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Microplastics in wastewater treatment plants: Sources, properties, removal efficiency, removal mechanisms, and interactions with pollutants

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ABSTRACT

Since wastewater treatment plants (WWTPs) cannot completely remove microplastics (MPs) from wastewater, WWTPs are responsible for the release of millions of MPs into the environment even in 1 day. Therefore, knowing the sources, properties, removal efficiencies and removal mechanisms of MPs in WWTPs is of great importance for the management of MPs. In this paper, firstly the sources of MPs in WWTPs and the quantities and properties (polymer type, shape, size, and color) of MPs in influents, effluents, and sludges of WWTPs are presented. Following this, the MP removal efficiency of different treatment units (primary settling, flotation, biological treatment, secondary settling, filtration-based treatment technologies, and coagulation) in WWTPs is discussed. In the next section, details about MP removal mechanisms in critical treatment units (settling and flotation tanks, bioreactors, sand filters, membrane filters, and coagulation units) in WWTPs are given. In the last section, the mechanisms and factors that are effective in adsorbing organic–inorganic pollutants in wastewater to MPs are presented. Finally, the current situation and research gap in these areas are identified and suggestions are provided for topics that need further research in the future.

Key words: mechanisms, microplastic, pollutant adsorption, sludge, wastewater, wastewater treatment plant

HIGHLIGHTS

- Millions of microplastics (MPs) are released into the environment through the effluent and sludge of wastewater treatment plants (WWTPs).
- MPs removal by primary and secondary treatments is limited in WWTPs.
- Tertiary treatment technologies need to be combined with primary and secondary treatment technologies for MP removal with higher efficiency in WWTPs.

1. INTRODUCTION

Plastic pollution is a global environmental problem that is getting more and more terrifying every day. A total of 9.2 billion tons of plastic was produced between 1950 and 2017, and only less than 10% has been recycled so far (Plastic Atlas 2020). Since plastics are resistant to biodegradation, their presence in the environment poses a significant problem when not recycled (Nkwachukwu *et al.* 2013). Figure 1(a) shows the global plastic production from 1950 to 2020 and the percentage distribution of global plastic production in 2020. Plastic production in the world reached 367 million tons in 2020, and Asia (China 32%, Japan 4%, Rest of Asia 17%) contributed more than half (53%) of this amount. North American Free Trade Agreement (NAFTA) 19%, Europe 15%, Middle East, Africa 7%, Latin America 4%, and the Commonwealth of Independent States (CIS) 3% of global plastics production (Plastics Europe 2021) (Figure 1(b)).

The lightness, flexibility, low cost, and durability of plastics have made them widely used in many fields (Chatterjee & Sharma 2019). The high consumption of plastic polymers, their low recycling rate, and their resistance to degradability make plastics a persistent pollutant in the environment. Polyethylene (PE), polypropylene (PP), polystyrene (PS), polyethylene terephthalate (PET), and polyvinyl chloride (PVC) are the most widely used plastic types. Table 1 shows the density, lifespan, and areas of use of commonly used plastics.

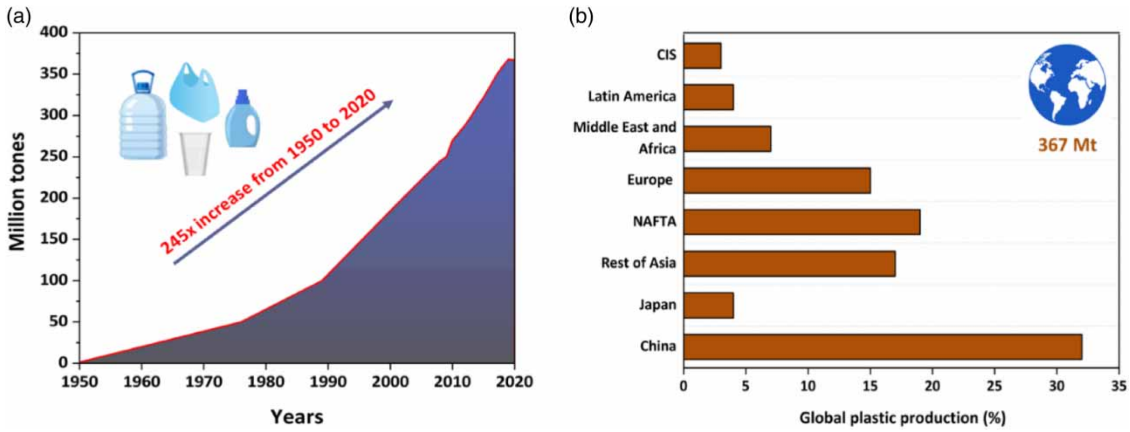

Figure 1 | (a) Global plastic production between 1950 and 2020. (b) Distribution of global plastic production (Adapted from Plastics Europe 2021).

Table 1 | Density, lifespan, and usage areas of commonly used plastics

	Density (g/cm^3) (Quinn *et al.* 2017)	Lifespan (years) (Mohanan *et al.* 2020)	Application (Barboza *et al.* 2018; Jones *et al.* 2020; Plastics Europe 2021)
Low-density polyethylene (LDPE)	0.91–0.92	10–600	Garbage bags, garbage bins
High-density polyethylene (HDPE)	0.94–0.97	>600	Freezer bags, detergent, juice and shampoo bottles, rigid pipes
Polypropylene (PP)	0.83–0.92	10–600	Food packaging, chips packages, automotive parts, pipes
Polystyrene (PS)	1.04–1.10	50–80	Food packaging (dairy products), disposable cutlery, knives and cups, toys, CD cases, electronic equipment
Polyethylene terephthalate (PET)	0.96–1.45	450	Bottles of water, juice, and cleansers
Polyvinyl chloride (PVC)	1.16–1.58	50–150	Window frames, credit cards, food packaging, pipes, garden hoses, cosmetic containers, blood bags, cable insulation

In addition to MPs (<5 mm), which are deliberately produced for use in the production of personal care products and large plastic products, large-size plastics also break down into MPs (<5 mm) when exposed to various factors such as mechanical abrasion and UV exposure (Song *et al.* 2017). Many studies are reporting that MPs are found in drinking water (Wong *et al.* 2021), freshwater (Yahaya *et al.* 2022), seawater (Núñez *et al.* 2021), landfill leachate (Sun *et al.* 2021), sludge of WWTPs (Mahon *et al.* 2017), atmosphere (Dris *et al.* 2015), soil (Zhao *et al.* 2021), sediments (Yahaya *et al.* 2022), food (Diaz-Basantes *et al.* 2020), and the body of aquatic organisms (Núñez *et al.* 2021).

WWTPs, where MP-containing wastewater is collected and relatively removed, are mainly designed and operated to remove inorganic and organic substances from the water and to make the water microbially suitable and discharge it to the receiving environment. Therefore, since WWTPs are not specifically designed for MP removal, although MP removal seems to occur with high efficiency, millions of MP are released from WWTPs to the receiving environment in a day (Murphy *et al.* 2016; Ziajahromi *et al.* 2017; Gündoğdu *et al.* 2018; Conley *et al.* 2019; Franco *et al.* 2021). Not only the effluent of WWTPs but also WWTP sludges cause the release of MPs into the environment. In WWTPs, high amounts of MP of different polymer types, different shapes, and sizes are accumulated in the sludge of primary settling tanks, secondary settling tanks, and membrane sludge (Lares *et al.* 2018; Ren *et al.* 2020; Pittura *et al.* 2021).

Millions or more MPs are released into the environment through the disposal of tons of sludge produced in WWTPs or their use as fertilizer on agricultural lands (Magni *et al.* 2019; Ren *et al.* 2020; Harley-Nyang *et al.* 2022). Except for Germany, which states that the plastic content in fertilizers cannot exceed 0.1% by weight (Weithmann *et al.* 2018) many countries have not stipulated a limit value for the plastic content in fertilizers. However, it is worth noting that plastics smaller than 2 mm are not taken into account in Germany's regulation on the limit value of plastics that may contain fertilizers (Weithmann *et al.* 2018). As a result of the lack of strict regulations regarding the plastic content of WWTP sludge used in agricultural areas, MPs spread uncontrollably from WWTPs to the terrestrial environment and become a major environmental problem (Harley-Nyang *et al.* 2022).

In this review, the sources of MPs, the quantities, and properties of MPs in WWTPs in different locations around the world, and the MP removal efficiency of WWTP units separately were investigated. The results of the studies on the polymeric types, shapes, sizes, and colors of WWTPs as well as the amounts of MPs in both the water and sludge phase are also mentioned separately. In addition, the MP removal efficiency of primary, secondary, and tertiary treatment methods in WWTPs and the mechanisms that are effective in MP removal in these treatment methods are also focused. Mechanisms and factors that are effective in adsorbing organic–inorganic pollutants with MPs are presented. After evaluating all the issues listed above, the current situation and deficiencies regarding MPs in WWTPs are determined and suggestions are made for future studies.

2. SOURCES OF MPS IN WWTPS

Plastics are classified according to their size as megaplastics, macroplastics, mesoplastics, microplastics, and nanoplastics. The classification of plastics according to their sizes and the corresponding examples is given in Table 2. MPs are synthetic polymers with the longest dimension of 5 mm. MPs can be divided into two as large MPs (1–5 mm) and small MPs (1 μm–1 mm). MPs are divided into two classes, primary and secondary, according to their sources. Examples of sources of primary and secondary MPs are shown in Figure 2. Primary MPs are synthetic polymers with fiber or spherical shape, smooth surfaces, and deliberately produced micro-size (Crawford & Quinn 2016; Chatterjee & Sharma 2019). Primary MPs are used in personal care products (such as facial cleansing gel, toothpaste, shower gel, soap, shampoo, and sunscreen), cleaning products, make-up, the manufacture of synthetic clothing, and dyes (Lassen *et al.* 2015; Crawford & Quinn 2016; Chatterjee & Sharma 2019; De Falco *et al.* 2019; Sun *et al.* 2020). For example, it has been reported that 0.05 g/g (2,450 particles/g) MP is found in facial cleansers, while 0.02 g/g (2.15 particles/g) MP is found in shower gels (Sun *et al.* 2020). In the study conducted by De Falco *et al.* (2019) it was reported that 48.6–307.6 mg/kg microfiber was released as a result of washing different commercial clothes containing polyester (PEST) with a washing machine, which corresponds to microfibers in the range of 640,000–1,500,000.

Secondary MPs are plastics that are formed by the fragmentation of macro-sized plastics into smaller-sized pieces by various environmental factors and have a more random appearance (Crawford & Quinn 2016). UV radiation from the sun contributes to the oxidation of the matrix of macroplastics, damaging its chemical structure (Plastic Atlas 2020). In addition, factors such as waves, wind, and sand cause the breakdown of macroplastics into MPs by physical abrasion (Plastic Atlas 2020).

