

## Interactions between microplastics and unit processes of wastewater treatment plants: a critical review

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### ABSTRACT

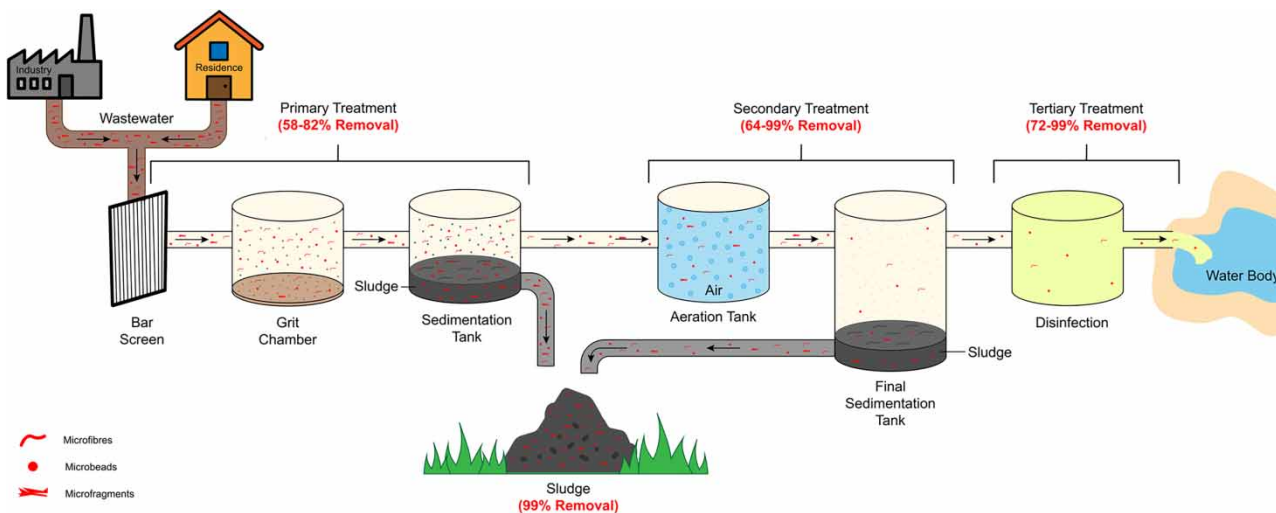
Microplastics are classified as emerging pollutants of the aquatic environment, necessitating a comprehensive understanding of their properties for successful management and treatment. Wastewater treatment plants (WWTPs) serve as point sources of microplastic pollution of the aquatic and terrestrial (eco)systems. The first part of this review explores the basic definitions of microplastics, sources, types, physical and chemical methods of identifying and characterizing microplastics in WWTPs. The next part of the review details the occurrence of microplastics in various unit processes of WWTPs and sewage sludge. Followed by this, various methods for removing microplastics from wastewater are presented. Finally, the research gaps in this area were identified, and suggestions for future perspectives were provided.

**Key words:** microplastics, unit process, wastewater, wastewater treatment plant, water pollution

### HIGHLIGHTS

- Wastewater treatment plants act as point sources of microplastic pollution.
- Microplastics in wastewater treatment plants are diverse in composition, size, shape and origin.
- Microplastics are seen in all unit processes of wastewater treatment plants.
- Wastewater treatment plants remove bulk of microplastics entering them.
- Majority of microplastics enter the environment from sludge generated in wastewater treatment plants.

## GRAPHICAL ABSTRACT



## ABBREVIATIONS

A/A/O	Anaerobic/Anoxic/and Aerobic treatment
AOB	Ammonium-Oxidizing Bacteria
ASP	Activated Sludge Process
ATR-FT-IR	Attenuated Total Reflection Fourier Transform Infrared Spectroscopy
DAF	Dissolved Air Flootation
DF	Disc Filter
FPA-FT-IR	Focal Plane Array-based Transmission Micro-FT-IR imaging
FT-IR Spectroscopy	Fourier Transform Infrared Spectroscopy
MBR	Membrane Bioreactor
NOB	Nitrite Oxidizing Bacteria
OD	Oxidation Ditch
OEP	Oil Extraction Protocol
PAO	Polyphosphate Accumulating Organisms
Py-GC-MS	Gas Chromatography coupled to Pyrolysis
RSF	Rapid Sand Filter
SEM	Scanning Electron Microscopy
STM	Scanning Tunnelling Microscopy
TDS-GC-MS	Gas Chromatography coupled to Thermal Desorption
TEM	Transmission Electron Microscopy
TF	Trickling Filter
UF	Ultra-filtration
WWTP	Wastewater Treatment Plant

## 1. INTRODUCTION

Globally the annual production of plastics is over 380 million tonnes (Plastics Europe Publications 2020). However, plastic waste is barely treated but mixed with municipal solid waste and discarded in the dump yards, landfills, and water bodies, which results in plastic pollution in various matrices of the environment. Changes that plastics undergo when they are in the environment has led to a new type of plastic pollutants termed microplastics.

Some of the earliest investigations that have set the pace for microplastic research include the studies of Carpenter *et al.* (1972) on polystyrene spherules in coastal waters of south New England, USA, Colton *et al.* (1974) on plastic pollution in the North Atlantic, and Ryan & Moloney (1990) on plastic debris in the beaches of South Africa. The word 'microplastics' was first used in 2004 for plastics of size <5 mm, as reported by Thompson *et al.* (2004). Despite the common usage of the term,

there is a lack of consensus on its definition. The accepted size range for the lower limit is 1–20  $\mu\text{m}$ , and the upper limit is 500  $\mu\text{m}$  to either 1 mm or 5 mm (Frias & Nash 2019). There have been a few attempts to define them based on their physicochemical characteristics (Verschoor 2015). However, it is imperative to understand the fundamental properties such as source, chemical composition, and solubility of microplastics to devise a standard classification method.

Based on their production, microplastics are classified into primary and secondary microplastics. Primary microplastics, such as microbeads commonly used in the cosmetic industry, are manufactured within the specified size range. Secondary microplastics are plastic debris that fragment/break down into the size range of microplastics due to photochemical, biological, or mechanical processes. Their classification into types is also a matter of contention, as there are no fixed criteria based on which they are classified. The most abundant types of microplastics in decreasing order are pellets, fragments, fibres, films, ropes, filaments, sponges, foams, rubber, and microbeads.

It is crucial to study microplastics in natural environments because of the risks that microplastics pose to environmental compartments. Low density, high persistence, and a wide range of occurrences are the unique characteristics of (micro)plastics, making them quite different from most other common pollutants. Their size and non-biodegradable nature make them pollutants that are almost impossible to treat/remove, especially from aquatic ecosystems. Their persistence in various environments and ubiquitous existence qualifies them as a planetary boundary threat (Villarrubia-Gómez *et al.* 2018).

Microplastics can cause detrimental effects in two ways – they can be harmful by themselves and serve as carriers of other contaminants. Microplastics can easily be internalized by biota and accumulated in the food chain (Anbumani & Kakkar 2018). Over 50% of commonly used plastics contain at least one hazardous component (Rochman *et al.* 2013a). Once they enter aquatic ecosystems, their high specific surface area and hydrophobic nature make them act as carriers of hydrophobic organic contaminants (Rochman *et al.* 2013b). Microplastics thus facilitate the entry of hazardous chemicals used in their manufacturing and other contaminants (mainly persistent organic pollutants) into the food chain. Hence, there is an imminent need to develop efficient microplastic removal and disposal methods to ensure that they do not find their way into aquatic ecosystems and the food chain. To do this, sources, types, and characteristics of microplastics in aquatic ecosystems, including point sources such as WWTPs, need to be extensively studied. WWTPs act as a point source of microplastics as the microplastics produced in/disposed of in household and industrial wastewater streams, and oftentimes in the stormwater drain system, make their way into WWTPs. This makes WWTPs important in the study of microplastics. Across the globe, the interest to study microplastics in WWTPs is catching up and an overview of the key publications reported in the literature from various parts of the world is presented in Table 1.

