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Chapter 11 - Microplastics in wastewater treatment plants

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Abstract

This chapter reviews recent developments in microplastic-related research; identifies major sources of microplastics to wastewater treatment plants (WWTPs); investigates the detection methods, concentration levels, and removal efficiency of microplastics at selected WWTPs; and presents a discussion on microplastics removal from wastewater and sludge using biological wastewater treatment processes. The findings are subsequently summarized to suggest potential improvements and future directions for research and development addressing the issue of microplastic pollution. The research identified that supportive policy measures coupled with effective WWTP designs are important for minimizing microplastic-related pollution. In addition, knowledge of the origins of microplastics is useful for WWT system designers, practitioners, policymakers, product designers, and other stakeholders. While sampling and analytical methods continue to evolve, several recommended best practices include focusing on the minimization of contamination, reproducibility, and the applicability of methods in WWTP facilities. Because microplastics can include complex composite polymers with additives, adsorbents, and biofilm, further research on toxicity, chemical leachability, and pathogenic biofilm properties is needed to understand the potential impact on biological wastewater treatment systems.

Keywords

Microplastic; wastewater treatment; down the drain; street runoff; microplastics in sludge; analytical methods; combined sewer; sampling

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CHAPTER

11

Microplastics in wastewater treatment plants

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11.1 Introduction

p0010 Plastic debris are found in all shapes and sizes (Fig. 11.1); debris larger than 1 μm and smaller than 5 mm in diameter (or about the size of a sesame seed) are called microplastics. According to the literature, the definition of microplastics varies to some extent. Initially, microplastics were defined as plastics less than 5 mm in length. However, with continued scientific inquiry into the topic, nanoplastics were also identified, leading to an expanded definition, including a lower size limit of 1 μm (Encyclopedia Britannica, 2020). On rare occasions, microplastics have also been defined in the size range of 0.3–5.0 mm (Wu et al., 2019). The basis of this definition may be associated with the common sampling with nets around 0.3 mm in size.

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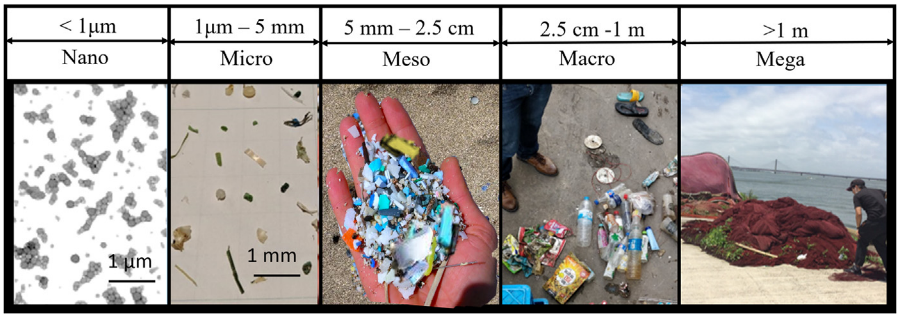


f0010 **FIGURE 11.1** Size-based classification of plastics.

However, in this chapter, the size range illustrated in Fig. 11.1 will be used, as this is the most widely recognized definition of microplastics within the scientific community.

p0015 Microplastics can also be categorized into two groups based on origin: primary microplastics, which enter the environment directly as microplastics, and secondary microplastics, which originate from the breakdown of larger plastics such as meso-, macro-, and mega-plastics in the environment. Microplastics in a water environment (including wastewater) can be subclassified into several categories such as floating, suspended, settled on the bed, associated with solids, and in biota (Abeynayaka et al., 2020). This classification depends on the specific properties of the plastic, such as composition, specific gravity (SG), and shape, as well as the hydraulics of the water body (Anderson et al., 2016). In addition, plastic particles can change not only in size but also possibly in SG, by aggregation, degradation, or biofilm growth (Kowalski et al., 2016).

s0015 11.2 Impacts of plastic pollution on ecosystems, food chain, and human health

p0020 More than 600 marine species have been found to be negatively affected by plastic ingestion, including 86% of all sea turtle species and about half of all seabird species (Gall and Thompson, 2015). Ingestion of plastic debris is widespread because birds and other marine species often mistake it for prey. Ingesting plastic can have many serious consequences, including false satiety leading to starvation and suffocation (Stelfox et al., 2016). Microplastics can also be bioaccumulated and biomagnified up the aquatic food chain, with concomitant impacts on human health (Akhbarizadeh et al., 2019). Although the effects of microplastic ingestion on human health are not yet fully understood, they are known to travel through the human digestive tract and into vital organs. In addition, microplastics can contain toxic contaminants (e.g., bisphenol A, phthalate plasticizers, carcinogens, polybrominated flame retardants, and heavy metals), which are either derived from the plastic itself or absorbed from the surrounding environment. Ingestion of these toxic chemicals has been found to cause health complications including cancer, cardiovascular disease, and diabetes (Gallo et al., 2018).

p0025 Microplastic-related studies have grown over the past decade with a monotonically increasing trend, especially in the last 10 years. Compared to the general research topic of microplastics, there is less research focused on microplastics in wastewater. Until 2020, wastewater- and microplastic-related research comprised less than 10% of the total published research discussing the issue of microplastics. At present, however, examinations of microplastics in wastewater treatment plants (WWTPs), targeting the sampling and analytical methods of microplastics in wastewater matrices, and the removal efficiency of microplastics at various levels/configurations in relation to different types of WWTPs are attracting greater research attention. The growing focus on the development of analytical methods is understandable, given that microplastics in wastewater remain a relatively novel topic, and the complexity of wastewater matrices has led to complications in sampling, recovery, and detection of microplastics (Parrish and Fahrenfeld 2019). Apart from the peer-reviewed publications, in recent years, the European Union (EU)- (Sabbah et al., 2019) and the United States Environmental Protection Agency (USEPA)-funded projects (Cook and Allen, 2020) have issued the best practice recommendations for detecting and analyzing microplastic concentrations in wastewater. Recent developments in terms of sampling/analytical methodologies and identification of best practices for the collection, preparation, and analysis are expected to incentivize continued research on microplastics in wastewater.

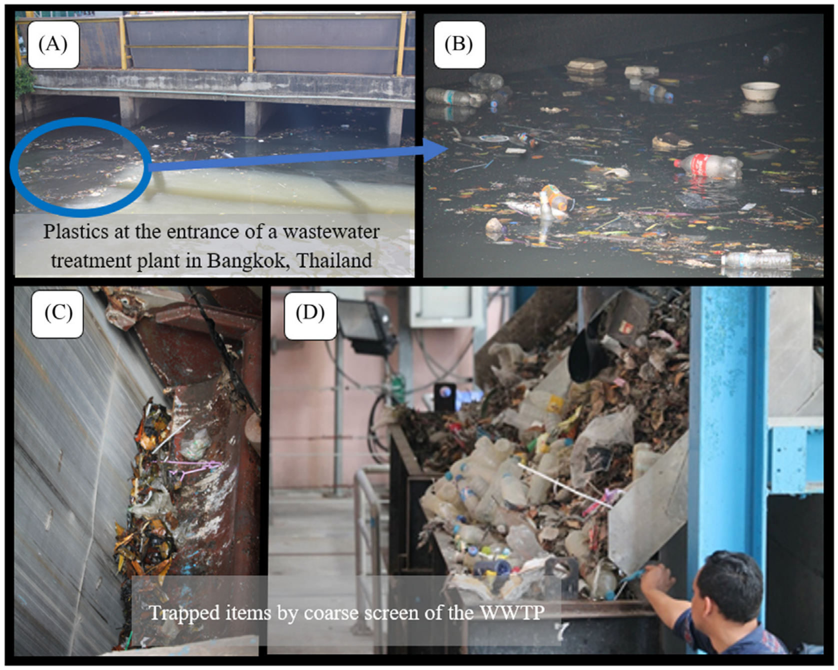
p0030 Nevertheless, at present, a limited number of studies are focused on the biodegradability of microplastics in WWTP and chemical leakage of microplastics in wastewater treatment systems. Despite this, the growing attention on bio-based and biodegradable plastics in the plastic industry may potentially lead to further examination of biodegradability in WWTP and sludge disposal options, including land applications (Keller et al., 2019, Lakhawat et al., 2020).

p0035 Microplastic-related pollution studies were initially focused on marine environments where plastic accumulation and degradation mostly occurs (Min et al., 2020). Sampling by a vessel dragging manta nets in marine environments provided evidence for the presence of small-sized anthropogenic particles (microplastics). Moreover, land-based primary or secondary microplastics have also been found to contaminate ocean water (Abeynayaka et al., 2020; Lebreton et al., 2017). In this context, studies on land-based microplastics emissions have focused on marine pollution, terrestrial and freshwater, or atmospheric and marine. The microplastic-related research is often multicompartmental (due to properties such as high mobility and longevity), and the development of sampling and analytical methods to detect microplastics in soil, freshwater, and other biota is highly relevant to microplastic-related research. Wastewater-related studies often focus on the origins of microplastics (mostly land based), wastewater treatment plant operations (in view of microplastic discharge into water environments), and microplastic transfer from wastewater to sludge (intercompartmental movement) in WWTPs.

s0020

11.3 Plastics and wastewater treatment plants

p0040 Plastics from domestic and commercial activities can enter sewerage systems. Especially in developing countries, macroplastics frequently enter sewerage systems on a mass scale (Fig. 11.2). However, coarse and fine screens can trap macroplastics and mesoplastics (as defined in Fig. 11.2) at the preliminary/primary treatment stages of the wastewater treatment process (as well as a fraction of microplastics, depending on the screen size). Consequently, potential leakage of macroplastics and mesoplastics into secondary



f0015 **FIGURE 11.2** Macroplastic and mesoplastic debris retained in a coarse screen at a WWTP in Bangkok.

