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The Effect of Wastewater Treatment Plants on Retainment of Plastic Microparticles to Enhance Water Quality—A Review

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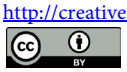


Abstract

Microplastics, plastic pieces of ≤5 mm in size, are ubiquitous in ther environment and can be found in both terrestrial and aquatic ecosystems. This manuscript reviews the literature on the fate of microplastics in wastewater treatment and briefly highlights novel developments in the removal of microplastics from aqueous systems.

Keywords

Microplastics, Nanoplastics, Wastewater Treatment Plants, Sludge Treatment, Plastic Retainment

1. Introduction

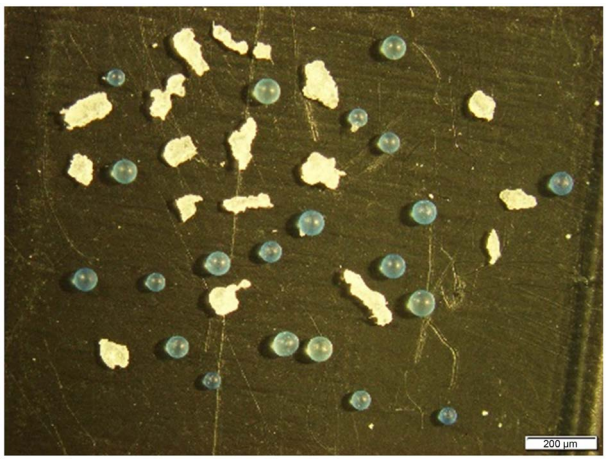
One year has passed since our first review on microplastics and wastewater treatment plants [1]. Since then, a number of other reviews on the topic have appeared [2]-[11] that complement reviews that had been there previously [12] [13] [14] [15] [16]. Nevertheless, the authors felt the need to extend the scope of the earlier review as it focused too much on plastic microbeads, an area of our research [17] at the time of writing, and too little on other forms of microplastics such as microtires [18] [19] and textile fibers [20] [21]. In addition, the contribution of the different sources of microplastics as found in the wastewater is changing over time, especially with the gradual phase-out of microplastic content in rinse-off cosmetics [22] [23] [24] and the ban in certain regions of plastic bags [25] and single use plastics [26], both potential materials for microplastics due to subsequent degradative fragmentation processes. Furthermore, there is continuous development and refinement of technologies to separate microplas-

tics from wastewater that warrant reporting. It is for these reasons that the current review was drafted.

Microplastics (MPs) can be defined as plastic particles of ≤5 mm in size [27] [28] [29]. Also, the term nanoplastic (NP) has been used for particles ≤ 1 μm in size [27] [30], although some authors define NPs as particles of up to 100 nm in size [31]. Here, the discussion is still ongoing [32]. These plastic particles have different sources. Some of the MP particles originate from the abrasive degradation of larger plastics such as from tyre materials and road-wear [33], clothing [34], plastic bags [35] and packaging [36], degradable over longer periods of time or even at the time of opening, and other larger pieces of plastic that are exposed to wear or weathering [37] [38] (**Figure 1**). These are called secondary MP. Then, there is the primary MP, which is materials that are produced at this small size for a specific purpose, micropellets in cosmetic formulations and in facial cleaners and body scrubs (median size of 0.2 - 0.4 mm) [16] [39], microspherules in toothpastes (2 - 5 μm in size) [40], microparticles in scrubbers used for air-blasting surfaces to remove paints and rust [41] [42] in paints and coatings [43], in detergents [43] and in drilling fluids in oil and gas exploration [1]. Recently, plastic micro-/nanoparticles have also been used in drug delivery systems [44] [45]. The amounts of materials used as primary MP and secondary MP stemming from the degradation of meso- and macroplastics on-land have been estimated in different studies commissioned by different European countries [43] [44] [46] [47] [48] and by the European Community [49]. The release of such materials, incl. of tyres, into sewage systems and as run-off into river systems or directly into the oceans has been estimated by different models, which sometimes took the known distribution of products other than MP as the basis for the models. Recently, the realization that such commonplace activities as using a tea-bag [50] or unwrapping a product [36] also lead to MP opens up further sources of such particles that need to be taken into account. Also, the atmospheric transfer of MP [51], which has been largely neglected until relatively recently, has been found to contribute to their accumulation in rivers, lakes [52] and oceans [53]. Much of the overall data on MP has been gained from overall a limited number of sampling studies at different locations, from which overall quantities of MP were derived using different models. It has been shown that different sampling techniques can lead to quite different results in the detection and quantification of MP in the environment [54] [55] and not in all areas is a consistent sampling technique for a matrix in use. In recent times, micro-Raman detection has been used to detect MP particles as small as 1 μm in size [56]. This compares well with Fourier-transform infrared spectroscopy, where the size limit of the MP is said to be 10 μm. In former times, small microparticles and nanoparticles were less easy to detect. Z. Wang *et al.* have shown that in certain regions of the Yangtze delta, 1 - 5 μm sized MPs make up to 58% of total, which were formerly left undetected. So, there is still a lot of uncertainty about the distribution of MP in the environment, both qualitatively and quantitatively.

