA: Polylactic Acid; PMMA: Polymethyl Methacrylate; PP: Polypropylene; PS: Polystyrene; PTFE: Polytetrafluoroethylene; PUR: Polyurethane; PVC: Polyvinyl Chloride

rial attached to the microplastic surface, WW density, and the WW motion produced by the system, the MPs might be retained and removed by skimming of fats, oil and grease if they are found in the surface; or in the grit and solids settled at the bottom of the tanks. For example, the density of the raw wastewater is 1.0 g/cm^3, and for the microbial biomass in a biological reactor can vary from ~1.02 to 1.05 g/cm^3, thus the final density of the WW might be altered by the biomass present in the system (Schuler and Jang, 2007; Xu et al., 2014). Furthermore, this parameter may vary according to the stage at which the composition of the WW is being quantified. The density of the WW and its components, in addition to the density of the particle itself, influence the fate of the MPs. Neutrally buoyant MPs are more likely to escape both skimming and settling processes (Carr et al., 2016).

Bayo et al., (2020) reported that ~74% of MPs were retained during the primary stage in a WWTP located in Spain; similar results were observed by Rasmussen et al., (2021) in Denmark. Gies et al. (2018) found that most of the MPs removed in these stages were fibres. The latter was also observed by Hidayaturrahman and Lee (2019) and Blair et al. (2019), as they found that MP fragments are removed in the next treatment stages. Talvitie et al. (2017) found that ~95% of MPs were eliminated by DAF, although MPs concentrations were relatively low. Fibres in the influent are dominant over other MPs, and usually are retained during the primary stage, possibly because these fibres become easily attached to flocs. However, Tadsuwan and Babel (2022) found a large portion of fibres in the effluent from the secondary stage: in this case the WWTP studied did not have a primary sedimentation stage. From the previous examples, it is demonstrated that the performance and availability of the primary stage is a key factor in the removal of MPs from WW.

Biomass accumulation onto MPs surfaces, i.e. biofouling, can increase the density of the particles, causing them to sink (Kaiser et al., 2017). Biofouling might completely encapsulate these MPs and affect their properties along the treatment process, with small MPs the most affected (Carr et al., 2016; Lee and Kim, 2018). As Yu et al., (2016) explained, the solid retention time (SRT) is a crucial parameter that affects biofouling of a MBR as it can modify biomass behaviour. Therefore, it is expected that the SRT has an influence on the MPs, alongside the hydraulic retention time (HRT). It is important to highlight the differences between the SRT and HRT. The SRT indicates the average period the bacteria last in the reactor, whilst the HRT indicates the time the wastewater remains in the treatment tank or unit (Clara et al., 2005; Eggen and Vogelsang, 2015). According to Tadsuwan and Babel, (2022), sedimentation of larger MPs is influenced by longer SRT during the conventional ASP. Further studies are needed to understand more fully how SRT and HRT may modify the characteristics and fate of the MPs in the WWTPs.

Biological treatments, usually a secondary treatment stage, strongly influence the concentration of MPs in a WWTP (Hidayaturrahman and Lee, 2019; Bui et al., 2020). In secondary settlement tanks, MPs may adhere to suspended solids that settle and become trapped in the sludge (Bui et al., 2020). Murphy et al., (2016) and Blair et al., (2019) reported MPs removal efficiencies from the activated sludge process of approximately 93% and 98%, respectively. Magni et al. (2019) reported a concentration decrease of ~50% in MPs from the secondary to the final effluent, contributing to an overall 84% removal efficiency. Ziajahromi et al., (2017) detected in one of three selected WWTPs a decrease from the primary effluent of 66% after the secondary process, which includes secondary aeration and sedimentation. Hidayaturrahman and Lee, (2019) evaluated three different WWTPs and concluded an average removal of MPs after secondary treatment of ~58%.