(biological WWT) systems is negligible. Taken together, the treatment, disposal, and associated management of macroplastics and mesoplastics fall under more general solid waste management. Recovery of plastics and subsequent recycling and waste-to-energy applications are recommended treatment options. From this point onward, this section focuses only on the microplastics fraction of plastics entering WWTPs.

p0045 Microplastics that enter WWTPs are partially removed at primary treatment facilities (Fig. 11.3). Higher plastic polymer density (in polymers such as PVC and PET), composite material properties (elastomers with additives, multilayered plastic with metal layers), and aggregation into other high-density particles can be the main reasons for microplastics settling in primary settling tanks (Carr et al., 2016). Subsequently, in the common activated sludge process (ASP), a portion of microplastics can be associated with biomatrices and transferred into the sludge phase (microplastics associated with sludge are discussed in Section 11.4). The free-floating portion of microplastics entering tertiary treatment units could be further removed based on the tertiary treatment options (such as filtration). The reported removal efficiencies are given in detail in Section 11.3.

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11.4 Microplastics in biological wastewater treatment

p0050 Microplastics that enter centralized wastewater treatment systems can be traced to multiple origins. For simplicity of understanding the entrance pathway can be divided into two origins:

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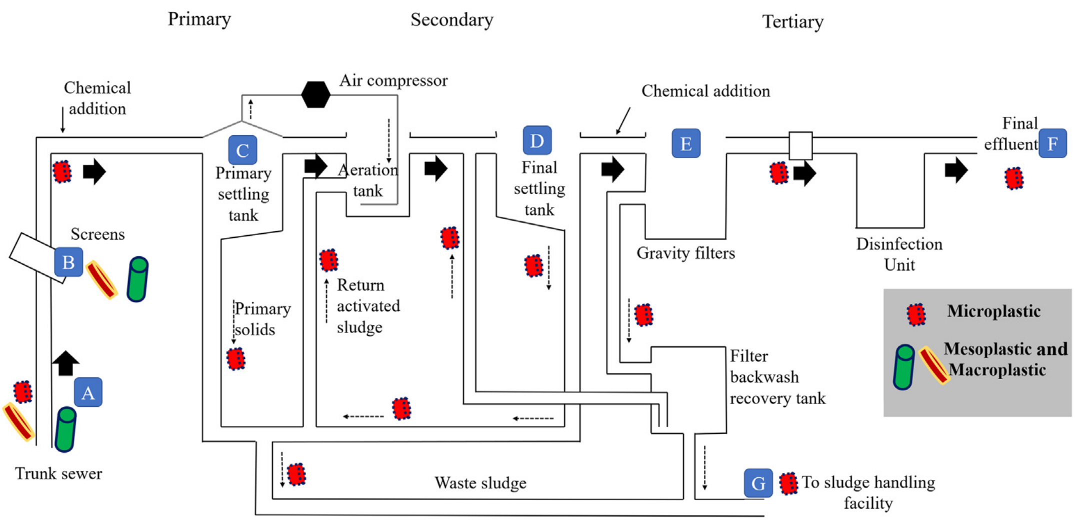

f0020 **FIGURE 11.3** Typical wastewater treatment process and removal of plastics in various units of WWTP.

items that go "down the drain," which includes microplastics from kitchen sink wastewater, bathroom wastewater, and toilet flushes. The second origin concerns "street wash" (the street runoff) reaching the WWTPs through combined sewers (Fig. 11.4).

p0055 The "down-the-drain" category includes primary microplastics such as microbeads from personal-care products (microbeads are tiny pieces of plastics added as exfoliants, to health and beauty products, such as some cleansers and toothpaste), and textile microfiber from washing machines or other clothes washing activities (such as handwashing). These types of microplastics enter WWTPs through either combined sewers or separate sewer systems. There can also be smaller fractions of secondary microplastics that originate from plastics in WWT systems or from household items. "Street wash," or stormwater runoff, carries primary microplastics such as tire abrasion particles, broken road-marking paint, decomposed plastic household materials due to prolonged exposure to the sun, and secondary microplastics that originate from macroplastic debris.

p0060 Through either combined or separate sewers, WWTPs act as major receivers of microplastics. The diversity of plastic polymers and plastic-associated chemicals (Rochman et al., 2019) can vary depending on the sewer network catchment characteristics. Knowledge of the origin of microplastics is useful for WWT system designers, practitioners, policymakers, product designers, and other stakeholders (including educators, who can advise people on how to recycle plastics). System design approaches can be top-down, bottom-up, or hybrid. For example, textile microfibers that are trapped at the household level can be addressed with technological interventions such as redesigning washing machines or improving the removal efficiency at WWTPs through advanced processes. An additional example concerns broken tire abrasion particles: environmental contamination can be minimized through speed control, promotion of nontire-based vehicle transport modes (i.e., trains), or expanding combined sewer networks, especially in high-traffic-density areas (coupled with effective wastewater treatment technologies and sludge management approaches in WWT systems).

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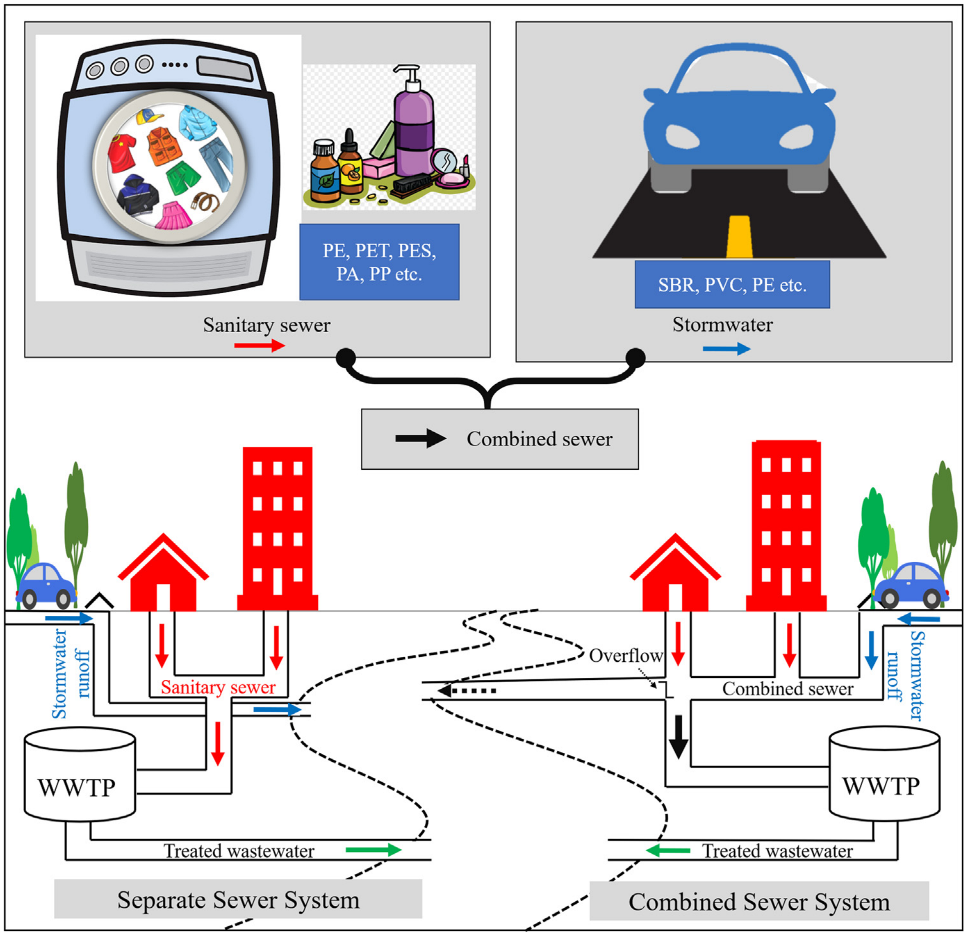

f0025 **FIGURE 11.4** Microplastic inflow to WWTPs through separate and combined sewer systems. [Polyethylene (PE); polyethylene terephthalate (PET); polyethersulfone (PES); polyamide (PA); polypropylene (PP); styrene-butadiene rubber (SBR); polyvinyl chloride (PVC)].