(A)

(B)



(C)

Figure 1. Various sources of microplastics that may end up in the municipal sewage system. (A) Microplastic from a meat-cutting board; (B) Microplastic from road dust; (C) Microplastic from rinse-off cosmetics.

Nevertheless, there are numbers in regard to MP that one can work with. One of the most extensive studies on MP in the environment has been forwarded by

Eriksen *et al.* [57], where empirical data from 1571 locations taken in the years 2007-2013 has led to the estimation of 5 trillion (5×10^{12}) plastic pieces floating on the world's oceans, amounting to 268,940 tons, with 485×10^{10} particles of less than 5 mm in size. Plastic particles, even initially positively buoyant particles [58] [59], can sink through the water column to the ocean floor [60], where it is generally accepted that the ocean floor serves as a sink for marine plastic [61] [62]. There could be as many as 35 times more MP on the ocean floor than on the ocean's surface, amounting to 4.4 million tons of MP in the top 9 cm of sediment throughout the world's oceans [63]. Between 1.15 and 2.41 million tons of plastic waste enter the oceans every year from rivers [64], with an estimated 4.8 to 12.7 million tons of plastic waste entering the oceans from land via all methods of transport, as of 2010 [65]. This number has been predicted to increase by an order of magnitude by 2050 [65]. Plastics degrade and mineralize very, very slowly [66]. It is known that microbes colonize plastics in an aquatic environment [67] [68]. Nevertheless, while they do help to biodegrade plastics, especially through the excretion of enzymes that catalyze chemical transformations in the plastic, even microorganisms (*Zalerion maritimum* [69], Bacillus cereus [70], *B. gottheili* [71]) specially selected through screening processes lead to a weight loss of various MPs consisting of PE, PET, PP or PS by less than 8% in 40 days. However, MPs can also serve as a transport for microorganisms that can damage ecosystems and/or affect living organisms negatively. In this context, it is interesting to note that the composition of microorganisms on MPs can change as they move through wastewater treatment plants (WWTPs) [71].

What is without doubt is that one major stream that carries MP and can pass them potentially on to the environment is municipal and industrial wastewater. Here, WWTPs act as a defense of last resort. Therefore, it is essential to know in how far WWTPs can retain MP under different treatment regimes. Published, extensive studies on the MP retaining ability of wastewater treatment plants started in earnest with the work of Browne *et al.* in 2011 [72], with data from the year before. In the years 2016-2020, a large body of work was published on the MP retaining ability of wastewater treatment plants from around the world.

Sewage sludge as a by-product of the water treatment, released into the environment, often carries a significant load of MP as well. It has been estimated that between 63,000 and 430,000 tons of MP are added via sewage sludge to European farmlands, annually [73] [74]. Already in 1998, Habib *et al.* reported on the presence of synthetic fibers in agricultural soils treated with sewage sludge [75]. This work has been expanded by other groups to non-fiber MP and the distribution and fate of MP in soils is a matter of pressing interest.

The current paper aims to review the effect of wastewater treatment plants on MP abundance in treated water dis-charged into the environment with a focus on the retaining ability of different types of wastewater treatment methods. Also, the fate of MPs in wastewater treatment plants is looked at. Furthermore, the review takes into account new methodologies to retain MP from aqueous media, developed in laboratories.