However, a SCOPUS search done on 16 October 2021, showed only 382 and 16 documents on microplastics in WWTPs and microplastics in unit processes of WWTPs, respectively. Hence there is a dire need to study how the microplastics interact with each and every unit process of WWTPs. This review provides a comprehensive understanding of interactions of microplastics with various unit processes of WWTPs, which paves the way for their removal from and modelling as pollutants in WWTPs. This review has explored the basic definitions of microplastics, sources, types, physical and chemical methods of identifying and characterizing microplastics in WWTPs. The study focuses on the occurrence of microplastics in various unit processes of WWTPs and sewage sludge. Next, methods for removing microplastics from wastewater are presented. Gaps in this research area were identified, and some suggestions were provided for future perspectives.

## 2. IDENTIFICATION AND CHARACTERIZATION OF MICROPLASTICS IN WWTPs – CHEMICAL (ANALYTICAL) AND PHYSICAL METHODS

Identification and characterization of microplastics in WWTPs are necessary to understand how they act as point sources and make them more efficient in microplastic removal.

The main steps involved in the characterization process are (i) Sampling and separation, (ii) Digestion, and (iii) Identification.

### 2.1. Sampling and separation

Proper sampling is critical for the characterization of microplastics. Sampling methods vary according to the environment from which the microplastics are to be collected. For *in situ* sampling of microplastics from marine and other large aquatic environments, usually plankton, neuston and manta nets are used. However, for smaller aquatic environments and WWTPs, either *in situ* or *ex situ* sampling strategies are followed, using various types of sieves and pumps

**Table 1** | Overview of the reported studies on microplastics in WWTPs around the globe

Country	Wastewater treated	Unit processes studied	Type of microplastics recovered	Size of microplastics	Concentration of microplastics in the influent	Cumulative removal of microplastics from WWTP	References
Iran	Domestic wastewater	<ul style="list-style-type: none"> <li>• Bar screen</li> <li>• Grit chamber</li> <li>• Primary settling tank</li> <li>• Anoxic tank</li> <li>• Aeration basin</li> <li>• Clarifier</li> </ul>	<ul style="list-style-type: none"> <li>• Fibres</li> <li>• Particles</li> </ul>	37 µm – 5 mm	12,667 ± 668 MP/m <sup>3</sup>	96.7%	Alavian Petroody <i>et al.</i> (2020)
Spain	Domestic wastewater	<ul style="list-style-type: none"> <li>• MBR</li> <li>• RSF</li> </ul>	<ul style="list-style-type: none"> <li>• Fragments</li> <li>• Fibres</li> <li>• Microbeads</li> <li>• Films</li> </ul>	210 µm – 6.3 mm	4.40 ± 1.01 MP/L	79%	Bayo <i>et al.</i> (2020)
United Kingdom	Domestic and industrial wastewater	<ul style="list-style-type: none"> <li>• Coarse screen</li> <li>• Grit chamber</li> <li>• Primary settling tank</li> <li>• ASP and clarification tank</li> <li>• Nitrification tank</li> </ul>	<ul style="list-style-type: none"> <li>• Fragments</li> <li>• Fibres</li> <li>• Films</li> </ul>	60 – 2,800 µm	8.1 × 10 <sup>8</sup> MP/day	96%	Blair <i>et al.</i> (2019)
South Korea	Domestic and industrial wastewater	<ul style="list-style-type: none"> <li>• Grit chamber</li> <li>• Primary settling tank</li> <li>• MBR, ASP, and settling tank</li> <li>• Coagulation tank</li> <li>• Membrane DF</li> <li>• RSF</li> </ul>	<ul style="list-style-type: none"> <li>• Fragments</li> <li>• Fibres</li> <li>• Microbeads</li> <li>• Sheets</li> </ul>	100 µm – 5 mm	8,400 MP/L 62,800 MP/L 11,680 MP/L	92–99%	Hidayaturrahman & Lee (2019)
China	Domestic wastewater and pretreated industrial wastewater	<ul style="list-style-type: none"> <li>• Primary treatment</li> <li>• Secondary treatment</li> <li>• Seasonal tertiary treatment</li> </ul>	<ul style="list-style-type: none"> <li>• Fragments</li> <li>• Fibres</li> <li>• Pellets</li> <li>• Granules</li> </ul>	250 µm – 5 mm	1.57–13.69 MP/L	79.3–97.8%	Long <i>et al.</i> (2019)
China	Domestic and industrial wastewater	<ul style="list-style-type: none"> <li>• Aerated grit chambers</li> <li>• OD</li> <li>• MBR</li> <li>• Secondary settling tank</li> <li>• UV disinfection chamber</li> </ul>	<ul style="list-style-type: none"> <li>• Fragments</li> <li>• Fibres</li> <li>• Films</li> <li>• Foam</li> </ul>	100 µm – 5 mm	4.0 MP/L	97–99%	Lv <i>et al.</i> (2019)
Italy	Domestic wastewater	<ul style="list-style-type: none"> <li>• ASP and sedimentation tank</li> <li>• RSF and disinfection tank</li> </ul>	<ul style="list-style-type: none"> <li>• Fragments</li> <li>• Films</li> <li>• Lines</li> </ul>	0.01 – 5 mm	2.5 ± 0.3 MP/L	84%	Magni <i>et al.</i> (2019)
China	Domestic and industrial wastewater	<ul style="list-style-type: none"> <li>• Primary aerated grit treatment tank</li> <li>• A/A/O</li> <li>• Denitrification, UF, Ozonation, UV tanks</li> </ul>	<ul style="list-style-type: none"> <li>• Fibres</li> <li>• Particles</li> </ul>	100 µm – 5 mm	12.03 ± 1.29 MP/L	95%	Yang <i>et al.</i> (2019)
Canada	Domestic wastewater and storm water from combined sewers	<ul style="list-style-type: none"> <li>• Bar screen</li> <li>• Primary clarifier</li> <li>• Trickling filters</li> <li>• Solids contact tanks</li> <li>• Secondary clarifiers</li> </ul>	<ul style="list-style-type: none"> <li>• Fragments</li> <li>• Fibres</li> <li>• Pellets</li> </ul>	250 µm – 5 mm	31.1 ± 6.7 MP/L	94%	Gies <i>et al.</i> (2018)
Finland	Domestic wastewater	<ul style="list-style-type: none"> <li>• MBR</li> <li>• ASP</li> </ul>	<ul style="list-style-type: none"> <li>• Fragments</li> <li>• Fibres</li> </ul>	250 µm – 5 mm	1.5 × 10 <sup>8</sup> MP/d	98.3%	Lares <i>et al.</i> (2018)

(Continued.)