s0030 11.4.1 Down-the-drain microplastics

p0065 A major fraction of "down-the-drain" microplastics is primary microplastics from the textile washing process. Addressing fibrous microplastics derived from textiles is challenging not only due to the microplastic content but also associated chemicals such as perfluoroalkyl acids (PFAAs) (Henry et al., 2019). Research conducted using domestic washing simulations in Italy suggests that significant amounts of microfibers are released from polyester and polypropylene fabrics (De Falco et al., 2018). Microfiber released during washing with detergent can vary from 650 to 3500 fibers per gram of cloth. Even though many studies focus on cases that involve washing machine–based clothes washing, handwashing is also a common method practiced in developing countries. Hence, it is also interesting to investigate potential microfiber leakage into the water during the process of washing clothes by hand. Tian et al. (2021) identified that the average length of microfiber from either hand or machine washing is approximately 600 μm. While microfiber lengths fall in a similar range, the quantity of microfiber released was found to be significantly different. The median values of the number of microfibers released were 10,500 fibers per item from hand-washed new clothes and 75,200 fibers per item for machine-washed new clothes (Tian et al., 2021). Considering these differences, the inflow of textile microfibers into WWTPs can vary in a large range based on the “wastewater catchment” characteristics, such as the methods of washing clothes.

p0070 Microbeads, an ingredient added to personal-care and cosmetic products (PCCPs), which are also discharged into WWTPs through daily human activities, are another major “down-the-drain” microplastic material (Ding et al., 2020). In China, it is estimated that more than 80% of microbeads emitted into the aquatic environment originate from incomplete removal in WWTPs (Cheung and Fok, 2017). Despite the lower mass, due to the AU:4 smaller size range of microbeads, their estimated quantity is projected to be enormous (Gouin et al., 2015). Microbeads used in the cosmetics industry are often made of polyethylene (PE) or polypropylene (PP). It is estimated that 10,000 tons of microbeads per year are released into the environment through the use of personal-care and cosmetic products (Ryberg et al., 2019). Hence, microbeads can also be considered as major microplastics entering WWTPs. With regard to “down-the-drain” microplastics (mainly coming from washrooms), WWTPs can be considered as potential barriers to microbeads. Although WWTPs may potentially be effective in preventing environmental contamination, direct human contact while using PCCPs is unavoidable. Hence, upstream measures such as regulating the use of microbeads are widely discussed at the policymaking level. Many developed countries (The Netherlands, Australia, Canada, Italy, South Korea, New Zealand, Sweden, United Kingdom, and United States) have regulated the use of microbeads in personal-care products (OECD, 2021).

s0035 11.4.2 Microplastics from street wash runoff

p0075 Tire abrasion was found to be the largest source of microplastics contaminating the environment. Globally, such microplastics are estimated to make up 1.4 million tons per year (Ryberg et al., 2019). Life cycle analyses suggest that roughly 20% of synthetic rubber in a tire is displaced its lifetime (Boucher and Friot, 2017). For urban areas, where half of the world population lives, tire abrasion particles are likely to enter sewage systems. This can either reach WWTPs through combined sewers or be released directly to aquatic environments with stormwater runoff. While the SG is 0.94 for tire elastomers, such as SBR, the vulcanized SBR used in tires has an SG of about 1.2 (Bondan, 2019), which makes these rubber particles nonbuoyant in water environments. Moreover, the size of microplastics from tire abrasion might be below the detection limit in water environments, as the typical opening size of sampling nets is about 300 μm. In addition, there is also the possibility that microplastics associated with tires are captured WWTPs before reaching water environments or being released into terrestrial environments.

p0080 Fragments of broken road markings are another common form of microplastic associated with stormwater runoff originating from streets. These particles can enter WWTPs through combined sewers. Fragmented road markings were reported at high concentrations in cities where there were large amounts of road markings and high traffic density. At the same time, combined sewers are often connected to city sewer networks. There were reported cases of fragmented road markings detected in city dust [e.g., in Japan, Nepal, and Vietnam (Yukioka et al., 2020) and Norway (Vogelsang et al., 2018)]. Various acrylic resins, including methyl 2-methyl propanoate (PMMA), were detected in road dust samples (Kitahara and Nakata 2020; Yukioka et al., 2020). Properties such as good weatherproofing and paintability of acrylic resins allow for wide application of such resins for various road markings such as lane markings, road crossings on general roads, urban roads, and highways. Toxic chemicals associated with some resins are substantial, and their effects on biological WWTPs need to be considered (see Section 11.5 for further discussion).

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11.5 Microplastic detection methods, presence, and removal efficiency in wastewater treatment plants

p0085 Section 11.3 discusses the microplastic detection methods, especially focusing on WWTPs, the reported information on microplastics in various steps of the wastewater treatment process, and the removal efficiency of microplastics in WWTPs.

s0045

11.5.1 Microplastic sampling in wastewater treatment plants

p0090 Several studies conducted over the past decade have focused on developing analytical methods for sampling, extraction, and identification of microplastics in complex environmental media. However, at present, no standardized sampling or analytical protocols have been agreed upon for international use. Sampling methods can be divided into two categories: grab sampling of smaller volume and on-site filtration sampling (using nets or sieves) of comparatively large volume. Grab sampling mostly focuses on sediment, sludge, or (comparatively) smaller sized microplastic analysis. On-site filtration sampling using nets or sieves for floating and suspended microplastics can be conducted by dragging a net (often a plankton net) or pumping water through a net (or a sieve). Nets (commonly referred to as plankton or manta nets) with mesh opening sizes of about 100 and 300 μm are conventionally used for sampling microplastics in marine or freshwater environments (Lima et al., 2015; Maes et al., 2017a; Zhang et al., 2020). However, dragging such nets is impractical in WWTPs. To overcome this, motorized units coupled with nets and flow measuring devices (Fig. 11.5B) have been introduced. Nevertheless, large volume sampling with nets is impeded by the higher amounts of suspended solids in WWTPs, which often results in clogging. Increasing the opening size of the net to minimize clogging would adversely affect the capture of microplastics. This is especially important when considering the size range and the shape of microfibers derived from textiles, as such nets have been found to be ineffective at capturing small- to medium-sized fibers (discussed further in Fig. 11.6).

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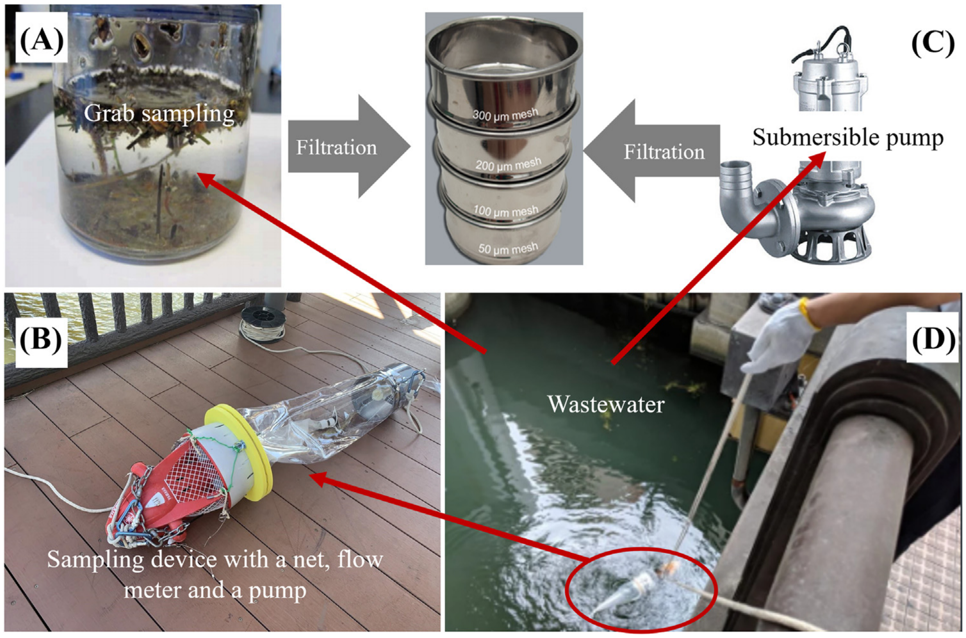
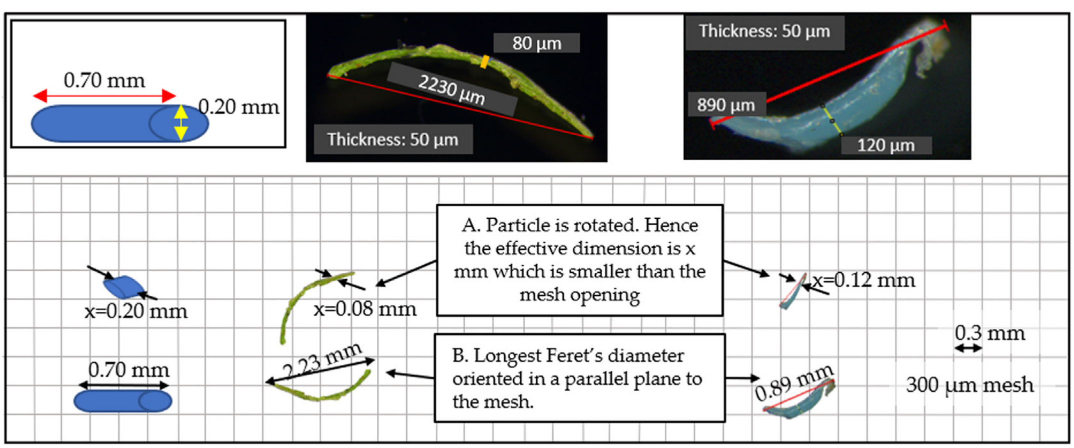


