Approaching the environmental problem of microplastics: importance of WWTP treatments

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Abstract:

The undeniable presence of microplastics (MPs) in soil, air and, especially, in the aquatic environment has revealed them to be an emerging pollutant. One of the main sources contributing to the release of these microplastics into the environment is wastewater treatment plants (WWTPs). During the treatment of wastewater, these microparticles undergo incomplete retention, which leads to their discharge in huge amounts into water masses. The microplastics removed from the wastewater during the treatment processes usually become entrained in the sewage sludge, which is commonly employed as organic fertilizer. Alarming data regarding the occurrence of MPs in nature and the increasing public awareness of environmental concerns have led to the appearance of numerous studies on this topic in recent years. So, this work is focused on providing an overview of available processes for the removal of microplastics from water and also from sediments. Social demand for the correct and effective management of microplastics is constantly increasing and should be given careful consideration before future action is taken.

Recycling is a good option, and policies might be developed in this direction, moving towards a circular and sustainable economy for plastics.

Keywords: microplastics, wastewater treatment, WWTP, processes, removal, recycling.

Acronyms: ABS, Acrylonitrile butadiene styrene; ACRYL, Acrylate; ADWTP, Advanced drinking water treatment plants; BAF, Biological active filter; BAW, Bulk acoustic wave; BPL, Biopolymer; CAS, Conventional activated sludge; DAF, Dissolved air flotation; DM, Dynamic membranes; DWTP, Drinking water treatment plants; EPS, Exopolymer substances; FTIR, Fourier transform infrared spectroscopy; FTIR-ATR, Attenuated total reflectance-Fourier transform infrared spectroscopy; GHG, Greenhouse gas; HDPE, High density polyethylene; HRT, Hydraulic retention time; LDPE, Low density polyethylene; MBR, Membrane bioreactors; MCR, Methacrylate; MPs, Microplastics; MPSS, Munich plastic sediment separator; NOAA, National Oceanic and Atmospheric Administration; NYL, Nylon; PAHs, Polycyclic aromatic hydrocarbons; PAM, Polyacrylamide; PBT, Polybutylene terephthalate; PC, Polycarbonate; PCBs, Polychlorinated biphenyls; PCLs, Polycaprolactones; PE, Polyethylene; PEP, Polyethylene-propylene; PES, Polyester; PET, Polyethylene terephthalate; PFE, Pressurized fluid extraction; PHAs, Polyhydroxylalkanoates; PHBs, Polyhydroxybutyrates; PMMA, Polymethyl methacrylate; PLA, Polylactic acid; POM, Polyoxymethylene; PP, Polypropylene; PS, Polystyrene; PUR, Polyurethane; PVAL, Polyvinyl alcohol; PVC, Polyvinyl chloride; PVS, Polyvinyl stearate; RO, Reverse Osmosis; RSF, Rapid sand filtration; SEM, Scanning electron microscope; SMI, Sediment-Microplastic Isolation; UF, Ultrafiltration; UV, Ultraviolet irradiation; WPO, Wet peroxidation; WWTP, Wastewater treatment plant.

1. Microplastics characteristics and occurrence

In the last five years, a large number of studies have reported the presence of MPs in nature, especially in aquatic environments, i.e., oceans, rivers, estuaries, lakes, arctic water and estuaries, among others (Bellasi et al., 2020; Li et al., 2020a; Xu et al., 2020). Nevertheless, few studies have analysed the presence, fate or effect of MPs in terrestrial ecosystems (Hurley and Nizzetto,

2018; Peng et al., 2017). As can be seen in Figure 1, in general, the number of publications on microplastics has dramatically increased from 2011 to now, which clearly indicates that, nowadays, this is a topic of great interest. Additionally, a progressive increase in the number of articles can also be observed when more specific terms related to wastewater treatment are included in the search (Figure S1).

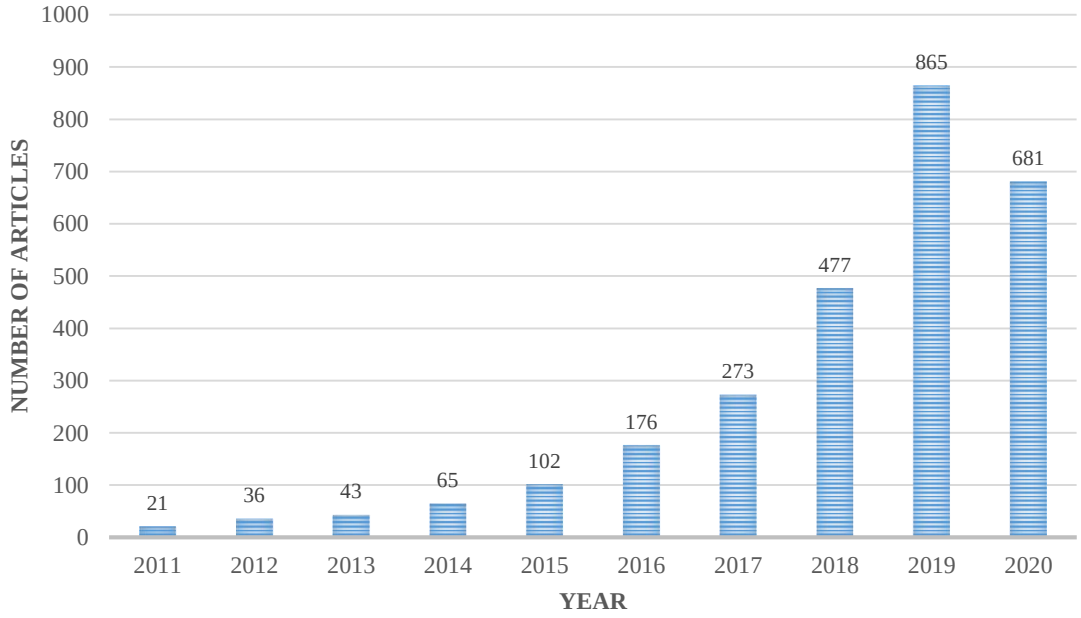

Figure 1. Number of articles found on Scopus database between 2011 (January) and 2020 (April), inclusive, when searching using "microplastic" as keyword.

Microplastics are particles of varying origin that are mainly characterised by their small size (less than 5 mm) (Law and Thompson, 2014; Picó and Barceló, 2019). They are considered "secondary microplastics" when they originate from the physical, chemical or biological degradation of larger plastics. "Primary microplastics" on the other hand, can be found as components of cleaning and hygiene products, cosmetics, paints, detergents, etc. Regardless of their origin, the vast majority of microplastics end up in the environment (Auta et al., 2017; Galafassi et al., 2019; Xu et al. 2020). The ubiquitous presence of these contaminants in different natural environments entails a potential risk (Alimi et al., 2018; Chen et al., 2020; Waring et al. 2018), not only for animals, but also for humans, mainly due to entry of microplastics into the food chain (Barboza et al., 2020; Barceló and Picó, 2019; Carbery et al., 2018; Prata, 2018a). Certainly, microplastics have been

found in a wide variety of animals, such as molluscs, crustaceans, seabirds, marine mammals and more than 150 species of fishes (Chang et al., 2019). Their size is very similar to that of zooplankton, which can lead to the direct ingestion of these pollutants by aquatic animals. In addition, the interaction between microplastics and phytoplankton leads to the formation of heteroaggregates in the gut of prey organisms which potentially increase the probability of trophic transfer of MPs to predators (Egbeocha et al., 2018).

Ecotoxicological studies have shown that some species may suffer from obstruction of the digestive tract, oxidative stress, damage to different organs, weakening and even death due to the ingestion of microplastics (de Sá et al., 2018; Franzellitti et al. 2019; Lu et al., 2019; Wang et al., 2019a). For example, MPs have been found in the gills and liver of European sea bass (*Dicentrarchus labrax*), generating a greater bioconcentration of mercury in the fish body and producing oxidative damage to lipids present in the gills (Barboza et al., 2018). Microplastics have also been found in European perch larvae (*Perca fluviatilis*), causing reproductive problems and increasing its mortality risk (Lönnstedt and Eklöv, 2016).

In the terrestrial environment, MPs have scarcely been studied. There are a variety of sources of MPs pollution in soils, such as sewage sludge from wastewater treatment plants, organic fertilizers from composting, plastic film and greenhouse covering in agricultural applications, atmospheric deposition, irrigation with polluted water or flooding and illegal waste dumping (Xu et al., 2019a; Wang et al., 2019b). It has been reported that microplastics accumulation can affect the germination and growth of some floral species (Qi et al., 2018). Besides, MPs have been found in earthworms, inhibiting their growth, producing gut damage and even death (Huerta Lwanga et al., 2016). Additionally, it has been reported that earthworms and collembola can act as vectors of MPs (Maaß et al., 2017; Rillig et al., 2017), so it is to be expected that other animals, including birds or mammals, could also transport microplastics.

The presence of MPs has extended to everyday foods, such as common salt, sugar, honey, beer, bottled water and even tap water, but also to other frequently consumed foods such as fish, lobsters, mussels, oysters and scallops, which entails the subsequently human ingestion of

microplastics (Chang et al., 2019; Zhang et al., 2019). Thus, although today there are insufficient studies to determine the specific effects of MPs on human health, they can be considered as a potential risk. But the ingestion route is not the only one that could be harmful to human health, since the inhalation of MPs present in the air and even dermal contact must also be taken into account (Prata et al., 2020). For example, from data reported on commonly consumed food, Cox et al. (2019) evaluated the potential intake of MPs per year by an American citizen. These authors estimated that a person may ingest 39000-52000 microplastics per year, values that depend on factors such as age and sex. This value increases if it is considered that bottled water could contribute to the intake of MPs with another 90000 particles annually, whereas tap water could produce an increment of 4000 MPs. Additionally, authors reported that the number of MPs that enter the human body is higher if the inhalation effect is considered, since this increases the MPs by 74000-121000 particles. Finally, and, although Revel et al. (2018) have suggested that nanoplastics could cross the epidermal barrier, dermal contact is not a major entry route for MPs into the human body. An additional problem related to microplastics is that contaminants could be retained on their surface, thus increasing their toxic effect (Prata et al., 2020).

2. WWTPs role in microplastics performance

There are different sources of plastic pollution to the environment, such as the mismanagement of plastics, the degradation of large plastics deposited in the environment or in landfills, the atmospheric transport of plastic particles deposited in soils, maritime activities (fishing and transport by ship), agricultural activities (greenhouses films, fertilizers, etc.), industrial activities directly related to plastics production, urban activities (personal and cosmetic products, paints, washing of textile, detergents, etc.) and, finally, wastewater treatment plants (WWTPs) (Boucher and Friot, 2017; Rochman 2018; Ziajahromi et al., 2016).