Table 1 | Continued

Country	Wastewater treated	Unit processes studied	Type of microplastics recovered	Size of microplastics	Concentration of microplastics in the influent	Cumulative removal of microplastics from WWTP	References
Finland	Domestic wastewater	<ul style="list-style-type: none"> <li>• DF</li> <li>• RSF</li> <li>• MBR</li> <li>• DAF</li> </ul>	<ul style="list-style-type: none"> <li>• Fragment</li> <li>• Fibres</li> </ul>	10 µm – 5 mm	6 MP/L	97%	Talvitie <i>et al.</i> (2017a)
Denmark	Domestic and industrial wastewater	<ul style="list-style-type: none"> <li>• Coarse screen</li> <li>• Grit removal</li> <li>• Primary settling tanks</li> <li>• Aeration tank</li> <li>• Clarification tank</li> </ul>	<ul style="list-style-type: none"> <li>• Fragments</li> <li>• Fibres</li> <li>• Microbeads</li> <li>• Films</li> <li>• Foam</li> </ul>	100 µm – 5 mm	15.70 ± 5.23 MP/L	98.41%	Murphy <i>et al.</i> (2016)
United States of America	Domestic wastewater and storm water	<ul style="list-style-type: none"> <li>• Bar screen</li> <li>• Grit chamber</li> <li>• Primary settling tank</li> <li>• ASP</li> <li>• TF and ASP</li> <li>• RSF</li> <li>• Anaerobic MBR</li> </ul>	<ul style="list-style-type: none"> <li>• Fragments</li> <li>• Fibres</li> <li>• Microbeads</li> </ul>	100 µm – 5 mm	133.0 ± 35.6 MP/L	97–99%	Michielssen <i>et al.</i> (2016)

(Prata *et al.* 2019). Samples are to be collected from all relevant points in the WWTPs, mainly at the start and end of each unit process. Sampling is done in conjunction with the separation step. Separation can be done through filtration, using a series of sieves, and through density separation, using denser solutions such as NaCl or ZnCl<sub>2</sub>. The latter method is followed if the sample has a high concentration of organics, which would lead to clogging of the pores during filtration (Long *et al.* 2019).

The oil extraction protocol (OEP) takes advantage of microplastics' oleophilic properties to cost-effectively extract them in an FTIR-compatible method. While OEP has been used to study microplastics in sediments, it can also be used for studying microplastic in WWTPs (Crichton *et al.* 2017).

## 2.2. Digestion

Digestion removes the organic matter from the sample and ensures that only inorganic microplastic particles remain. Digestion of microplastics is done using acid, alkali, oxidizing agents, or enzymes, as described below.

Various kinds of acids are used to digest the organic matter present in the microplastic samples. At low concentrations and room temperature, mineral HCl is inefficient and inconsistent (Cole *et al.* 2014). Higher concentrations or stronger acids like HNO<sub>3</sub> cannot be used because they may affect the microplastic polymers, which have a low resistance to acids (He *et al.* 2018). Conversely, low concentrations of stronger acids may not have the desired effect on organics. Hence it is imperative to perform acid digestion at optimum concentration and temperature (Prata *et al.* 2019).

When alkali agents are used in microplastic digestion, the polymers may become discoloured or affected by high concentrations. It has the added issue of the possibility of organics being deposited over the polymers (Prata *et al.* 2019).

Oxidizing agents like hydrogen peroxide/Fenton's reagent are widely used to oxidize the organic matter present in the sample. Hydrogen peroxide can digest organic matter more efficiently than most acids and alkalis, without causing any changes to the microplastic polymers (Prata *et al.* 2019). Hydrogen peroxide can be directly used (Ziajahromi *et al.* 2017), or organic matter may be separated by chemical settling and decantation, then dissolved in hydrogen peroxide (Bretas Alvim *et al.* 2020). A combination of hydrogen peroxide, heat, and catalysts such as Fenton's reagent can be used to speed up the digestion process and yield the desired results (Lares *et al.* 2018; Prata *et al.* 2019; Bretas Alvim *et al.* 2020).

Various enzymes can be employed for digestion, and this accomplishes the task with minimal damage to polymers. However, the process is a multistep process, hence may cause contamination of samples (Bretas Alvim *et al.* 2020), and it is also expensive (Prata *et al.* 2019).

## 2.3. Identification

Identification of microplastics is the final step, in which microplastics present in the samples are classified according to their physical and chemical characteristics.

### 2.3.1. Physical characteristics

Physical characteristics include size, shape, colour, and type (foam, granules, pellets, etc.). Physical characterization is done visually, using microscopes, a time-consuming but cost-effective process to quantify microplastics in the sample. (Prata *et al.* 2019).

For studying the morphology of smaller sized microplastics, electron and scanning probe microscopes such as scanning electron microscopy (SEM), transmission electron microscopy (TEM), scanning tunnelling microscopy (STM) are used (Barbosa *et al.* 2020). SEM is well suited for spotting and differentiating impurities and microplastics in the sample due to its potential to generate high-resolution images. This approach is not best suited for analyzing multiple samples because it is time consuming (Barbosa *et al.* 2020).

### 2.3.2. Chemical characteristics

For studying the chemical composition of microplastics, the most commonly used technique is Fourier transform infrared (FT-IR) spectroscopy (Renner *et al.* 2018; Silva *et al.* 2018). FT-IR spectroscopy has the added advantage of observing the ageing process of microplastics by surface oxidation and visual characterization by transmission mode (Silva *et al.* 2018). As more microplastics can be identified than visual inspection and the results are accurate and reliable, FT-IR spectroscopy is widely used (Prata *et al.* 2019). Shortcomings of FT-IR spectroscopy include lower sensitivity and the incapability of detecting smaller microplastics (nano-size) at lower concentrations (Barbosa *et al.* 2020). However, attenuated total reflection Fourier transform infrared (ATR-FT-IR) spectroscopy and focal plane array-based transmission micro-FT-IR imaging (FPA-FT-IR) spectroscopy allow the identification of polymers to a size of up to 20  $\mu\text{m}$  (Mintenig *et al.* 2017).

Raman spectroscopy with a better size resolution, up to 1  $\mu\text{m}$ , has also been used to characterize microplastics (Nguyen *et al.* 2019). However, the fluorescence background from filter papers can interfere with the results. This drawback has been overcome by using cellulose filters but at an increased cost of the analysis.

Thermal methods of analyzing the chemical compositions of microplastics are gas chromatography coupled with pyrolysis followed by mass spectroscopy (Py-GC-MS) and gas chromatography coupled with thermal desorption followed by mass spectroscopy (TDS-GC-MS). In Py-GC-MS, pyrograms of known standards serve as references to measure the production of thermal degradation due to pyrolysis in a mass spectrometer. This process's limitations include high reliance on sample preparation and manual introduction of particles into the pyrolysis tube. Testing only one particle per cycle limits its application only to smaller sample sizes (Barbosa *et al.* 2020). In TDS-GC-MS mode, the sample is mounted and heated to 1,000  $^{\circ}\text{C}$  in a thermo-gravimetric balance, and then the degradation products are first adsorbed in a solid phase, then desorbed and analyzed by mass spectroscopy. However, this method is less sensitive than Py-GC-MS (Nguyen *et al.* 2019).

A summary of separation, digestion, and identification steps involved in microplastics' characterization is presented in Table 2.

The ubiquitous nature of microplastics has paved the way for them to enter into sewage streams and consequently into WWTPs. As an integral part of the sanitation system, WWTPs have become prominent sources of microplastic release into aquatic systems, that receive treated effluent (Talvitie *et al.* 2015; Ziajahromi *et al.* 2016; Kalčíková *et al.* 2017; Kay *et al.* 2018; Conley *et al.* 2019; Bretas Alvim *et al.* 2020), and terrestrial ecosystem, when sewage sludge is applied as soil amendment/fertilizer (Corradini *et al.* 2019; Rolsky *et al.* 2020). Studies that quantify and characterize microplastics in WWTPs have been done by comparing the influent and effluent for calculating the removal efficiencies (Talvitie *et al.* 2017a; Blair *et al.* 2019).

Considering WWTPs as sources of microplastics to freshwater ecosystems, studies have been conducted in river catchments, which show that concentrations of microplastics are found to increase downstream from WWTPs (Kay *et al.* 2018). Further, WWTPs serving a larger population equivalent have shown more significant fluctuations in the downstream concentrations of microplastics. WWTPs serve as a source of microplastics that enter marine environments as well. Studies on the Baltic Sea (Talvitie *et al.* 2015) and the sea in Greece have identified WWTPs as a route for microplastics present in the seawater and ocean sediments (Talvitie *et al.* 2015; Mourgogiannis *et al.* 2018).