The combination of different technologies to achieve a better effluent quality, alongside tertiary processes, has been studied. One study has compared two WWTPs with different treatment systems (Lv et al., 2019). The first system employed aerated grit chambers, oxidation ditch, secondary settling tank and a UV disinfection chamber; the second system used rotary grit chambers and a A^{2}O (Anaerobic-Anoxic-Aerobic)-ASP unit combined with a Membrane Bioreactor. The removal efficiency results were as follows: MBR (~84%), >secondary settling tank (~77%) > oxidation ditch (~17%) > A^{2}O unit (15%). It was found that the A^{2}O-ASP MBR-based unit traps MPs in the sludge and removes them via membrane filtration (Lv et al., 2019). According to Peeva and Livingston (2019), the addition of a membrane module within the ASP reactor increases the clarification and quality of the treated water. Although these results showed efficient removal of MPs from WW, to combine different technologies to further decrease MPs concentration needs further attention as outcomes may not be as expected.

Talvitie et al., (2015) concluded that a biologically active filter as part of a tertiary treatment does not decrease MPs concentration. Likewise, Michielssen et al. (2016), by comparing secondary and tertiary WWTPs, and analysing a novel anaerobic MBR pilot-scale added to the tertiary treatment plant, found that the integration of a second process in the secondary stage does not remove more of the of MPs. However, the addition of a MBR outperformed MP removal rates from both the WWTPs studied Michielssen et al., (2016). The secondary plant applied two secondary treatment processes in series, a trickling filter followed by an ASP; and the tertiary stage included slow sand filtration within its process (Michielssen et al., 2016).

Wastewater treatment facilities implementing MBR technology might promote higher removal of MPs from WW. Lares et al., (2018) found that MBR technology is more efficient in removing MPs compared with ASP technology. Talvitie et al., (2017) compared four different tertiary technologies, rapid sand filtration (with removal efficiency of 97%), DAF (~95), disc filter (40-98%), and MBR. They concluded that MBR is the most efficient technology of those studied as they detected a high removal rate of ~99% from MBR grab and composite samples; the MBR had the smallest pore size (0.4 μm) of these three filtration methods. In contrast, Hu et al., (2022) found that for >40 WWTPs in three different regions in Shaanxi Province, China, MBR does not efficiently remove MPs from WW. The removal efficiency of these WWTPs was more related to differences in the characteristics of the MPs (shape and size) across regions.

Within the tertiary stage, some advanced filtration methods have been studied. Mintenig et al., (2017) studied the effluents from 12 WWTPs located in Germany, and one of them was a tertiary treatment plant with an additional post-filtration system installed. They found that the use of a filter unit provides for significant MP removal. In contrast, a study of 17 WWTPs in the U.S.A. concluded that the presence of an advanced filtration phase was not effective in terms of reducing MPs concentrations (Mason et al., 2016). Although a polymeric identification analysis was not performed, it was deduced that the usage of filters might promote microbial growth and discovered that these microorganisms might be misled as "fibres particles". The efficiency of the disc filter will depend on the pore size of filters, according to Talvitie et al., (2017), who reported a higher removal efficiency from 20 μm filters (~98%) than 10 μm filters (40%). Although the opposite result was expected, they assumed that this outcome might be due to membrane fouling by excessive flocculant polymers adhered to it. Hidayaturrahman and Lee (2019) stated a 79% MPs removal by a disk-shaped filter with 10-μm pore size. Within the granular filtration field, sand filtration is one of the most studied treatments. Slow sand filtration appears not to be the most suitable method for the removal of MPs, according to Wang et al. (2020), who only detected ~37% removal efficiency. These authors also discovered that beads/pellets and fibres were more efficiently retained than fragment-shaped MPs. A study in the U.S.A. involved a gravity-bed filter simulator which consisted of a column made of anthracite, sand and gravel (Carr et al., 2016). Although the filters contained high concentrations of anthracite fragments, the authors confirmed the efficiency of the gravity sand filtration as it successfully retained microbeads from 300μm to 10μm in size.