2. Wastewater Treatment Plants and Microplastic

2.1. Major Contributors to Microplastic Waste

A major share of the microplastics entering the environment does so through sewage water. Some of the main contributors are microbeads from cosmetic formulations [76], textile fibers [77], and microtires and road wear particles (TRWP) [19]. Plastic microbeads in cosmetics are trending downward, as many countries have banned MP in many cosmetic products, such as in rinse-off cosmetics [78]. Nevertheless, products with MP content are still exported to countries where no ban is in place, even from countries that already have banned MP in the formulations [17] [79]. Fibers for the most part come from the shedding of particles during the washing process of textiles [72] [77] [80] [81]. Textile fibers are ubiquitous in the environment. We have found them in almost all samples that we have analysed, be they from lake shorelines, soil samples or even from commercially sold meat [82]. Nevertheless, in many regions, released textile fibers are for the most part natural fibers. Thus, Talvitie *et al.* have reported that the most common fibers emitted from a large wastewater treatment plant (WWTP) in Finland were natural (66%), where (natural) cotton and (synthetic) polyester made the largest contribution with 44% and 33%, respectively [83]. While natural fibers tend to degrade more quickly, organic pollutants can be adsorbed equally on synthetic and on natural fibers, where often textile fibers are impregnated with flame retardants. This can lead to an additional environmental impact. Tire and roadwear particles constitute a main contributor of plastic microparticles in the environment. While tire and roadwear particles have been investigated for a long time from a road safety point of view, their environmental impact has come to the fore only relatively recently [19]. Models have shown that appreciable amounts of microplastics from TRWP are swept into surface waters [84] [85], where a recent calculation showed that 8700 - 19,800 tons per year of TRWP, amounting to 12% - 20% of the emitted total, reach surface water, alone in Germany [86]. Tyres are known to be very complex constructs, incorporating many small organic molecules. These are released [87] over time when tyre particles are discharged into the environment. A recent study in this regard has shown that the slow release of N-(1,3-dimethylbutyl)-N'-phenyl-p-phenylenediamine) (6PPD), a globally used tire rubber antioxidant, is most likely to blame for the acute mortality in adult coho salmon on the US west coast [88]. Phthalates are also often associated with microplastics as with plastics in general [87] [89]. Also of interest to the evaluation of wastewater treatment plants is that it has been estimated that 1400 - 2800 tons per year of TWP are currently deposited on agricultural areas. Again, this is for Germany, alone [86].

2.2. Studies of MP Retainment by Wastewater Treatment Plants around the World

Municipal wastewater treatment facilities are typically designed based upon a common schematic (**Figure 2**), though each facility will differ slightly in the exact

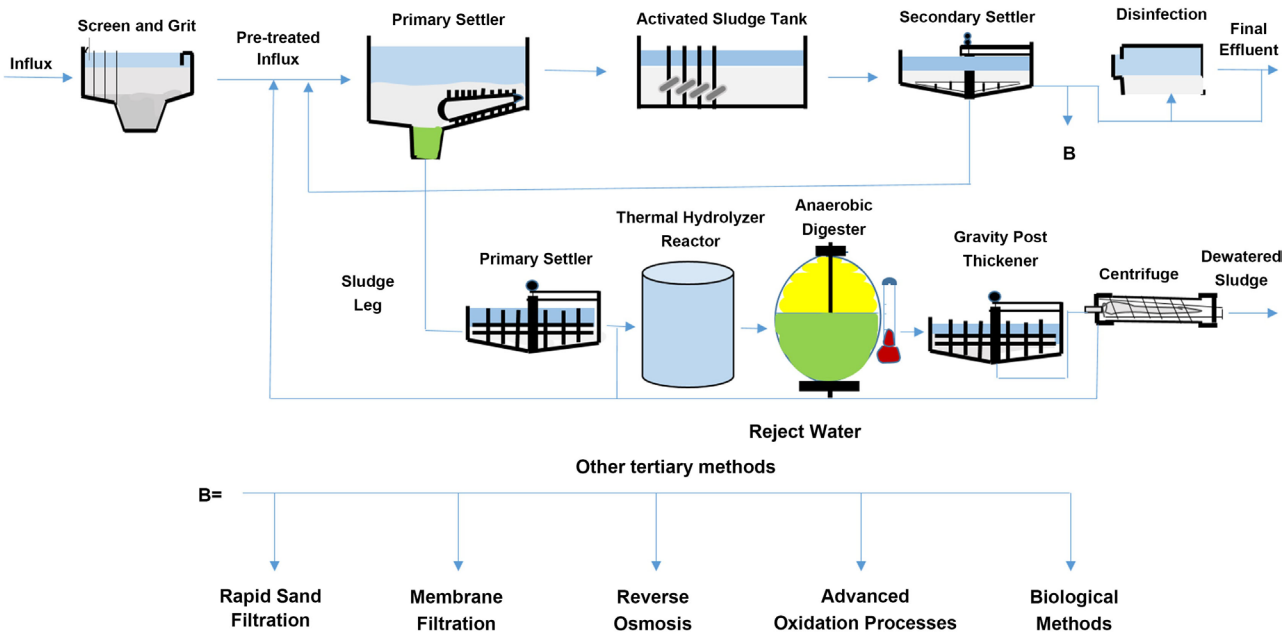


Figure 2. Possible set-up of a wastewater treatment plant.