While studies have shown that WWTPs have high removal efficiencies of microplastics (Talvitie *et al.* 2017a), a significant quantity is being released from WWTPs. Hence it is imperative to remove microplastics from WWTPs to reduce their release



**Table 2** | Separation, digestion, and identification steps in characterization of microplastics

Characterization Step	Process	Method	Reagent/Equipment	
Separation	Filtration	Size fractionation	Sieves in series	
	Density separation	High-density supernatants	ZnCl <sub>2</sub> , NaCl	
	Oil extraction protocol	Oils	Filtered canola oil	
Digestion	Acid digestion	Milder acids/in low concentration	HCl in low concentration	
	Alkaline digestion	Milder alkali/in low concentration	NaOH in low concentration	
	Oxidation	Mild, stable oxidizing agents	Hydrogen peroxide Fenton's reagent	
	Enzymatic digestion	Enzymes	Proteolytic enzyme (proteinase-K)	
Identification	Visual	Optical microscopy	Simple microscope	
	Morphology	Electron and scanning probe microscopy	SEM TEM STM	
		Chemical composition	Spectroscopy	FT-IR spectroscopy Raman spectroscopy
		Chemical composition	Thermal methods	TDS-GC-MS Py-GC-MS

into aquatic environments significantly. In this pursuit, it is essential to understand (i) the types, sizes, and distribution of microplastics in the wastewater treatment process, (ii) how each unit process in wastewater treatment contributes towards the removal of microplastics, (iii) the effect of microplastics on treatment mechanisms (physical and chemical), and vice versa. The above information would enable identifying crucial unit processes that remove microplastics and undertake necessary alterations in WWTPs to minimize the release of microplastics into the natural environment.

### 3. SOURCES AND TYPES OF MICROPLASTICS IN WWTPs

Microplastics that make their way into WWTPs are incredibly diverse, as depicted in Table 3. As microplastics originate from diverse sources, they are of different types, chemical composition, origin, and concentration (Michielssen *et al.* 2016; Gies *et al.* 2018; Blair *et al.* 2019; Hidayaturrehman & Lee 2019; Long *et al.* 2019; Lv *et al.* 2019; Magni *et al.* 2019; Yang *et al.* 2019; Bayo *et al.* 2020). Microplastics enter WWTPs from household and industrial sewage (Carr 2017). The cosmetics and personal care industries release more microplastics than others (Carr *et al.* 2016). The chemical composition of microplastics is dependent on the source industry. The quantity and type of microplastics in a WWTP vary according to the season as well.

Microplastics found in WWTPs are diverse in chemical composition, size, shape, and origin, hence generalizing their types and treatment is challenging. Secondary microplastics, including fibres, fragments, and films, are more commonly found in wastewater than primary microplastics (Blair *et al.* 2019). Fibres are thought to find their way into sewage streams primarily from household sewage (Carr 2017). Fragments are found in all stages of wastewater treatment and are of varied sizes and shapes (Ziajahromi *et al.* 2016). They are also harder to remove and can be found in higher concentrations in the treated effluent. Films are less common and are almost entirely removed in the skimming process in primary treatment (Blair *et al.* 2019).

The presence of primary microplastics in WWTPs cannot be neglected. Microbeads, commonly used in the personal health-care industry, are removed in primary treatment and sludge (Michielssen *et al.* 2016; Bayo *et al.* 2020).

Chemical composition-wise, the most commonly found microplastics in WWTPs are polyethylene, and polypropylene (Talvitie *et al.* 2015; Hidayaturrehman & Lee 2019; Magni *et al.* 2019), which is used in the packaging industry and has other household uses (Talvitie *et al.* 2015; Hidayaturrehman & Lee 2019; Magni *et al.* 2019; Bretas Alvim *et al.* 2020). Microplastics have a shorter residence time, resulting from fast fragmentation and degradation rates (Long *et al.* 2019). Smaller sized microplastics exhibit a higher removal rate, and unit processes influence the size distribution of microplastics in a WWTP (Lv *et al.* 2019).

**Table 3** | Different types of microplastics identified from WWTPs

Type of microplastics	Sources of microplastics	Specific microplastic type out of total microplastic sample, %	Presence in WWTP influent/ effluent	References		
Fragments	Household sewage, industry, produced in wastewater treatment process (Blair <i>et al.</i> 2019)	6.7	Influent	Bayo <i>et al.</i> (2020) Hidayaturrahman & Lee (2019) Blair <i>et al.</i> (2019) Lares <i>et al.</i> (2018) Magni <i>et al.</i> (2019) Michielssen <i>et al.</i> (2016) Gies <i>et al.</i> (2018) Long <i>et al.</i> (2019) Lv <i>et al.</i> (2019) Murphy <i>et al.</i> (2016)		
		10.8–53.4				
		15				
		18				
		21				
		22.9–25.9				
		28.1				
		30				
		65				
		67.3				
Fibre	Laundry discharges and household sewage (Carr 2017)	5.6–21.4	Effluent	Hidayaturrahman & Lee (2019) Michielssen <i>et al.</i> (2016)		
		13–33				
		14.6–46.7			Influent	Hidayaturrahman & Lee (2019) Long <i>et al.</i> (2019) Murphy <i>et al.</i> (2016) Lv <i>et al.</i> (2019) Michielssen <i>et al.</i> (2016) Bayo <i>et al.</i> (2020) Gies <i>et al.</i> (2018) Blair <i>et al.</i> (2019) Lares <i>et al.</i> (2018) Yang <i>et al.</i> (2019)
		17.7				
		18.5				
		21				
54.5–62						
61.09						
Microbeads	Cosmetics and personal care products (Kalčíková <i>et al.</i> 2017)	65.6	Effluent	Hidayaturrahman & Lee (2019) Michielssen <i>et al.</i> (2016)		
		67				
		82				
		85.4				
Films	Packaging and industries (Gündoğdu <i>et al.</i> 2018)	0.5–22.5	Influent	Hidayaturrahman & Lee (2019) Michielssen <i>et al.</i> (2016)		
		61–84.7				
		Negligible				
Foam	Industries (Murphy <i>et al.</i> 2016)	0.6	Effluent	Hidayaturrahman & Lee (2019) Michielssen <i>et al.</i> (2016)		
		3				
Sheet	Localized fragmentation of larger plastics (Kapp & Yeatman 2018; Hidayaturrahman & Lee 2019)	11.1–15.8	Influent	Murphy <i>et al.</i> (2016) Lv <i>et al.</i> (2019)		
		18.1–70.4				
Pellets	Industries (Gies <i>et al.</i> 2018)	55.7–93.2	Effluent	Hidayaturrahman & Lee (2019) Michielssen <i>et al.</i> (2016)		
		9.9				
Pellets	Industries (Gies <i>et al.</i> 2018)	12	Influent	Murphy <i>et al.</i> (2016) Lv <i>et al.</i> (2019) Blair <i>et al.</i> (2019) Bayo <i>et al.</i> (2020) Magni <i>et al.</i> (2019)		
		18				
Pellets	Industries (Gies <i>et al.</i> 2018)	31.5	Influent	Murphy <i>et al.</i> (2016) Lv <i>et al.</i> (2019)		
		73				
Pellets	Industries (Gies <i>et al.</i> 2018)	1.3	Influent	Murphy <i>et al.</i> (2016) Lv <i>et al.</i> (2019)		
		2				
Pellets	Industries (Gies <i>et al.</i> 2018)	3.8–4.5	Influent	Hidayaturrahman & Lee (2019)		
		0–3			Effluent	Hidayaturrahman & Lee (2019)
Pellets	Industries (Gies <i>et al.</i> 2018)	2.5	Influent	Long <i>et al.</i> (2019) Gies <i>et al.</i> (2018)		
		5.4				

(Continued.)