Hidayaturrahman and Lee (2019), by comparing different tertiary treatment technologies, determined that rapid sand filtration had the lowest removal rate (74%) compared with membrane disc filter (79%) and ozone (90%) technologies. They also concluded that MPs in WW might get trapped between the grains (medium-sized particles) or adhere to the surface of the grains, whilst the treated water is discharged to the bottom (Hidayaturrahman and Lee, 2019). Another study reported a high removal rate by advanced sand filters, however, was not high enough to compete with the efficiency of the MBR technology (Blair et al., 2019). Mason et al. (2016) stated that, for the removal of microbeads, rapid sand filters are not the most recommended technology to use.

In Spain, a WWTP removed ~93% of MPs from WW during a process that includes coagulation-flocculation tanks, lamellar decanter, rapid sand filter and UV disinfection system, according to Menéndez-Manjón et al., (2022). Ziajahromi et al. (2017) detected a 90% MPs removal efficiency during advanced treatment processes, such as flocculation, disinfection, ultrafiltration, and reverse osmosis. Particles from 1000 μm to 500 μm might be fully removed during ultrafiltration system (Tadsuwan and Babel, 2022).

In summary, the composition and concentration of MPs in effluent involves a variety of factors such as the type of wastewater (i.e. presence of combined sewers), loading, and the general treatment processes on site, particularly if tertiary treatment techniques are included (Mason et al., 2016; Gündoğdu et al., 2018; Blair et al., 2019). After the removal of MPs from WW throughout the treatment process, these are concentrated in the sludge.

Figure 6 summarises the stages of the treatment process, the accumulation removal rates of MPs and the factors involved in the MPs removal. However, it is important to highlight the necessity of a consensus regarding the terms used for the sampling points and avoid misunderstandings. For instance, the WW sampled after the bar screening is sometimes referred as the raw WW, when in fact it is the effluent from the screening stage. The first incoming water, the 'crude, is the raw WW. The use of a to-scale, fully labelled diagram illustrating exactly where the samples were collected is recommended.

4.2 Contribution of WWTPs to microplastic pollution

WWTPs might influence the release of microplastics or physically degrade the plastic particles into smaller pieces. In a WWTP located in Turkey, an increase in PES particles from the influent (62%) to the effluent (~69%) was detected, denoting a PES source within the treatment process (Gündoğdu et al., 2018). However, some authors found similar results but gave alternative explanations. For example, in Italy, Magni et al. (2019) found epoxy resin, PVC and styrene-isoprene-styrene MPs only in the effluent; concentrations of polyacrylate, polyamide (PA) and polyester