Depending on the diversity of wastewater coming to WWTPs, MP sources in WWTPs differ. It is well known that household MPs are transported to WWTPs through the use of personal care products and laundry wastewater. In a study, it was found that MPs vary in the range of 25.0–112.5 n/g in 10 types of toothpaste, in the range of 205–2,235 n/g in 10 types of facial cleaning products, and in the range of 2,900–7,100 n/L in laundry wastewater and it was determined that the biggest MP source in domestic wastewater is laundry wastewater (Tang *et al.* 2020). On the other hand, the population in the region where the water comes to WWTP, the lifestyle of the population, economic conditions, and seasonal changes are also effective in the number and characteristics of MPs originating from domestic wastewater in WWTPs. As for industrial

Table 2 | Classification and examples of plastics according to their sizes

	The longest size (Crawford & Quinn 2016; Lusher *et al.*2017)	**Item** (Lusher *et al.* 2017; Barboza *et al.* 2018).
Megaplastic	>1 m	Fishing nets and ropes, agricultural plastic films
Macroplastic	25 mm–1 m	Plastic bags, food packaging, balloons
Mesoplastic	5–25 mm	Bottle caps, plastic parts
Microplastic	5 mm–1 μm	Primary: resin pellets, micro-sized particles used in industrial products
Secondary: fibers from clothing		
Nanoplastic	<1 μm	Nanoplastics used in the pharmaceutical and medical device industries

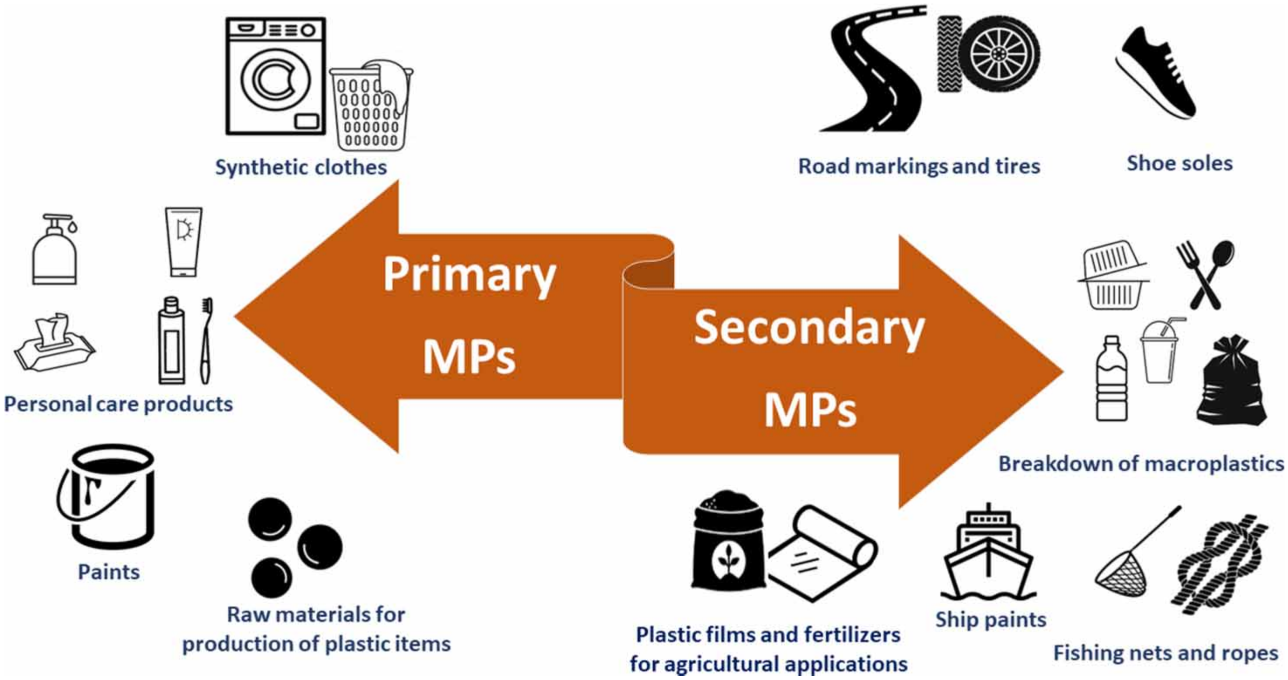

Figure 2 | Sources of primary and secondary MPs.

wastewater, the number, and type of industries from which the wastewater comes into WWTP affect the properties and concentration of MPs in the plant (Fältström *et al.* 2021). Since MPs cannot be completely removed even if leachate from solid waste landfills is treated with methods including advanced methods (Sun *et al.* 2021; Zhang *et al.* 2021b), the number of MPs in WWTPs increases, and their dominant characteristics may change when untreated or treated leachate to a certain extent in leachate treatment plant comes to WWTPs. Surface runoff also plays an important role in the differentiation of the amount and variety of MPs coming to WWTPs. Especially in the winter seasons when the runoff is higher, MPs coming to WWTPs (such as automobile tire wear, artificial grass, and cigarette filters) may cause a change in the number and properties of MPs in WWTPs. MPs in atmospheric fallout also contribute significantly to MPs in WWTPs via runoff with an average of 118 particles per m^2/day (Dris *et al.* 2015). In addition to MPs entering WWTPs, it is also suggested that paints used to prevent corrosion in tanks in WWTPs and some treatment units (especially filtration with polymeric membrane) may release MPs into wastewater and create an undesirable additional MP contamination in wastewater (Sun *et al.* 2021; Barbier *et al.* 2022).

Although current studies mostly focus on characterizing the MPs entering WWTPs and determining the polymer type, shape, size, and color, studies focusing on the sources of MPs in WWTPs are very limited. In studies dealing with the characterization of MPs in WWTPs, the sources of MPs are estimated based on the properties of MPs (especially polymer type and shape) from people's daily activities. Therefore, the sources and entry routes of MPs entering WWTPs are still not clearly understood. In future studies, further research on domestic wastewater only, industrial wastewater, combined domestic/industrial wastewater, and MPs in WWTPs with separated sewage systems will further improve understanding of the sources of MPs in WWTPs. In future studies, further research on domestic wastewater only, industrial wastewater, combined domestic/industrial wastewater, and MPs in WWTPs with separated sewage systems may help further improve understanding of the origins of MPs in WWTPs.

3. PROPERTIES OF MPS IN INFLUENT AND EFFLUENT OF WWTPS

MPs reaching WWTP from different sources differ in polymer types, shapes, sizes, and colors (Figure 3). Some studies in the literature on the percentage distribution of polymer types, shapes, and sizes of MPs in the influent and effluent of different WWTPs in the world are summarized in Table 3.

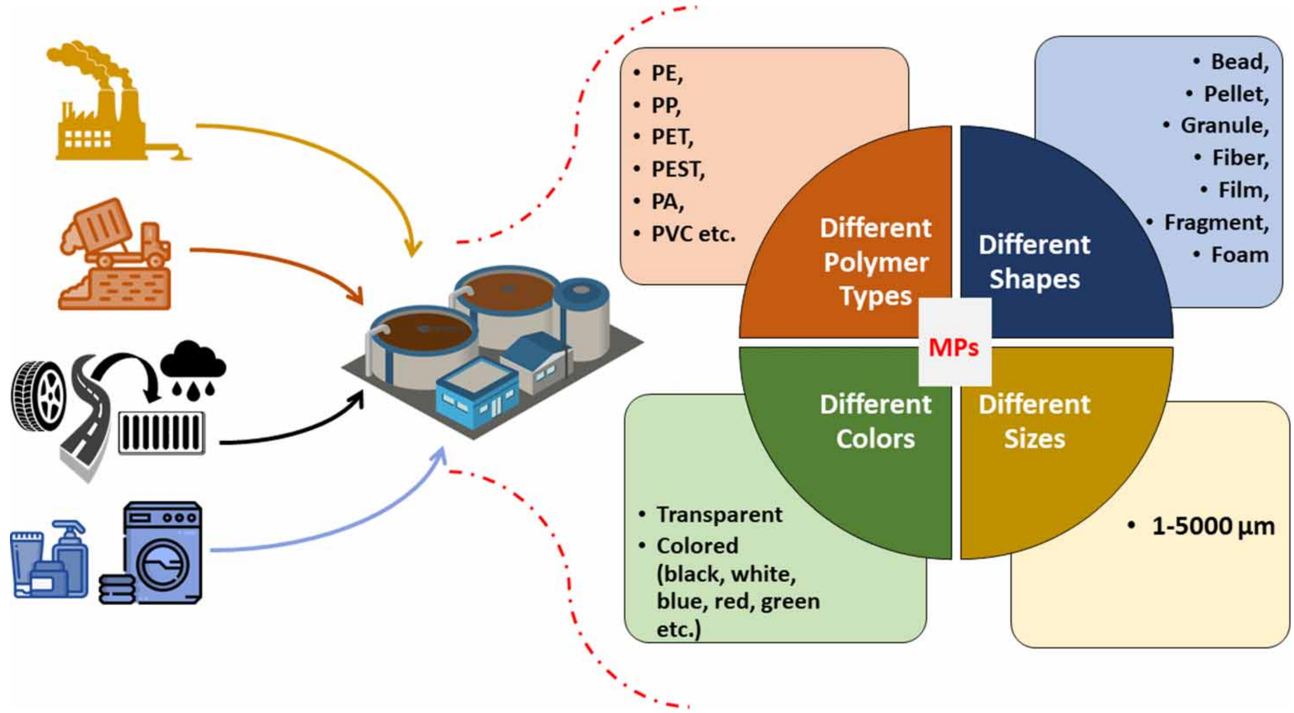

Figure 3 | Polymer types, shapes, sizes, and colors of MPs in WWTPs (PA: polyamide, PE: polyethylene, PEST: polyester, PET: polyethylene terephthalate, PP: polypropylene, PVC: polyvinylchloride).

3.1. Polymeric types of MPs in influent and effluent of WWTPs

Domestic wastewater treatment plants contain predominantly PA, PET, and PEST MPs released from clothes as a result of domestic washing (Gündoğdu *et al.* 2018; Yang *et al.* 2019; Franco *et al.* 2021). In addition, PE, PP, PS, and PVC are among the main polymers found in domestic WWTPs (Ziajahromi *et al.* 2017; Gündoğdu *et al.* 2018; Long *et al.* 2019; Yang *et al.* 2019; Alavian Petroody *et al.* 2020; Franco *et al.* 2021). It has been reported that polymers such as diallyl phthalate (DAP), polycaprolactone (PCL), and acrylonitrile styrene acrylate (ASA) are encountered in industrial WWTPs, unlike domestic WWTPs, due to their superior properties (such as stability, and resistance to solvents and oil) in industrial applications (Franco *et al.* 2021). Kim & Park (2021) reported that the MP species arriving at the treatment plant may be related to the density as well as the raw material of MP. They suggested that MP particles with lower density (such as PE and PP) reach the WWTP they studied in greater numbers because they tend to settle less from the source until they reach the treatment plant (Kim & Park 2021).