f0030 **FIGURE 11.5** (A) Grab sampling followed by filtration, (B) large water volume microplastic sampling device (Pirika Inc.), (C) submersible pump coupled with sieving system for filtration, and (D) sampling at a biological WWTP in Yokohama, Japan.



f0035 **FIGURE 11.6** Effect of size and shape of microplastics on retention with net-based sampling. Source: *Modified from Abeynayaka, A., Kojima, F., Miwa, Y., Ito, N., Nihei, Y., Fukunaga, Y., et al., 2020. Rapid sampling of suspended and floating microplastics in challenging riverine and coastal water environments in Japan. Water 12 (7), 1903. https://doi.org/10.3390/w12071903.* AU:7

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p0095 The best practices and methods for collection, preparation, and analysis recommend that sampling should be straightforward, simple to follow, reproducible, and designed to prevent contamination (Cook and Allen, 2020). To avoid contamination, materials such as stainless steel are recommended for sampling devices and equipment. This is an alternative to PA (polyamide or nylon) net-based sampling devices. It is recommended to use a submersible pump (Fig. 11.5C) or peristaltic pump with the sampling apparatus. The collection of sludge samples is often conducted using waste-activated sludge in the location illustrated (G) in Fig. 11.3 (Lares et al., 2018; Li et al., 2018), sludge from digesters (Bayo et al., 2016), or at sludge posttreatment facilities (Jiang et al., 2020). Composite and simple grab sampling is often practiced in the case of WWTP sludge sampling.

p0100 As mentioned above, Fig. 11.6 depicts the influence of size and shape on the retention of microplastics by nets. In this case, fibers have a higher potential to escape through the nets even though the length of the fiber exceeds the net opening. Hence, microplastic sampling activities at WWTPs may indicate less microfiber recovery from wastewater than is actually the case. Grab sampling presents another option but may limit the sample volume. Ultimately, grab sampling is also associated with filtration through sieves (or nets) at the lab to separate the suspended solids from the wastewater (Fig. 11.5A). In the case of lower volume the application of nets or filter papers with smaller openings is possible with vacuum filtration if needed (Ben-David et al., 2021; Sabbah et al., 2019; Schmiedgruber et al., 2019). In studies focusing on the lower range of microplastics or fibrous microplastics the preferred method is grab sampling followed by highly sensitive analytical methods (discussed in detail in Section 11.5.2). Nevertheless, it is possible to apply large volume methods in secondary or ter- AU:5 tiary treatment effluents where the suspended solid concentrations are comparatively low. However, due to the lower concentrations of large microplastics in such effluents, net-based sampling (Fig. 11.5B) has been found to be less productive. Large volume sampling with sieves (as described in USEPA, 2020; extensively discussed by Sun et al., 2019) is the recommended method (Fig. 11.5C) to detect low concentrations of small-sized (and/or fibrous) microplastics (Dyachenko et al., 2017).

p0105 Due to the challenges outlined above, the application of a uniform method at various levels of WWTPs and sludge treatment makes observations difficult, especially when comparing the removal efficiency and fate fractions of microplastics in field situations. To address these challenges, several studies have been conducted with metal-doped microplastics to improve removal efficiency and fate analyses (Frehland et al., 2020; Keller et al., 2019; Schmiedgruber et al., 2019).

p0110 Microplastic extraction from collected samples (i.e., samples retained on nets and sieves) and separation from other debris (such as biosolids) represents another important sampling step. In the case of environmental samples the often-used protocol for separating microplastics from other debris is density-based separation (using salt solutions of NaCl, $ZnCl_2$, KI, etc.) and degradation of organics (Fenton's degradation) (Tagg et al., 2017). The protocols are described extensively in environmental microplastic-related publications (Ben-David et al., 2021). Fig. 11.7 illustrates the experimental flow of sampling, extraction, and analysis.

p0115 Table 11.1 provides the SG values of typical plastic polymers and solutions used for density-based separation of microplastics. Common low-density polymer types can be readily separated using saturated NaCl solutions. However, for high-density polymer

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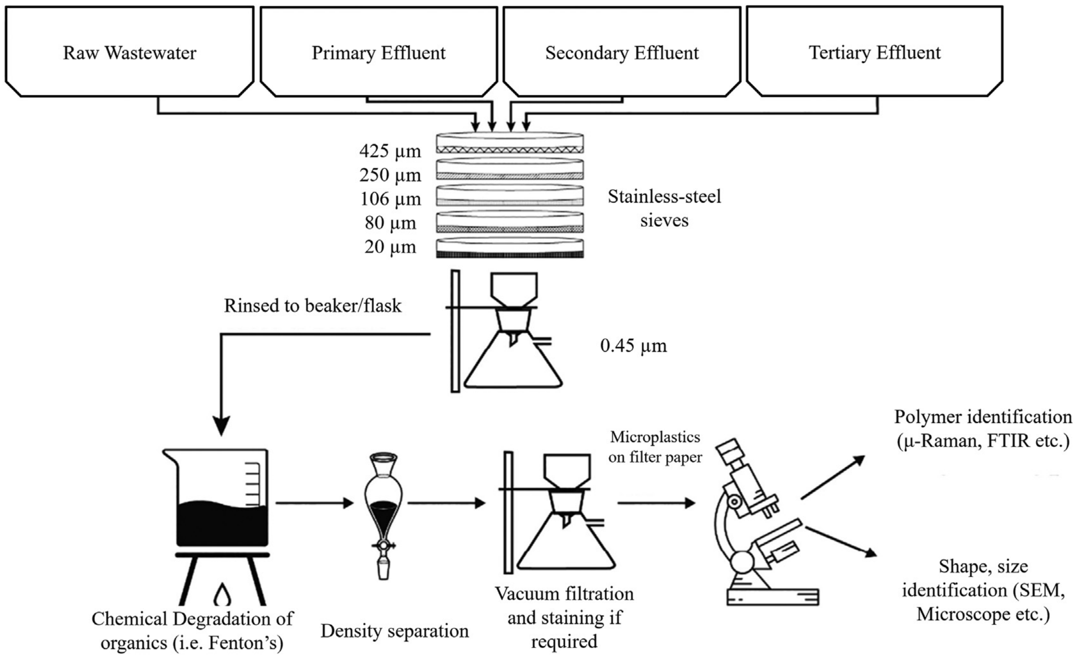

f0040 **FIGURE 11.7** Typical microplastic sampling and analytical process flow. Source*: Modified from Ben-David, E. A., Habibi, M., Haddad, E., Hasanin, M., Angel, D.L., Booth, A.M., et al., 2021. Microplastic distributions in a domestic wastewater treatment plant: removal efficiency, seasonal variation and influence of sampling technique. Sci. Total Environ. 752, 141880. https://doi.org/10.1016/j.scitotenv.2020.141880.*

t0010 **TABLE 11.1** Density of common plastic polymers and solutions commonly used for density separation.