Table 3 | Continued

Type of microplastics	Sources of microplastics	Specific microplastic type out of total microplastic sample, %	Presence in WWTP influent/ effluent	References
Particles	Cosmetics and personal care products, household sewage (Kalčíková <i>et al.</i> 2017)	14.6	Influent	Yang <i>et al.</i> (2019)
Lines	Laundry discharges and fishing industry (Carr 2017)	6	Influent	Magni <i>et al.</i> (2019)
Granules	Household sewage, industry (Carr 2017; Long <i>et al.</i> 2019)	49.8	Influent	Long <i>et al.</i> (2019)

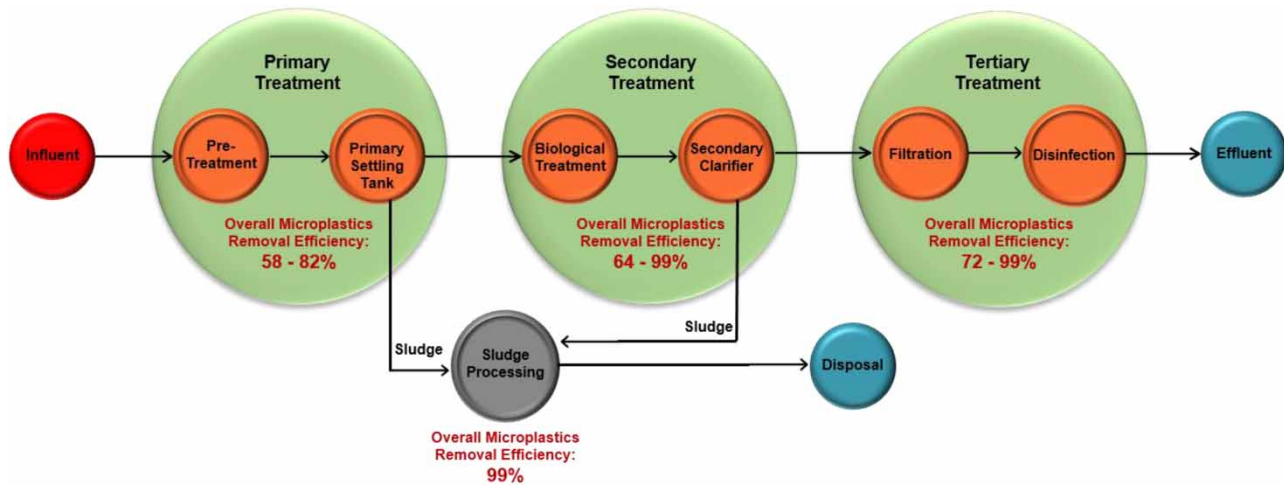


Figure 1 | Overall removal efficiencies of microplastics at various treatment stages in a typical WWTP.

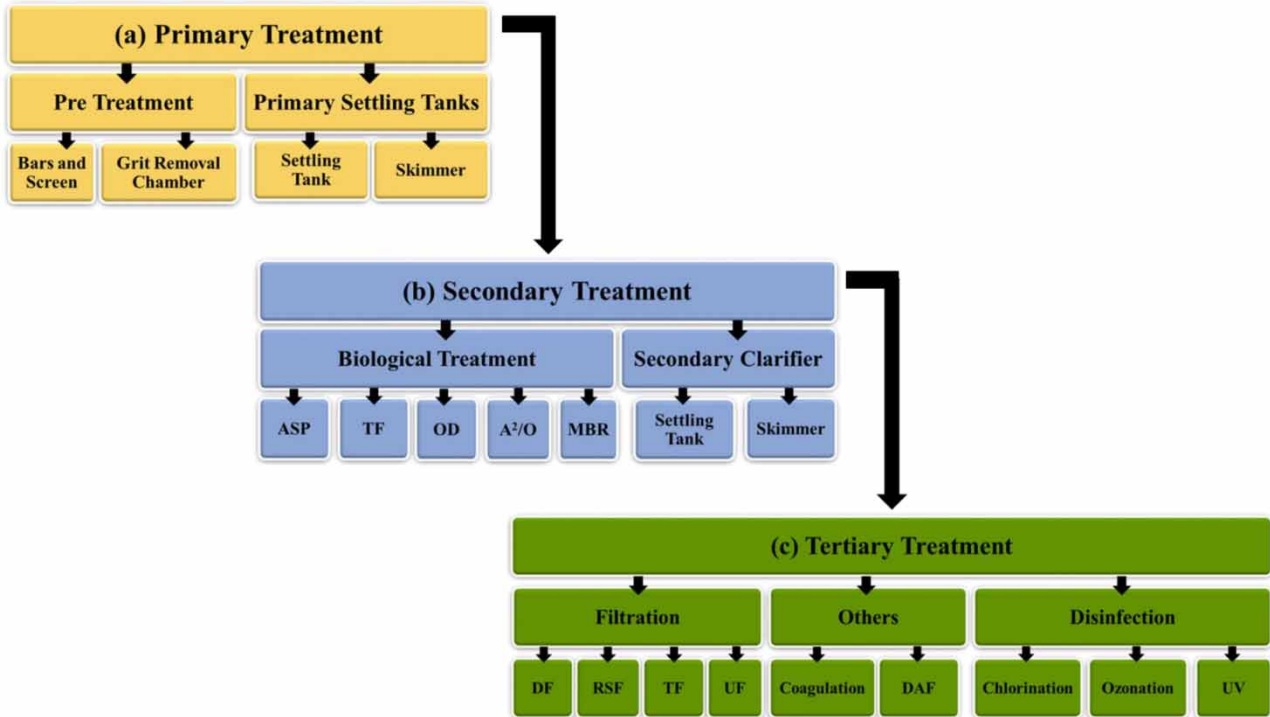
#### 4. MICROPLASTICS IN VARIOUS UNIT PROCESSES OF WWTPS

Microplastics have been recorded in all unit processes of WWTPs. Their presence is diverse in size, shape, type, and concentrations, and inconsistent across reported studies. To effectively control the release of microplastics into the environment by WWTPs, it is imperative to study the characterization, removal, impact, and fate of microplastics in each WWTP unit process. The unit processes employed by WWTPs to process wastewater play a critical role in deciding the types and concentrations of microplastics released into the environment (Talvitie *et al.* 2017a). However, such studies are lacking. It can be seen that each unit process responds differently to microplastics in terms of impact, removal, and fate. Understanding the fate of microplastics in unit processes is key to removing WWTPs and modelling microplastics as pollutants.

Figure 1 represents wastewater treatment stages and their overall removal efficiencies, which is the cumulative removal of microplastics up to a given treatment stage. However, the removal efficiency of the unit processes is calculated as the percentage of several microplastic particles or mass of microplastic particles or a combination of both; hence there is no uniformity.

##### 4.1. Primary treatment

Primary treatment is the first step in the wastewater treatment process. It consists of preliminary treatment and primary settling tanks. Figure 2(a) represents the standard primary treatment unit processes, where microplastic removal efficiencies



**Figure 2** | The unit processes of WWTPs associated with microplastics removal.

have been studied. The maximum removal of microplastics happens during the primary treatment in conventional WWTPs (Michielssen *et al.* 2016; Blair *et al.* 2019). Standard primary treatment unit processes and their microplastic removal efficiencies are tabulated in Table 4(a).

Most WWTPs employ pre-treatment to prepare the influent for further treatment by removal of large debris. Most of the studies reported in the literature focus on primary treatment as a whole, but pre-treatment removal efficiencies are not measured individually (Blair *et al.* 2019), except for a few studies (Michielssen *et al.* 2016; Murphy *et al.* 2016; Blair *et al.* 2019).