3.2. Shapes of MPs in influent and effluent of WWTPs

MPs can be found in wastewater in different morphologies such as spherical (beads, pellets, granules), lines (filaments, fibers), films, fragments, and foams (Paul-Pont *et al.* 2018; Rosal 2021). It has been reported by many researchers that the predominant MP morphology in WWTPs is especially fibers (Ziajahromi *et al.* 2017; Conley *et al.* 2019; Long *et al.* 2019; Franco *et al.* 2021). Lage found that fibers were the predominant MP type in samples taken from the influent and effluent of four different treatment plants in Norway (Lage 2019). In the study conducted by Gündoğdu *et al.* (2018), it was found that fibers are the dominant type at the influent and effluent of two different WWTPs in Turkey, and 44.4 and 86.5% of the MPs at the exit of the treatment plants are in fiber structure. Similarly, Conley *et al.* (2019) found that the microparticle removal efficiency ranged from 88.8 to 98.4% in three different treatment plants, while the fiber removal efficiency was lower (83.7–97.2%). The excess in the number of microfibers in the effluent of WWTPs is also an indication that the microfibers are not removed very effectively (Conley *et al.* 2019). Therefore, further research is needed to reduce the number of MPs in fiber structure, which is the predominant morphology in WWTPs, in the effluent of the plant.

Table 3 | Percentage distribution of polymer types, shapes, and sizes of MPs in different WWTPs

Location	Polymer type distribution in WWTPs		Shape distribution in WWTPs		Size distribution in WWTPs		Reference
	Influent	Effluent	Influent	Effluent	Influent	Effluent	
Cadiz, Spain	52.5% PVC, 22.5% EAA, 7.5% HDPE, 5.0% PA, 5% PE, 2.5% PP, 2.5% PMMA, 2.5% EVA	40.0% PVC, 40.00% PA, 13.3% PS, 6.67% HDPE	51.3% fiber, 23.1% flake, 19.1% fragment, 5.5% film, 0.7% sphere	44.6% fiber, 25.9% fragment, 24.3% flake, 3.8% film, 1.1% sphere	61.9% (355–100 μm), 32.2% (1,000–355 μm), 5.7% (5,000–1,000 μm)	57.2% (355–100 μm), 37.2% (1,000–355 μm), 5.5% (5,000–1,000 μm))	Franco *et al.* (2021)
Cadiz, Spain	45.4% PVC, 18.1% PE, 16.3% HDPE, 9.0% EAA, 3.6% PS, 3.6% PMMA, 1.8% PET, 1.8% PB	22.7% PS, 18.18% HDPE, 18.1% EAA, 13.64% PVC, 9.0% PCL, 9.09% DAP, 4.5% PP, 4.55% ASA	43.1% fiber, 30.9% fragment, 22.1% flake, 3.4% film, 0.4% sphere	45.8% fiber, 26.0% fragment, 22.3% flake, 3.8% film, 2.0% sphere	53.9% (355–100 μm), 22.4% (1,000–355 μm), 23.6% (5,000–1,000 μm)	71.8% (355–100 μm), 19.8% (1,000–355 μm), 8.3%(5,000–1,000 μm)	Franco *et al.* (2021)
Xiamen, China	30.2% PP, 26.9% PE, 10.3% PS, 7.5% PET, 6.3% PE + PP, 5.1% PP + PE, 3.3% PEST, 9.9% others	34.8% PP, 17.90% PE, 13.9% PP + PE, 9.6% PS, 7.5 PET%, 4.7% PE + PP, 1.1% PEST, 10.1% others	49.8% granule, 30.0% fragment, 17.7% fiber, 2.5% pellet	36.0% granule, 30.40% fiber, 28.0% fragment, 5.6% pellet	43.5% (125–63 μm), 23.7% (63–43 μm), 20.7% (355–125 μm), 12.1% (5,000–355 μm)	32.1% (355–125 μm), 28.0% (125–63 μm), 27.2% (5,000–355 μm), 12.7% (63–43 μm)	Long *et al.* (2019)
Adana, Turkey	50.8% PEST, 29.2% PE, 13.8% PP, and others	43.80% PEST, 31.30% PE, 18.80% PP, 6.30% Nylon-6	54.8% fiber, 26.8% fragment, 18.4% film	44.4% fiber, 30.2% film, 25.4% fragment	53.6% (1–5 mm), 23.0% (0.5–1 mm), 21.8% (0.1–0.5 mm), 1.7% (<0.1 mm)	34.9% (1–5 mm), 34.9% (0.5–1 mm), 27.0% (0.1–0.5), 3.2% (<0.1 mm)	Gündoğdu *et al.* (2018)
Adana, Turkey	61.9% PEST, 23.8% PE, 11.9% PP, and others	68.80% PEST, 18.80% PE, 12.50% PP	87.7% fiber, 10.0% fragment, 2.4% film	86.5% fiber, 10.8% fragment, 2.7% film	59.2% (1–5 mm), 24.6% (0.1–0.5 mm), 14.7% (0.5–1 mm), 1.4% (<0.1 mm)	40.5% (1–5 mm) 27.0% (0.5–1 mm) 29.7% (0.1–0.5) 2.7% (<0.1 mm)	Gündoğdu *et al.* (2018)
Glasgow, Scotland	28.7% alkyd, 19.1% PS acrylic, 10.8% PEST, 8.9% PU, 8.3% acrylic, 4.5% PE, 4.5% PA, 3.8% PET, 3.2% PVA, 2.6% PP, 2.6% PS and others	28.0% PEST, 20.0% PA, 12.0% PP, 12.0% acrylic, 8.0% alkyd, 4.0% PET, 4.0% PE, 4.0% poly aryl ether	67.3% flake, 18.5% fiber, 9.9% film, 3.0% beads, 1.3% foam	–	–	–	Murphy *et al.* (2016)
Across United States	–	–	–	59.00% fiber, 33.00% fragment, 5.00% film, 2.00% foam, 1.00% pellet	–	57.0% (125–355 μm) 43.0% (>355 μm)	Mason *et al.* (2016)

Note: ASA, acrylonitrile styrene acrylate; DAP, diallyl phthalate; EAA, ethylene acrylic acid; EVA, ethylene-vinyl acetate; HDPE, high density polyethene; PA, Polyamide; PB, polybutylene; PCL, polycaprolactone; PE, polyethylene; PEST, polyester; PET, polyethylene terephthalate; PMMA, polymethyl methacrylate; PP, polypropylene; PS acrylic, polystyrene acrylic; PS, polystyrene; PU, polyurethane; PVC, polyvinyl chloride; WWTP, wastewater treatment plant.

3.3. Colors of MPs in influent and effluent of WWTPs

To increase the attractiveness of plastic products for consumption and to improve their performance, dyes and pigments are used during production (Xu *et al.* 2020). Different colored MPs in the water are an indication that MPs are mixed into the aquatic environment from different sources. The presence of transparent or colored (white, black, blue, green, red, yellow, and other colors) MPs in treatment plants and aquatic environments has been reported by many researchers (Lage 2019; Martí *et al.* 2020; Montoto-Martínez *et al.* 2020; Dey *et al.* 2021; Van Do *et al.* 2022). Although it is thought to be insignificant considering the fact that the effect of the color factor on the MP removal efficiency cannot be determined, the dyes in MPs have a toxic effect on aquatic organisms. There are also studies showing that the surfaces of colored MPs can contain harmful compounds such as heavy metals and persistent organic pollutants (Xu *et al.* 2019). Since the colored or transparent MPs released from WWTPs are similar to food, ingestion by organisms in the aquatic environment accumulates in their bodies and eventually reaches humans via the food chain (Vivekanand *et al.* 2021; Sun *et al.* 2022). Moreover, MPs of different colors released into the aquatic environment by discharge from WWTPs affect the physiology of algae by changing the light absorption in the aquatic environment and creating a shading effect. In a recent study examining the effect of green, black, and white PET MPs on *Microcystis aeruginosa*, it was found that especially green MPs increased the growth and photosynthesis of *M. aeruginosa* due to their color close to cyanobacteria and black and white MPs were found to inhibit photosynthesis due to their higher shading effect (Lu *et al.* 2022). Moreover, in the study, it was determined that green colored MPs inhibited microcystin production, but white and especially black MPs caused a significant increase in microcystin production (Lu *et al.* 2022).

3.4. Sizes of MPs in influent and effluent of WWTPs

The sizes of MPs in the influent and effluent of WWTPs can be at the level of large MPs (1–5 mm) and small MPs (1 μm–1 mm) according to studies (Table 3). As can be seen from Table 3, it cannot be generalized that most of the sizes of MPs discharged from WWTPs to the aquatic environment belong to large or small MPs. The treatment methods/technologies used in WWTPs and the fragmentation of MPs in WWTPs during the treatment process affect the size of MPs discharged to the aquatic environment. In a study, it was found that for MPs examined in the 100–5,000 μm range, small MPs in the 355–100 μm range in the influent of WWTP correspond to 53.95% of the total MPs, while it corresponds to 71.81% in the effluent (Franco *et al.* 2021). Researchers have associated this with better removal of larger MPs in WWTP and fragmentation of MPs into smaller fragments during transport (Franco *et al.* 2021). Similarly, Alavian Petroody *et al.* (2020) reported that both fiber and particle MPs $\geq$500 μm in size exhibit higher removal efficiency in the primary settling tank compared to MP in the 300–37 μm range. MPs $<$100 μm have been examined in studies by some researchers (Gündoğdu *et al.* 2018; Long *et al.* 2019; Alavian Petroody *et al.* 2020), MPs $<$100 μm in size were not included in the study and were underestimated by most researchers in the literature. In the studies to be carried out to determine the removal efficiency of the treatment units in WWTPs according to the MP size, this deficiency in the literature should be eliminated by considering the small MPs.

4. PROPERTIES OF MPS IN SLUDGE OF WWTPS

The MPs in WWTPs are entrapped in high quantities in primary settling tank sludge (Lee & Kim 2018), secondary settling tank sludge (Lv *et al.* 2019; Pittura *et al.* 2021), and membrane sludge (Lares *et al.* 2018; Lv *et al.* 2019). The number of MPs in WWTP sludge varies depending on the characteristics of the wastewater coming to WWTP, the capacity of WWTP, and the different treatment technologies applied in WWTP (Lares *et al.* 2018; Lee & Kim 2018; Lage 2019; Lv *et al.* 2019), the amount of sludge coming out of WWTP and the different processes applied to the sludge (Lares *et al.* 2018; Edo *et al.* 2020; Harley-Nyang *et al.* 2022).

In Table 4, the results regarding the MP amounts determined in the sludges of different WWTPs in the world in recent years and their percentage distribution of polymer type, shape, and size are summarized. As seen in Table 4, the difference in MP concentration in WWTPs in different countries, in influent, treatment technology, and the treatment unit from which the sludge is sampled can result in a relatively low or relatively high (hundreds of MPs) MP content per gram of sludge. Lee & Kim (2018) reported that MP removal by sludge cake was 49.3, 44.7, and 49.0%, respectively, in three WWTPs in Korea where the A2O, sequence batch reactor (SBR) process, and Media process were applied. In a study by Lv *et al.* (2019), it was noted that MP removal efficiency was 83.5% with membrane tank, 76.5% with secondary settling tank, 16.5% with oxidation ditch, and 15% with A/A/O unit, depending on the water/sludge separation process of different treatment methods.