Water is the main vector for the spread of MPs and WWTPs are considered as a hotspot of contamination, despite often having microplastics removal efficiencies above 90% (Bayo et al., 2020a; Blair et al., 2019; Edo et al., 2020). WWTPs receive millions of MPs per day through the sewage system (Okoffo et al., 2019; Prata, 2018b) and the quantity and characteristics of these

microplastics depend mainly on the local agricultural and industrial activities (Eerkes-Medrano et al., 2015; Long et al., 2019). Thus, large amounts of microplastics that are not removed from wastewater during the treatment processes are discharged into rivers and oceans (Conley et al., 2019; Edo et al., 2020). For example, it has been reported that in a Scottish treatment plant serving a population of 650000 that, despite its having a removal efficiency higher than 98%, around 65 million microplastics were released to the environment each day (Murphy et al., 2016). In addition, MPs concentration is usually higher downstream of WWTPs compared to upstream (Kay et al., 2018; McCormick et al., 2014; Meng et al., 2020; Shruti et al., 2019). For example, Vermaire et al. (2017) found that the treated water (effluent) discharged into the Ottawa River (Canada) significantly increases the concentration of microplastics downstream of the WWTP (1.99 items per m^3) in comparison with upstream (0.71 items per m^3). Despite the fact that WWTP treatment processes achieve the removal of microplastics from wastewater mainly by entrapping them in sludge, these facilities increase the concentration of microplastics at the point of discharge, since 100% of removal efficiency is not achieved. Thus, it is clear that WWTPs are a significant source of microplastics release into the environment.

Additionally, it also should be considered that the concentration of MPs varies significantly depending on the season, being higher in hot periods (Bayo et al., 2020a). This variation is caused by higher levels of sun exposure which help fragmentation and degradation of plastics. Also, large amounts of microplastics are usually detected after rainfall due to the entrance into the sewage system of MPs washed from the soil. No noticeable differences were found during the day, the microplastics concentration in the mornings and afternoons being very similar.

Microplastics removed from the treated water are mostly accumulated in sludge, as reported in a study carried out in Canada, which found that 93% of MPs received by a WWTP with a 98% removal efficiency became entrained in sludge (Gies et al., 2018). Sludge can be managed in different ways, i.e., by landfill disposal, incineration and application to soil, the most frequent being the third one (around 50% of total sewage sludge generated in Europe and North America is employed as soil fertiliser) (Habib et al., 2020; Hurley and Nizzetto, 2018; Rolsky et al., 2020).

It is usually recognized that primary and secondary settling are the processes that entail the highest removal of microplastics, with average values from 78 to 98% and 7 to 20%, respectively (Prata, 2018b). On the contrary, Murphy et al. (2016) indicated that the highest removal efficiency of MPs (55%) was found during the pretreatment processes, specifically, during the grit and grease removal process. In general, it is considered that tertiary treatments have no noticeable effects on reducing microplastics concentration (Habib et al., 2020). Several studies have analysed the presence of MPs in mixed sludge, which are obtained from primary and secondary treatments, and the average concentrations were very variable, ranging between 400 and 7000 particles/kg in wet sludge and 1500 and 170000 particles/kg in dry sludge (Sun et al., 2019).

In all samples taken in wastewater treatment plants, MPs can be classified into six main shapes: fibres, granular, pellets, films, foams and fragments, fibres (57%) and fragments (34%) being the predominant shapes. Fibres are the most difficult particles to remove due to their morphological characteristics, namely, a high length-to-width ratio (Ngo et al., 2019). Furthermore, more than 30 polymers of different chemical compositions have been found in WWTPs (Sun et al., 2019). The abundance of one kind of polymer or another depends on the wastewater origin, i.e., industrial, agricultural or if the wastewater stream comes exclusively from the city (Ngo et al., 2019). Rolsky et al. (2020) indicated that the larger microplastics commonly settle with sludge during the primary and secondary settling processes. Additionally, although the smaller particles are more likely to remain in the water stream, many of them can also be absorbed in the sludge during the settling processes (Liu et al., 2019a).

It is important to note that MPs have the ability to adsorb chemical additives or toxic pollutants such as metals, polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), phthalates, pesticides, etc. (Rodrigues et al., 2019; Xu et al., 2019a). This can happen more easily during wastewater treatment because a variety of contaminants can be found in relatively high concentrations in the water stream. This entails a doubly adverse effect on environment and humans, since microplastics act as vector for these harmful contaminants (Caruso, 2019; Zhang and Chen, 2020).

It is clear from these data that research into technologies that allow the removal of MPs from sludge and also from environmental samples, especially those of marine origin, is a challenge that must be faced to enable us to move forward towards a circular economy for plastic wastes.

3. Efficiency of the different operations in WWTPs

In a wastewater treatment plant (WWTP) different processes can be distinguished and each treatment step has a specific objective in the overall purpose of cleaning the water flow. Sometimes, the composition of water, local circumstances or even the final fate of the effluent stream may require modifications to the conventional treatments. In addition, it is important to bear in mind that, at the present moment, these facilities are not designed for the specific removal of microplastics from the wastewater stream. One option to reduce the microplastic concentration before discharging treated water is the use of advanced treatments such as ozone, rapid sand filtration (RSF), dissolved air flotation (DAF), reverse osmosis (RO), dynamic membranes (DM), among others (Figure 2). Secondary treatments employing membrane bioreactors (MBR) can also improve MPs removal during wastewater treatment (Zhang and Chen, 2020). To a greater or lesser extent, all of these techniques contribute to reducing the amount of microplastics in treated water, reducing the subsequent pollution of water masses. A summary of the main data on MPs in WWTPs found in recent research can be seen in Table 1.

1 Table 1. Summary of main data on MPs in WWTPs found in literature from 2016 to 2020.

References	Stages sampled	Sampling	Size (µm)	MPs by size (µm)	MPs by shape	MPs by chemical composition (%)	Removal efficiency (%)
Carr et al., 2016	Influent, primary, secondary and tertiary	Surface filtration	45 – 400	90 – 100 (13%) 100 – 125 (25%) 150 – 200 (37%) 250 – 300 (25%)	Mainly fragments	Blue and white PE	Gravity filters: 99.9
Michielssen et al., 2016	Influent, pretreatment (grit and grease removal), primary, secondary and tertiary	Container	20 – 4750	<u>Pretreatment</u> 20 (8%) 106 (8%) 300 (64%) 850 (19%) 4750 (1%) <u>Effluent</u> 20 (8%) 106 (16%) 300 (32%) 850 (40%) 4750 (4%)	<u>Influent</u> Fibres (55 – 62%) Fragments (23 – 26%) Microbeads (11 – 16%) Others (4%) <u>Effluent</u> Fibres (61 – 89%) Fragments (11 – 33%) Others (0 – 6%)	–	CAS: 95.6 RSF: 97.2 MBR: 99.4
Murphy et al., 2016	Influent, pretreatment (grit and grease removal), primary and secondary	Container	> 65	598 (average on liquid fraction)	<u>All the liquid fractions</u> Flakes (67%) Fibres (19%) Films (10%) Beads (3%) Foams (1%)	<u>Influent</u> Alkyd (29%), PS acrylic (19%), PES (11%), PUR (9%), Acrylic (8%) <u>Effluent</u> PES (28%), PA (20%), Acrylic (12%), PP (12%), Alkyd (8%)	98.4

Leslie et al., 2017	Influent and effluent	Container	20 – 300	<u>Influent</u> 10 – 300 (39%) 300 – 5000 (42%) <u>Effluent</u> 10 – 300 (71%) 300 – 500 (27%)	Fibres, foils and spheres	–	11 – 94 MBR: 25
Mintenig et al., 2017	Effluent	Pumping	20 – 5000	<u>MPs > 500 µm</u> 500 – 2500 (94%) > 2500 (6%) <u>MPs < 500 µm</u> < 50 (9%) 50 – 250 (86%) > 250 (5%)	Fibres (61%)	<u>MPs > 500 µm</u> PE (59%), PP (16%) <u>MPs < 500 µm</u> PE (40%), PVAL (16%), PA (8%), PS (8%)	97
Talvitie et al., 2017a	Influent, effluent	Pumping and sieving	20 – 300	<u>Influent</u> 20 – 100 (39%) 100 – 300 (43%) ≥ 300 (18%) <u>Effluent</u> 20 – 100 (70%) 100 – 300 (28%) ≥ 300 (2%)	<u>All the samples</u>: mainly fibres Before treatment (39 – 81%) After advanced treatment (29 – 100%)	<u>All the samples</u> PES (60%), PE (14%), ACRYL (7%), PVC (5%), PS (4%), PP (3%)	RSF: 97 Disc-filter: 40 – 98.5 DAF: 95 MBR: 99.9
Talvitie et al., 2017b	Influent, primary, secondary and tertiary	Pumping and sieving	20 – 300	<u>Influent</u> 20 – 100 (39%) 100 – 300 (42%) ≥ 300 (19%) <u>Effluent</u> 20 – 100 (71%) 100 – 300 (27%)	<u>Influent</u> Fibres (68%) Fragments (10%) Flakes (14%) Films (8%) <u>Effluent</u> Fibres (29%)	<u>Effluent</u> Cotton (44%), PES (33%), linen (11%), viscose (9%), wool (2%), polyacryl (1%)	BAF: 97

				≥ 300 (2%)	Fragments (62%) Flakes (9%)		
Ziajahromi et al., 2017	WWTP – A: Primary and effluent WWTP – B: Primary and secondary WWTP – C: Primary and tertiary	Pumping and sieving	25 – 500	Mainly 20 – 190 (61 – 65%)	Mainly fibres and granules	<u>Primary effluent</u> WWTP – A: PET (80%), PE (20%) WWTP – B: PET (35%), NYL (28%), PE (23%), PP (10%), PS (4%) WWTP – C: PE (42%), PET (36%), PS (15%), PP (8%)	90
Gies et al., 2018	Influent, primary and secondary	Container	1 – 65	–	Fibres (66%) Fragments (28%) Pellets (5%) Others (1%)	NYL, PES, PVC, PS, etc.	97 – 99
Gündoğdu et al., 2018	Pretreatment (after screening) and effluent	Container	> 55	<u>Influent</u> < 100 (2%) 100 – 500 (24%) 500 – 1000 (18%) 1000 – 5000 (56%) <u>Effluent</u> < 100 (3%) 100 – 500 (28%) 500 – 1000 (31%) 1000 – 5000 (38%)	<u>Influent</u> Fibres (70%) Fragments (19%) Films (11%) <u>Effluent</u> Fibres (60%) Fragments (20%) Films (20%)	<u>Influent</u> PET (56%), PE (27%), PP (13%) <u>Effluent</u> PET (56%), PE (25%), PP (16%)	73 – 76
Lares et al., 2018	Pretreatment (after screening),	Container	250 – 5000	< 1000 (64%) > 1000 (37%)	Fibres (50 – 90%) Fragments (10 – 50%)	PES (79%), PE (11%), PA (4%), Others (6%)	CAS: 99 MBR: 99.4