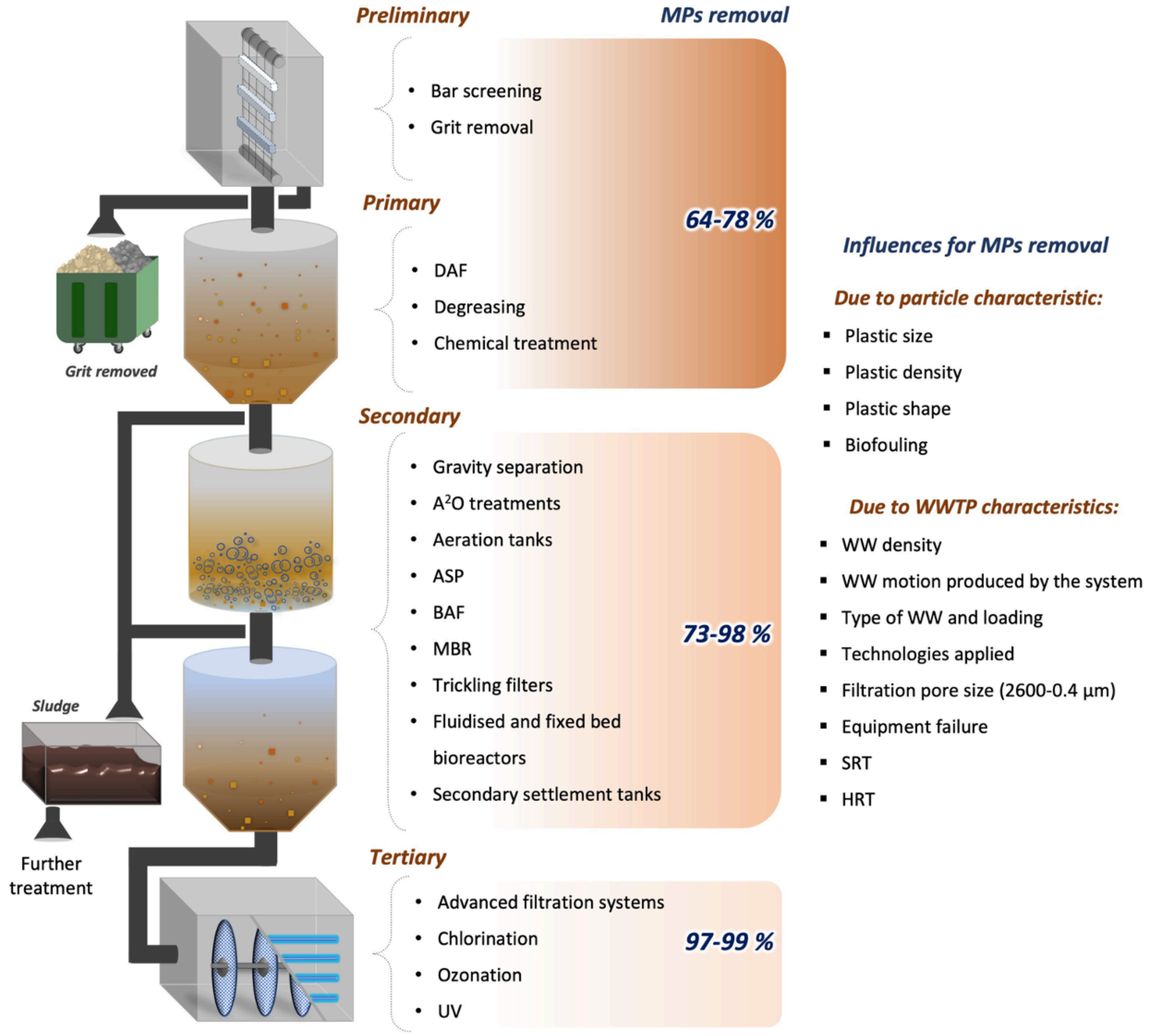

FIGURE 6: Schematic diagram of the processes included in a wastewater treatment plant, alongside the estimated microplastic removal throughout the system and the factors causing such effect.

(PES) particles increased by 7%, 17% and 35%, respectively. These particles may come from paints, adhesive agents and personal care and cosmetics products (PCCPs). Lv et al., (2019) found that the samples from the rotary grit chambers have higher MPs concentration compared with the aerated grit chambers. They assumed that mechanical stirring from the rotary grit chambers further disintegrate the microplastics into more, and smaller fragments. Furthermore, the same authors presumed that air introduction into the aeration systems can influence the MPs to breakdown due to air friction forces, and that the disinfection treatment by UV light, although it enhances MPs removal, it can further decompose the particles to smaller sizes and weakened properties of MPs (Bao et al., 2022).

Within a WWTP, some elements can be a potential direct source of microplastics. The filter medium in trickling filtration is usually made of stone, although they can be replaced by synthetic materials, such as PVC, as it provides a larger surface for growth of microorganisms (Vayenas, 2011). Furthermore, bio-beads, round-shaped particles, sometimes are used as filtering medium in certain treatment facilities, such as for industrial WW (Turner et al., 2019). For the MBR system, Talvitie et al. (2017) reported that MPs might be derived by the occasional breakthrough of the filters, and from the membrane cartridges made of polyvinylidene fluoride (Lv et al. 2019). High numbers of PA particles were detected after filtration from an advanced tertiary treatment system, and it is presumed that they come from the pile fabric itself (Mintenig et al., 2017). The filter materials might vary from PES, PA and PE (Mintenig et al., 2017).