Table 4 | The amount of MP in different WWTP sludges and their percentage distribution of polymer type, shape, and size

Location	WWTP capacity	WWTP units	MP concentration in influent	Sludge type	MP amount in sludge	Polymer type of MPs in sludge	Shape of MPs in sludge	Size of MPs in sludge	Reference
Spain	8,000 m^3/day	Screening system, grit and grease, biological reactor, double secondary clarifier, coagulation–flocculation, lamellar decanter, rapid sand filtration and UV irradiation	16.1 MPs/L	Mixture of double secondary clarifier and lamellar decanter	24.0 (MPs/g)	36.0% PET, 25.0% PS, 20.0% PA and 9.0% PVC	57.0% fragment, 33.0% fiber	–	Menéndez-Manjón *et al.* (2022)
England	1,000 L/s	–	–	Reception tank Thickened Digestate centrifuge feed tank Sludge cake Pre-limed Limed	107.5 50.2 180.7 286.5 97.2 74.7 37.7 (MPs/g dw)	39.8% PEST, 13.6% PVA, 13.1% PE and 33.5% others	57.5% particle and 42.5% fiber	In most locations the majority of MPs (except the limed and thickened samples) are in the 100–500 μm range.	Harley-Nyang *et al.* (2022)
Italy	18,000 m^3/day	Screen and grit, primary settler, activated sludge tank, secondary settler, and disinfection	3.6 MPs/L	Primary sludge Waste activated sludge Final sludge	1.6 5.3 4.7 (MPs/ gTS)	52.0% PE, ~30.0% PP, ~5.0% EEA and others ~30.0% PE, ~30.0% PP, ~5.0% PEST and others ~35.0% PP, ~25.0% PE, ~10.0% PEST and others	70.0% particle and 30.0% fiber 80.0% particle and 20.0% fiber 80.0% particle and 20.0% fiber	Most of MPs were between 0.5–5 mm in primary sludge. Most of MPs were between 0.1–1 mm in activated sludge and final sludge.	Pittura *et al.* (2021)
China	300,000 m^3/day	Inlet room, primary sedimentation tank, secondary sedimentation tank, V-type filtration pool, and outlet room	16.0 MPs/L	Dewatered sludge	2,920 (MPs/ kg)	–	~63.0% fiber and ~37.0% fragment	41.0% (0.08–0.55 mm) 51.0% (0.55–1.70 mm) 8.0% (1.70–5.00 mm)	Ren *et al.* (2020)

(*Continued.*)

Table 4 | Continued

Location	WWTP capacity	WWTP units	MP concentration in influent	Sludge type	MP amount in sludge	Polymer type of MPs in sludge	Shape of MPs in sludge	Size of MPs in sludge	Reference
China	50,000 m^3/day	Aerated grit chambers, oxidation ditch, secondary settling tank, and UV disinfection	0.2 MPs/L	Secondary settling tank sludge	0.7 (MPs/L)	–	Fibers are more dominant than films and fragments.	MPs >500 μm are dominant.	Lv *et al.* (2019)
China	70,000 m^3/day	Rotary grit chambers, anaerobic, anoxic and aerobic tanks, and membrane tank	0.2 MPs/L	Membrane tank sludge	1.6 (MPs/L)	–	Fragments are more dominant than films.	MPs >500 μm are dominant.	Lv *et al.* (2019)
France	80,000 m^3/day	–	244 MPs/L	Sewage sludge	16.1 (MPs/g)	~25.0% PS, ~20.0% PET, ~18% PE, ~15.0% PP, ~10.0% PA and others	~77.0% fiber and others	~55.0% (200–500 μm) ~20.0% (80–200 μm) ~20.0% (>500 μm) 5.0% (20–80 μm)	Kazour *et al.* (2019)
Italy	400,000,000 L/day	Screening, grit and grease removal stages, biological treatment, sedimentation (with recycled activated sludge), sand filter, and disinfection	2.5 MPs/L	Recycled activated sludge	113 (MPs/ g dw)	27.0% NBR, 18.0% PE, 15.0% PEST, 9% PP and others	51.0% film, 34.0% fragment and 15.0% line	54.0% (0.5–0.1 mm) 24.0% (0.1–0.01 mm) 12.0% (1–0.5 mm) 10.0% (5–1 mm)	Magni *et al.* (2019)
Korea	35,000 m^3/day	Coarse and fine screen, primary settling tank, A^2O tanks, secondary settling tank, and UV sterilization	29.9 MPs/L	Secondary settling tank sludge	14.9 (MPs/g)	–	3.6 fibers/g 11.2 fragments/g	13.2 MP/g (106–300 μm) 1.6 MP/g (>300 μm)	Lee & Kim (2018)
Korea	130,000 m^3/ day	Coarse and fine screen, primary settling tank, bioreactors and aerobic tanks, secondary settling tank, and UV sterilization	13.9 MPs/L	The mixture of primary and secondary settling tank sludge	13.2 (MPs/g)	–	6.0 fibers/g 7.1 fragments/g	10.6 MP/g (106–300 μm) 2.5 MP/g (>300 μm)	Lee & Kim (2018)

(*Continued.*)

Table 4 | Continued

Location	WWTP capacity	WWTP units	MP concentration in influent	Sludge type	MP amount in sludge	Polymer type of MPs in sludge	Shape of MPs in sludge	Size of MPs in sludge	Reference
Canada	180,044 ML/ year	Screening bars, primary clarification, trickling filters and solids contact tanks, secondary clarifiers, and chlorination	31.1 MPs/L	Primary sludge Secondary sludge	14.9 4.4 (MPs/g)	–	9.7 fibers/g, 5.1 fragments/g, 0.0 foams/g, 0.0 pellets/g 3.6 fibers/g 0.9 fragments/g	–	Gies *et al.* (2018)
Finland	10,000 m^3/day	Screening, grit separation, primary clarification, activated sludge, secondary sedimentation, and disinfection.	57.6 MPs/L	Activated sludge Digested sludge MBR sludge	23.0 170.9 27.3 (MPs/g dw)	~ 95.0% PEST and 5% PE ~85.0% PEST, 7.0% PA and others ~80.0% PEST, 10.0% PE and others	21.7 fibers/g 1.3 particles/ g 161.0 fibers/ g 9.8 particles/ g 24.1 fibers/g 3.3 particle/g	~67.0 (<1 mm) ~70.0% (<1 mm) ~85.0% (<1 mm)	Lares *et al.* (2018)
Norway	–	Screening, sand/fat removal, chemical dosing, and sedimentation Screening, sand/fat removal, pre-sedimentation, chemical dosing, and post sedimentation Screening, sand/fat removal, chemical dosing, and sedimentation	445 MPs/L 289 MPs/L 525 MPs/L	Anaerobic treatment process sludge Raw dewatered sludge Raw dewatered sludge	37,502 13,770 14,419 (MPs/kg dw)	–	50.3% fragment 46.2% fiber and others ~93.0% fiber ~5.0% fragment and others ~93.0% fiber ~5.0% fragments	–	Lage (2019)

Note: dw, dry weight; EEA, ethylene-ethyl acrylate copolymer; NBR, acrylonitrile–butadiene; PA, polyamide; PE, polyethylene; PEST, polyester; PET, polyethylene terephthalate; PP, polypropylene; PS, polystyrene; PVA, polyvinyl acetate; PVC, polyvinyl chloride; TS, total solids.

Considering that tons of sludge come out of WWTPs, a significant amount of MP is released into the environment with the use of sludge with high MP content as fertilizer in agricultural areas and improper management. Magni *et al.* (2019) reported 113 ± 57 MPs/g (dw) MP in the recycled activated sludge of WWTP in Italy and estimated that 3.4×10^9 MPs accumulated per day in the sludge of this plant, from which 30 tons/day of sludge was produced. Ren *et al.* (2020) reported that the MP concentration in the dewatered and dried sludge in a WWTP of 300,000 m^3/day in China was 2.92×10^3 MP/kg and 3.15×10^8 MP would be released into the environment from WWTP producing 108 tons of sludge. Similarly, Harley-Nyang *et al.* (2022) found that 1.61×10^{10} and 1.02×10^{10} MP would be released into the environment each month, respectively, with the use of anaerobic digested and lime-stabilized sludge of a WWTP in the UK as fertilizer on agricultural land, and they estimated that this was the equivalent of >20,000 plastic debit cards. Since the uncontrolled use of sludge in WWTPs in agricultural lands causes the distribution of MPs in large quantities to the environment, scientific studies should be given priority to monitoring the MPs in the sludge in WWTPs and examining the effects of the processes applied to the sludge on MP removal.

4.1. Polymeric types of MPs in sludge of WWTPs

Since the type of polymer of MPs is a factor that directly affects the density of MP, it affects the precipitation of MPs in WWTPs and their deposition in the sludge. Studies have shown that PESTs (Lares *et al.* 2018; Kazour *et al.* 2019; Pittura *et al.* 2021; Harley-Nyang *et al.* 2022; Menéndez-Manjón *et al.* 2022), PS (Kazour *et al.* 2019; Menéndez-Manjón *et al.* 2022), and PA (Lares *et al.* 2018; Kazour *et al.* 2019) MPs accumulate more in WWTP sludge due to their high densities. On the other hand, there is information in the literature that lower-density MPs such as PE (Lares *et al.* 2018; Pittura *et al.* 2021; Harley-Nyang *et al.* 2022) and PP (Kazour *et al.* 2019; Magni *et al.* 2019; Pittura *et al.* 2021; Zhang *et al.* 2021a) are also found in WWTP sludge. Menéndez-Manjón *et al.* (2022) found that after secondary and tertiary treatment of wastewater from a WWTP in Spain, the predominant MP types in the wastewater were specifically PE and PP, while the high-density PET, PS, and PA MP types predominate in the dewatered secondary and tertiary treatment sludge mixture. Consistent with the results of Menéndez-Manjón, Zhang *et al.* (2021a) also found that PET (37.62%) was the predominant MP type in the dewatered sludge of modified SBR (MSBR). It was also found that PA significantly increased in sludge compared to wastewater influent and MSBR effluent.