	primary and tertiary						
Lee and Kim, 2018	Influent and effluent	Container	> 106	Influent 106 – 300 (80%) > 300 (20%) Effluent 106 – 300 (76%) > 300 (24%)	Influent Fibres (50%) Fragments (50%) Effluent Fibres (60%) Fragments (40%)	–	Media process: 98 A^2O: 98.4 SBR: 99.1
Simon et al., 2018	Influent, secondary and effluent	Auto-sampler	1 – 500	Mainly < 130	Mainly fragments	Influent PP (39%), ACRYL (27%), PVC (11%), PE (7%), PUR (6%) Effluent ACRYL (34%), PP (19%), PE (9%), PES (20%)	99.3
Blair et al., 2019	Influent, pretreatment (grit and grease), primary, secondary and tertiary	Container	60 – 2800	–	Fibres (73%) Fragments (20%) Films (6%) Pellets (1%)	PP (23%), PVS (7%), PE (4%), Copolymer (3%), POM (1%)	Nitrification tanks: 96
Conley et al. 2019	Influent and effluent	Container	> 60	60 – 178 (46%) 178 – 418 (24%) > 418 (30%)	Fibres (78%) Particles (22%)	–	75 – 98
Hidayaturrahman and Lee, 2019	Influent, primary, secondary,	Container	> 1.2	–	Influent WWTP – A:	–	Coagulation-flocculation: 47 – 82

	coagulation-flocculation effluent and effluent				Microbeads (18%), fibres (47%), sheets (4%), fragments (31%) WWTP – B: Microbeads (70%), fibres (15%), sheets (4%), fragments (11%) WWTP – C: Microbeads (24%), fibres (18%), sheets (4%), fragments (54%) <u>Effluent</u> WWTP – A: Microbeads (79%), fibres (12%), sheets (3%), fragments (6%) WWTP – B: Microbeads (93%), fibres (5%), sheets (1%), fragments (1%) WWTP – C: Microbeads (56%), fibres (23%), fragments (21%)		RSF: 74 Disc-filter: 79 Ozone: 90
Kazour et al., 2019	Influent and effluent	Pumping and sieving	20 – 500	<u>Influent</u> < 20 (14%) 20 – 200 (60%) 200 – 500 (18%) > 500 (8%) <u>Effluent</u> < 20 (5%) 20 – 200 (62%)	<u>Influent</u> Fragments (75%) Fibres (25%) <u>Effluent</u> Fragments (60%) Fibres (40%)	<u>Influent</u> PS (38%), PE (20%), PA (17%), PUR (12%), PC (8%), PVC (5%) <u>Effluent</u> PE (40%), PS (15%), PP (12%), PA (10%), PET	98.8

				200 – 500 (20%) > 500 (13%)		(10%), PUR (5%), PVC (4%), ABS (4%)	
Liu et al., 2019a	Influent, primary, secondary and tertiary	Container	> 47	<u>Influent</u> 20 – 300 (50%) 300 – 1000 (35%) 1000 – 5000 (15%) <u>Effluent</u> 20 – 300 (72%) 300 – 1000 (18%) 1000 – 5000 (10%)	<u>Influent</u> Fragments (43%) Fibres (39%) Microbeads (4%) Ellipse (6%) Films (4%) Foams (4%) <u>Effluent</u> Fibres (45%) Fragment (45%) Films (6%) Microbeads (4%)	PA (53%), PE (13%), PP (9%), PVC (4%)	64 (Chlorination increase a 7%)
Long et al., 2019	Pretreatment (after screening) and effluent	Pumping and sieving	43 – 355	<u>Influent</u> 43 – 63 (24%) 63 – 125 (43%) 125 – 355 (21%) 355 – 5000 (12%) <u>Effluent</u> 43 – 63 (27%) 63 – 125 (32%) 125 – 355 (28%) 355 – 5000 (13%)	<u>Influent</u> Granules (50%) Fragments (30%) Fibres (18%) Pellets (2%) <u>Effluent</u> Granules (36%) Fibres (30%) Fragments (28%) Pellets (6%)	<u>Influent</u> PP (30%), PE (27%), PS (10%), PET (8%), PEP (11%), PES (3%), PA (0.3% <u>Effluent</u> PP (35%), PE (18%), PEP (19%), PS (10%), PET (8%)	79 – 98
Lv et al., 2019	Influent, pretreatment (grit and grease), oxidation ditch	Container and sieving	25 – 500	25 – 63 (8%) 63 – 125 (29%) 125 – 250 (12%) 250 – 500 (11%) > 500 (40%)	Fragments (65%) Fibres (21%) Films (12%) Foams (2%)	PET (47%), PS (20%), PE (18%), PP (15%)	Oxidation ditch: 97 MBR: 99.5

	effluent, secondary and tertiary						
Magni et al. 2019	Influent, primary, secondary and tertiary	Container	63 – 5000	Influent 10 – 100 (12%) 100 – 500 (36%) 500 – 1000 (17%) 1000 – 5000 (35%) Effluent 10 – 100 (27%) 100 – 500 (52%) 500 – 1000 (7%) 1000 – 5000 (14%)	Influent Films (73%) Fragments (21%) Fibres (6%) Effluent Fibres (41%) Films (38%) Fragments (21%)	Influent Acrylonitrile-butadiene (40%), PE (17%), PEP (14%), PES (4%), PP (4%), PUR (3%), PA (2%) Effluent PES (35%), PA (17%), PE (10%), ACRYL (7%), PUR (7%), PVC (3%)	Sand filter: 84
Yang et al., 2019	Pretreatment (after screening), primary, secondary and tertiary	Container	50 – 5000	–	Effluent Microfibres (86%) Microparticles (14%)	Effluent PET (42%), PES (19%), PP (13%)	A^2O: 95
Akarsu et al., 2020	Influent and effluent	Container	> 26	< 500 (34%) > 500 (66%)	Influent Fibres (79%) Soft plastics (13%) Hard plastics (8%) Effluent Fibres (46%) Hard plastics (33%) Soft plastics (19%) Others (2%)	PE, PP, Acrylic, PS and cellulose acetate	48 – 73

Bayo et al., 2020a	Influent, primary and secondary	Container	0.45 – 5000	< 200 – 400 (20%) 400 – 600 (35%) 600 – 1000 (28%) 1000 – 5000 (17%)	Fragments (47%) Films (34%) Beads (12%) Fibres (7%) Foams (0.2%)	LDPE (52%), PP (11%), HDPE (9%), ACRYL (5%), BPL (3%), NYL (2%), PEP (3%), PET (2%), PS (2%)	90.3
Bayo et al., 2020b	Pretreatment (grit and grease) and tertiary	Container	0.45 – 5000	200 – 400 (4%) 400 – 600 (24%) 600 – 1000 (31%) 1000 – 2000 (31%) 2000 – 5000 (10%)	Fibres (61%) Films (31%) Fragments (7%) Beads (1%)	LDPE (71%), HDPE (5%), ACRYL (5%), PP (5%), PS (4%), NYL (3%), MCR (2%)	RSF: 75 MBR: 79
Edo et al., 2020	Primary and secondary	Container and sieving	25 – 375	<u>Primary effluent</u> 25 – 104 (54%) 104 – 375 (34%) 375 – 5000 (12%) <u>Secondary effluent</u> 25 – 104 (48%) 104 – 375 (28%) 375 – 5000 (23%)	<u>Primary effluent</u> Fragments (60%) Fibres (28%) <u>Secondary effluent</u> Fragments (80%) Fibres (20%)	<u>Primary effluent</u> PES, PE, dyed cotton, PP and cellophane fibres <u>Secondary effluent</u> PE, PET, PP and cellophane	A^2O: 94
Park et al., 2020	Pretreatment (grit and grease) and effluent	Container/ pumping and filtration	100 – 5000	–	<u>Pretreatment</u> Fragments (68%) Fibres (32%) <u>Effluent</u> Fragments (82%) Fibres (18%)	<u>Influent</u> PP (40%), PE (26%), PET (21%) <u>Effluent</u> PP (63%), PE (14%), PET (13%)	99.4 – 99.9 (CAS, A^2O and MBR)
Raju et al., 2020	Influent, pretreatment (grit and grease) and	Container	1.5 – 1000	<u>Influent</u> 1.5 – 38 (40%) 38 – 125 (23%) 125 – 250 (19%) 250 – 1000 (10%)	<u>Influent</u> Fragments (39%) Fibres (36%) Films (12%) Glitter (6%)	<u>Influent</u> PP (36%), PES (25%), PET (11%), PA (9%), PVC (7%), PS (5%)	76.6

	effluent (after UV treatment)	> 1000 (8%) Effluent 1.5 – 38 (17%) 38 – 125 (21%) 125 – 250 (40%) 250 – 1000 (13%) > 1000 (9%)	Foams (3%) Beads (1%) Others (3%) Effluent Fibres (58%) Fragments (25%) Films (8%) Glitter (2%) Beads (3%) Others (4%)	Effluent PES (46%), PP (24%), PA (11%), PE (9%), PMMA (3%), PBT (3%), rayon (3%)

2

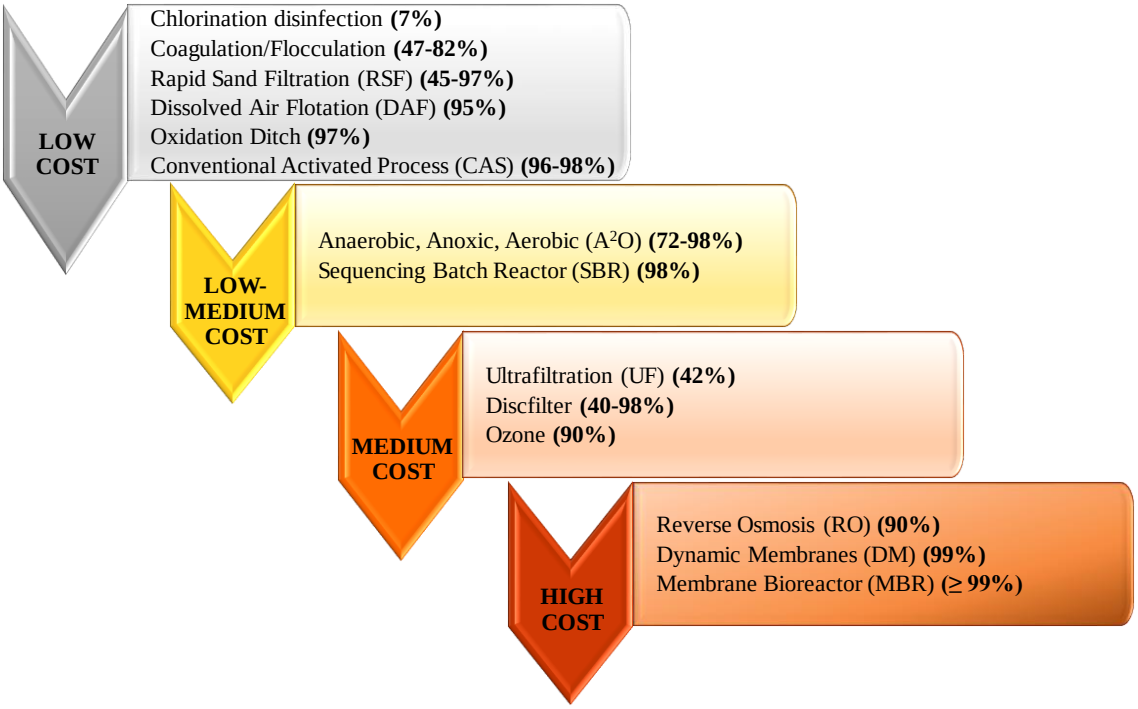

Figure 2. Removal efficiency of microplastics during different wastewater treatment processes in WWTPs and overview of the costs of the technologies employed (Hidayaturrahman and Lee, 2019; Lares et al., 2018; Lee and Kim, 2018; Liu et al., 2019a; Lv et al., 2019; Magni et al., 2019; Michielssen et al., 2016; Murphy et al., 2016; Simon et al., 2019; Talvitie et al., 2017; Yang et al., 2019; Ziajahromi et al., 2017).