According to Chandrasekaran (2010), rubber is any easy target for ozone, as it can react with the polymer molecules and break the polymer chain, leaving it prone to cracking. However, in a study at a drinking water treatment plant, researchers found that ozonation degrades larger MPs to smaller sizes, which can increase the number of MPs in the effluent (Wang et al., 2020). Fitri et al., (2021) revealed that the ozonation process can impact the chemical structure, in this case of the PE (841 μm in size), influenced by the environment pH, contact time and ozone dosage variation. Li et al., (2022) tested the deterioration of PS plastics by chlorination and ozonation processes, discovering that O_3 is more effective to further breakdown the plastic particles. However, even if the previous study was performed under laboratory-controlled condition using drinking wastewater instead, it can be concluded that disinfection technologies do not eliminate MPs but create smaller particles, including nanoplastics. Further research is needed when it comes to study the disinfection methods in the sewage treatment process with different types and sizes of MPs.

WW treatment facilities can be a source of MPs to effluent, and not only by the purification technologies applied, but also by the degradation of plastic equipment over time (Blair et al., 2019). Therefore, regular maintenance of the treatment facilities might reduce the MPs flow to discharge (Lv et al., 2019). From another point of view, there are other factors that might show an increase of MPs or lead to the detection of new plastic particles from the influent to the effluent, for example, wear and use of synthetics fabrics (e.g. clothing made out PES) when sampling or when experiments are conducted in a laboratory. We emphasise the importance of using blank and control samples to detect any irregularity and correct results. Over- and under-counting the number of particles due to human error (e.g. lack of expertise or fatigue) are other reasons for likely miscalculated values.

4.3 Microplastics in Effluent and Sewage Sludge

The presence of MPs in the final effluent has been reported globally, despite the previous sanitisation process the WW has received. Reported removal efficiencies vary from 73% to 99%, yet a complete removal has not been detected. Information about previous investigations is shown in Table S1 in the supplementary material.

Table S1 also exposes the differences in terms of the sampling points (e.g. raw WW), collection period techniques (grab and composite samples) and duration of sampling campaign, that goes from 1 day to 1 year. Grab sampling consists of one single collection point in a certain time, whilst the composite samples are collected at fixed intervals over an extended period of time. According to Conley et al., (2019), grab sampling is a good technique only for a specific or short period of time, but it may not provide a representative scenario of the wastewater stream. Seasonal variation is a particularly important factor regarding MPs in WWTPs. For example, increases of MPs during warmer periods have been observed, due to either high evaporation of water affecting its dilution, plastic fragmentation due to sunlight radiation or clothing preferences and higher washing frequency (Bayo et al., 2020; Bao et al., 2022; Jiang et al., 2022; Menéndez-Manjón et al., 2022).

Magnusson and Norén (2014), in Sweden, calculated an inflow of 3.25 x10^6 MPs but an outflow of less than 2,000 MPs each per hour based on a single day of sampling, whilst Alavian Petroody et al. (2020), in Iran, reported a total emission of ~ 9 million MPs per day, with a 24-hour composite sampling during a week. A study testing one treatment plant in Spain through a monthly sampling campaign in a year, revealed that ~0.7 MPs/L are released via the effluent (Menéndez-Manjón et al., 2022). However, the minimum size limit they analysed was 100 μm, probably significantly underestimating the total number of MPs in smaller size ranges. The selection criteria to classify MPs in WWTPs according to their sizes must be justified in order to reach an agreement. For example, Lee and Kim (2018) selected 106 μm and 300 μm considering the average value of microzooplankton, plankton and floating waste. Table 2 highlights the different size filters used in the study of MPs in WWTPs.

It is acknowledged that the selection of the different techniques to conduct a study regarding the presence of MPs in WWTPs occurs according to the aims/objectives, limitations and resources available. Nevertheless, the lack of consensus in respect of how the results are reported makes it difficult to compare data across studies. For example, in Thailand, a study testing two WWTPs reported 16.55-77 MPs/L release via the effluent, but in the absence of the outflow data is not possible to modify the units to MPs per day (Tadsuwan and Babel, 2022). Inflow and outflow fluctuations happen hourly and seasonally, therefore, such information must be added to get better quality information about the number of MPs entering the WWTPs and released via the final effluent. Flow variation fluctuates according to the geographical location and the area served, and might determine the pollutant load (Vesilind, 2003). The different sampling techniques are not further discussed in this paper.