Plastic or solution	Specific gravity	Reference
Polypropylene (PP)	0.85–0.94	Lambert and Wagner (2018)
Polyethylene (PE)	0.93–0.97	Alaerts et al. (2018)
Polystyrene (PS)	0.96–1.05	Lambert and Wagner (2018)
Polyethylene terephthalate (PET)	1.35–1.39	Alaerts et al. (2018)
Polyvinyl chloride (PVC)	1.10–1.45	Alaerts et al. (2018)
Polymethyl methacrylate (PMMA)	1.09–1.18	Grigorescu et al. (2019)
Styrene-butadiene rubber (SBR)	0.94–1.295	Bondan (2019)
Polyamide (PA)	1.12–1.14	Lambert and Wagner (2018)
NaCl solution (saturated)	1.20	Moffitt Schall and Myerson (2019)
$ZnCl_2$ solution (60%–65% saturation)	1.60–1.80	Rodrigues et al. (2020)
KI solution (saturated)	1.67	Moffitt Schall and Myerson (2019)
Bulk sludge	1.4	Tang and Sillanpää (2018)

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types such as PVC and PET, a saturated NaCl solution may not be effective for purposes of separation. In such situations, $ZnCl_2$ or KI solutions are effective. However, the high cost (more than tenfold more expensive) and the special disposal measures required are among the drawbacks of KI and $ZnCl_2$ solutions. In view of these issues, it is suggested that researchers carefully select the appropriate solution for the density separation based on the objective of the study (i.e., the target polymers to be detected).

p0120 Microplastic separation/extraction from sludge matrices is challenging, considering that the bulk density of sludge is approximately 1400 mg/L (Tang and Sillanpää, 2018). On the other hand, microplastics associated with biofilms may have adverse effects on simple physicochemical separation activities. In this context, degradation of organics should be conducted as a first step (as illustrated in Fig. 11.7), after which, if required, density separation is recommended to separate the plastic polymers from the inert material. Thereafter, polymer identification can be conducted. Li et al. (2018) reported 22,700 MP/kg sludge weight by density separation using NaCl solution followed by organic degradation with 30% H_2O_2 method with sampling. The authors reported steering the NaCl−sludge solution for the separation of microplastics from biomass.

p0125 Several researchers have used tweezers to examine and pick microplastics from sludge matrices (Lares et al., 2018). In the case of dried sludge, researchers have added small amounts of distilled water to break up the sludge material and then identified microplastics using an optical microscope. The authors have used additional steps such as changing the background color to enhance the recovery of microplastics during the analysis. This background color variation helps to distinguish microplastics from sludge based on color. However, the full recovery can be affected, and it depends on the skills of the analyst. In this context, total macroplastic concentrations reported for sludge samples may be underestimated.

p0130 As outlined above, sampling methodologies are diverse and continuing to evolve. Researchers need to assess their research objectives and identify the scope on a case-by-case basis and select the appropriate methods to obtain clear results.

s0050 11.5.2 Analytical methods for microplastics in wastewater and sludge

p0135 Once the sampling has been done, certain properties of microplastics can be analyzed. Rochman et al. (2019) reviewed the physical and chemical properties of microplastics: the common physical properties are mass, shape, and color and the chemical properties are polymer-type and associated chemicals. In the context of WWTPs, both the physical and chemical properties are useful information. Table 11.2 summarizes the important features of the analytical equipment used in microplastic-related research. μ-Raman and FT-IR-based analytical methods have often been used by researchers for polymer identification. For polymer identification using microscopy, often fluorescence staining (such as Nile Red) is used (Erni-Cassola et al., 2017; Maes et al., 2017b). Sierra et al. (2020) reported microplastics detection in wastewater samples with a polarized optical microscope. Apart from polymer identification, the detectable size range, affordability, and time needed for the analysis are important considerations in the selection/use of equipment.

p0140 The detection limit (the smallest particles to be detected) is also an important parameter to consider. The detection limit depends not only on the equipment but also on the

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t0015 **TABLE 11.2** Features of analytical equipment.

Features	μ-Raman	μFT-IR	ATR-FT-IR	Microscope-based	Scanning electron microscope	Pyrolysis GC–MS
Possible equipment price range[a]	3		2	1	5	3
	Prices vary from USD 20,000 to 1,000,000. Ranking is done from 1 to 5 based on relative price (lower number = cheaper equipment).					
Type of polymer	Yes				No	Yes
Detectable additives	Pigments	No				Yes
Particle surface chemical	Yes	No	Yes	No		
State of degradation	Surface oxidation	No	Surface oxidation	No		
Suitable sampling sites	Wastewater, water environment, drinking water		Wastewater, water for larger microplastics	Wastewater, water environment	Nanoplastics and small microplastics with known polymers	Wastewater, water environment
Dimension of specimen mass	ng-μg		μg-mg	ng-μg	ng	mg
Number of measurable particles per sample	$10^2–10^5$		One at a time	Microscopic visibility	Visibility	Depends on sample mass
Preparation and measuring time	Hours to days		minutes	hours		minutes to hours
Detection level	>5 μm		>80 μm	>5 μm	>1 nm	Depends on sample mass
Example reference[a]	Wolff et al. (2019)	Mintenig et al. (2017)	Simon et al. (2018)	Sierra et al. (2020)	Nguyen et al. (2021)	Hermabessiere et al. (2018)

[a] *Equipment prices were obtained through personal communication with the leading manufacturers (as of 2021) and available information on manufacturers' homepages. Reference is given for further reading as a case study of equipment usage. Tabulated information does not necessarily represent example reference content.*

analytical skills of the operators. Research related to a smaller range of microplastics (1–100 μm) is hindered due to the unavailability of analytical equipment and a robust method. Moreover, there is no acceptable method of detecting plastic polymers in the nanoplastic range. There were reported studies of nanoplastics detected in marine environments with pyrolysis GC–MS, however, the sample quantity is an influential factor (Ter-Halle et al., 2017). Confirmation of the particles extracted from wastewater and sludge matrices in the range of nanoplastics has not yet been done. However, in special situations such as polymer degradation (with known polymers), the size reduction observation type of analysis is possible with SEM (von der Esch et al., 2020). Another challenge is analyzing plastic-related chemicals, such as toxic metal analysis. Common metal analysis methods

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such as inductively coupled plasma mass spectroscopy (ICP-MS) require a sample weight of several grams. However, the weight of a microplastic is less than a milligram (mg). This limits the analysis of toxic metals in microplastics. However, there have been several efforts in toxic metal analysis of larger-sized microplastics using X-ray fluorescence spectroscopy (Abeynayaka et al., 2021; Turner, 2017).

p0145 The selection of analytical equipment needs to be done based on various factors. The research objectives should be in line with the available facilities to provide a meaningful outcome. For example, if the available analytical equipment is ATR-FT-IR, studying microplastic removal efficiency by a membrane bioreactor (MBR) or analyzing grab samples with small volumes of tertiary treated effluent would not provide a meaningful scientific output.

s0055 11.5.3 Microplastic concentrations in wastewater treatment plants

p0150 The reported microplastic concentrations in WWTPs should be analyzed with care to avoid misunderstandings about their magnitude. For example, as discussed in Section 11.3, there are different sampling and analytical methods. Based on the sampling method, the range of macroplastics capture can vary. Moreover, the lower limit of analytical equipment can affect the measured concentrations and values reported. Fig. 11.7 compares microplastic concentrations in WWTP effluent based on analytical limitations and the level of treatment. The analytical limitations affect the reported effluent concentrations. Concentrations measured with methods incompatible with smaller particle detection (limit over 45 μm) had a mean concentration of 0.2 microplastic particles/L, while samples analyzed with methods capable of detecting a smaller range of microplastics (such as μ-Raman and μFTIR; Table 11.2) indicated a mean concentration of 9.5 microplastic particles/L. WWTP effluents have a lower content of microplastics than larger-sized microplastics.

p0155 Considering the level of treatment, with both analytical conditions the tertiary treatment effluent indicates a lower microplastics concentration. However, a comparison between tertiary treatment effluent measured by smaller size microplastic detection methods and secondary treatment effluent with larger-sized microplastic detection gave misleading results (SE detection limit of >45 (0.4) vs TE detection limit of <45 (5.3); Fig. 11.8). Hence, the importance of having detection limitations when comparing microplastic concentrations from different studies is highlighted.

p0160 Mean effluent microplastic concentrations from SE and TE (with smaller microplastic detection) are 24 and 5.3 particles/L respectively. It is estimated that 85 km^3 of wastewater is generated in North America annually (UNU, 2013). A simple calculation would show emissions of more than 1000 billion microplastic particles from WWTPs annually. Considering the amount of untreated wastewater and the level of treatment, the actual environmental contamination is even greater.

p0165 A study by Talvitie et al. (2017) compares the removal efficiency of different treatment options (Table 11.3). Compared to RSF and DAF, MBR provides an effective barrier to microplastics due to the membrane cutoff. The higher percentages of microplastics in wastewater are transferred to the sludge phase in the three treatment options given in