3.1. Traditional operations

Rapid sand filtration (RSF) is a popular technology for water treatment because it provides rapid and efficient removal of pollutants at a low cost. The highest MPs removal percentage achieved with RSF was 97% (Talvitie et al., 2017a), but its main drawback is that this technology can fragment microplastics into smaller particles (Prata, 2018b).

Dissolved air flotation (DAF) is a technique consisting in saturating the water with air at high pressure so that a stream of air bubbles (20-70 µm) are generated into the wastewater. The air bubbles adhere to suspended solids, causing the flotation of the air-solid suspension. Finally, solids and MPs can be removed by skimming (Palaniandy et al., 2017; Wang et al., 2005). In comparison with the typical sedimentation processes that occur in the primary and secondary

clarifiers, DAF is less sensitive to flow variations and it shows high efficiency removal in a short retention time (5-10 min), especially of low-density particles which are less likely to settle. The highest removal efficiency achieved with DAF was 95% (Talvitie et al., 2017a).

According to the published data, the most effective technique for removing MPs consists in employing a membrane bioreactor (MBR), achieving efficiencies of near to 100% (99.9%). This system treats primary effluent, containing suspended solids and also dissolved organic matter, by means of the combination of membrane filtration processes with suspended growth biological reactors (Talvitie et al., 2017a). Only the smallest particles can pass through the system, so that it can achieve a high effluent quality (Poerio et al., 2019). The main disadvantages are the membrane costs, the high energy demand, control of fouling and the low flux (Ersahin et al., 2012).

Additionally, dynamic membranes (DM) are considered to be a promising technology in wastewater treatment processes for the removal of low-density microplastics, which cannot be removed by conventional settling (Li et al., 2018). There are a variety of factors that affect DM performance, including the membrane materials, membrane pore size, deposited materials (size, concentration) and operating conditions (pressure, cross-flow velocity, hydraulic retention time (HRT), oxidation-reduction potential, aeration, temperature, etc.) (Ersahin et al., 2012; Li et al., 2018; Ma et al., 2013). The high removal efficiency for microplastics (99.5%), the low cost of the filter module and the low energy consumption compared with traditional membranes, are some of the advantages of DM. The main disadvantage is the problem of blocking of the filter (Li et al., 2018).

3.2. Tertiary operations

Ozone is a strong oxidizer that is usually used in common disinfection processes by injecting a bubble flow containing ozone through the wastewater in a tank. Ozone reacts with polymers by the formation of radical oxidizing species, causing degradation of polymer chains. Low efficiencies are obtained when low pressures are used, since ozone is only slightly soluble in water (Rodríguez et al., 2008). Few studies have analysed the effect of ozone in MPs removal, but one

recent investigation showed that ozone technology can remove 90% of microplastics, using an average dose of 12.6 mg/L and a treatment time of approximately 1 min (Hidayaturrahman and Lee, 2019). This efficiency can even be higher (~ 98%) if the treatment lasts for 60 min and takes place at 35-45ºC, as Chen et al. (2018) showed in another study. This indicates that MPs can be degraded by ozone oxidation (Zhang et al., 2020). The main disadvantages of employing ozone are the high energy consumption and the formation of oxidizing by-products that may have toxicity equal to or greater than their precursor. Ozone has a strong affinity for organic pollutants, such as phenol, aniline or deprotonated-amine compounds, generating their respective derivatives, i.e., carboxylic acids or aldehydes. Additionally, an inorganic by-product, such as bromate, can be formed too (Ahmed et al., 2017; Benner et al., 2013; Wert et al., 2007).

Reverse Osmosis (RO) is another important technology in which water is pushed under pressure through a semi-permeable membrane and which is employed to remove large amounts of contaminants from water. Ziajahromi et al. (2017) reported a removal efficiency of 90% for MPs in a WWTP that employed RO. These authors observed that smaller microplastics were not removed by the RO process due to membrane defects and/or small openings in pipework. The main disadvantages of reverse osmosis are high energy demand, membrane fouling and waste management (Ahmed et al., 2017).

Finally, it should be taken into account that the recovery of microplastics in the tertiary treatment of WWTPs that employ bioreactors would imply that MPs would be obtained practically clean, since most organic and inorganic matter would have been removed in previous steps. This would be of interest if MPs are going to be revalorised, for example, by means of recycling processes.

4. Microplastics sampling in WWTPs

Collecting water samples from WWTPs requires the use of specific devices that include container collection, automatic sample collection, surface water filtration and pumping and filtration. Container collection implies collecting a spot sample, i.e., a specific volume of water is taken in a drum or a container and after that the sample is filtered through screens or meshes. Automatic sample collection is similar to container collection, but, in this case, the device pumps water

samples from time to time and stores them (a mixed sample is obtained), until they are filtered. Surface water filtration uses a net over the surface of the water to recover the particles, its main drawback is that a homogeneous sample is not collected (only surface water is sampled). Finally, pumping and filtration is the method most frequently employed in WWTPs, unlike the methods described for marine water samples, it allows large volumes of water to be processed. The sample is pumped through filtration equipment, consisting of several meshes and sieves whose pore sizes range between 20 and 4750 μm. In this way, microplastics are separated into several groups by size depending on the number and pore range of the meshes used in the device. The MPs obtained are subsequently washed from the meshes with distilled water to store them in glass bottles until further processing in laboratory (Alvim et al., 2020; Sun et al., 2019).

The extraction of MPs from sewage sludge is more difficult because they are usually embedded in the sludge. Further investigations into techniques and/or methods that allow the separation of MPs from organic matter are undoubtedly challenging, but are of great importance to reduce the subsequent microplastic pollution when sludge is employed for soil improvement.

5. Processes for microplastic removal

In this section, different processes will be mentioned, some of which have been studied in real plants, while the vast majority have been evaluated for MPs removal at laboratory scale. The future objective is to analyse, quantify and identify microplastics in wastewater, sediment or seawater samples after a pretreatment stage that serves to ensure the subsequent extraction of microplastics without impurities.

5.1. Pretreatment

As a general rule, the MPs extracted from samples, especially from those that come from wastewaters, are accompanied by impurities (organic and inorganic particles) which can interfere with the identification of microplastics. Different technologies have been employed to separate microplastics from impurities, though almost all of them have been employed at lab scale. The

elimination of impurities requires a pretreatment stage, which can consist of organic matter oxidation or the breaking down of the sludge to release the MPs entrained inside it.

5.1.1. Oxidation

The oxidisers most frequently employed are H_2O_2 and NaClO (Okoffo et al., 2019; Stock et al., 2019). The US National Oceanic and Atmospheric Administration (NOAA) recommends Fenton's reagent for aquatic samples (Masura et al., 2015). This is a catalytic wet peroxidation (WPO) that is usually carried out with a solution of hydrogen peroxide (H_2O_2) with ferrous iron (typically iron(II) sulphate, $FeSO_4$) as a catalyst. The Fenton's reagent allows the decomposition of almost 87% of organic matter in just 2 hours at room temperature without modifying the morphology and chemical composition of microplastics (Hurley et al., 2018; Tagg et al., 2017) and the process can be carried out in only 30 minutes if the reaction takes place at 70°C. The use of solutions of acids or bases to remove organic matter can damage microplastics if high concentrations are employed and offers poorer performance in terms of reducing impurities and needs longer treatment times (around 24 hours). A recent procedure employs enzymatic degradation, using a mixture of enzymes such as amylase, lipase, chitinase, proteinase and cellulase. The major advantages of enzymes lie in their ability to react specifically with individual components of a mixture, so that no alterations are made to the microplastics, whereas the main problem is that the exposure time for the total removal of organic matter is very long (13 days) (Liu et al., 2020; Sun et al., 2019).

5.1.2. Post-Breaking down

The extraction of MPs in sludge samples is difficult (Li et al., 2020b) and, in some circumstances, it may require a more specific and exhaustive pretreatment than the oxidation, e.g., like breaking up sludge using a low speed peristaltic pump.

Once the samples are pretreated, specific techniques should be employed at laboratory level to purify microplastics that will be subsequently identified by instrumental analysis (Huppertsberg and Knepper, 2018; Shim et al., 2017; Silva et al., 2018; Wirnkor et al., 2019). Firstly, this

identification is carried out by optical techniques (morphological and physical analysis) to determine the main shapes, colours and sizes of MPs and, secondly, by spectroscopic techniques (like FTIR or its equivalent FTIR-ATR) to assess the chemical composition of microplastics.

Techniques developed so far to selectively separate microplastics from organic and inorganic wastes that could not be removed during the pretreatment stage will be discussed in following sections. An overview of the main processes for MPs removal found in literature is shown in Table 2. Although most of the techniques have been developed and used for marine samples of water and sediments, they could be equally employed to treat samples that come from WWTPs, namely, wastewater and sludge.