The quantity of microplastics are usually published in particles per L/day, however, it can also be calculated as particle mass, as Simon et al., (2018) reported for their final treated outflow, and Rasmussen et al., (2021) for their sampled WW and sludge. The mass measurement can describe the presence of a certain polymer less affected by size ranges and might be more helpful for analytical procedures. However, using particles as a unit might allow the production of information for projects with technological and financial difficulties, as reported by Gimiliani and Izar (2022). We suggest that the number of particles (L/day or kg/day) should be consistently used as units based on the flow rate data from the sampling dates or studies should obtain the annual average flow rate to estimate an annual value.

There are other key factors affecting MPs released to the environment alongside the on-site engineering: the degree of urbanisation surrounding the facilities, economic level, lifestyle and habits of the public, current population served, tourism activities, type of receiving water (e.g. domestic vs. industrial WW), amount of water resources ruled by weather conditions and season, strongly influence the quality and composition of WW (Mason et al., 2016; Murphy et al., 2016; Sutton et al., 2016; Gündoğdu et al., 2018; Lares et al., 2018; Lee and Kim, 2018; Alavian Petroody et al., 2020; Ben-David et al., 2021; Hu et al., 2022; Jiang et al., 2022). In China, even though Hu et al., (2022) did not analyse the type of polymers of the detected MPs, they concluded that the more developed the region, the fewer

TABLE 2: Collection methodology and size categories selected per study.

Number of WWTPs	WW	SS	Type of Sampling	Sampling Duration	Pore filter size (µm)	Reference
3	X		Grab	Monthly, 1 year,	26	Akarsu et al., 2019
1	X		Composite (24 h, every hour)	1 week	500-300-37	Alavian Petroody et al., 2020
1	X	X	Composite (24 h)	2 months, 3 days each	380-180-75-25	Bao et al., 2022
1	X		Grab	5 days within 9 months	2800	Blair et al., 2019
1	X		Composite (24 h)	5 months	5000-1000-355-125	Dyachenko et al., 2017
2	X		Composite (24 h)	5 days	55	Gündoğdu et al., 2018
3	X		Grab	3 days within 2 months	-	Hidayaturrahman & Lee, 2019
3	X	X	Grab	3 months	4750-330	Hongprasith et al., 2020
48	X		Grab	3 days	75	Hu et al., 2022
5	X		Composite (24 h)	2 months	-	Jiang et al., 2022
3	X	X	Grab	2 days within 3 months	300-106	Lee & Kim, 2018
1	X	X	Composite (24 h)	7 days	1000	Lofty et al., 2022
1	X	X	Grab	Once	500-250-125-62.5-25	Lv et al., 2019
1	X	X	Grab	Monthly, 1 year,	500-250-100-20	Menendez-Manjon et al., 2022
2	X		Grab	4 days within 3 months	4750-850-300-106-20	Michielssen et al., 2016
1	X	X	Composite (24 h)	5 days within 5 months	500-20-10	Rasmussen et al., 2021
2	X		Grab	Monthly, 1 year	500-10	Roscher et al., 2022
2	X	X	Grab	1 day	1000-500-50	Tadsuwan & Babel ,2022

The X marks if wastewater (WW) or solid/sludge (SS) samples were collected. The sorting of microplastics by sizes were performed on site or during the laboratory procedures. Information about the WW and SS sampling points is shown in Table S1.

fibres, more foams and more microbeads are found. They concluded that the higher the economic level in the region, alongside the shortage of water resources, a higher number of MPs arrive to the WWTPs. Conversely, Mason et al. (2016) suggested that the number of MP fragments in WW are not associated with the population served by the treatment plant, but there is a direct relationship between the population and the number of fibres. The latter is based on the usage of washing machines (Blair et al., 2019). These fibres might escape into the effluent due to their morphology, which allows them to pass longitudinally through filtration treatments (Ziajahromi et al., 2017).