configuration of this same basic design. Pre-treatment and primary treatment remove large debris items with screen mesh sizes of 6 mm or larger. Screen of mesh size between 3 and 10 mm is called fine screening. This together with the grit and primary clarifier can reduce the MP content of the influent by up to 45%, especially catching MPs with density different from water. Therefore, the pre-treatment and primary treatment play a significant role in the removal of MPs, especially those of 100 μm - 5000 μm in size [16] [90] [91] [92]. Secondary treatment removes suspended and dissolved organic material and nutrients, largely through the incorporation of microorganisms within large aeration tanks. Extracellular polymer substances (EPS) from microorganisms are capable of catching MPs that then become entrapped in the sludge. Flocculates and settling tanks help separate the sewage sludge from the post-processing effluent (hereafter simply "effluent") prior to any disinfection, polishing or advanced (tertiary) treatment. Here, about 50% of MPs are caught, trapped by sinking flocculates or in floating oil and grease [16] [83] [93] [94]. The tertiary stage can include further coagulation or filtration steps. Usually, the resulting effluent is discharged into a nearby waterbody. Thus, most MPs end up the sewage sludge [14], which in many cases undergoes a digestion process.

Over the last 7 years, studies on the retainment and release of MP by wastewater treatment plants have been published for close to 70 locations worldwide (**Figure 3** and **Table 1** [72] [83] [90]-[146]). Again, sampling strategies and analytical methods used have an effect on the quantification of MP fluxes through WWTPs [147] [148]. Thus, Murphy *et al.* collected bulk samples and then used a sieving step (65 μm mesh size) [90], while Carr *et al.* [99] fixed stacked sieves (400 - 445 μm) in a wastewater stream to sample for MPs. Sampling can vary from grab sampling [72] [83] [90] [96] [100] [101] [103] [127],

Table 1. Studies of microplastics (MP) in wastewater treatment plants (WWTPs) around the world.

Ref.	MP conc. in influents	MP conc. in effluent	Lower size limit for fractionation µm	WWTP type	Retention/ Efficiency	Analytical Method	Country
Lares *et al.*, 2018 [95]	57.6 MP/L	1.05 part./L	250	Primary and secondary	98.3%	FTIR and Raman Microscope	Finland (Mikkeli)
Dyachenko *et al.*, 2017 [97]	n.a	0.02 part./L	125	Primary, secondary and tertiary	n.a.	n.a	USA
Mason *et al.*, 2016 [98]	n.a	0.05 part./L	125	17 WWTPs, Tertiary	n.a.	Microscope	USA
Murphy *et al.*, 2016 [90]	15.70 MP/L	0.25 part./L	11	Secondary	98.4%	FTIR	UK
Carr *et al.*, 2016 [99]	1.10×10^9/day (681 million L./day)	0.88 part./m^3	45	Secondary and Tertiary	99.9%	Visual sorting, Microscope FTIR	USA
Ziajahromi *et al.*, 2017 [93]	n.a	0.28 part./L	25	Primary, secondary and tertiary	92% - 99%	ATR-FT-IR	Australia
Michielssen *et al.*, 2016 [100]	367 MP/L	0.5 MP/L	20	Tertiary (AnMBR)	99.4%	Visual sorting, Microscope	USA (Northfield)
Mintenig *et al.*, 2014 and 2017 [94] [101]	n.a	0.1 to 10.1 MP/L	20	12 WWTPs: mostly secondary and tertiary	97%	micro-FT-IR	Germany (Oldenburg)
Talvitie *et al.*, 2015 [102]	610 MP/L	13.5 MP/L (incl. all textile fibers)	20	Primary, secondary and tertiary	97.6%	Visual sorting, Stereo-microscope	Finland Viikinmäki
Talvitie *et al.*, 2017 [114]	6.9	0.005 MP/L	20	4 tertiary WWTPs	99.9%	Visual sorting, Stereo-microscope	Finland
Leslie *et al.*, 2017 [104]	73 MP/L	9 to 91 MP/L	0.7	7 WWTPs	72%	Visual sorting FTIR	Netherlands
Browne *et al.*, 2011 [72]	n.a	1 MP/L	(filtered)	Primary, secondary and tertiary	n.a.	n.a	Australia
Dris *et al.*, 2015 [91]	n.a	14 to 50 MP/L	100	Secondary	83% - 95%	Visual sorting	France
Carr *et al.*, 2016 [99]	1 MP/L	90 MP/L	90 - 300	Primary, secondary and tertiary	95% - 99%		USA
Talvitie and Heinonen 2014 [105]	627 MP/L	70 MP/L	20	n.a	95.6%	μ-Raman spectroscopy Microscope	Russia
Gűndoğdu *et al.*, 2018 [106]	4,825,697/day	7.02 MP/L	n.a.	Secondary	73%	Visual and μ-Raman spectroscopy	Turkey (Seyhan)
Gűndoğdu *et al.*, 2018 [106]	2,040,639/day	4.11 MP/L	n.a.	Secondary	79%	Visual and μ-Raman spectroscopy	Turkey (Yűreğir)