134 Table 2. An overview of main processes for MPs removal found in literature.

Process	Process type	Procedure	Matrix	Removal efficiency (%)	Particle size (µm)	Treatment time	References
Froth Flotation	Physical	Injection of air bubbles in a liquid phase	Sediment samples, mixture of plastic wastes	55	1000 – 5000	–	Imhof et al., 2012
Coagulation/Flocculation	Chemical and physical	Addition of coagulants to generate flocs	Wastewater stream	47 – 82	64 – 1500	–	Hidayaturrahman and Lee, 2019
Elutriation	Physical	Density separation: saturated salt solution	Sediments	NaCl solution: Fibres (75), PE (61), PVC (0) NaI solution: Fibres (98), PE (100), PVC (100)	250 (granules) Different sizes (fibres)	1 h	Claessens et al., 2013
Munich Plastic Sediment Separator (MPSS)	Physical	Water flow	Marine sediments	50	5000 x 5000	10 min	Zhu (2015)
		Water column with sieving separation	Sediments (sand)	92 – 97 (PA, PVC)	125 – 2000	–	Kedzierski et al., 2016
		Density separation: $ZnCl_2$ solution		100	1000 – 5000	1 – 2 h	Imhof et al., 2012
				96	≤ 1000		
		Density separation: $ZnCl_2/CaCl_2$ (50/50)		13 – 39	–	1 h	Zobkov and Esiukova, 2017

Sediment-Microplastic Isolation (SMI)	Physical	Density separation: $ZnCl_2$ solution	Sediments	92 – 98	200 – 1000 PE, 200 – 1000 NYL, 100 – 800 PVC, 400 –1000 LDPE	10 – 120 min	Coppock et al., 2017
				88 – 92	> 200 PVC	10 – 120 min	Nel et al., 2019
Pressurized fluid extraction (PFE)	Chemical and physical	Solvents at supercritical pressure and temperature conditions	Oils, sediments, municipal waste	84 – 94	~ 50 HDPE, ~ 50 PVC, 1000 PS, > 1000 PET, > 1000 PP	–	Fuller and Gautam, 2016
				83 – 87	2000 PE > 200 PP	< 7 h	Dierkes et al., 2019
Electrostatic separator	Physical	Sample is dried, sieved and added to the drum for electrostatically charged	Sediments, quartz sand, beach sand, suspended particles	90 – 100	63 – 100, 200 – 630, 630 – 2000, 2000 – 5000 (HDPE, LDPE, PET, PP, PS, PVC, PMMA, PLA, PE, tire wear)	3 – 4 h	Felsing et al., 2018
Magnetic extraction	Physical	Hydrophobic interaction between iron nanoparticles and MPs	Seawater samples	92 (PE, PS)	10 – 20	–	Grbic et al., 2019
				93	≥ 1000		
				78 – 98	200 – 1000		

5.2. Froth flotation

Froth flotation (Figure 3A) is a physical process based on the hydrophilic and/or hydrophobic character of particles. Air is introduced in the bottom of the reactor while an agitator homogeneously disperses the air. Air bubbles are hydrophobic and interact with MPs (also hydrophobic), so that MPs-air bubbles go upwards to the surface, whereas hydrophilic particles remain in liquid phase. The supernatant, where MPs are located, is collected in a container. This procedure has not been tested for small plastics, only for microplastics between 5 and 1 mm in size, giving a removal efficiency of 55% (Imhof et al., 2012; Nguyen et al., 2019). In addition, its performance depends on different factors, such as the surface free energy of MPs, the surface tension of the liquid and the critical surface tension. It is well-known that microplastics, due to their hydrophobic characteristics, have the ability to adsorb different chemical pollutants. In that sense, the presence of contaminants adsorbed to MPs can negatively affect the separation of microplastics by the froth flotation process. In this context, additives that are adsorbed selectively by one kind of MPs but not to another can be employed to separate different plastics by this method (Crawford and Quinn, 2017; Fraunholcz, 2004).

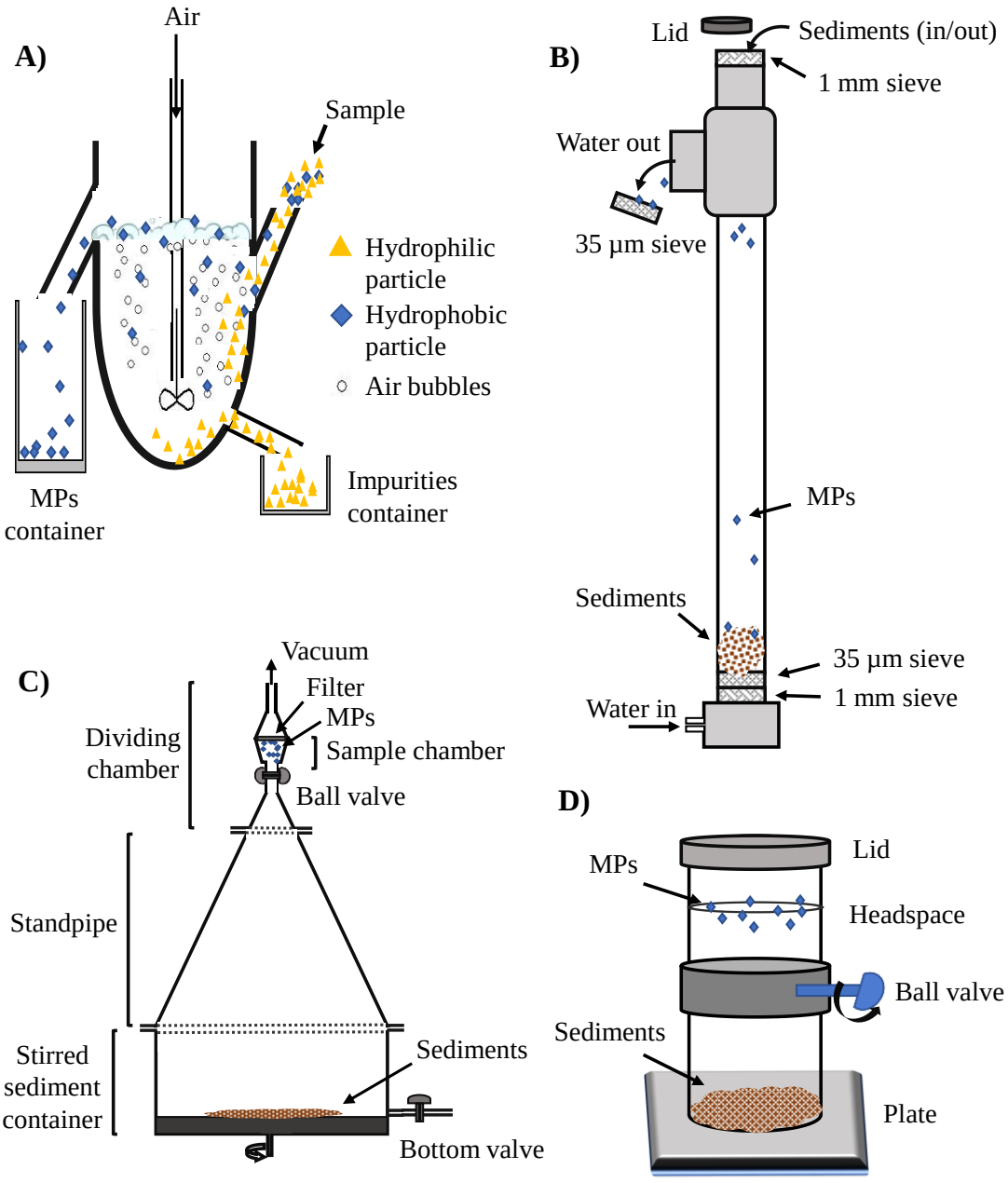

Figure 3. Schematic representation of different processes for microplastic removal. A) Froth flotation process, B) First elutriation system based on Claessen's device, C) MPSS device and D) Sediment-Microplastic Isolation (SMI) device. Adapted from Claessens et al. (2013), Coppock et al. (2017), Crawford and Quinn (2017) and Imhof et al. (2012).

It should be considered that in WWTPs with activated sludge treatments, an air stream is injected to supply oxygen to the biomass, so that MPs could be removed when a supernatant layer is formed.

5.3. Sedimentation

5.3.1. Importance of Coagulation-Flocculation

Coagulation-flocculation is a chemical water treatment process typically applied before a physical separation (usually sedimentation or filtration) in order to improve its performance. Coagulation consists in the destabilization of colloidal particles, whereas flocculation is the agglomeration of these destabilized particles into microfloc and then into bulky floccules by the addition of chemical reagents (coagulants and flocculants) (Baptista et al., 2015; Bratby, 2016; Lee et al., 2012; Ma et al., 2018; Te et al., 2016). Some factors, mainly pH and concentration of chemical reagents, are determining for optimising the coagulation-flocculation process.

The use of aluminium and iron salts, such as $AlCl_3 \cdot 6H_2O$ and $FeCl_3 \cdot 6H_2O$, as coagulants is very common in water treatment to eliminate microplastics. The first offers better performance but removing high quantities of MPs in the wastewategr stream implies using high dosages of coagulants and if $AlCl_3 \cdot 6H_2O$ is employed this can produce concentrations of Al-salts in water that could be harmful for human health (Li et al., 2020c).

The coagulation process has been studied in three WTTPs in South Korea using Al-based reagents as a coagulant to remove MPs, achieving efficiencies between 47 and 82% (Hidayaturrahman and Lee, 2019). Electrocoagulation is an innovative technology that involves an electrochemical reaction by liberating metals ions into water which originates flocs via electrolysis. Perren et al. (2018) have evaluated this technology by using artificial wastewater containing polyethylene microbeads of different concentrations. The wastewater was tested in a 1 L stirred-tank batch reactor and the effect of pH, NaCl concentration and density on removal efficiency were studied. Removal efficiencies higher than 90% were observed in all experiments and the optimum value (99%) was found at pH 7.5, indicating that electrocoagulation is an effective method for removing MPs from wastewater streams.

Coagulation is also commonly applied to drinking water. For example, several advanced drinking water treatment plants (ADWTP) in China have recently been analysed by Wang et al. (2020) and

186 a coagulation-sedimentation process allowed a reduction of 40-55% in MPs in treated water in
187 comparison with raw water. Pivokonsky et al. (2018) have studied the coagulation-flocculation
188 process, together with others, in three drinking water treatment plants (DWTPs) in the Czech
189 Republic and reported that it was possible to remove between 70 and 80% of microplastics.
190 Additionally, Katrivesis et al. (2019) found that when a coagulation-flocculation process was
191 applied in the production of potable water, the best MPs removal efficiencies were achieved in
192 the case of microplastic fibres (51-61%) and polyethylene terephthalate (PET) (59-69%).

193 Ma et al. (2019) reported that small particles of polyethylene (PE) ($d < 0.5$ mm) have the best
194 removal efficiencies (61%) when using Al-based coagulants in comparison with Fe-based ones
195 in ultrafiltration processes. On the contrary, large particles of PE ($2 \text{ mm} < d < 5 \text{ mm}$) reach only
196 18% of removal efficiency after the subsequent ultrafiltration.

197 Recent work (Cunha et al., 2019) has evaluated the interactions of microplastics and exopolymer
198 substances (EPS) excreted by marine/freshwater microalgae. Cultivated EPS were observed by
199 Scanning Electron Microscope (SEM) and it was found that the exopolymers formed a dense,
200 thick and viscous mesh. This leads to microplastics aggregation and adhesion to the surface of
201 exopolymers, generating hetero-aggregates, i.e., EPS + MPs. Even microplastics smaller than 106
202 μm can adhere to EPS.

203 Thus, coagulation-flocculation is an interesting technology for microplastic removal from water.
204 Further investigations should be carried out to widen the application of this process in wastewater
205 treatment plants with the aim of reducing the release of MPs, especially fibre particles, to the
206 environment (Ngo et al., 2019; Novotna et al., 2019; Wang et al., 2020).

207 *5.3.2. Density separation*

208 This method is based on density separation by flotation, which requires the use of a saturated
209 saline solution (a brine solution). It is a physical process and can be widely applied to samples
210 obtained from water and sediments. Microplastics float on the surface of the solution and are
211 isolated by filtration on screens, meshes or filter papers.