Alavian Petroody et al., (2020) discovered the presence of microbeads made of PE that are likely to come from PCCPs. This study was in Iran, a country with no current regulations to control or avoid microbeads from PCCPs. Michielssen et al. (2016) found that microbeads were effectively removed during preliminary, primary and the secondary treatment processes. However, some authors did not observe any microbeads in the final effluent but only in the sludge from the 'grit and grease' stage (Magnusson and Norén, 2014; Murphy et al., 2016), or in any other stage (Lv et al., 2019). For tyre particles, it is predicted that ~85% are removed during the primary stage; however, this depends on the load of particles arriving at the WWTP (Sundt et. al., 2014).

The final treated effluent can be used for groundwater recharge and, most commonly, as an indirect water supply through discharge into rivers, lakes and estuaries (Qasim, 2017). It can be reused for irrigation of highways, city parks and farmlands. In some countries treated WW is used for agriculture and forestry irrigation (Table 3) (FAO, 2017). However, the outflows from WWTPs cause concern because of the likely content of microorganisms, nutrients and toxic components, such as MPs, and their consequent effects on the environment. Therefore, the effluent has been studied as a source of MPs to rivers, lakes and oceans.

Sewage sludge is the biosolid fraction generated throughout the WW treatment process. The sludge is further processed in a sewage sludge treatment plant, following these steps: 1) preliminary treatment, 2) primary

TABLE 3: Direct use of treated WW for irrigation purposes.

Country	Irrigated water (x10^{9}m^3/annual)
Australia	0.14
Brazil	0.008
Chile	0.138
China	1.26
Egypt	0.29
Greece	0.069
Iran	0.328
Italy	0.087
Japan	0.0116
Mexico	0.401
Morocco	0.002
Peru	0.114
South Africa	0.006

Source: FAO, 2017

thickening, 3) liquid sludge stabilization, 4) secondary thickening, 5) conditioning, 6) dewatering, 7) final treatment (Kacprzak et al., 2017). This by-product is a mixture of inorganic and organic matter that contains nutrients and helps to improve soil properties and enhance agricultural crops (Singh and Agrawal, 2008; Abdul Khaliq et al., 2017; Fijalkowski et al., 2017). This material is also used for energy recovery purposes from the anaerobic digestion, pyrolysis and gasification processes (Đurđević et al., 2019). Landfill is another option to discard the sludge coming from the treatment plants (Yang et al., 2015).

However, the incineration of dewatered sludge and landfilling are of great concern due to their potential for air and groundwater pollution (Qasim, 2017). Furthermore, as most MPs from WW are partitioned to the sewage sludge, this product might represent a direct source of high concentrations of MPs to soil if it is applied as fertiliser (Fijalkowski et al., 2017; Hurley and Nizzetto, 2018). In Denmark, 70% of dewatered sludge is used for agricultural purposes and the rest is destined for energy recovery purposes (Simon et al., 2018). In North America and Europe, it is estimated that there is an annual input of 63,000–430,000 and 44,000–300,000 tonnes of MPs from the sludge to farmlands, respectively (Nizzetto et al., 2016). South Korea and Switzerland are examples of countries that have banned biosolids usage for agricultural fields, diverting the by-product instead to incineration for energy recovery (Outotec, 2016; Rolsky et al., 2020).

Mahon et al., (2017) analysed sludge samples from anaerobic digestion, thermal drying and lime stabilization processes for seven WWTPs, reporting an average abundance of MPs of 10,000 particles per kg of sludge. They discovered that a lime stabilisation process breaks the MPs into smaller pieces. A study evaluating 12 WWTPs in China concluded that the sludge retained was ~ 23,000 particles per kg dry sludge (Li et al., 2018). Leslie et al., (2017) selected two WWTPs to assess the content of MPs from the influent, effluent and sludge, and they found that the content of MPs in the sludge was ~ 630 particles per kg wet weight. Magni et al. (2019) concluded that 1 tonne dry weight of sludge from an Italian WWTP can transfer ~113 million plastic microparticles to the soil. Nevertheless, the sludge pointed to as a possible source of MPs to soil is sometimes the pre-treated solid that, depending on prevailing legislation, still has to undergo additional treatment to be used in the environment. MPs in this biosolid might be further modified or removed. More studies are needed in the treated sludge used as fertiliser and not from WWTPs directly.