Continued

Estahbanadi and Fahrenfeld 2016 [118]	n.a	0.028 to 0.44 MP/L	250-500	Primary and secondary	n.a.	Visual sorting, Microscope	USA
Kalčiková *et al.*, 2017 [107]	n.a	0.021 MP/L	37 to 95	Primary (Mechanical and Biological)	87%	FT-IR	Slovenia
Simon *et al.*, 2018 [113]	7216 MP/L	54 MP/L	10 to 500	-	98.3%	FPA-FTIR	Denmark
Sutton *et al.*, 2016 [92]	n.a	0.086 MP/L	125	Primary and secondary	n.a.	Dissection microscope and Raman spectroscopy	USA
Michielssen *et al.*, 2016 [100]	133.0 MP/L	5.9 MP/L	20	Primary and secondary	93.8%	Visual sorting, Microscope	USA (Detroit)
Michielssen *et al.*, 2016 [100]	367 MP/L	2.6 MP/L	20	Primary, secondary and tertiary	97.2%	Visual sorting, Microscope	USA (Northfield)
Gies *et al.*, 2018 [119]	31.1 MP/L	0.5 MP/L	64	Primary and secondary	98.3%	Microscopy and FT-IR	Canada
Wisniowska *et al.*, 2018 [120]	$19.4 \cdot 10^3$ to $552.2 \cdot 10^3$ MP/1 m^3	0.028 to 0.96 MP/L	n.a.	n.a.	95% - 99%	n.a	Poland
Yang *et al.*, 2019 [121]	12.03 MP/L	0.59 MP/L	50	Primary and secondary	95%	Microscopy and FT-IR	China (Beijing)
Bayo *et al.*, 2019 [122]	15.70 $MP \cdot L^{-1}$	0.25 MP/L	n.a	Primary	90.3%	Microscopy and FT-IR	Spain (Cartagena)
Long *et al.*, 2019 [123]	1.57 - 13.69 MP/L	0.20 - 1.73 MP/L	28.3	Primary and secondary	97.8%	Visual sorting and micro-Raman spectroscopy	China
Blair *et al.*, 2019 [111]	3 and 10 $MP \cdot L^{-1}$	<1 and 3 MP/L	300	Tertiary	96%	FT-IR	UK
Xu *et al.*, 2019 [124]	196.00 MP/L	9.04 MP/L	1000	Primary and secondary	97.2%	ATR-FTIR	China (Changzhu)
Lv *et al.*, 2019 [125]	0.28 mp/L	0.13 and 0.05 MP/L	25	n.a.	MBR 99.5%,	FTIR	China
Liu *et al.*, 2019 [126]	80 MP/L	28.4 MP/L	100	Primary and secondary	64.4%	Visual inspection and FTIR	China
Wolff *et al.*, 2019 [127]	n.a	59 and 30 MP/L	10	Primary and secondary	n.a	Raman spectroscopy	Germany
Conley *et al.*, 2019 [112]	147, 126, 146 MP/L	3.7, 17.6 and 17.2 MP/L	23	Primary and secondary	97.6%, 85.2%, 85.5%	Visual Observation and stereomicro-scope	USA
Magni *et al.*, 2019 [115]	2.5 MP/L	0.4 MP/L	8	Primary, secondary and tertiary	84%	FTIR	Italy
Ren *et al.*, 2020 [108]	16.0 MP/L	2.9 MP/L	0.08 - 0.55 mm	Primary, secondary and tertiary	81.9%	Visual inspection and Microscope	China (Zhengzhou)