As bar screens and grit chambers are commonly used in WWTPs, the studies on removing microplastics seem to be limited to mainly these pre-treatment processes, and others like coagulation and flocculation (to remove turbidity) and equalization tanks have been overlooked.

Racks and bar screens are used to filter out large debris when raw wastewater passes through them. The effect of such screens on microplastics depends on the size of the gaps between the bars. Mostly spaced at 2–6 cm, they easily allow microplastics to pass through them. Grit chambers, used to remove grit (denser inorganics), are where microplastic removal happens in the pre-treatment process.

Preliminary treatment has shown a removal efficiency of 39–58% (Michielssen *et al.* 2016; Murphy *et al.* 2016; Blair *et al.* 2019). Microfibers with higher densities and settling behaviour seem to be the microplastics group most influenced by grit chambers (Alavian Petroody *et al.* 2020).

Owing to their high specific surface area and hydrophobic nature, microplastics may adsorb chemical agents used in wastewater treatment, which would require an additional dosage of these chemicals (Zhang & Chen 2020). Microplastics may also adsorb additional pollutants, which may hinder the treatment process.

The primary settling tank is the next phase of primary treatment. The objective of having a primary settling tank is to remove suspended solids (both organic and inorganic) by sedimentation and flotation. Most primary settling tanks involve a surface skimming mechanism, along with sedimentation. Here, along with oil and grease, most microplastics in the wastewater stream rise to the surface and are removed by skimmers, as reported in some studies (Michielssen *et al.* 2016; Ziajahromi *et al.* 2016; Kay *et al.* 2018; Conley *et al.* 2019; Hidayaturrehman & Lee 2019; Yang *et al.* 2019).

**Table 4** | Different types of microplastics removed from various unit processes of WWTPs

Treatment process	Removal efficiency (%)	Type of microplastic removed	References
<b>(a) Primary Treatment</b>			
Screened by grit removal and primary settling tank	62.7–64.4	Fragments	Hidayaturrahman & Lee (2019)
Coarse screening (12 mm) and grit removal, primary settling tanks	82	Pellets, fragments, fibres	Blair <i>et al.</i> (2019)
Bar screen (20 mm), an aerated grit chamber, a flow metering, primary settling tank	72.3	Low-density microplastics, fibres	Alavian Petroody <i>et al.</i> (2020)
Bar screen, grit chamber, primary settling tank	84.1–88.4	Microbeads, fibres	Michielssen <i>et al.</i> (2016)
Primary aerated grit treatment	58.84 ± 8.05	Not mentioned	Yang <i>et al.</i> (2019)
Coarse screening (20 mm) and grit removal, primary settling tanks	78.34	Microbeads	Murphy <i>et al.</i> (2016)
<b>(b) Secondary Treatment</b>			
MBR, ASP, and settling tank	83.1–91.9	Fragments	Hidayaturrahman & Lee (2019)
ASP and clarification	92	Fragments, fibres	Blair <i>et al.</i> (2019)
Anoxic tank, aeration basin, clarifier	24.4	Smaller microplastics	Alavian Petroody <i>et al.</i> (2020)
ASP	93.8	Microbeads	Michielssen <i>et al.</i> (2016)
TF and ASP	89.8	Microbeads	
MBR	79.01	Fibres, polypropylene, polystyrene	Bayo <i>et al.</i> (2020)
A/A/O	71.67 ± 11.58	Not mentioned	Yang <i>et al.</i> (2019)
ASP, sedimentation	64	Fibres	Magni <i>et al.</i> (2019)
Aeration and clarification treatment	98.4	Not mentioned	Murphy <i>et al.</i> (2016)
MBR	99	Fragments, fibres	Lv <i>et al.</i> (2019)
OD	97	Fragments, fibres	
<b>(c) Tertiary Treatment</b>			
Coagulation	92.2–95.7	Not mentioned	Hidayaturrahman & Lee (2019)
Ozone	99.2	Not mentioned	
Membrane DF	99.1	Not mentioned	
RSF	98.9	Not mentioned	
Nitrification on plastic media TF	96	Fibres, fragments	Blair <i>et al.</i> (2019)
RSF	97.2	Fibres	Michielssen <i>et al.</i> (2016)
Anaerobic MBR	99.4	Microbeads, fragments	
RSF	75.49	Polypropylene, polystyrene	Bayo <i>et al.</i> (2020)
Denitrification, UF, ozonation, UV	95.16 ± 1.57	Not mentioned	Yang <i>et al.</i> (2019)
RSF and disinfection	84	Fibres, fragments	Magni <i>et al.</i> (2019)
DF (pore size = 10 µm)	40	All size fractions (20 µm – 5 mm)	Talvitie <i>et al.</i> (2017a)
DF (pore size = 20 µm)	98.5	All size fractions (20 µm – 5 mm)	
RSF	97.1	All size fractions (20 µm – 5 mm)	
MBR	94	All size fractions (20 µm – 5 mm)	
DAF	95	All size fractions (20 µm – 5 mm)	

The skimmers remove low-density microplastics in the primary treatment process as they float to the surface. Further, [Alavian Petroody \*et al.\* \(2020\)](#) stated that the skimming mechanism could remove a wide range of low-density microplastics of size >500 µm. Microbeads, a significant category of primary microplastics with low density, are removed in the primary treatment process ([Murphy \*et al.\* 2016](#)). Another explanation could be that most microbeads are made of polyethylene and associate themselves with grease and fat floating on the water surface, only to get skimmed out.

[Mourgkogiannis \*et al.\* \(2018\)](#) studied 101 WWTPs in Greece and noted that 34% did not have any primary treatment, especially the smaller WWTPs designed to cater to population sizes of 10,000–15,000. The lack of primary treatment can radically increase the microplastic release into the environment.

## 4.2. Secondary treatment

Secondary treatment aims to treat the wastewater emanating from primary treatment and eliminate the residual organics and suspended solids. In this stage, aerobic or anaerobic biological treatment methods are employed to remove dissolved and colloidal biodegradable organic matter. During secondary treatment, often a secondary clarifier is put to use. [Figure 2\(b\)](#) represents the standard secondary treatment unit process, where microplastic removal efficiencies have been studied. Microplastic removal efficiencies of standard secondary treatment unit processes have been tabulated in [Table 4\(b\)](#).

Biological treatment is not known to remove microplastics directly. Significant microplastic removal at the secondary treatment stage happens due to microplastics settling with organic matter as flocs. Microplastics thus find their way into sludge streams from the biological treatment systems.

Biological treatment processes are pretty diverse, and each one responds to various types of microplastics uniquely. Hence, the microplastic type that is removed from each unit process also varies. For example, MBR technology and rapid sand filters remove particulates more efficiently than fibres ([Bayo \*et al.\* 2020](#)). Some processes are found to be more efficient than others in removing microplastics. MBR technology expressed a marginally better removal efficiency than the conventional activated sludge process ([Lares \*et al.\* 2018](#)).

The efficiency of the sequencing batch reactor was not affected by microplastics ([Kalčíková \*et al.\* 2017](#)). In the active filter process, microplastics provided surfaces for the attachment of microorganisms. However, they can lead to an uneven distribution of water due to the formation of spheres with suspended solids ([Talvitie \*et al.\* 2017b](#)). Microplastics can decrease volatile solids destruction, leading to an increase in waste sludge production. In the study done by [Wei \*et al.\* \(2019\)](#), waste sludge production increased by 9%.

Studies on the effect of microplastics on microorganisms showed a facilitating effect on biological treatment processes. However, activities of ammonium-oxidizing bacteria (AOB), nitrite oxidizing bacteria (NOB), denitrifiers, polyphosphate accumulating organisms (PAOs) were seen to be unaffected by microplastics ([Liu \*et al.\* 2019](#)).