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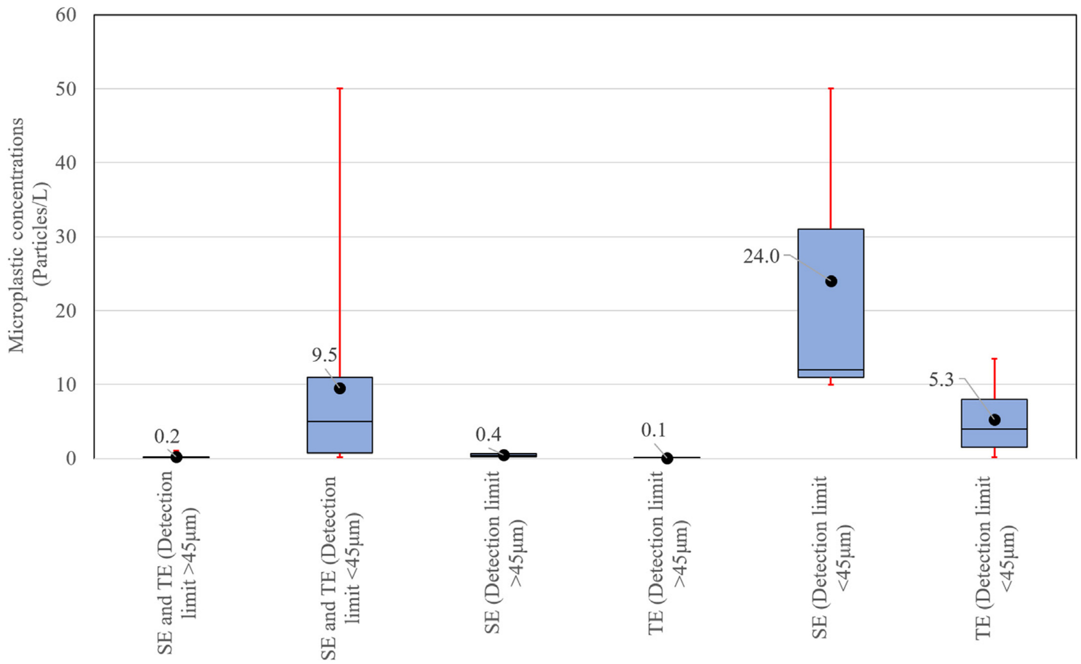

f0045 **FIGURE 11.8** Microplastic concentrations in wastewater treatment plant effluents (*meta*-analysis of values from reported literature). Detection limit comparison is based on sampling and detection method; secondary treatment effluent (SE) and tertiary treatment effluent (TE) comparisons assess treatment level effects. Top edge of box and bottom of box are upper and lower quartile, respectively (box spans the interquartile range). Horizontal lines inside the box and point marker with number represent median and mean value, respectively. Whiskers represent the highest and lowest observations.

t0020 **TABLE 11.3** Microplastic removal efficiency according to treatment option followed by activated sludge process in WWTPs.

Treatment	Influent type	Influent microplastics (particles/L)	Effluent microplastics (particles/L)	Removal (%)
Rapid sand filtration (RSF)	Secondary	0.7	0.02	97.1
Dissolved air flotation (DAF)	Secondary	2.0	0.1	95
Membrane bioreactor (MBR)	Primary	6.9	0.005	99.9

Adapted and modified from Talvitie, J., Mikola, A., Koistinen, A., Setälä, O., 2017. Solutions to microplastic pollution—removal of microplastics from wastewater effluent with advanced wastewater treatment technologies. Water Res. 123, 401–407. doi: 10.1016/j.watres.2017.07.005.

Table 11.3. However, a considerable portion remains in the water phase in the case of RSF and DAF. In all three cases, sludge management and disposal play important roles in counteracting environmental contamination by microplastics.

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t0025 **TABLE 11.4** Average concentrations of microplastic particles and fibers in wastewater (shape-based comparison).

Sampling point	Average concentration of microplastic fragment (particles/L)	Average concentration of microplastic fiber (particles/L)	Average concentration of microplastic (particles/L)
Influent	5 (± 1.3)	52.6 (± 11.3)	57.6 (± 12.4)
Effluent from primary clarifier	0.2 (± 0.1)	0.3 (± 0.1)	0.6 (± 0.2)
Final effluent	0.5 (± 0.2)	0.5 (± 0.3)	1.0 (± 0.4)
Membrane bioreactor permeate	0.1 (± 0.1)	0.2 (± 0.1)	0.4 (± 0.1)

Adapted and modified from Lares, M., Ncibi, M.C., Sillanpää, M., Sillanpää, M., 2018. Occurrence, identification and removal of microplastic particles and fibers in conventional activated sludge process and advanced MBR technology. Water Res. 133, 236−246. doi: 10.1016/j.watres.2018.01.049.

p0170 Table 11.4 provides information related to the shape of microplastics. A shape-based comparison of microplastics in WWTPs indicates higher amounts of fiber-shaped microplastics in wastewater. As microfiber derived from textile washing is a major source of microplastics in WWTPs (Schellenberger et al., 2019), the higher presence of fibers can be understood. The primary clarifier is effective in the removal of both shapes of microplastics (fragments and fibers).

s0060 11.6 Microplastics in biological wastewater treatment plant sludge

p0175 This section discusses microplastics in biological wastewater treatment plant sludge and the environmental relevance concerning sludge treatment/disposal options. As previously discussed, a larger fraction of microplastics is removed from the water phase at WWTPs. This removed fraction is transferred to the biomass and withdrawn from wastewater treatment units with the sludge. Hence, the higher removal efficiency of microplastics at WWTPs means that the macroplastics are highly associated with the sludge. Therefore sludge treatment and disposal options play important roles in reducing the environmental contaminants of macroplastics. Lares et al. (2018) reported that dry sludge from the activated sludge process, digested sludge, and MBR contained 23, 171, and 27 microplastic particles per gram of sludge, respectively. These values are significantly higher compared to the values reported in WTTP microplastic effluent. However, considering the balance of microplastic particle numbers in a WWTP, the numbers do not add up (assuming 3 g of mixed liquor suspended solids (MLSS) per liter of wastewater and one liter of wastewater in an aeration tank consisting of 8−10 microplastics). The sludge of this aeration tank should produce more than 1000 microplastic particles per one gram of sludge. This difference can be due to the practical difficulties of detecting microplastics in sludge samples (Corradini et al., 2019) and the breaking of macroplastics into smaller pieces due to mechanical forces in WWTPs (where there is minimal potential biological degradation), resulting in the limited detectability.

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p0180 Table 11.5 indicates the shape-based concentrations of microplastics in wastewater treatment plant sludge, showing higher amounts of fiber-shaped microplastics. MBR sludge indicates a slightly higher microplastic concentration compared to conventional ASP sludge. This can be due to the membrane retention and hence accumulation of microplastics within the MBR reactor transferring it to the biosolids.

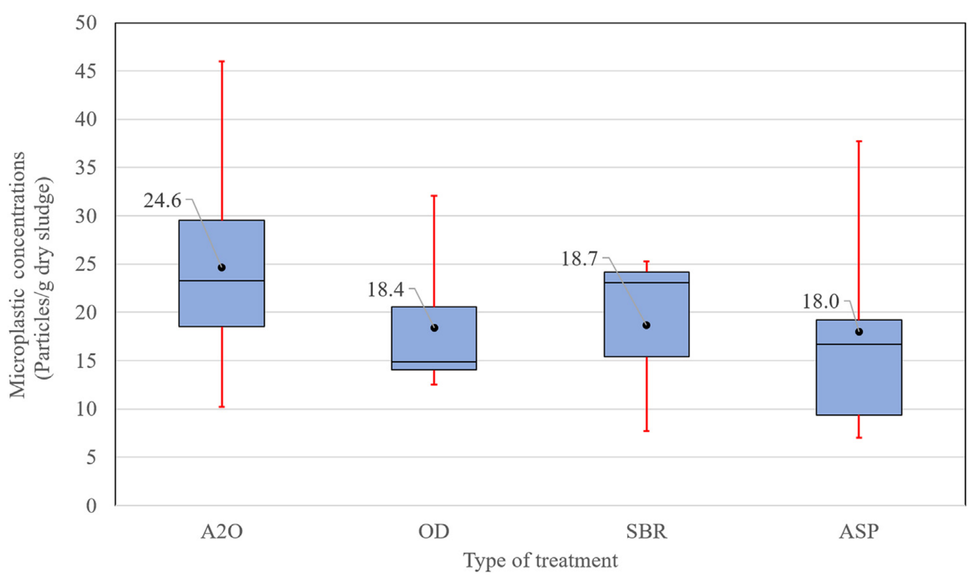
p0185 Fig. 11.9 summarizes the presence of different microplastics in the sludge of different treatment systems. The mean values are in the range of 18.0–24.6 particles per gram of dry sludge. Considering the quantities of sludge produced in a year (Japan, 2000 t/year; United States,

t0030 **TABLE 11.5** Average concentrations of microplastic fragments and fibers in wastewater treatment plant sludge samples.