4.2. Shapes of MPs in sludge of WWTPs

In studies conducted in different WWTPs around the world, it has been determined that a significant amount of MPs in the form of fibers (Gies *et al.* 2018; Lee & Kim 2018; Kazour *et al.* 2019; Lage 2019; Lv *et al.* 2019; Ziajahromi *et al.* 2021) and fragments (Gies *et al.* 2018; Lee & Kim 2018; Lv *et al.* 2019; Magni *et al.* 2019; Ren *et al.* 2020) are found in the sludge. Pittura *et al.* (2021) reported that while the percentage of microfiber and microparticles in the inlet of WWTP showed an almost equal distribution, the percentage of microparticles in primary sludge, aerated waste sludge, and dewatered sludge increased to 70, 80, and 80%, respectively. Moreover, they noted that fragment-type MPs formed the dominant MP shape in all samples. On the other hand, fiber-shaped MPs, which reach WWTPs as a result of washing synthetic clothes and are found significantly even in the effluent of WWTP, also accumulate significantly in sludge. For instance, Lares *et al.* (2018) in their analysis of activated sludge, digested sludge, and MBR sludge, determined that fiber-type MPs constitute approximately 94.3, 94.2, and 88.2% of total MPs, respectively. Tadsuwan & Babel (2021) reported that in the sludge sample taken from the final clarifier after secondary treatment in a WWTP in Thailand, the dominant MP shape was fiber (53%), followed by films (29%) and fragments, respectively.

4.3. Sizes of MPs in the sludge of WWTPs

In studies examining the size of MPs in WWTP sludges, it was found that MPs with a size <1 mm were dominant in general (Lares *et al.* 2018; Kazour *et al.* 2019; Magni *et al.* 2019; Ren *et al.* 2020; Pittura *et al.* 2021). In more detail, it has been determined by many researchers that MPs smaller than 0.5 mm are much more abundant in WWTP sludge (Kazour *et al.* 2019; Magni *et al.* 2019; Tadsuwan & Babel 2021; Harley-Nyang *et al.* 2022). Generally, the absence of large-size MPs in WWTP sludges is also due to the coarse and fine screens used in wastewater pretreatment, typically with gap sizes of 6–150 mm and <6 mm, respectively (Carr *et al.* 2016; Liu *et al.* 2019b). Di Bella *et al.* (2022) found that the number of MPs <1 mm in the secondary sludge of three different WWTPs with pre-treated CAS, non-pre-treated CAS, and MBR was higher than the number of MPs in the 1–5 mm range. Tadsuwan & Babel (2021) reported that MPs of 0.05–0.5 mm in size were predominant (~70%) in the sludge taken from the final clarifier and passed through fine screening, grit trap, aeration tank, and final clarifier purification units. As the size of MPs increased, their percentage in the sludge decreased, i.e. 0.5–1 mm MPs were found to be ~20%, while 1–5 mm MPs were found to be ~10% (Tadsuwan & Babel 2021). Lares *et al.* (2018) examined MPs in activated sludge, digested sludge and MBR sludge in the size ranges from <0.25 to 5 mm and found that MPs in the 0.25–1 mm range were predominant in all three different sludge samples. Zhang *et al.* (2021a) noted that in the WWTP sludge containing 12.73 MP/g, MPs were dominant in the 0.9–0.45 mm range, with MPs in this size range corresponding to 7.76 MP/g.

5. MP REMOVAL PERFORMANCE OF TREATMENT UNITS IN WWTPS

In WWTPs, water generally goes through primary, secondary, and tertiary treatment stages. The water coming to WWTP firstly passes through coarse and fine screens, sand/oil chamber for primary treatment, and reaches the primary settling tank. At this stage, depending on the characteristics of the wastewater, a flotation unit can also be used (de Sena *et al.* 2009). In secondary treatment, suspended or attached-growth biological processes are used to remove organic matter by microorganisms, and then the wastewater is given to the secondary settling tank. Methods such as sand filtration and membrane filtration applied after the final settling tank are the methods used for the tertiary treatment of wastewater. The different concentrations and characteristics of MPs in the influent of WWTPs, the application of different treatment technologies in WWTPs, the lack of a standard method for MP analysis, and the analysis of MPs with different sizes in studies led to different MP removal efficiencies in different WWTPs. Figure 4 shows the sources of MPs in WWTPs and the treatment stages/units of WWTPs. In this section, MP removal efficiencies of the methods/technologies used in WWTPs are examined.

5.1. MP removal by flotation and primary settling

Primary settling tanks are used in wastewater treatment for the removal of high efficiency suspended solids under the effect of gravity before biological treatment. Unlike sedimentation, flotation is a method that allows substances with a lower density than water to be raised to the surface of the water against the direction of gravity using gas bubbles, and then to the surface, and then to separate these substances from the water environment by skimming (Kwak *et al.* 2005). Low-density MPs tend to float in water, while high-density MPs tend to settle. Therefore, it is reasonable to remove MPs with a higher density than

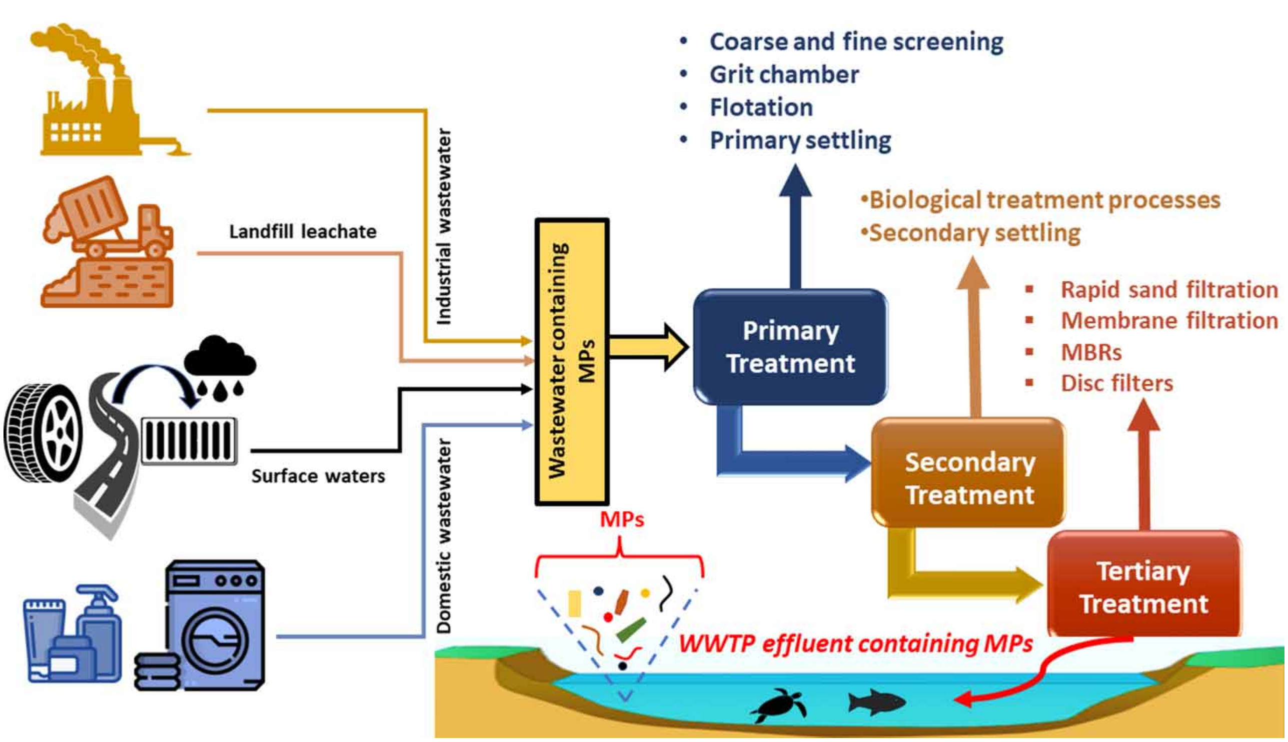

Figure 4 | MP sources in WWTPs and treatment stages in WWTPs (MPs: microplastics, WWTP, wastewater treatment plant).

wastewater (such as PET) from the wastewater by precipitation. In addition, the flotation method can be considered a suitable method for the removal of low or medium-density MPs that cannot be precipitated. Talvitie *et al.* (2017) reported that 95% of MPs in wastewater were removed by dissolved air flotation (DAF) (from 2.0 MP/L to 0.1 MP/L). Long *et al.* (2019) reported that the removal rate of PP, PE, PS, and PET type MPs in WWTP increased with increasing density and they noted that removal efficiencies of 92.0, 87.8, 94.8, and 96.4% were achieved for PP, PE, PS, and PET, respectively. On the other hand, the accumulation of pollutants or biofilm formation on the MP surface can also cause an increase in the density of MPs independent of the polymer structure and different positioning of MP in the water column than expected, and different removal efficiencies than expected can be obtained.

Table 5 presents the MP removal efficiencies of WWTPs by the methods used for primary treatment, including primary settling and flotation. It has been reported by different researchers that MP removal efficiencies of 40.7% (Liu *et al.* 2019b) and 58.8% (Yang *et al.* 2019) are achieved after the primary settling tank following the pretreatment units. Although the MPs removed by precipitation do not reach the receiving water from the WWTP effluent, these MPs eventually accumulate in the sludge from the precipitation units (Pittura *et al.* 2021). With the disposal of WWTPs sludge in landfills, MPs mix with leachate and eventually return to WWTP again (Freeman *et al.* 2020). As another possibility, when WWTPs sludge is used as fertilizer in agricultural activities, MPs are dispersed into the environment. For this reason, it is of great importance to focus more on studies on the management of MPs trapped in sludge.

5.2. MP removal by biological treatment and secondary settling

Biological treatment is a process that is included in the secondary treatment stage and ensures the removal of organic materials from wastewater by microorganisms in a controlled environment after primary treatment (Sonune & Ghate 2004). In anaerobic, anoxic, and aerobic processes, microorganisms provide the removal of nutrients and organic matter. MPs are also removed during the removal of dissolved organic matter by the activity of microorganisms (Kwon *et al.* 2022). The removal of MPs in aeration tanks can be explained by their attachment to microorganisms and sludge due to their hydrophobic structure (Hongprasith *et al.* 2020).

Table 6 includes studies examining the removal efficiency of MPs of treatment methods used for secondary treatment (i.e., biological processes, and secondary settling tanks) in WWTPs. Liu *et al.* (2019b) reported that 16% MP removal efficiency was achieved with anaerobic + anoxic + oxic processes while Yang *et al.* (2019) reported that 54.47% MP removal was achieved with anaerobic + anoxic + aerobic processes. Similarly, it was reported that 60.0% (Pittura *et al.* 2021) and 74.8% (Bretas Alvim *et al.* 2020) MP removal efficiencies were achieved with the secondary settling tank following the activated sludge tank. Therefore, even if the same biological treatment technology is applied, the characteristics of WWTP operation and MPs can lead to differences in removal efficiencies. Therefore, even if the same biological treatment technology is applied, the characteristics of WWTP operation and MPs can lead to differences in removal efficiencies.

In the secondary settling tanks following the biological treatment, MPs accumulate in the settled sludge and the number of MPs reaching the outlet decreases. Therefore, like primary settling tank sludges, secondary settling tank sludges also contain significant MPs (Gies *et al.* 2018; Lage 2019; Lofty *et al.* 2022). That is, reducing the number of MPs released from the WWTP effluent to the aquatic environment is not the only focus. There should also be a focus on the management of MPs accumulated in primary and secondary settling tank sludges.