Studies on the effects of microplastics on biological unit processes have not exhibited consistent results. It was also found that microplastics in wastewater have minimal effects on nitrification and denitrification processes ([Li \*et al.\* 2020b](#)). However, they caused a significant increase in N<sub>2</sub>O emission during denitrification. Nevertheless, this result is not consistent, as [Zhang & Chen \(2020\)](#) claim that microplastics inhibit the nitrification process and cause ammonium accumulation in water.

There has been no research on removing microplastics in secondary clarifiers because the studies reported in the literature combine secondary clarifiers with biological treatment. Secondary clarifiers, due to their sedimentation mechanisms, could significantly contribute towards the removal of microplastics.

## 4.3. Tertiary treatment

Tertiary (advanced) treatment is the final step in the wastewater treatment process. Here, left-over contaminants are present after previous treatment steps are removed. [Figure 2\(c\)](#) represents the standard tertiary treatment unit process, in which microplastic removal efficiencies have been studied. Tertiary treatment at WWTPs increases microplastic removal efficiency, and newer advanced treatments that could entirely remove microplastics are being researched. It is only in the recent past that advanced treatment processes to remove microplastics have gained momentum, as reported previously ([Talvitie \*et al.\* 2017a](#); [Bayo \*et al.\* 2020](#); [Padervand \*et al.\* 2020](#)). Microplastic removal efficiencies of standard tertiary treatment unit processes have been tabulated in [Table 4\(c\)](#).

Recent studies have shown that advanced treatment technologies are highly efficient in the removal of microplastics. Removal efficiencies of 75% ([Bayo \*et al.\* 2020](#)), 90% ([Hidayaturrahman & Lee 2019](#)), and 98% ([Talvitie \*et al.\* 2017a](#)) were

reported. Combining MBR, ultrafiltration (UF), and granular activated carbon has been shown to remove microplastics below the deductible levels (Baresel *et al.* 2019).

It was seen that unit processes that involve filtration mechanisms have performed better in the removal of microplastics. However, microplastics have irregular shapes that may wear the filters (Zhang & Chen 2020) and pose a high risk of membrane fouling due to their surface properties (Ma *et al.* 2019b).

Some studies consider nitrogen and phosphorous removal mechanisms and MBRs to be the tertiary treatment. These studies have also shown significant microplastic removal efficiencies (Bayo *et al.* 2020).

Coagulation is a commonly used method in wastewater treatment to remove suspended solids. It has been found that coagulation before the final advanced treatment increases the microplastics removal efficiency (Hidayaturrehman & Lee 2019). Microplastics' high surface area allows them to adsorb reagents used for coagulation, meaning a higher concentration must be used to achieve desired results. Similar problems will arise in air flotation because microplastics will adsorb other pollutants and agglomerate, which will not match the tank's design parameters.

Disinfection processes are a crucial part of tertiary treatment. Chlorination and ozonation are commonly used disinfection processes in wastewater treatment processes. However, little research has been done on the effect of chemical treatments on microplastics. Kelkar *et al.* (2019) studied the effects of chlorination on microplastics in WWTPs and concluded that common microplastic polymers have the potential to be affected by chlorination. The microplastic polymer most affected by chlorination is polystyrene. However, high-density polyethylene and polypropylene proved to be quite resistant to chlorine concentrations used in conventional WWTPs. Ozone treatment causes an alteration in the physical properties of microplastics and influences polymer degradation. They seem to increase the rate of degradation and affect the end products (Hidayaturrehman & Lee 2019). However, the long-term effects of ozonation have not been extensively studied. There is also a possibility of microplastics hindering the disinfection process by acting as a protective surface for the targeted microorganisms.

#### 4.4. Processing of sewage sludge

Sewage sludge processing encompasses the steps to manage and dispose off the sewage sludge produced during wastewater treatment. Sludge is produced in the primary settling tank, biological treatment processes, and secondary clarifiers.

All the microplastics removed during the wastewater treatment process as skimmings and settled sludge from primary settling tank and secondary clarifier, as waste activated sludge from biological treatment, end up in sewage sludge. Studying microplastics in sludge processing is expensive and time-consuming due to the difficulty in extraction.

Microplastics in the sewage sludge are received from varying sources in the sewage treatment process, so they vary widely in properties (size, shape, composition), making their removal complicated (Talvitie *et al.* 2017a). Gravity flotation as a pre-treatment process for sludge processing is the most commonly used method for their removal.

On studying the effect of microplastics on the anaerobic digestion of sludge, it was observed that methane production potential decreased due to the sludge's incomplete digestion because microplastics hinder its biochemical processing. However, there seemed to be no effect on the microbial community (Li *et al.* 2020a). The presence of microplastics also showed an increase in the dewaterability of sludge. Anaerobic digestion decreases the microplastic concentration in sewage sludge because it may be causing their breakdown (Mahon *et al.* 2017).

There is a paucity of research on the fate, behaviour, and removal of microplastics during sewage sludge processing, which is a matter of concern. High concentrations of microplastics present in the sewage sludge could find their way into landfills and enter the environment, if left untreated.

## 5. SPECIFIC METHODS FOR REMOVAL OF MICROPLASTICS IN WASTEWATER

The existing methods of removing microplastics in wastewater streams are either expensive, inefficient, or ineffective or use chemical reagents (coagulants), which eventually add to the sewage treatment process. If the coagulants are not appropriately treated, then they could be detrimental to the receiving environments. There have been developments in recent times on methods to remove microplastics from the wastewater stream. These can be broadly categorized into filtration-based and coagulation-based. However, recent times have seen the development of other methods (Table 5).



**Table 5** | Specific methods for removal of microplastics from wastewater

Classification	Method	Type of microplastic removed
Filtration-based methods	Membrane filtration	Fragments, microbeads
	A gravity-powered filtration system	Microplastics harnessed from generic face scrub
	Dynamic membrane filtration	Micro-particles
	Density separation	Polyethylene, polypropylene, polyester, polyethylene terephthalate, polystyrene, polyamide
Coagulation-based methods	Electrocoagulation	Polyethylene
	Sol-gel method	Polyethylene, polypropylene
Other methods	Magnetic recovery	Polyethylene and polystyrene beads
	Biodegradation	Polyethylene, polypropylene, polyethylene terephthalate

### 5.1. Filtration-based methods

Filtration has been reported to be the most cost-effective and relatively simple way to remove microplastics. This method does not use any external chemical reagents. However, the smaller size of microplastics can lead to the clogging of filters making their maintenance complicated (Zhang & Chen 2020).

A gravity-powered filtration system for removing microplastics from the secondary effluent in WWTPs was developed and tested (Beljanski 2016). It includes both filtration and backflush. However, the system was tested only for a synthetic microplastic-water solution and not evaluated for actual wastewater. Dynamic membrane filtration (DMF) uses a physical barrier (e.g., cloth or mesh) on which a cake layer is formed. DMF can be used to remove small-sized and low-density inorganics. Their ability to remove micro-particles from wastewater has been studied by Li *et al.* (2018). DMF has been shown to be more efficient, cost-effective, low energy-intensive, and poses a low risk of fouling than MBR (Ersahin *et al.* 2012).

Enfrin *et al.* (2019) suggest that the density separation method can separate microplastics by altering the density of wastewater using chemicals like  $ZnCl_2$ . However, it may not be feasible to employ density separation in WWTPs due to the volume of wastewater treated and the fact that the wastewater stream contains many other pollutants.

### 5.2. Coagulation-based methods

Electrocoagulation produces coagulants with metal electrodes. Here, the suspended microplastics are collected on a layer formed by coagulants. Electrocoagulation is a cost-efficient way of removing microplastics and has been studied in detail (Ma *et al.* 2019a; 2019b) to remove microplastics in drinking water treatment. Nevertheless, this method could be modified and adapted for the wastewater treatment process. It was also found that aluminium-based coagulants showed better performance than iron-based coagulants. The sol-gel method uses synthetic amorphous silica (which is chemically stable and acts as an adsorbent) to induce the agglomeration of microplastics so that they can be removed easily. This process can be used in a wide range of pH and can result in a floating flocculate (Herbort *et al.* 2018; Zhang & Chen 2020).