A large number of saline solutions can be used, such as sodium chloride (NaCl; d = 1.2 $g \cdot mL^{-1}$), zinc chloride ($ZnCl_2$; d = 1.5-1.7 $g \cdot mL^{-1}$), sodium iodide (NaI; d = 1.6-1.8 $g \cdot mL^{-1}$), calcium chloride ($CaCl_2$; d = 1.5 $g \cdot mL^{-1}$) and sodium polytungstate ($H_2Na_6O_{40}W_{12}$; d = 1.4 $g \cdot mL^{-1}$). In general, NaCl and $ZnCl_2$ solutions are the most commonly employed (Picó et al., 2019; Ruggero et al., 2020).

Sodium chloride solution is cheap and non-toxic to the environment, and is mainly employed for the extraction of microplastics from surface waters (freshwater and seas). Sometimes, it is also used for wastewater samples from WWTPs. Its major drawback is that it does not allow the extraction of some polymers, namely, polyvinyl chloride (PVC) and polyethylene terephthalate (PET) due to their high densities (between 1.14-1.56 and 1.32-1.41 $g \cdot mL^{-1}$, respectively) (Alvim et al., 2020; Sun et al., 2019). So, in that case, it is necessary to use denser saturated solutions, such as sodium iodide or zinc chloride, whose main disadvantages are high cost and toxicity. Comparatively, $ZnCl_2$ solution shows better performance than NaI, since it has a lower cost and it can be reused up to five times without losing efficiency in MPs extraction (above 95%). Obviously, this solution should be filtered through a micron pore size filter to be reused (Rodrigues et al., 2020).

Sludge or sediments accumulate MPs by settling, so they contain a certain amount of microplastics that, in general, have higher density than water. For this reason, solutions with higher density ($ZnCl_2$ or NaI) are needed to extract MPs from sludge or sediments (Picó et al., 2019; Rodrigues et al., 2018).

Microplastics float on the saline solution, so that they can be skimmed and filtered, being retained in the filter. The filter materials most commonly used are nitrocellulose, polycarbonate, glass microfiber and aluminium oxide (Alvim et al., 2020). The filter must satisfy some specific requirements such as water resistance, a pore size that allows water to pass through and with minimal spectral interference to avoid errors during the subsequent visualization and characterization of microplastics. In this sense, the best materials are glass microfiber and

aluminium oxide, because they exhibit great transparency in the infrared spectrum (Xu et al., 2019b).

5.3.3. Elutriation

The elutriation system is another methodology that has been designed at lab scale to separate heavier particles from lighter ones based on their sedimentation rate, when they are suspended in an upward stream flow (often a liquid). This process depends on the size, density and shape of the particles and is generally used to separate microplastics from sediments. A stream of liquid, usually water, is injected on the bottom of the elutriation column, whose size is usually between 147-186 cm in height and 106-150 mm in width. Particles go upwards in the column and at the top there is the storage and filtration device, which consists of several meshes where microplastics are retained. Optimised conditions allow the separation of MPs from the rest of the sediments. Its main advantages include low cost, low processing time (1 hour per sample) and high efficiency for large quantities of sediments, which makes it possible to process representative samples (Prata et al., 2019a).

The first elutriation system to separate plastic particles was developed by Claessens et al. (2013) (Figure 3B). Three parts can be distinguished in this system, the elutriation column, the pump and the storage and filtration device. In this first design, the column has a size of 147 cm in height and 15 cm in width. The sediment sample is introduced at the top of the column where a 1 mm sieve is fitted to avoid the entry of large particles. At the bottom of the column there are two screens: the 35 µm mesh has the function of a sample holder and is supported on a 1 mm mesh. In addition, under the screens are deposited three air-stones, whose main role is to homogenise the distribution of air bubbles generated by air pumping, improving aeration and recovery efficiency of MPs. It is suitable for 500 mL of sample and the mixture of air and water flows favours the separation of lighter particles from heavier ones. The optimal conditions of extraction are achieved by employing a flux of 300 L/h of water for 15 min. Particles go upwards and at the top of the column there is a sieve of 35 µm to retain MPs. Subsequently, solids retained in sieves are washed out

and separated with a saline solution (NaI; d = 1.6 $g \cdot mL^{-1}$) based on a density separation by flotation. The MPs removal efficiency of the process is 93-98%.

Later, Zhu (2015) developed a new elutriation system based on the Claessen device, with the aim of obtaining the maximum recovery efficiency of microplastics by modifying water flow and column diameter. In this case, the column is 50 cm in height and 5.06 to 10.16 cm in width and it contains an air-stone inside. At the bottom a water flow is injected and at the top there is a 3 mm sieve. To analyse the performance of this device, the authors employed as a control sample a mixture of 500 mL of sand and 50 pieces of plastic 5x5 mm in size. They found that the lower the water flow, the fewer MPs were recovered. In addition, these authors reported that the smaller the diameter of the MPs, the greater was the recovery efficiency. The optimal conditions were 385 L/h and 5.06 cm in column width, achieving in 10 minutes a recovery of MPs of 50%. In comparison with Claessen's device, the maximum recovery is much lower, but it should be considered that in this case, it is not necessary to employ NaI or NaCl solutions that could be toxic to the environment. Moreover, although its performance should be improved, Zhu's device showed great potential for microplastic pollution clean-up on beaches.

A new designed that involved further improvements was developed by Kedzierski et al. (2016). Four parts can be distinguished in this system, the storage and filtration device, the injection and flow control system, the elutriation column and the water temperature control system. In this case, the column has a height of 186 cm and 106 mm in width. It is a closed circuit to avoid wasting water and it has a flowmeter and a thermostat to regulate the water temperature (20ºC) and to minimise dynamic fluctuations of particles. In addition, several hundred grams of sediments can be processed. At the top of the column there are two meshes of 63 and 32 µm. These authors used this device to study the recovery efficiency of microplastics when water flow is varied. They found that 92% of PVC can be recovered when water flow is 0.013 $m \cdot s^{-1}$ and if flow increases to 0.019 $m \cdot s^{-1}$ the recovery efficiency reached 97%, but the amount of impurities (sand) is increased to 9%. In later work, in-depth studies to optimise this elutriation system were carried out (Kedzierski et al., 2017 and Kedzierski et al., 2018).

Finally, it should be remarked that the major removal of microplastics during wastewater treatment in WWTPs is achieved precisely by sedimentation processes. Specifically, during the grit and grease removal and the primary and secondary settling, it being possible in some cases to eliminate up to 98% of MPs (Habib et al., 2020; Hidayaturrahman and Lee, 2019; Prata, 2018b).

5.3.4. Combined sedimentation processes

Munich Plastic Sediment Separator (MPSS)

A particular elutriation system that involves a combination of sedimentation and flotation processes is the Munich Plastic Sediment Separator (MPSS) (Figure 3C). Three different parts can be distinguished, the sediment container with a rotor, the conical standpipe in which microplastics float and the dividing chamber with ball valve and filter holder to retain MPs. Using the bottom valve, a solution of zinc chloride (d = 1.6-1.7 $g \cdot mL^{-1}$) is injected and the density is adjusted with the aerometer. While the rotor is stirring, solids are slowly added at the top of the device to carry out the first separation, in which MPs float and solids settle. The mixture is stirred for 15 minutes and then it is allowed to rest for 1-2 hours. After that, the dividing chamber is placed on the device and using the bottom valve, more solution is introduced, allowing the MPs to rise to the open ball valve at the top of device. The valve is closed, and 68 mL of solution and MPs are retained inside. The bottom valve is opened to drain all the liquid from the standpipe and sediment container and finally, the chamber is rotated to filter the sample through a 0.3 µm mesh by means of a vacuum pump (Imhof et al., 2012). This system is suitable for samples of 5-6 kg. For natural marine sediments, the extraction efficiency by MPSS is 100% for microplastics in size 1-5 mm and 95.5% for MPs smaller than 1 mm.

Zobkov and Esiukova (2017) evaluated the MPSS device using a 50/50% mix of $ZnCl_2$ and $CaCl_2$ solution d = 1.48 $g \cdot mL^{-1}$), but in comparison to the data obtained for sediments the MPs extraction was much lower, between 13-39%.

Sediment-Microplastic Isolation (SMI)

316 The Sediment-Microplastic Isolation device (SMI) is a small and portable MPs extraction unit
317 (130 mm in width, 130 mm in diameter, 380 mm in height and weighing 1.5 kg) and is represented
318 in Figure 3D. It has been designed to isolate microplastics from sediments of environmental
319 samples (Coppock et al., 2017; Nel et al., 2019) and is based on the Munich Plastic Sediment
320 Separator (MPSS). Manufactured in PVC, it consists of a two-pipe system connected by a ball
321 valve fixed to a plate for stability. A solution of zinc chloride (around 700 mL) is poured inside
322 the device and after that the sample (30-50 g of sediments) is added. The sample and $ZnCl_2$
323 solution in the SMI unit are mixed by a magnetic stirrer and after 10 minutes the mixture is
324 allowed to settle. Sediments pass to the lower pipe and the supernatant, which contains the MPs,
325 floats in the upper pipe. Then the valve is closed, and after that the content of the upper pipe is
326 filtered on a micron pore size mesh to recover the microplastics. It can be applied in the field and
327 laboratory and its main advantages are the rapid, simple and efficient extraction of microplastics,
328 but it can only be used for small amounts of sample.

329 **5.4. Other physical processes**

330 *5.4.1. Pressurized fluid extraction*

331 Pressurized fluid extraction (PFE) has emerged as an alternative technique for separating volatile
332 organic compounds from solid materials. PFE uses solvents at supercritical pressure and
333 temperature conditions and the process depends on factors such as temperature, pressure, flow
334 rate, extraction time and different matrix parameters (dispersants, solvents, etc.). Different PFE
335 applications have been developed for the extraction of organic pollutants, bioactive compounds
336 from seaweed or microalgae, antioxidants, antibiotics and other pharmaceutical compounds
337 (Alvarez-Rivera et al., 2020; Turner and Waldebäck, 2013). In the search for alternatives to obtain
338 MPs in high purity from oils, sediments and wastes, PFE has been proposed as a possible option.
339 The sample is pumped and enters the furnace, in such a way that by varying the solvent,
340 temperature and pressure conditions, the process can allow the extraction and separation of
341 partially emulsified microplastics. Additionally, different plastics can be stored in different
342 collectors, depending on their chemical compositions. Fuller and Gautam (2016) obtained high

343 removal efficiencies for different microplastics, i.e., HDPE (87%), PP (84%), PVC (94%), PS
344 (90%), PET (89%), whereas Dierkes et al. (2019) achieved removal efficiencies for PE (87%) and
345 PP (83%). So PFE could be an interesting alternative for recovering MPs because it implies
346 simplicity, low cost, short processing time and data uniformity, while its main drawback is that it
347 is only accurate for small samples (10-20 mg).

348 *5.4.2. Electrostatic separator*

349 Recently, a new approach for microplastics separation in samples based on the electrostatic
350 behaviour of these particles has been developed. Felsing et al. (2018) used a hamos KWS type
351 separator to isolate plastics from different samples, in particular, from quartz sand, suspended
352 particles and sediments from freshwater and beach sand. It begins by drying the sample, which is
353 essential to sift it. Then, the sample is poured into a rotating metal drum and transported to a
354 corona electrode, where each particle is electrostatically charged. Because of the conductive
355 capacity of certain sediments and particles, they can be separated from MPs, which are not
356 conductive. Materials with higher conductivity are discharged more quickly than non-conductive
357 materials so that the rotational movement of drum separates the particles and sends them to
358 different collectors. This system has the capacity to process several tonnes of sample per hour.