5.3. MP removal by filtration

Membrane filtration is a widely used method in the treatment of drinking water and wastewater. Membranes produced from different polymers such as PE, PP, PA, polyethersulfone (PES), polyvinylidene fluoride (PVDF), and polycarbonate (PC) are

Table 5 | MP removal efficiencies of WWTPs by the methods used for primary treatment; including primary settling and flotation

Treatment units	Removal efficiency (%)	Referencess
Primary settling tank	47.8	Pittura *et al.* (2021)
Aerated grit trap + primary settling tank	58.8	Yang *et al.* (2019)
Coarse screen + fine screen + grit chamber + primary settling tank	40.7	Liu *et al.* (2019b)
DAF	95.0	Talvitie *et al.* (2017)

Note: DAF, dissolved air flotation.

Table 6 | MP removal efficiencies of WWTPs by the methods used for secondary treatment, including biological processes and secondary settling tanks

Treatment units	Removal efficiency (%)	References
Bioreactor + secondary settling tank	72.5–91.0	Kwon *et al.* (2022)
Activated sludge + secondary settling tank	60.0	Pittura *et al.* (2021)
UASB	52.6	Pittura *et al.* (2021)
Primary settling + subsequent biological treatment steps	68.3	Kim & Park (2021)
Aerobic biological reactor + secondary settling tank	74.8	Bretas Alvim *et al.* (2020)
Aeration tank + secondary settling tank	84.0	Hongprasith *et al.* (2020)
Anaerobic + anoxic + aerobic processes	54.4	Yang *et al.* (2019)
Anaerobic + anoxic + oxic processes	16.0	Liu *et al.* (2019b)

Note: UASB, upflow anaerobic sludge blanket.

widely used in the treatment of drinking and wastewater due to their ease of production, cost-effectiveness, and superior properties (Himma *et al.* 2016; Li *et al.* 2021; Pizzichetti *et al.* 2021; Acarer *et al.* 2021). Pressure-driven membranes are ranked microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) in order of decreasing pore size. Considering that the pore size of the MF membrane with the highest pore size among these four pressure-driven membranes is in the range of about 100 nm–10 μm, it can be predicted that it can retain MPs (<5 mm). Pizzichetti *et al.* (2021) showed that a membrane made of three different polymers with a pore size of 5 μm can retain PA MPs in the range of 99.6–99.8% and PS MPs in the range of 94.3–96.8%. While such polymeric membranes separate MPs from water, they can also cause MPs to migrate to water by fragmentation or rupture, as they are themselves made of polymers (Tang & Hadibarata 2021). Therefore, this issue needs to be addressed further by researchers.

Membrane bioreactors (MBRs) are systems that combine biological treatment with membrane filtration (usually MF and UF) (Mabrouki *et al.* 2020). MBRs provide superior MP removal efficiency compared to other treatment methods used in water and wastewater treatment and provide MP removal efficiency of over 99% (Talvitie *et al.* 2017; Lares *et al.* 2018). On the other hand, Bayo *et al.* (2020) reported that MP removal efficiency with MBR is 79.01%. Many factors such as the structure of the MP removed, its morphological properties, membrane material, membrane properties, the interaction between the membrane and MP, the presence of other pollutants in the wastewater, and membrane contamination affect the MP removal efficiency of the membrane (Dey *et al.* 2021). Therefore, although it seems that the same treatment technology is used, different MP removal efficiencies with MBR can be encountered in the literature, since many factors influence the change of MBR and MP removal efficiency.

RO membranes separate contaminants from wastewater that MF, UF, and NF membranes cannot separate due to smaller pore sizes (<1 nm) and lower molecular weight separation limits (MWCO) (<200 Da). However, studies in recent years have revealed that wastewater may contain significant amounts of MPs even after passing through RO membranes used as tertiary treatment (Ziajahromi *et al.* 2017; Sun *et al.* 2021; Cai *et al.* 2022). Cai *et al.* (2022) reported that MPs in the influent of a WWTP in which primary sedimentation, biological treatment, MBR and RO processes were applied achieved 93.2 and 98.0% MP removal efficiency after MBR and RO, respectively. On the other hand, as a remarkable point, Cai *et al.* (2022) stated that non-fiber MPs larger than 0.5 mm will be completely removed from wastewater with MBR and RO, but MPs with fiber structure, especially <200 μm in size, can pass through RO and remain in wastewater. Similarly, in the study of Ziajahromi *et al.* (2017), PET fibers accounted for 88% of total MPs in wastewater filtered from RO. The use of RO membranes after membranes with larger pore sizes and higher MWCO in WWTPs contributes to the presence of fewer MPs in the WWTP effluent under normal conditions. However, some studies suggest that MP migration through RO membranes may be released through membrane defects and small openings in piping (Ziajahromi *et al.* 2017) or worn polymeric membranes (Sun *et al.* 2021). Therefore, there is still a need to clarify this issue and take precautions by conducting more extensive research on whether the MPs in the effluent of the polymeric membranes used in WWTPs originate from the membrane material.

Relatively small-sized MPs are likely to pass through rapid sand filtration (RSF) systems used in wastewater and water treatment. For example, Na *et al.* (2021) reported that PS MPs larger than 20 μm were largely retained by the sand filter (98.8% and higher removal efficiency), but MPs smaller than 20 μm largely passed through the sand medium (83.4% removal efficiency). It has been noted in the literature that 83.4% (Na *et al.* 2021), 75.4% (Bayo *et al.* 2020), 73.8% (Hidayaturrahman & Lee 2019), and 97.0% (Talvitie *et al.* 2017) of MPs were removed by using RSF. However, when sand filtration and MBR filtration are compared, MBRs show much superior performance in terms of MP removal.

Disc filters (DFs) are units made of cloth, consisting of several discs, the filter size of which is generally in the range of 10–40 μm, and they are generally used in WWTPs for polishing water after biological treatment. DFs also exhibit lower MP removal performance (40.0–98.5%) than MBRs. (Talvitie *et al.* 2017; Simon *et al.* 2019).

In Table 7, studies investigating MP removal efficiencies from wastewater by filtration techniques are summarized. When MP removal is evaluated by filtration, more MP removal is provided, especially with MBRs. On the other hand, one of the most important problems in the separation process with membranes is the clogging of the surface and pores of the membrane with filtration (Türkoğlu Demirkol *et al.* 2021).

5.4. MP removal by coagulation

Coagulation is the process of adding chemical substances to the water to neutralize the charge of colloidal substances that cannot settle in water to facilitate precipitation. Since coagulation is a process used especially in drinking water treatment, studies on the removal of MPs from water by coagulation have generally been studied in surface water such as river and lake water (Lapointe *et al.* 2020; Na *et al.* 2021; Xue *et al.* 2021), and deionized water (Na *et al.* 2021) matrices. Coagulation can also be used as a tertiary treatment for the removal of total phosphorus that cannot be completely removed in WWTPs. However, the number of studies addressing MP removal in WWTPs by coagulation process and jar tests with wastewater is still very limited. Kwon *et al.* (2022) investigated MP removal efficiency by coagulation using polyaluminium chloride, which was applied as tertiary treatment after physical and biological treatment in two different WWTPs that treat domestic/industrial and domestic wastewater only and they determined MP removal efficiencies of these WWTPs as 42.26 and 15.79%, respectively. Kwon *et al.* (2022) reported that the total MP removal efficiencies in wastewater treated until secondary treatment was 91.63 and 97.74% for domestic–industrial and domestic wastewaters, and after coagulation, these removal efficiencies reached 96.33 and 98.1%, respectively. In another study by Hidayaturrahman & Lee (2019), MP removal efficiencies of the coagulation process used as a tertiary treatment in three different WWTPs were determined as 47.1, 53.8, and 81.6%. The overall removal percentage of MPs increased from 83.1 to 92.2%, from 75 to 95.4%, and from 91.9 to 95.7% with the application of coagulation after secondary treatment in three different WWTPs (Hidayaturrahman & Lee 2019).

Since a significant percentage of MPs in WWTPs are removed by primary and secondary settling, relatively lower MP removal percentages are observed in the treatment processes applied after these treatment processes. However, the decrease

Table 7 | MP removal efficiencies of WWTPs by the methods used for tertiary treatment, including RSF, membranes, and DFs

Treatment units	Removal efficiency (%)	References
RO	98.0	Cai *et al.* (2022)
PC membrane	99.6 and 96.8	Pizzichetti *et al.* (2021)
CA membrane	99.8 and 94.3	
PTFE membrane	99.6 and 96.0	
Sand filter	83.4–100.0	Na *et al.* (2021)
AnMBR	88.4	Pittura *et al.* (2021)
MBR	79.0	Bayo *et al.* (2020)
RSF	75.4	
DF	89.7	Simon *et al.* (2019)
RSF	73.8	Hidayaturrahman & Lee (2019)
MBR	99.4	Lares *et al.* (2018)
MBR	99.9	Talvitie *et al.* (2017)
RSF	97.0	
DF	40.0–98.5	

Note: AnMBR, anaerobic membrane bioreactor; CA, cellulose acetate; DF, discfilter; MBR, membrane bioreactor; PC, polycarbonate; PTFE, polytetrafluoroethylene; RSF, rapid sand filter.

in MP removal efficiency in different processes such as coagulation after secondary settling in WWTPs does not necessarily mean that these processes exhibit low MP removal efficiency. The number and characteristics of MPs in the wastewater sample taken from the sampling point, the amount of sample collected, and whether there is a difference in sampling/analysis methods and the effect of this on MP removal efficiency should also be evaluated.

Jar tests with surface water and deionized water matrices showed that MP type and properties, coagulant type and dosage, mixing speed and water quality (pH, ionic strength, presence of contaminants in the water) affect MP removal efficiency from waters by coagulation/flocculation. Therefore, MP removal efficiency by coagulation is different in WWTPs with different wastewater properties and operating conditions. Based on the literature, MP removal by coagulation in wastewater matrix was first studied by Rajala *et al.* (2020). Rajala *et al.* (2020) in their laboratory study with 1 μm PS particles in the secondary effluent of a WWTP in Finland, with ferric chloride and polyaluminum chloride, found that the dosage required for 90% MP removal at pH 7.3 was 0.37 and 0.16 mmol/L for iron and aluminum, respectively. In addition, in the study of Rajala *et al.* (2020), it was found that less coagulant was required for the removal of larger-sized PS MPs than smaller-sized MPs in coagulation experiments performed with ferric chloride. However, the lack of research should be eliminated by increasing studies on MPs and MP removal efficiencies of different polymeric types, shapes, and sizes with wastewater samples taken from the secondary treatment outlet of WWTPs with jar tests in the laboratory. Similarly, it is necessary to contribute to limited studies by investigating the removal percentages in wastewater samples collected from the inlet and outlet of the coagulation process in WWTPs.