Sampling point	Concentration of microplastic particles (particles/g dw[a])	Concentration of microplastic fibers (particles/g dw[a])	Total concentration of microplastic (particles/g dw[a])
Activated sludge	1.3 (±1.3)	21.7 (±4.6)	23.0 (±4.2)
Membrane bioreactor sludge	3.3 (±2.4)	24.1 (±6.1)	27.3 (±4.7)

[a]*Particles/g dw: Microplastic particles per gram of dewatered sludge.*

Adapted from Lares, M., Ncibi, M.C., Sillanpää, M., Sillanpää, M., 2018, Occurrence, identification and removal of microplastic particles and fibers in conventional activated sludge process and advanced MBR technology. Water Res. 133, 236–246. doi: 10.1016/j.watres.2018.01.049.



f0050 **FIGURE 11.9** Microplastics in sludge of wastewater treatment systems. Top edge of box and bottom of box are upper and lower quartile, respectively (box spans the interquartile range). Horizontal lines inside box and point marker with number represent median and mean value, respectively. Whiskers represent highest and lowest observations. [Types of treatment are as follows: anaerobic, anoxic and oxic (A2O), oxidation ditch (OD), sequencing bioreactor (SBR), and activated sludge process (ASP)].

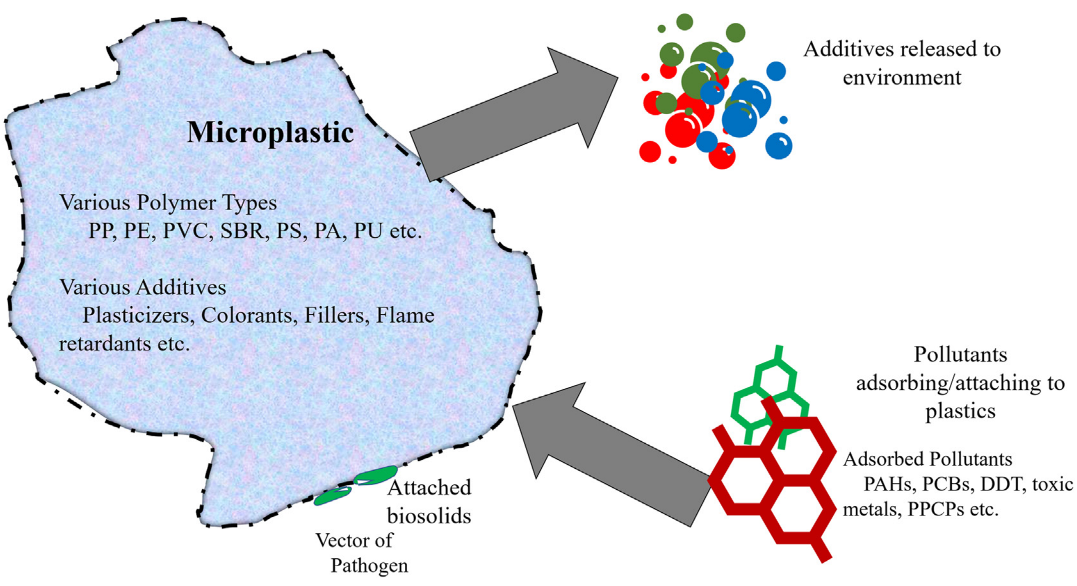
6500 t/year; EU, 8900 t/year) (Mateo-Sagasta et al., 2015), the quantity of microplastics associated with sludge is in the millions (e.g., Japan, 40,000,000,000 particles/year). Hence, sludge treatment and disposal play important roles, as mentioned previously. The common practices for sludge disposal are land disposal and incineration. In Japan, over 80% of sludge is incinerated, restricting a major portion of microplastics from entering the environment. However, the other common disposal method, applying sludge on agricultural soils as a fertilizer (Corradini et al., 2019), provides a pathway for microplastics to reenter the environment.

s0065 11.7 Effect of microplastics in biological wastewater treatment

p0190 This section discusses the potential effects of the presence of microplastics on biological wastewater treatment processes. Also, this initiates the discussion on the physicochemical and biological degradation of certain types of plastics (such as biodegradable plastics), focusing on future directions for WWTPs to address microplastic contamination.

s0070 11.7.1 Microplastic as a composite particle

p0195 Microplastic is not simply defined by its plastic polymer; it is made up of a diverse suite of chemicals. Composite materials with one-to-many polymer types and various additives (Rochman et al., 2019) are used in many plastic products. Hence, microplastics should be considered as a complex material (Fig. 11.10). Additives such as plasticizers,



f0055 **FIGURE 11.10** Microplastic as a diverse suite of chemicals and biosolids. Source*: Significantly modified from Rochman, C.M., Brookson, C., Bikker, J., Djuric, N., Earn, A., Bucci, K., et al., 2019. Rethinking microplastics as a diverse contaminant suite. Environ. Toxicol. Chem. 38 (4), 703–711. https://doi.org/10.1002/etc.4371.*

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colorants, fillers, and flame retardants, and adsorbed products, such as polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), dichlorodiphenyltrichloroethane (DDT), toxic metals, and pharmaceuticals and personal-care products (PPCPs), should not be ignored by wastewater treatment researchers and practitioners (Schellenberger et al., 2019; Zhou et al., 2020).

p0200 As mentioned previously, textile microfibers are a major microplastic contaminant flowing into WWTPs. Hence, textile-associated chemicals need to be considered as potential leakage elements at WWTPs. Fluorotelomer-based polymers (FTPs) are among the common products frequently used in the textile industry as durable water-repellents. Many poly-fluoroalkyl substances (PFASs) have the potential to partially breakdown in the environment (Lo et al., 2018) to form persistent perfluoroalkyl acids (PFAAs) such as perfluoro-octane sulfonate (PFOS) and perfluorooctanoate (PFOA). Some of the PFAAs are bioaccumulative and toxic (Goldenman et al., 2017). Based on the properties mentioned, some PFAAs are either included (PFOS, PFOA) or under consideration for inclusion in the United Nations Stockholm Convention on Persistent Organic Pollutants (Schellenberger et al., 2019; Wang et al., 2009).

p0205 Additives in road markings, plasticizers, and flame retardants are other contaminants that reach WWTPs (Kitahara and Nakata, 2020). Flame retardants such as 2-ethylhexyl diphenyl phosphate have been implicated as potential hormone mimetic compounds (Li et al., 2020). Bis(2-ethylhexyl) phthalate is a compound with human toxicity and ecotoxicity. The behavior of these chemicals at WWTPs and their interaction in the biological wastewater treatment process must be evaluated.

p0210 Microplastics are a potential medium for attached growth microorganisms to grow in wastewater systems. Biofilm growth on macroplastics has been reported in WWTP (Parrish and Fahrenfeld, 2019). Biofilm-associated pathogens can travel with microplastics far from points of emission such as WWTPs. In addition, even from diffuse origins where (in most cases) the level of treatment is comparatively lower, microplastics can be hazardous and a survival medium for pathogens. The potential benefits/hazards of biofilm communities on microplastics have been characterized by several researchers (Harrison et al., 2018; Yang et al., 2020). Parrish and Fahrenfeld (2019) reported that biofilm communities varied by source water (indicating potential utility for source tracking) and microparticle type, but not size. Kruglova et al. (2018) reported that bacterial communities attached to microplastics include phyla *Proteobacteria, Acidobacteria, Firmicutes, Actinobacteria*, and *Bacteroidetes*. Bacteria from classes *Leptospiraceae, Enterobacteriaceae*, and *Staphilococcaceae* were identified on microplastics after all treatment stages (including effluent). Bacteria from class *Mallicutes* containing pathogenic species were also found in effluent samples. Atugoda et al. (2020) indicated the potential of microplastics as a vector for antibiotics such as ciprofloxacin. Hence, microplastics can be a vector for chemicals to escape from WWTPs, and in case of escaping antibiotics, can lead to superbugs resistant to known antibiotics. These studies indicate that the situation of microplastics escaping from WWTPs does not just represent plastic-related pollution but can also escalate into a pathogen and toxic chemical—related matter. As previously discussed, further studies are needed to investigate the vector behavior of microplastics associated with attached pathogens and chemicals.