359 *5.4.3. Magnetic extraction*

360 Grbic et al. (2019) developed a method that magnetically extracts microplastics, taking advantage
361 of their hydrophobic properties. These authors synthesized hydrophobic iron nanoparticles
362 capable of binding to MPs, thus allowing their recovery by means of the magnetization process.
363 It has been applied to seawater samples and was able to extract 92% of polyethylene and
364 polystyrene particles whose size was between 10 and 20 μm. In addition, several microplastics
365 (polyethylene, polystyrene, polyurethane, polyethylene terephthalate, polyvinyl chloride and
366 polypropylene) with a size greater than 1 mm were also recovered from aquatic samples with a
367 removal efficiency of 93%, although the efficiencies were rather lower (78-84%) in microplastics
368 with an average size of between 200 μm and 1 mm. So, magnetic extraction can be widely applied
369 to recover MPs with different sizes, densities and chemical compositions.

5.5. Bioremediation

Bioremediation is another approach to eliminating microplastics that is being given increasing attention. Biodegradation of MPs using bacteria, fungi and algae has been recently investigated (Shahnawaz et al., 2019; Wilkes and Aristilde, 2017). All of these organisms have in common the slowness of the degradation process, so, today, employing them in WWTPs to remove MPs from wastewater and sludge is still unrealistic due to the long time period that the process would need (Caruso, 2015; Paço et al., 2017; Wu et al., 2017). For example, polyethylene succinate (PES), which is a biopolymer, can be degraded at a rate of 1.65 mg per day by the secretion of enzymes by the strain *Pseudomona sp. AKS2* (Tribedi et al., 2012). In order to extend this study, these authors used the same strain for low-density polyethylene (LDPE) degradation, finding that *Pseudomona* can degrade 5 ± 1% of LDPE films at 30ºC in 45 days. This degradation can be increased up to 14 ± 1% by adding mineral oil to the growth medium (Tribedi and Sil, 2013). Another interesting example is *Idonella sakainensis 201-F6*, which is capable of completely degrading films of 6 mm in diameter of one of the most widely manufactured plastics worldwide, namely, polyethylene terephthalate (PET), in 6 weeks at 30ºC. This means a degradation rate of 0.13 $mg \cdot cm^{-2} \cdot day^{-1}$ (Yoshida et al., 2016).

Eukaryotic species have been the focus of much less research, despite the fact that they are able to accumulate microplastics and later expel them. The problem with eukaryotes is that they are not able to degrade MPs. For example, Graham and Thompson (2009) found that several sea cucumbers can selectively ingest a large number of PCB-contaminated microplastics (93-149 particles for an individual). PCBs are accumulated in lipid-rich tissues and passed along to predators, including humans. Thus, this proved that MPs ingestion may be a risk for land-dwelling and pelagic vertebrates, but also for other marine communities like benthic invertebrates. On the other hand, higher plants offer the advantage that there is no evidence of their being damaged by MPs, so microalgae or macrophytes could be a good option for the removal of microplastics from the environment (Shahnawaz et al., 2019). In addition, recent work has reported bioremediation as a promising strategy for removing MPs from WWTPs, employing for example annelids,

echinoderms or seagrasses, although further research on this topic should be carried out (Masiá et al., 2020).

6. Global framework and alternatives

6.1. Policy guidelines

The global production of plastic was 359 million tonnes in 2018 and this amount increases each year. The World Economic Forum (WEF, 2016) estimated that plastic production will continue increasing at an annual rate of 3.8% until 2030, and after that at 3.5% until 2050. Some calculations estimated that the global production of plastics would reach between 1124 and 1900 million tonnes in 2050. China is the largest worldwide plastic manufacturer (51%), followed by North America (18%) and Europe (17%). The most widely produced plastics in the world are polyethylene (PE, 36%), polypropylene (PP, 21%) and polyvinyl chloride (PVC, 12%), followed by polyethylene terephthalate (PET), polyurethane (PUR) and polystyrene (PS) (less than 10% each) (Geyer et al., 2017). Plastic waste management can be carried out in different ways, i.e., recycling, incineration, landfill disposal and others (Rhodes, 2019; Shen et al., 2020). Considering post-consumer plastic waste generation in Europe, 32.5% is recycled, 42.6% is employed for obtaining energy and 24.9% is disposed of in landfill (PlasticsEurope, 2019).

Globally, around 80% of plastics are deposited in landfills or end up in the oceans (4-12 million tonnes), contributing to environmental pollution (Brooks et al., 2018). Recycling or reusing plastics is essential to prevent the dispersion of these wastes in the environment, but also to achieve a circular economy of plastic materials. Although since 2006 the amount of plastic wastes that are recycled has doubled in Europe (PlasticsEurope, 2019), globally, only 9% of plastics are recycled, and this percentage varies substantially depending on the country. For example, 7% in India, less than 10% in United States, 20% in France, 25% in China and 50% in Germany, Spain and Sweden (Ogunola et al., 2018; PlasticsEurope, 2019).

In Europe, plastic demand by sectors is as follows, 39.9% for packaging, 19.8% for building and construction, 9.9% for the automotive industry, 6.2% for the electrical and electronic sector, 4.1%

for household, leisure and sport activities, 3.4% for agriculture and 16.7% for others (mechanical engineering, furniture, medical sector, etc.). Evidently, packaging is the sector that consumes most plastic, generally employing low-density polyethylene (LDPE), high-density polyethylene (HDPE), polypropylene (PP) and polyethylene terephthalate (PET) (PlasticsEurope, 2019). In addition, it is estimated that plastics packaging production will double by 2030 and quadruple by 2050, because most products are packaged in this material. The main role of packaging is prevention, avoiding the risk of damage and contamination of products before consumption. Regarding plastic packaging, globally, 14% is recycled, 14% is incinerated, 40% is deposited in landfills and 32% is released to the environment. So, plastic packaging is a real concern because it is manufactured for single use, so it is ubiquitous in trash, and in addition, its recycling is difficult and implies high costs.

Plastics deposited in landfills (packaging materials being the plastic waste most commonly deposited) can be degraded by chemical, biological or physical processes. For example, it is well-known that UV irradiation can degrade plastic to small particles, including microplastics. MPs can be airborne or even be responsible for greenhouse gas (GHG) emissions. Certainly, MPs can become embedded in soil aggregates, modifying soil structure, which can increase GHG release because many processes in soil are highly sensitive to soil structure (Ren et al., 2020). In some cases, if landfills do not meet the requirements in terms of protection and security, plastics could cause environmental damage due to possible leaks or spills. Plastic and electronic wastes are often open burned, which releases a variety of contaminants to the atmosphere and to the soil (Gullett et al., 2007). Plastics and microplastics occurrence can be accompanied by the presence of toxic contaminants, since, as was mentioned above, they have the capacity to adsorb and concentrate different pollutants (Al-Odaini et al., 2015). These contaminating species may be degraded, together with the MPs to which they adhere, thus contributing to the emission of toxic gases to the atmosphere. In addition, if security measures in the landfill are not sufficiently effective, these toxic contaminants could be responsible for environmental risks due to possible illegal waste dumping (Hale et al., 2020). Therefore, landfill disposal must be the last option in plastic

450 management, due to its environmental impact, high pollution risks, and the land requirements and
451 the loss of resources it implies (Liu et al., 2018).

452 Incineration is a useful way to reduce large-scale plastic pollution and, simultaneously, to obtain
453 energy from wastes. Nevertheless, considering that each ton of plastic waste contains around 79%
454 of carbon, the emission of carbon dioxide to the atmosphere is an important drawback of this
455 management alternative (Prata et al., 2019b; Shen et al., 2020). In this sense, Hamilton et al.
456 (2019) estimated that each ton of plastic packaging waste incinerated emits 2.9 tonnes of CO_2,
457 but if the power generated and energy recovered by the burning process is considered, it is
458 calculated that net GHG gases can be reduced to 0.9 tons of CO_2. In 2015, plastic packaging
459 wastes generated 16 million tonnes of GHG emission and, given the tendency of packaging
460 production to grow, this will lead to 84 and 309 million of tonnes of GHG emission in 2030 and
461 2050, respectively (Shen et al., 2020).

462 Plastic marine litter harms activities such as tourism, fishery and sailing. It is estimated that 80-
463 85% of marine litter in the oceans are plastics and, in particular, a high percentage of them (70%)
464 corresponds to single-use plastic. The environmental impacts of plastic wastes are increasing each
465 year. Nowadays, plastic residues can be found in many marine species (turtles, seals, birds and
466 several species of fish), entering the food chain and becoming a potential risk to human health.
467 For these reasons, some policy guidelines have been introduced to tackle plastic pollution (Table
468 3).

469

Table 3. An overview of main strategies of European Commission to improve plastic waste management.

Year	Directive	Aim of legislative procedure	Measures*
2002	**Directive 2002/96/EC**	Prevention of waste electrical and electronic equipment	**Plastic containing brominated flame-retardant materials** must be removed and selectively collected from electrical and electronic machines
2004	**Directive 2004/12/EC**, amending Directive 94/62/EC	Preventing and minimising the environmental impacts of packaging waste	**Recycling at least 22.5% of plastic by 2008**
2008	**Directive 2008/98/EC**	Waste and repealing certain Directives (waste management)	**Obligation to collect waste separately** (at least paper, metal, plastic and glass) **by 2015** **Recycling 50%** of paper, metal, plastic and **glass by 2020**
2015	**Directive (EU) 2015/720**, amending Directive 94/62/EC	Reducing the consumption of lightweight plastic carrier bags	**Reducing and levying charges on plastic bags by 2019** Maximum annual consumption of **90 lightweight plastic bags per person by 2020** Maximum annual consumption of **40 plastic bags per person by 2025** **Recycling 50% of plastic bags by 2020 and 75% by 2025**
2018	**Directive 2018/850**, amending Directive 1999/31/EC **Directive 2018/851**, amending Directive 2008/98/EC **Directive 2018/852**, amending Directive 94/62/EC	Landfilling wastes and packaging management	**Recycling** at least **65% of packaging waste by** 31 December **2025** **Recycling** at least **50% of plastic waste by** 31 December **2025** **Recycling** at least **70% of packaging waste by** 31 December **2030** **Recycle** at least **55% of plastic waste by** 31 December **2030**
2019	**Directive (EU) 2019/904**	Reducing the impact of certain plastic products on environment	**Banning single-use plastic by 2021** Recycling 15% of fishing gear by 2025 Recycling 50% of cigarette filters by 2025 and 80% by 2030 **Collecting separately plastic bottles 77% by 2025 and 90% by 2029** **Recycling 25% of PET bottles by 2025 and 30% by 2030**
2019	**TA/2019/0071**	**PROPOSAL** of limit values of MPs for treated water and sewage sludge	**Address MPs performance in WWTPs, i.e., pollution in treated water and in sewage sludge (applied to soils)**

*The percentages are based on weight, with respect to total amount of wastes generated.