6. MP REMOVAL EFFICIENCY IN WWTPS AND MILLIONS OF MPS RELEASED INTO THE ENVIRONMENT

WWTPs generally consist of pretreatment, primary treatment, secondary treatment, and tertiary treatment units. While designing WWTPs, MP removal efficiency in the plant is not taken into consideration. However, it reaches many MP WWTPs in different polymeric structures, morphologies, sizes, and colors from daily used personal care products, washing machine wastewater, and leachate from solid waste landfills. Even if the MP removal efficiency is high in WWTPs and/or the MP concentration in the effluent is low, considering the treatment capacity of the WWTPs, very large volumes of MP-containing water are discharged into the aquatic environment and MPs accumulate in the aquatic environment.

In Table 8, the concentration of MPs in the influent and the effluent, the removal efficiency, and the daily amount of MP released from WWTP to the aquatic environment in some treatment plants located in different countries are summarized. For example, Murphy *et al.* (2016) stated that after the increase in WWTP of wastewater containing 15.7 MP/L, even though the MP amount decreased by 0.25 MP/L with 98.41% removal, 65 million MP was released into the aquatic environment daily. Similarly, Ziajahromi *et al.* (2017) reported that in a 308 ML capacity WWTP, wastewater contains 1.5 MP/L after primary treatment and 4.6×10^8 plastic particles will be released into the receiving environment per day. Conley *et al.* (2019) reported that 291–596 million MPs would be released into the receiving environment per day, even if the MP removal efficiency was 97.6% in WWTP with a capacity of 136×10^6 L/day. As a result, even if primary treatment, secondary treatment, or tertiary treatment is applied after primary and secondary treatment in WWTPs, millions of MPs reach the receiving environment depending on the WWTP capacity and pose a danger to the receiving environment. Therefore, it is necessary to develop new treatment technologies for more controlled management of MPs in WWTPs or to switch to WWTP applications that will keep 100% of MPs by sequential application of existing technologies.

7. REMOVAL MECHANISMS OF MPS IN DIFFERENT TREATMENT UNITS IN WWTPS

7.1. Settling and flotation tanks

The effective mechanisms for the removal of high-density and low-density MPs in settling tanks in WWTPs are gravitational settling and flotation, respectively (Kwon *et al.* 2022). MPs float or sink in wastewater depending on the density of the polymer type. Polymers such as PET, and PVC, which have a higher density than wastewater, are suitable for settling, while polymers such as PE and PP are suitable for floating. While the air bubbles given to the wastewater in flotation rise toward the wastewater surface against the direction of gravity, they carry the suspended MPs to the surface with them and the MPs on the surface are separated from the wastewater by skimming. It should be noted that the properties of wastewater, the physical properties of MPs (such as density, size, and shape) (Melkebeke *et al.* 2020), and the accumulation of pollutants on the surface of MPs (Kaiser *et al.* 2017) can change the sedimentation/floating behavior of MPs.

Table 8 | MP removal efficiencies of different WWTPs and the amount of MP released daily from these WWTPs to the receiving environment

Location	Wastewater type treated in WWTP or WWTP type	Treatment processes	Treatment capacity	Influent concentration (MP/L)	Effluent concentration (MP/L)	MP removal efficiency (%)	MP released into the receiving environment (MP/day)	Reference
Cadiz, Spain	Urban WWTP Industrial WWTP	Primary and secondary Primary and secondary	19,100,000 m^3/year 30,000 m^3/year	645.03 1,567.49	16.40 131.35	97.20 91.62	1.49–1.94 × 10^9	Franco *et al.* (2021)
USA	Residential, commercial and industrial, Residential and commercial, Residential and commercial	Primary, secondary, and disinfection (NaOCl) in all three plants	136,000,000 L/day 22,700,000 L/day 14,000,000 L/day	~ 100–240 ~ 90–190 ~ 110–230	~2–6 ~6–27 ~6–28	97.60 85.20 85	291–596 × 10^6 104–578 × 10^6 86–308 × 10^6	Conley *et al.* (2019)
Wuhan, China	Mainly contains the municipal WWTP	Primary, secondary, and chlorination	20,000 m^3/day	79.90	28.40	64.40	5.70 × 10^8	Liu *et al.* (2019b)
Beijing, China	Sewage treatment plant	Primary, secondary, and series of advanced treatments	1,000,000 m^3/day	12.03	0.59	95	0.59 × 10^9	Yang *et al.* (2019)
Xiamen, China	Seven WWTPs	Secondary WWTP	–	1.57–13.69	0.20–1.73	79.30–97.80	~6.50 × 10^8	Long *et al.* (2019)
Vancouver, Canada	Municipal wastewater and stormwater from	Primary and secondary	180,044 ML/year	31.10	0.50	97.10–99.10	3 × 10^{10} (annually)	Gies *et al.* (2018)
Adana, Turkey	Municipal WWTPs	Secondary Secondary	200,02 m^3/day 87,49 m^3/day	26,555 (MP/m^3) 23,444 (MP/m^3)	6,999 4,111	73 79	1.25 × 10^6 3.51 × 10^5	Gündoğdu *et al.* (2018)
Sydney, Australia	WWTP	Primary Primary and secondary Primary, secondaiy, and tertiary	308 ML/day 17 ML/day 13 ML/day	–	1.50 0.48 0.28	–	4.60 × 10^8 8.16 × 10^6 3.60 × 10^6	Ziajahromi *et al.* (2017)
Glasgow, Scotland	Municipal WWTP	Primary and secondary	260,954 m^3/day	15.70	0.25	98.41	65 × 10^6	Murphy *et al.* (2016)

Note: WWTP, wastewater treatment plant.

7.2. Bioreactors

In bioreactors where biological treatment takes place, MP is removed by two main mechanisms: the binding of MPs to organisms/sludge due to their hydrophobic structure (Hongprasith *et al.* 2020; Wei *et al.* 2020) and the sedimentation of MPs (Wei *et al.* 2020). The type of biological treatment process (anaerobic/aerobic) and the treatment process-specific conditions (hydraulic retention time, aeration) may also affect the improvement of the effective MP removal mechanism. In anaerobic processes, due to the higher settling velocity of large MPs (0.1–5 mm) compared to small MPs (<0.1 mm) in MP removal, large MPs are removed with higher efficiency by sedimentation mechanism (Wei *et al.* 2020). In aerobic processes, interception and sludge adsorption are the dominant mechanisms in the removal of small-sized MPs (<0.1 mm), and it can also improve MP removal by interception and sludge adsorption with the effect of turbulence caused by aeration (Wei *et al.* 2020). Studies on the degradation of MPs by microorganisms have revealed that plastics need periods of weeks/months to degrade by microorganisms (Kathiresan 2003; Yoshida *et al.* 2016). Therefore, the degradation of MPs by microorganisms is not an effective MP removal mechanism in activated sludge tanks used in conventional WWTPs with a hydraulic retention time of hours. In addition, MP concentration and properties (size, shape, polymer type) in the bioreactor may change as a result of the trapping of MPs in the sludge in the secondary settling tanks and then returning the secondary sludge to the bioreactor at a certain rate. This may lead to changes in MP removal efficiency and dominant removal mechanism.

7.3. Sand filtration

The main mechanism in MP removal by sand filtration is mechanical straining. The porosity and pore size of the filter media and the size of the MPs significantly affect MP removal by sand filtration (Sembiring *et al.* 2021). Relatively larger MPs are more easily retained on the filter surface and between the sand particles by straining during filtration (Na *et al.* 2021; Sembiring *et al.* 2021), which prevents larger MPs from reaching the outlet of the sand filter. Another mechanism in MP removal by sand filtration is the attachment of smaller particles to the grain surface in the filter media caused by van der Waals forces. Na *et al.* (2021) associated the absence of MPs $\geq$ 45 μm in size at the outlet of the sand filtration, with the strain being a predominant factor in the retention of large MPs. On the other hand, Na *et al.* (2021) found the removal efficiency of 10 μm MPs smaller than the maximum pore size in the sand filter by over 80% and they confirmed by X-ray computed tomography analysis that the attachment mechanism to the grain surface in the sand filter is effective. In wastewater, pollutants clogging the spaces between the grains and the filter surface during filtration can also lead to more retention of MPs. The effect of attraction–repulsion effects between MPs and between MPs and filter material on MP removal should also be investigated.

7.4. Membrane filtration

The main mechanism of MP removal by membranes is size exclusion and theoretically, MPs larger than the membrane pore size are retained by the membrane. However, in a study by Pizzichetti *et al.* (2021), it was determined that the physical properties of MPs and the larger-sized MPs pass through the pore sizes of the membranes due to the mechanical properties of the membrane. The adsorption of MPs to the membrane surface and pores is another effective mechanism for the removal of MPs by membranes. In particular, the hydrophilicity and zeta potential of the MP and the membrane affect the repulsive and attractive forces between the membrane surface and the MP. Breite *et al.* (2016) reported that the negatively charged PES membrane surface (−43 mV at pH 7) was contaminated by positively charged PS beads (+74 mV at pH 7) due to the electrostatic attraction, resulting in a decrease in flux permeability. In contrast, it was noted that fouling did not occur due to electrostatic repulsion between the negatively charged PS beads (−90 mV at pH 7) and the surface of the PES membrane (Breite *et al.* 2016). In addition, it was determined in the study that PS beads with different charges on the membrane surface completely blocked the membrane surface, and PS beads with the same charge did not adsorb (Breite *et al.* 2016).

A cake layer is formed on the membrane surface as a result of the accumulation of MPs and other pollutants in the wastewater on the membrane surface. Cake layer formation causes flux reduction, which is an undesirable phenomenon in membranes, but the cake layer can also act as a second membrane, increasing the removal efficiency of MPs and other pollutants. For instance, Enfrin *et al.* (2020) found that the water flux of the polysulfone UF membrane was reduced by 38% due to the interaction of NPs/MPs with the surface and pores of the membrane. On the other hand, Enfrin *et al.* (2020) also note that after filtration of PE NPs/MPs from polysulfone UF membranes for 4 h, the concentration of NPs/MPs in the permeate remained constant and after 4 h the NPs/MPs in the permeate decreased due to membrane surface fouling.

Although studies in general provide a numerical result for MP removal efficiencies with membranes in WWTPs, the number of studies evaluating MP removal of membranes concerning the properties of MPs and membranes is quite limited. In addition, in the studies conducted, MPs in WWTPs were examined under a microscope, and their shapes (Gündoğdu *et al.* 2018; Franco *et al.* 2021) were characterized, but studies on the hydrophilicity, roughness, and zeta potential of MPs in WWTPs were underestimated. Therefore, in future studies, it is necessary to examine in detail the zeta potential, hydrophilicity, roughness, and mechanical properties of different polymeric membrane materials used in WWTPs and MPs in WWTPs, and to determine the effects of these factors on MP removal efficiencies by membranes.