Continued

Ziajahromi *et al.*, 2021 [128]	n.a	22.1×10^6 to 133×10^6 per day	>25 µm	n.a	99.8% - 98.2%	Visual Observation and FT-IR	Australia
Wei *et al.*, 2020 [129]	430 - 2154 MP/m^3.	430 - 2154 MP/m^3	0.0308 - 0.1 mm	RD-WWTFs	84%	Micro-Raman spectroscopic	China (Hangzhou)
Petroody *et al.*, 2020 [130]	12,667 MP/m^3	12667 ± 668, 3514 ± 543 and 423 ± 44.9 MP/m^3	37 - 500 µm	n.a	96.7%	Visual Observation and Micro-Raman spectrometry	Iran (Sari)
Edo et al, 2020 [109]	n.a	12.8 ± 6.3 MP/L	25 to 104 µm	Primary and secondary	>90%	Visual inspection and FT-IR	Spain (Madrid)
Ben-David *et al.*, 2021 [131]	28.28 MP/L	1.97 MP/L	≥20 µm	Primary, Secondary and tertiary	97%	optical microscopy, SEM and µ-Raman microscopy	Israel (Karmiel)
Tagg *et al.*, 2020 [117]	n.a	1.5 MP/L	392 µm	Primary, Secondary and tertiary	76.9%	micro-FT-IR	UK (East Midlands)
Akarsu *et al.*, 2020 [132]	1.1 and 3.6 MP/L	0.9 MP/L	1057 and 1095	Primary, Secondary and tertiary	55% - 97%	FT-IR	Turkey Mersin Bay
Bayo *et al.*, 2020 [110]	15.70 MP/L	13.04 MP/L	400 to 600 µm	Primary	90.3%	Visual inspection and FT-IR	Spain (Cartagena)
Naji *et al.*, 2021 [133]	74 (±11.01, SD) and 67 (±18.35, SD) MP 35/L	70.66 MP/L	> 250 mm	Primary and secondary	n.a	SEM and X-ray EDX	Iran (Bandar Abbas City)
Tang *et al.*, 2021 [136]	23.3 MP/L and 80.5 MP/L	23.3 to 7.9 MP/L	0.636 to 0.803	Primary and secondary	66.1% and 62.7%,	Raman spectroscopy	China (Wuhan City)
Park *et al.*, 2020 [137]	10 to 470 MP/L	10 to 470 MP/L	45 µm to 5 mm	Primary, Secondary and tertiary	98.7% - 99.99%	FT-IR	Korea
Rajala *et al.*, 2020 [138]	n.a	0.1 mg/L, 6.7 mg/L (used)	1 µm and 6.3 µm	Secondary	99.4%	coagulation/ flocculation	Finland
Nguyen *et al.*, 2021 [139]	n.a	n.a	>45 µm	Primary, Secondary and tertiary	80%	Microscopic and FT-IR	Korea (Seoul)
Alvim *et al.*, 2020 [140]	n.a	11.1 MP/L	<1 mm	Primary, Secondary	n.a	Visual/ μ-ATR-FTIR	Spain (Valencia)
Yuan *et al.*, 2020 [141]	n.a	10.30 MP/L, 6.10 MP/L	141 to 665 µm	Primary, Secondary and tertiary	97.67% and 98.46%	n.a	China (Nanjing)
Pittura *et al.*, 2021 [142]	(12,170,000 MP/h) 3.6 MP/L	1,730,000 MP/h	<1 mm	Primary, Secondary	94%	Stereomicroscope and μ FT-IR	Italy
Zou *et al.*, 2020 al. [143]	n.a	1.719 ± 1.035 MP/L	n.a	n.a	n.a	n.a	China (Guangzhou)
Mak *et al.*, 2020 [134]	n.a	10,816 MP/m^3	90 to 189 µm	Primary, Secondary	86.4%		Hong Kong (Victoria Harbor)

Continued

Zhou *et al.*, 2020 [135]	54,100 MFs/L	537.5 MFs/L (MF)	200 - 300 µm	Primary, Secondary and tertiary	85%	ImageJ software microscopy	China (Keqiao industrial park)
Hidayaturrahman *et al.*, 2020 [116]	13,813 MP/L	132 MP/L	<65 µm	Primary, Secondary and tertiary	>98%	Microscope	South Korea (Daegu)
Raju *et al.*, 2020 [144]	11.80 ± 1.10 MP/L	2.76 ± 0.11 MP/L	>1.5 µm to >1 mm	Secondary	76.61%	Visual sorting and ATR-FTIR	Australia (New south wales Hunter Region)
Ferreira *et al.*, 2020 [145]	n.a	0.24 ± 0.07 MP/m^3 (Laucala Bay) and 0.09 ± 0.02 MP/m^3 (Suva Harbour)	125 µm	79 WWTPs'	N.a	ATR-FTIR	Fiji (Suva)
Schmidt *et al.*, 2020 [146]	n.a	4×10^0 and 4.5×10^5 MP/m^3	10 to 5000 µm	Secondary	n.a		Germany