In 2015, the EU published Directive (EU) 2015/720 that restricted and regulated the use of plastic bags. This document included the use of plastic bags of less than 50 μm in thickness and also required charges to be levied on light plastic bags at points of sale. The directive aims at the reduction of consumption of light plastic bags to a maximum of 90 per person a year by 31 December 2019 and of 40 by 31 December 2025. The target is to achieve the recycling of 50% of plastic wastes by 2020 and of 75% by 2025.

In 2019, the European Commission adopted some additional arrangements to reduce plastic wastes (Directive (EU) 2019/904). One of the main strategies consisted in banning throwaway plastics (single-use plastic cutlery, cotton buds, straws and stirrers) by 2021. The main objective of this initiative is the prevention and reduction of plastic marine litter. Other strategies proposed are that at least 50% of lost or abandoned fishing gear containing plastic is collected per year, with a recycling target of at least 15% by 2025 (fishing gear represents 27% of waste found on European beaches) and cigarette filters containing plastic would have to be reduced by 50% by 2025 and by 80% by 2030. Countries must apply levy systems that cover the costs of cleaning the wastes such as tobacco filters and fishing gear. Additionally, measures to reduce the consumption of plastic food and drink containers should be introduced. Member states will have to achieve a 90% collection target for plastic bottles by 2029, and plastic bottles will have to contain at least 25% of recycled content by 2025 and 30% by 2030. The European Commission considers that the new measures will entail both environmental and economic benefits, for example: avoiding the emission of 3.4 million tonnes of CO_2 equivalent, avoiding environmental damage which would cost 22 billion euros by 2030 and saving consumers 6.5 billion euros by 2030.

Hence, it is clear that, from a policy perspective, the best way to reduce MPs pollution seem to be limiting plastic consumption (Shen et al., 2020). Research has shown that microplastics can affect animal health (Carbery et al., 2018; Chang et al., 2019; Franzellitti et al. 2019) and, although there is still no scientific evidence, MPs could also be a risk for human health. For these reasons, some countries such as Canada, Ireland, the UK and USA have introduced specific regulations on microplastics, such as the ban on the use of microbeads (primary MPs) in personal care and

cosmetic products (Lam et al., 2018; Prata, 2018b; Kentin and Kaarto, 2018). In the case of the USA, the legislation that banned the addition of microbead particles in these products has been in force since 2018 (The Microbead-Free Waters Act of 2015, 2015). Aware of the environmental concern and following the same line, the European Chemicals Agency (ECHA) has submitted a proposal to ban the addition of microbeads to personal care and cosmetic products. It is expected to be ready in June 2020 to be subsequently evaluated by the European Commission. This law could avoid the release of approximately 400 thousand tonnes of MPs into the environment during the next 20 years (ECHA, 2019). Additionally, the European Parliament submitted a proposal (TA/2019/0071) on MPs pollution in treated water and sewage sludge (European Parliament, 2019). If the European Commission accepts this request, the member countries would have a period of two years to regulate the presence of MPs in WWTPs.

The implementation of different strategies in WWTPs to reduce the release of MPs to the environment would be a worthwhile measure to reduce the pollution associated with microplastics. In the near future, WWTPs will have to face a change in their operating systems and procedures in accordance with the restrictive measures that will probably be imposed. Educating society on reducing, recycling and reusing plastics correctly would also be key elements in reducing the environmental risks arising from MPs.

6.2. Minimising the presence of microplastics in the environment

6.2.1. Production of microplastics wastes

As previously discussed, wastewater treatment plants (WWTPs) are not designed to remove microplastics from the wastewater stream, but they are capable of eliminating these pollutants with removal efficiencies of even higher than 90%. Different studies indicate that the vast majority of MPs that are removed from wastewater at different stages are concentrated in the sludge. It is not possible to know exactly how many microplastics are expelled from these facilities or are entrapped in sludge, but some specific studies have tried to estimate this value. For example, in a WWTP in Vancouver (Canada), assuming that it received 1.76 trillion microplastics annually, it was estimated that 1.28 trillion MPs went to primary sludge, 0.36 trillion

to secondary sludge and 0.03 trillion were discharged into the river (Gies et al., 2018). In addition, Edo et al. (2020) calculated that around 300 million microplastics per day were discharged into the Henares river from a WWTP located in Madrid (Spain), although a removal efficiency of 93% was achieved during the wastewater treatment process. In addition, it is well-known that 50% of sewage sludge generated in North America and Europe is used in agriculture as fertilizer, which means that annually total amounts of 63000-430000 and 44000-300000 tons of microplastics are released to the soil, respectively (Hurley and Nizzetto, 2018).

Washing machines are considered an important source of microplastic released to the environment, since it is estimated that, from a single garment, more than 1900 fibres are discharged into the drains during laundry (Browne et al., 2011). Specifically, a study carried out in Finland estimated that, in this country, the annual emission of polyester and cotton microfibres to wastewater facilities was between 154000 and 411000 kg (Sillanpää and Sainio, 2017). In addition, 35% of MPs found in oceans are fibres whose main origin is the laundry. So, in that sense, an interesting option for reducing microplastic pollution is using a bulk acoustic wave (BAW) system to filtrate and remove MPs in washing machines (Akiyama et al., 2020). This device achieves removal values of 99% and 95% for Nylon 6 and PET, respectively.

6.2.2. Transformation of microplastics waste

Given the previously mentioned data regarding the millions of microplastics that arrive at wastewater treatment plants (WWTPs), it would be of great interest to develop techniques to recover MPs for their subsequent reuse or utilisation. Several strategies employed for plastic management are described below. Although specific procedures to treat recovered microplastics are yet unrealistic, these techniques could be applied to MPs in the near future.

There are different procedures to convert plastics into fuel (Kunwar et al., 2016; Rajmohan et al., 2020). For example, the conversion of plastics to fuel can be suitable for LDPE, HDPE, PP, PET, PVC and PS, allowing the recovery of energy by a co-pyrolysis process. Additionally, syngas (a mix of H_2 + CO), an alternative energy source, can be obtained from various types of plastics (LDPE, HDPE, PS, PET, PP) by catalytic pyrolysis (Saad and Williams, 2016). Plastics can also

be used as a construction material as self-compacting concrete (Almeshal et al., 2020; Gu and Ozbakkaloglu, 2016; Faraj et al., 2020) or in clay composites (Istrate and Chen, 2018; Velásquez et al., 2019).

A program known as Operation Clean Sweep® is an international project that has been implemented in North America, the United Kingdom and Europe. Its main objective is to prevent plastic pollution in the marine environment. A manual of good practices has been written with the aim of helping plastics industry operations managers reduce the loss of resin pellets, flakes, and powder to the environment at all stages of the plastic chain, including production, handling, transportation and recycling (Operation Clean Sweep, 2018). In addition, transforming microplastics into reusable products would be an eco-friendly strategy, and in fact, this is an alternative that is today employed for plastic wastes. As an example, Precious Plastic by Hakken (2013), an open hardware plastic recycling project, relies on a series of machines and tools which grind, melt, and inject recycled plastic, allowing for the creation of new products out of recycled plastic on a small scale. There are also new technologies to produce textiles, ECONYL© (Econyl, 2016) recover nylon waste from landfills and oceans to obtain regenerated nylon that can be employed in various applications, closing the loop of the circular economy. Nevertheless, it should be remembered that most of these alternatives are limited to large plastic fragments and have not been developed specifically for microplastics management.

6.2.3. Substitution of conventional plastics by other alternatives

Production of biodegradable plastics could be a real solution as an alternative to substituting plastics in general. These synthetic polymers can be decomposed by the action of microorganisms in the environment, producing CO_2 and H_2O (Faris et al., 2014; Quecholac-Piña et al., 2020). A biodegradable plastic must meet some requirements such as: 50% of the mass should be organic, heavy metal limits should not be exceeded, up to 90% of plastic should be degraded in less than 6 months under soft conditions and the by-products obtained should be eco-friendly and not affect fauna and flora negatively. For example, some studies have shown alternatives that would reduce the amount of plastic employed in food packaging using biodegradable plastics with polylactic

580 acid (PLA). In addition, biomaterials such as polyhydroxylalkanoates (PHAs),
581 polyhydroxybutyrates (PHBs), polycaprolactones (PCLs), starch and cellulose have also been
582 studied, offering interesting characteristics for packaging applications (Din et al., 2020).
583 Bioplastics, like polyhydroxyalkanoates (PHAs) can be produced from sewage sludge and,
584 although these materials cannot be employed in the food sector, they can be used, for instance, in
585 the textile industry or the agricultural sector (Liu et al., 2019b). In addition, polyester-based
586 biodegradable plastic can be broken down by several microbial species with very efficient results.
587 For example, poly(ε-caprolactone) (PCL) can be degraded by a polymer-degrading bacterium
588 isolated from coastal water (strain TKCM 64) at a rate of 1.39 ± 0.09 mg·cm^{-2}·day^{-1} (Suzuki et
589 al., 2018), whereas lipases (5 mg·mL^{-1}) from *Lactobacillus plantarum* (MTCC 4461) can degrade
590 approximately 60% of PCL films in 10 days (Khan et al., 2017). Satti and Shah (2020) have
591 reported that biological degradation depends on temperature, pH, nutrients and the microorganism
592 population. In this context, to avoid long-term environmental damage, biodegradation is the best
593 option to reduce the impact of plastic wastes (Yogalakshmi and Singh, 2020).

594 Finally, it should be mentioned that in order to minimise the negative impacts derived from plastic
595 pollution, different strategies would have to be considered simultaneously, not only a reduction
596 in the use of plastics and their replacement by other alternatives such as cardboard or
597 biodegradable polymers, but also, eco-design, environmental education, improvements in waste
598 management and others (Ogunola et al., 2018; Prata, 2018b).

599 **7. Conclusions**

600 Recent work has proved that the presence of MPs in the environment is ubiquitous, and in
601 addition, these pollutants have potentially harmful effects, mainly through the food chain, on
602 animals and also humans. The release of microplastics to nature is a concern of increasing interest
603 to society, which has led to legislation aimed at reducing MPs pollution. For example, the use of
604 microbead particles in personal care and cosmetic products has been banned in the USA since
605 2018, and is expected to be forbidden in Europe before the end of 2020. WWTPs are capable of
606 removing from wastewater more than 90% of MPs, which are mainly entrapped in sludge.

607 However, these facilities still represent an important hotspot for the emission of microplastics into
608 aquatic ecosystems. In 2019, the European Parliament submitted a proposal to regulate MPs
609 pollution in treated water and sewage sludge. Thus, in the coming years WWTPs are likely to
610 face the implementation of strategies to reduce the amount of MPs in effluent and to manage the
611 microplastics retained in sludge. Additionally, research into new technologies to revalorise the
612 MPs recovered from different origins also represents an important challenge that must be met in
613 order to comply with future legislation.

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