### 5.3. Other methods

However, it is interesting to note that methods that do not use either filtration or coagulation principles are also proposed to remove microplastics from aquatic environments. Microplastics' hydrophobic nature has been exploited in magnetic recovery by binding hydrophobic Fe nanoparticles to microplastics (Grbic *et al.* 2019).

There have been suggestions to increase the degradation of microplastics using biological agents that use polymers as a carbon source (Enfrin *et al.* 2019). There have been studies that demonstrated the biodegradation of polyethylene (Auta *et al.* 2018), polypropylene (Caruso 2015), and polyethylene terephthalate (Yoshida *et al.* 2016). However, there is a lack of research regarding the effect of microplastics on these organisms and the environment.

## 6. RESEARCH GAPS AND SUGGESTIONS

Research on microplastics and their removal has been studied since their identification as a pollutant in 2004 (Thompson *et al.* 2004). There are still many areas that require further understanding. Some of the key aspects have been reviewed in this section.



### 6.1. Definitions and characterization

- Finding an **all-inclusive definition** for microplastics, including their properties such as chemical composition, and solubility would contribute to a standard characterization (Verschoor 2015; Frias & Nash 2019).
- There is a lack of **standard experimental procedures** to be followed during sampling, characterization, and identification of microplastics (Silva *et al.* 2018; Prata *et al.* 2019).
- **Removal efficiency** definitions have not been consistent over different studies – total removal, removal in each treatment stage, based on total plastic mass, based on the number – are ways to measure removal efficiency, making comparative studies complicated (Ziajahromi *et al.* 2016).
- The removal efficiencies of various unit processes of WWTPs vary because of the varied nature and concentration of microplastics in the samples/influents used in each study. The differences introduced by the experimental setup and design parameters of the treatment systems cannot be ignored (Talvitie *et al.* 2015; Carr *et al.* 2016; Michielssen *et al.* 2016; Bayo *et al.* 2020).
- **The size distribution** of microplastics in unit processes of WWTPs has not been extensively studied. Size distribution over the wastewater processing stream could be connected to unit processes, size, composition, and type of microplastics (Lv *et al.* 2019).

### 6.2. Unit processes of wastewater treatment plants

- Although presently established wastewater treatment methods may be incapable of efficiently tackling microplastics, it would be beneficial (economically and technology-wise) to alter them to cater to microplastic removal.
- As the maximum microplastic removal happens at the primary treatment stage (Carr *et al.* 2016), there could be an attempt to study the influence of operational parameters such as loading rate on microplastic removal. Optimizing flotation and sedimentation in the primary settling tank would directly contribute to removing microplastics from this step. Flocculants that promote microplastics accumulation may be considered a possible means of increasing removal efficiency (Herbort *et al.* 2018; Zhang & Chen 2020). **Pre-treatment processes** studied have been limited to bar and screens and grit chamber. Other pre-treatment processes, such as equalization tanks and comminuting devices, have to be explored (Blair *et al.* 2019).
- **Skimmings** from primary settling tanks are often landfilled (Alavian Petroody *et al.* 2020). However, landfilling is not a permanent solution to microplastic pollution because the microplastics could find their way into soil and water from landfills.
- **As the biological treatment** processes are very diverse, research gaps exist on the behaviour and removal of microplastics through the biological treatment process, especially on attached growth processes.
- Despite being a vital component of the wastewater treatment process, **secondary clarifiers** have been overlooked. They have a sedimentation-skimming mechanism, which could potentially be significant in microplastic removal. There is only a handful of studies on the removal of microplastics by secondary clarifiers.
- Research on the impact of **chemical agents** on microplastics during wastewater treatment is imperative because many chemical agents are used in the treatment process, such as coagulants and disinfectants (Hidayaturrehman & Lee 2019; Kelkar *et al.* 2019; Zhang & Chen 2020).
- In the **filtration methods**, altering the membrane's material to suit the hydrophobicity and negative charge of microplastics can improve removal efficiency. Adjusting the size of bubbles in air flotation may be conducive to removing microplastics (Zhang & Chen 2020).
- **Agglomeration** between microplastics and other contaminants in the wastewater stream has not been studied extensively. Better insight into this may prove quite valuable for coagulation-based removal of microplastics (Hidayaturrehman & Lee 2019).
- **The separation** of microplastics from sewage sludge is a relatively unexplored area and challenging because it is complicated, time consuming, and expensive (Talvitie *et al.* 2017b).
- All microplastics removed from WWTPs end up in the sludge, yet, there is no comprehensive study on microplastics during **sludge processing**. The few studies that have been conducted are isolated and not corroborated by further research (Mahon *et al.* 2017; Li *et al.* 2020a; Zhang & Chen 2020). **Pyrolysis** could be an effective treatment method in permanently removing microplastics so that they do not re-enter the environment (Li *et al.* 2020a; Zhang & Chen 2020).

### 6.3. Microplastic-specific removal technologies

- Beyond altering existing WWTPs for microplastic removal, specific technologies targeted at microplastic removal must be explored. Some of the promising technologies have been listed below.
- **Filtration-based removal** has been explored, but they have filter clogging and membrane fouling issues (Ersahin *et al.* 2012; Beljanski 2016).
- **Coagulation-based removal** requires the addition of other chemical reagents into wastewater streams. The effects of these chemicals, if released into the ecosystem, are yet to be keenly studied (Herbort *et al.* 2018; Zhang & Chen 2020).
- **The biological treatment** of microplastics, although explored at the laboratory scale, has not been studied from all aspects, especially on the impact of plastic degradation on microorganisms and the formation of intermediate products and by-products and their effects on the environment (Caruso 2015; Tiwari *et al.* 2020). Adaptability of biological treatment at the actual industrial scale WWTPs remains unanswered (Caruso 2015; Beljanski 2016; Grbic *et al.* 2019; Tiwari *et al.* 2020).
- Specific **industries** such as cosmetics, personal health care, and packaging contribute disproportionately to microplastic production (Carr *et al.* 2016). Drawing policy frameworks for responsible usage and arresting microplastics' release into the environment would be conducive for reducing microplastic production itself.

## 7. CONCLUSIONS

Despite extensive research, there exists a lack of consensus on the definition, characterization, and identification of microplastics. Studies on the impacts of microplastics on the unit processes of WWTPs and vice versa are imperative, but rare. Their unique properties make microplastics pollutants that challenge their treatment and removal from WWTPs. From the available studies on microplastics in WWTPs, it can be concluded that although WWTPs remove the bulk of microplastics entering them, yet they act as a point source of microplastics pollution to the aquatic environment, through treated wastewater and to the terrestrial environment, through sludge. Understanding the fate of microplastics in unit processes is crucial in modelling the impact of microplastics as pollutants in each unit process. Microplastics entering and leaving WWTPs are exceedingly diverse in their size, type, shape, chemical composition, and concentration, which further vary based on the unit process, time of the day and year, population equivalent the WWTP caters to and the influent (domestic/industrial). WWTPs serve as epicentres for converting primary microplastics entering with the wastewater into secondary microplastics that emanate out with the sludge and treated wastewater. Microplastic-specific removal technologies remain an unexplored area of research. However, the currently available technologies can be broadly categorized into filtration based, coagulation based, and biological degradation. This review would serve as a ready reckoner for a comprehensive understanding of interactions between a variety of microplastics and the unit processes of WWTPs.

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## DATA AVAILABILITY STATEMENT

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

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