Various reports concurred that fibres are the dominant form of MPs detected in the WWTP and tend to be more fully retained in the sewage sludge (Magnusson and Norén, 2014; Gewert et al., 2017; Ziajahromi et al., 2017; Gies et al., 2018; Lee and Kim, 2018; Corradini et al., 2019; Lares et al., 2019; Yang et al., 2019). Some of the most common polymer found are PES, PP and polyethylene terephthalate, which arise predominately from washing, laundry and industrial processes (Browne et al., 2011; Murphy et al., 2016; Magni et al., 2019). The presence of MPs has become an indicator of previous sewage sludge applications on land (Zubris and Richards, 2005; Corradini et al., 2019). Sometimes, microplastics might never leave the treatment facilities, as they could sink and be retained during the process. For example, Rasmussen et al., (2021) detected that ~ 12% of MPs was missing from the plastic (based on a mass balance calculation), due to sampling and measuring uncertainties. However, these MPs also might get trapped in the digested and activated sludge tanks and remain unaccounted. Many factors are thus involved with the behaviour and release of MPs to either effluent or sludge.

5. CONCLUSIONS

The impact of WW treatment stages and the existing technologies applied, have been critically discussed and reviewed in terms of removal of microplastics. Every facility has a specific design, performance, characteristics, and thus should be assessed accordingly. The removal of MPs from different stages in WWTPs depends upon a range of factors, for example, the STR and HRT, which might interfere with the integrity of the plastic particles. In general, most MPs (up to 78%) are removed in the preliminary and primary stages of WWTPs. The reasons for this are that the particles found in oil and grease on the surface are skimmed out, whilst the ones located in the grit and solids settle at the bottom of the tanks and get removed. The secondary stage is the second most important in terms of MP removal at WWTPs as when the process is complete it eliminates up to 99% of MPs found in the raw WW, with variations in removal rate according to technology. MBR in particular has been shown as an effective system to remove MPs from WW. The combination of different technologies during advanced treatment process, i.e. tertiary stage, has revealed better removal performance, however, it has been suggested that sand filtration technologies alone are the less efficient to eliminate MPs. Additional research to evaluate the function of the systems applied during the tertiary stages is required.

Only a small fraction of the MPs entering a WWTP leave via the final effluent, down to 1% from total in the raw WW, therefore, MPs typically end up being concentrated in the sludge. Their environmental fate depends upon whether the effluent is discharged back into fresh/marine watercourses or used for e.g. irrigation purposes, or they are spread on land, incinerated or landfilled if MPs are found in sludge. MPs may create a great impact in the environment, although their concentration levels of concern are still unclear. The sewage sludge must be treated to be safely used on grassland, therefore, the investigations aimed to the sludge directly from the WW treatment process might not report a proper estimation of the occurrence and appearance of MPs found in the biosolid that is spread in agricultural soil.

Although it is confirmed the presence of MPs in wastewater and sewage sludge, the comparison of data from global studies is complicated as researchers use a variety of means to report the quantity of MPs and to denominate the different WW treatment stages. If mass balance and number of particles are considered, kg/day and L/day are the units we recommend for reporting purposes, according to the flow rate data from the time of the sampling campaign. If MPs are classified based on their size, the size selection criteria must be justified to ease the comparison data to other studies and work on a consensus. We recommend that the incoming water to the WWTPs should be referred as raw WW, crude sewage, or influent, and to avoid calling "influent" the WW that already has started the treatment process, for instance, the bar screening/grit chamber effluent. Furthermore, if the WW of interest is coming from a certain system, the sampling point label should add the term "effluent", not considering the following step. A to-scale, fully labelled diagram illustrating exactly where the samples were collected is advised. Overall, this work aims to provide an insight in the field of MPs and WWTPs together, highlighting some gaps where further investigation is required.

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