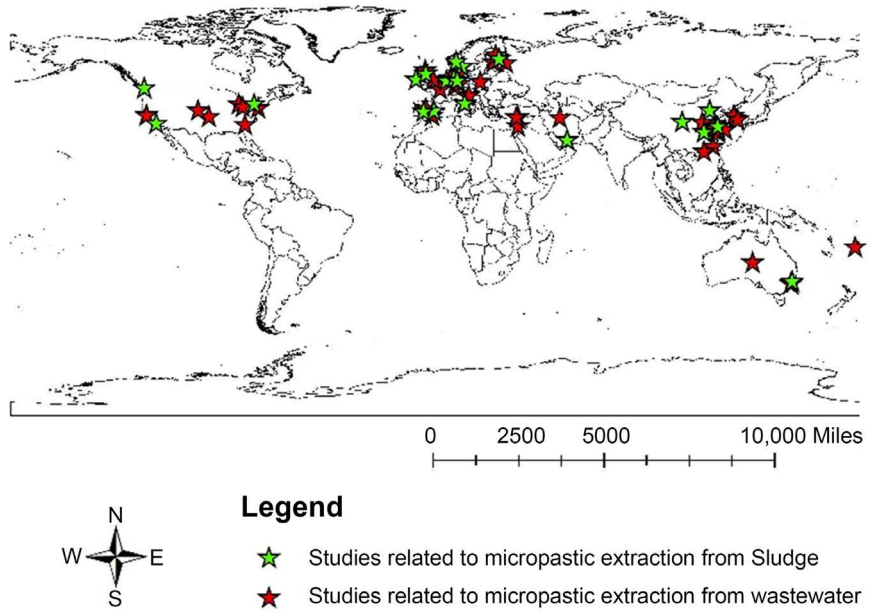


Figure 3. World map showing the locations of the studies carried out on MP in WWTPs (**Table 1** and **Table 2**).

the use of extraction pumps [83] [92] [97] [98] [99], the use of neuston/plankton nets [99] [118] to analysing composite samples [83] [91] [97] [106] [112]. From wastewater, microplastics can be separated by visual selection, but can also be detected by spectroscopic imaging such as by focal plane array (FPA)-based micro-Fourier transform infrared (micro-FTIR) imaging [94] [147] [148] [149] after filtration onto a membrane filter. Micro- and nanoplastics, while having been studied by the use of scanning electron microscopy, X-ray photoelectron spectroscopy (SEM-XPS), can also be identified by micro-Raman imaging techniques [56] [150] [151]. Wastewater is a biogenic organic matter-rich matrix, which

makes MP detection, isolation and quantification difficult [117]. There are but few studies that address the MP flux through a WWTP over a long period of time [91] [111]. Additionally, most studies have solely looked at MP concentrations in the WWTP influent and final effluent, but only few studies have investigated the MP concentrations at the different treatment stages [90] [117]. Nanoplastics (NPs) have also not been assessed thoroughly.

Figure 2, possible set-up of a wastewater treatment plant (partially adopted from [142]).

Nevertheless, it is clear that WWTPs cannot completely retain MPs, and that the effluent from WWTPs contributes to the MP load in receiving waters [83] [90] [93] [95] [99] [101] [102] [113] [114] [152]. In many cases, there is a significantly higher concentration of MPs downstream of a WWTP as compared to upstream [89] [153]. MPs ranging in size from 20 to 1000 μm have been found in WWTP effluents [16], in amounts of between 9.3×10^5 to 4.0×10^9 particles per day [16]. Many WWTPs also receive stormwater run-off, which can be discharged in cases of severe weather after only basic physicochemical treatment or with no treatment at all, and in such cases larger amounts of MPs can be released from WWTPs into the environment [154]. Nevertheless, there is still a high uncertainty in regard to overall MP emissions from WWTPs [44] [47] [96] [113] [153]. It has been noted, however, that there is a greater chance of smaller microplastics to break through WWTPs [101].

An important variable in the MP emission to the environment is the nature of the tertiary stage of the WWTP that can include advanced separation methods such as ultrafiltration and reverse osmosis. Rapid sand filters (RSF) [110] can remove up to 97% of microplastics from the effluent of a secondary stage of a WWTP. As part of the last stage of a tertiary WWTP, they are deemed to be economical and can be operated at a reasonable scale [114]. However, microplastics between 0.02 μm and 2 μm in size have been reported to travel through sand [155]. In this regard, spherical microbeads have the highest mobility through sandfilters as compared to fibers, microplastic fragments, microtyres and plastic films, and it has been shown that in certain locations these microbeads constitute the largest portion of microplastics released with the effluent of tertiary WWTPs [116]. Nevertheless, also small fibers have been reported to travel through RSFs, where in one study the removal efficiency for small fibers by RSF was only 53.8%. Also, sand filtration has been noted to fragment microplastics into even smaller particles [4] [114]. Ultrafiltration (UF) is often used for the removal of particulates and macromolecules from raw water to produce potable water, and is more and more replacing secondary treatment methods such as coagulation, flocculation, and sedimentation and more classical tertiary filtration such as sand filtration. While UF, typically having polymeric or ceramic membranes with a pore size between 1 - 100 nm, is laid out to retain large organic molecules such as proteins as well as bacteria, protozoa, and viruses, UF is not specifically designed to retain micro- or nanoplastics [156]. Often-

times, pre-treatments are necessary for the successful use of UF as UF membranes can be fouled easily. To that effect, a coagulation step as pretreatment with iron-based coagulants has been advocated, especially in combination with an addition of polyacrylamide (PAM), which increased the removal efficiency of small-sized polyethene particles (d < 0.5 mm) significantly from 13% to 91% [157] [158].