7.5. Coagulation

Charge neutralization and sweep flocculation are mechanisms that are effective in MP removal from wastewater by coagulation. Many factors such as the type and physical properties of MPs, coagulant type and dosage, pH, hydrolysis products distribution of the coagulant depending on pH, the surface charge of MPs, and the characteristics of the flocs formed play a role in the effective mechanisms in MP removal by coagulation (Ma *et al.* 2019; Lapointe *et al.* 2020; Na *et al.* 2021). Lapointe *et al.* (2020) added PE MPs before coagulation and 2 minutes after flocculation in the jar test, and almost the same MP removal efficiency was detected after precipitation, 81 ± 3 and $83 \pm 3\%$, respectively. This finding showed that the effective mechanism in the removal of PE MPs is incorporation into floc rather than the affinity of MPs with the coagulant (Lapointe *et al.* 2020). In another study by Ma *et al.* (2019), using 0.5 mM $AlCl_3.6H_2O$ and 0.5 mM $FeCl_3.6H_2O$ coagulants at pH 7, average floc sizes were found to be 258.6 ± 20.8 and 474.8 ± 25.6 μm, respectively. It was found that especially small-sized PE MPs were better captured by the flocs with the use of $AlCl_3.6H_2O$ coagulant due to the higher specific surface area of the smaller floc size (Ma *et al.* 2019). Na *et al.* (2021) also reported that in the removal of PS MPs by coagulation, the $AlCl_3$ coagulant exhibits superior MP removal efficiency than the $FeCl_3$ coagulant by neutralization of the surface charge, due to the stronger binding affinity of Al^{+3} to PS. Na *et al.* (2021) reported that the zeta potential of PS MPs, which was negative before the addition of $AlCl_3$, reached its maximum aggregation, with the zeta potential becoming close to zero (1.9 ± 4.1 mV), especially in slightly acidic conditions (pH = 6.0), after the addition of $AlCl_3$ (Na *et al.* 2021). However, unlike Ma *et al.* (2019) finding that smaller-sized PE MPs were removed with higher efficiency, Na *et al.* (2021) found that larger-sized PS MPs were removed with higher efficiency because they precipitated more easily after coagulation. Therefore, more studies should be conducted on the removal of MPs with different polymer types and different properties from wastewater by coagulation. In addition, there is a need to investigate the factors that are effective in the MP removal mechanism in detail.

8. INTERACTION OF MPS WITH POLLUTANTS IN WASTEWATER

Microplastics co-exist with pollutants such as heavy metals, pesticides, antibiotics, polyaromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), and bisphenols in different treatment units and effluents of WWTPs. Due to their large surface area, MPs adsorb pollutants in the effluent of WWTPs and MP-pollutants discharged from WWTPs to the receiving aquatic environment have a synergistic toxic effect on organisms. In Figure 5, pollutants sorption by MPs, sorption mechanism, and factors affecting sorption are shown.

Studies have shown that pollutant type (Llorca *et al.* 2020; Mao *et al.* 2020; Wang *et al.* 2020), pollutant concentration (Zon *et al.* 2018), MP type (Godoy *et al.* 2019; Guo *et al.* 2020; Puckowski *et al.* 2021), MP concentration (Wang *et al.* 2020), properties of MP (Fang *et al.* 2019; Mo *et al.* 2021; Yao *et al.* 2022), pH (Fang *et al.* 2019; Guo *et al.* 2020), ionic strength (Guo *et al.* 2020), and organic matter concentration (Godoy *et al.* 2019; Guo *et al.* 2020) are effective on the adsorption of different pollutants to MPs in the aquatic environment. In addition, studies have shown that many different mechanisms such as electrostatic interactions (Guo *et al.* 2019; Sharma *et al.* 2020; Puckowski *et al.* 2021; Yao *et al.* 2022), hydrogen bonds (Zhang *et al.* 2018; Guo *et al.* 2019; Yao *et al.* 2022), hydrophobic interactions (Puckowski *et al.* 2021; Yao *et al.* 2022), and $\pi - \pi$ interactions (Liu *et al.* 2019a; Sharma *et al.* 2020) are effective in the adsorption of pollutants to MPs, depending on MP and pollutant properties. Most of the existing studies on the adsorption of MP and pollutants to date have been carried out in distilled water (Fang *et al.* 2019; Godoy *et al.* 2019; Guo *et al.* 2019) and surface waters (Mai *et al.* 2018; Godoy *et al.* 2019; Ta & Babel 2020; Selvam *et al.* 2021). Studies on the adsorbing of pollutants by MPs in wastewater samples collected from WWTPs or in synthetically prepared wastewater are very limited. Nikpay (2022) investigated the adsorption of pollutants on two types of PP-based polymers (atactic PP and isotactic PP) in synthetic wastewater solutions containing organic, inorganic, and organic–inorganic fines and proved that the adsorption depends on the polymer type, the polymer surface, and the wastewater type. Godoy *et al.* (2019) found that PE, PP, PET, PS, and PVC MPs in urban wastewater

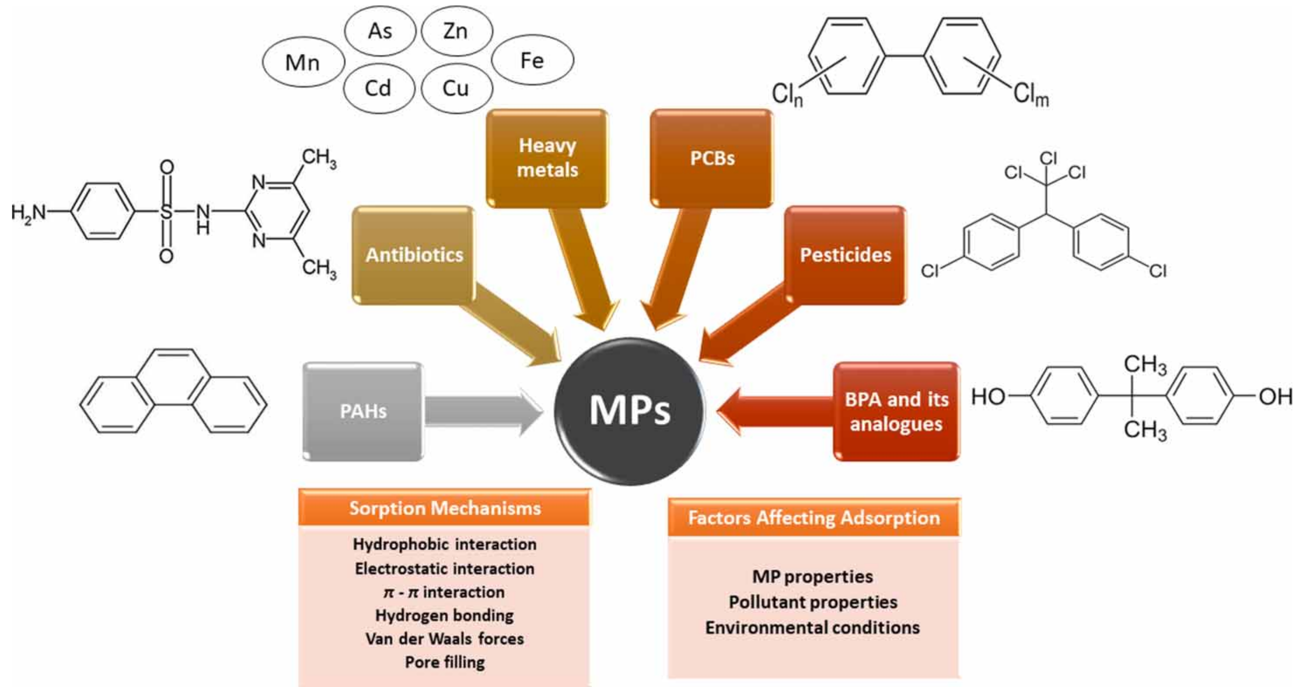

Figure 5 | Pollutants are sorbed by MPs, sorption mechanisms, and factors affecting sorption.

adsorbed Pb more than MPs in seawater and pure water. Godoy *et al.* (2019) suggested that this is due to the fact that metal and organic pollutants interact with hydrophobic interaction or complexation, and the organic matter competes for the adsorption sites of MP. Since WWTPs have MPs in different amounts and properties, different wastewater properties, and different operating conditions, the concentration and properties of MPs and organic/inorganic pollutants in their effluents also differ. Since the effluent of WWTP is responsible for the transfer of MPs and other pollutants in WWTPs to the aquatic environment, more MP-pollutant adsorption studies should be carried out, especially in real wastewater samples collected from effluents of different WWTPs. Thus, it can be understood which pollutants are more adsorbed to MPs in the effluent of WWTPs and pose more danger in the aquatic environment, and new strategies can be developed in WWTPs for precautionary purposes.

9. CONCLUSIONS AND FUTURE PERSPECTIVES

This paper reviews research in the literature examining the sources, properties (type, shape, size, and color) of MPs in WWTPs, and the MP removal efficiencies and removal mechanisms of treatment units in WWTPs. As a result of the examination of the studies in the literature, the current situation and the areas that need further research are summarized below:

- Conditions such as the concentration and distribution of the properties (polymer type, shape, size) of MPs in the influent of WWTPs are different, and the treatment technologies applied in WWTPs are different, causing the MP removal efficiency of WWTPs to differ from each other. In addition, the lack of a standard sample preparation method for MP analysis in wastewater and the different MP size ranges that researchers evaluated in MP analysis also cause different MP removal efficiencies in WWTPs.
- In the influent and effluent of WWTPs, PVC, PEST, PE, PP, and PA types of MP, which are used more frequently in daily life, are more common. Particularly, fiber-structured MPs released from synthetic clothes washed in the washing machine are the most dominant MP shape type in WWTPs. In addition, in general, the removal of fiber-structured MPs in WWTPs is more difficult than the removal of MPs in other shapes, and a significant amount of fiber-structured MP is released into the receiving environment. For this reason, studies on higher efficiency removal of the above-mentioned polymers and fiber-structured MPs from WWTPs should be given priority in future research.

- Although the size distribution of MPs in WWTPs has been studied by many researchers, the examination of MPs <100 μm in size has mostly been neglected. The possibility that MPs are gradually fragmented into smaller sizes and that small MPs can pass through the treatment units more easily should be taken into account, and information on the percentage ratio of especially small MPs (1 μm–1 mm) in samples taken from WWTPs should be considered more in future studies.
- MPs trapped in the sludge of the primary settling and secondary settling tank are generally high density, fiber and fragment form MPs <1 mm in size. Therefore, studies dealing with the removal of MPs from sludge should focus more on MPs with these properties.
- In WWTPs, in addition to primary and secondary treatment, tertiary treatment technologies should be applied to ensure better MP removal efficiency. However, 100% MP removal efficiency cannot be achieved in WWTPs where even tertiary treatment is applied, and millions of MP reach the receiving environment even in 1 day, depending on the WWTP capacity. Therefore, after the polymer type, size, and shape of the MPs that are planned to be removed from WWTPs have been thoroughly determined, there is still a significant need to determine and develop the most appropriate treatment methods/technologies for the removal of these MPs.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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