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s0075 11.7.2 Microplastic degradation in wastewater treatment plants

p0215 Since biological treatment methods are still unable to degrade microplastics, coupling them with physicochemical treatments is a more pragmatic solution to intercept microplastics in WWTPs. As indicated in Section 11.1, screens are effective in removing microplastics and mesoplastics before primary treatment. Improving the screen capacity range by introducing technological developments such as microfilter screens (15−30 μm) (Mena, 2020) could lead to the removal of an effective portion of microplastics at the initial WWT stage.

p0220 Photocatalysis degradation is another potential posttreatment option researched as a measure for degrading microplastics (Uheida et al., 2021). A research team at the Swedish Royal Institute of Technology (Tofa et al., 2019) developed a nanotechnology-based treatment method that has shown a positive influence on degrading low-density polyethylene (LDPE) microplastic residues. The application was based on zinc oxide nanorods and visible light. Nabi et al. (2020) also demonstrated a nanotechnology-based microplastic degradation mechanism. Both studies discussed enhanced degradability of microplastics under the tested conditions. Further, the authors mentioned that the byproducts of PP photodegradation have relatively low levels of ecotoxicity and human toxicity. Although this is still in the early stage of research, nanotechnology-based microplastic degradation with the help of sunlight could be a potential technology to remove microplastics in WWTP effluents at a relatively low cost (Tofa et al., 2019).

p0225 MBR is one of the most popular techniques for high-strength wastewater treatment due to its high organic removal rate and high retention of biomass. The combination of elevated biodegradation and micro- or nanosized filtration lets only small molecules pass through. Other materials such as solid particles, biomass, and macromolecules are captured by the membrane filter and removed with the sludge from the system (Seow et al., 2016). Due to this high particle removal efficiency, MBR can remove more than 99% of microplastics from wastewater (Lares et al., 2018; Talvitie et al., 2017). MBR filters are supposed to have the smallest pore size (around 0.08 μm) compared to other currently used filters in wastewater treatment, which can prevent most microplastics from passing through (Ngo et al., 2019). Hence, MBR can be identified as the best available technology so far among the common WWT methods in terms of removing microplastics from the water phase of wastewater. There may be variations in MBR efficiency in microplastic removal due to the restraining factors after longer periods of operation. However, the accumulation of microplastics in the bioreactor and the effect on treatment are yet to be studied.

p0230 Polymeric membranes (plastic polymer−based membranes) are a common type of membrane. Membrane aging, air scouring, and physicochemical cleaning methods can weaken the integrity of membranes (Huang et al., 2020), and if precautions are not taken, membranes can be a potential source of microplastic contamination of wastewater. However, this has not been studied or reported in the literature. In that case, MBR can be a source of microplastics. Microplastic removal efficiency studies and composition analysis studies could focus on this aspect as well. Moreover, different MBR configurations, such as polyvinyl alcohol (PVA) gel MBR (Chaikasem et al., 2014), use certain polymeric materials as attached growth media, and the potential microplastic leakage in such cases also remains to be studied. The impact of aging and the loss to the sludge are important considerations. The other concern associated with MBR is the extended sludge retention

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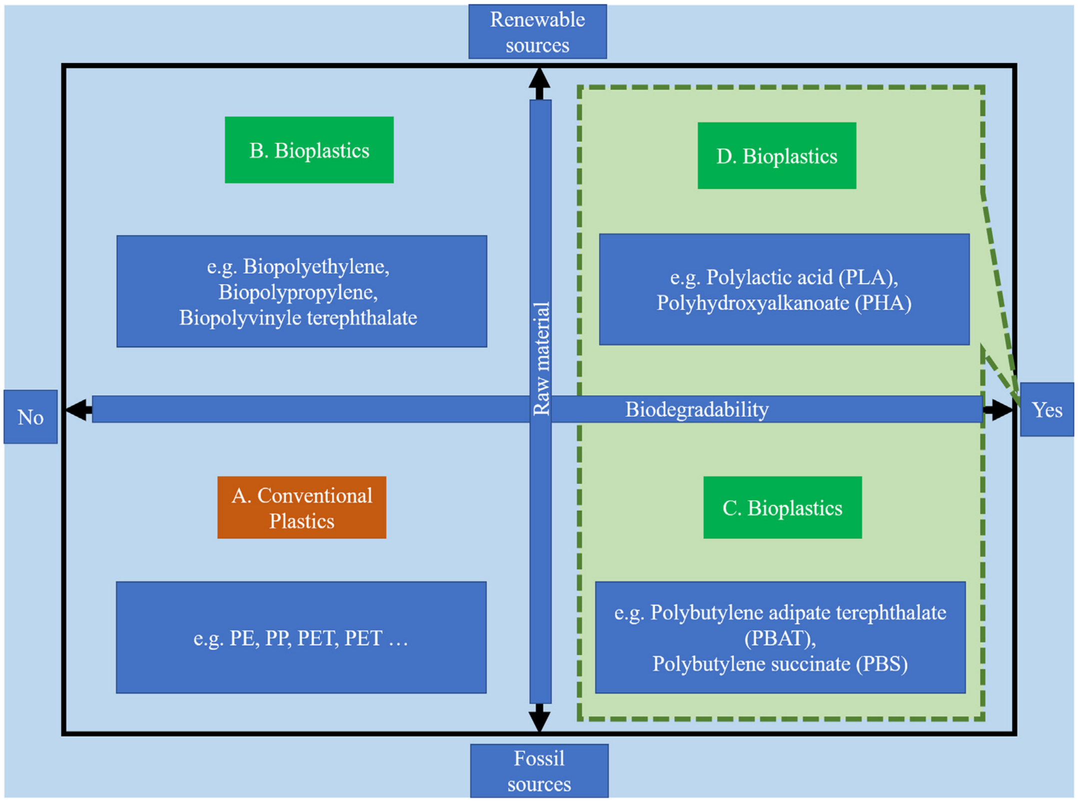

f0060 **FIGURE 11.11** Bioplastics, biodegradable plastics, and bio-based plastics. Source: *Modified from EU, 2018. What are the Bioplastics?. European Bioplastics e.V., Berlin. Retrieved from https://bioplasticseurope.eu/about.*

period and the leaching of chemicals into the biological wastewater treatment system. Hence, further studies are recommended on microplastic removal, polymeric membranes as a microplastic source, and chemical leaching from microplastics in biological reactors.

s0080 11.7.3 Biodegradable plastics in wastewater treatment

p0235 The topic of bioplastics has been discussed extensively in the literature (Polman et al., 2020). Bioplastic categories are shown in Fig. 11.11. Despite the growing attention to the topic, fossil fuel–based plastics still have more than 95% market share (Statistica, 2021). The share of bio-based plastics is expected to grow in the future and reach 40% by 2030 (Statistica, 2021). Biodegradable plastics are produced from fossil fuel–based or biomass-based sources (Fig. 11.11C and D) (EU, 2018). An increased market share of bioplastics will probably increase the biodegradable plastic share. The portion of biodegradable plastics entering WWTP poses additional concerns for the wastewater domain. There are very few studies focused on the biodegradation of plastics in wastewater environments. Biodegradation of polyvinyl alcohol by Thai indigenous mixed microbial culture has been reported recently (Kanjanasopa et al., 2020). The biodegradable aliphatic polyester poly (lactic acid) (PLA) was reported to be biodegraded by *Actinomadura keratinilytica* strain T16−1 in a 5 L stirred tank bioreactor (Panyachanakul et al., 2019). The biodegradability of plastics at WWTPs may be an area where research and development is needed, and biodegradation efforts and research activities should consider specialized microbial communities, enzymes, and environmental conditions for particular plastics.

s0085 11.8 Conclusions and perspectives

p0240 Microplastics in wastewater treatment systems are receiving increasing attention from researchers and professionals in the field. With the development of sampling and analytical methods, the potential of microplastic-related studies in wastewater systems is also increasing. For separate sewer systems the microplastics that enter WWTP can be mainly traced to kitchen sink wastewater, bathroom wastewater, and toilet flushes. For the combined sewers systems, the street runoff also carries microplastics into the WWTP. Therefore WWTPs are considered as the major receptor of microplastics as well as major point sources of microplastic emissions into the environment. In these plants, the transfer of microplastics from the water phase to the sludge phase is the dominant (or presently only considered) removal mechanism due to low polymer biodegradability. This "removal" efficiency for secondary and tertiary treatment levels exceeds 99%. However, the "removal efficiencies" need to be interpreted with care since the sampling and analytical methods affect the microplastic concentrations reported. Based on the methods employed, the size range detecting and reporting can be very. Hence, comparisons of removal efficiencies reported by different sampling and analytical methods need to be done with care.

p0245 Since the majority of microplastics are transferred into sludge at wastewater treatment plants, sludge treatment, disposal, and effective decision-making are critical to protecting the environment from microplastic (re)contamination. Therefore supportive policy measures coupled with effective wastewater treatment plant designs are important for minimizing microplastic-related pollution. Plastic-related chemicals and biofilms with attached pathogens are other factors to consider. As the release of toxic chemicals from microplastics has not been studied in great detail, reconciling the limitations of analytical capabilities requires continued work. The increased introduction of bioplastics such as biodegradable polymers worldwide may open new opportunities for biodegradability-related research and development studies in wastewater in the near future.

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