Membrane bioreactors, in which the action of biological catalysts (bacteria, enzymes), is coupled to a separation process have been seen to remove 99.9% of MPs (from 6.9 to 0.005 $MP \cdot L^{-1}$), while disc filters have a removal efficiency of 40% - 98.5% (from 0.5 - 2.0 to 0.03 - 0.3 $MP \cdot L^{-1}$) [114]. However, fouling also of MBRs through MPs has been noted, where PVC MPs artificially added at a concentration of 10 MPs/L inhibited the removal performance of the MBR towards polluting organic matter and ammonia [159]. Also, the process is quite expensive, both in investment (membrane) as well as in running costs (high energy consumption) [3] [4].

S. Fortin [151] has compared the efficiency of two other advanced tertiary treatment methods, namely of reverse osmosis nanofiltration and of activated carbon filtration systems, where in a pilot program was fed secondary effluent water with return activated sludge added to it to increase the turbidity. This water was branched and fed into the two different advanced treatment methods. The carbon train system was composed of a flocculation/sedimentation step followed by an ozonation step and treatment with bacterial activated carbon and granular activated carbon columns, before undergoing UVD/UVA disinfection. The other train consisted of a reverse osmosis nano-filtration step followed by UVD/UVA disinfection [151]. It was found that the reverse osmosis nano-filtration produced effluent that still showed microplastics with a relatively narrow size distribution of 1 μm and 10 μm, though over 90% of the particles were smaller than 10 μm, while the filtration on activated carbon gave effluent with much larger particles remaining, with 54% of the total particles larger than 10 μm. Many of the particles were not plastic but inorganic particles, some stemming from a prior flocculation step. Overall, the study found more small microparticles than previous studies have had, most likely due to a better quantification of the microparticles by Raman microspectroscopy [151]. All of the membrane processes above are of interest and continue to be developed. In order to be commercially viable, especially for larger WWTPs, the methods need to be cost-effective and have a high through-put.

2.3. Microplastics in Sludge

As much of the retained MP material (about 90% on the average) ends up in the sludge of WWTPs [14], high concentrations of MPs in WWTP sludge samples have been described in many studies [47] [76] [96] [98] [160] [161] [162]. Mahon *et al.* [161] found microplastic in concentrations of 4.20 to 15.4×10^3 particles kg^{-1} dry sludge. Lassen *et al.* [47] reported that sludge samples from Ger-

man WWTPs contain 1.00 to 24.0 × 10^3 MP particles (10 mm) per kg of dry sludge. A comparison of MP concentrations in sludge collected at different WWTPs from around the world is given in Table 2. Sludge from 8 WWTPs in Norway (Oslo, Stavanger, Tromsø, Federickstand, among others), serving altogether 1,500,000 inhabitants and producing about 100,000 tons of sludge, revealed an overall average plastic abundance of 6077 particles kg^{-1} (dw), of which 37.6% were beads, 31.8% fragments, and 28.9% fibers. The most common constituents were found to be polyethylene (PE, 30.5%), polyethylene terephthalate (PET, 26.7%) and polypropylene (PP, 20.3%) [163]. Sujathan *et al.* reported a very high microplastic particle count of 4.95 × 10^5 per kg (dw) in return activated sludge [164]. In the study, microplastics as small as 0.48 µm could be identified, which may mean that many microplastic particles in WWTP sludge are indeed small and escape detection [163].

Table 2. Studies on microplastics (MP) in accumulated sludge from wastewater treatment plants (WWTPs).

Type of WWTP's	Country	MP size range	MP concentration (particles kg^{-1} d.w.[a] and w.w[b])	Analytical Method	References
(7WWTPs,)	Ireland (7WWTPs)	250 µm - 4 mm	4196 - 15,385	Optical and FT-IR	Mahon *et al.*, 2017 [161]
(7WWTPs,)	Netherlands (Amsterdam)	0.7 µm - 5 mm	370 - 950	FT-IR	Leslie *et al.*, [104]
n.a	Sweden (Lysekil)	300 µm - 5 mm	16.7[a] × 10^4	FT-IR	Magnusson & Norén, 2014 [96]
n.a	(USA) Ithaca, New York	No Data	About 1000 - 4000	Filtration techniques	Zubris and Richards, 2017 [165]
Primary, Secondary and Tertiary	USA (Los Angeles County)	<5 mm	5000	Visual sorting, Microscope FT-IR	Carr *et al.*, 2016 [99]
	China (28WWTPs,)	37 µm - 5 mm	1565 - 56,386[a]		Li *et al.*, 2018 [166]
12 WWTPs: mostly secondary and tertiary	Germany (Oldenburg	<5 mm	1000 to 24,000[a]	micro-FT-IR	Mintening *et al.*, 2014, 2017 [94] [101]
	Germany (Seelze)	20 to 100 µm	495,000[a]	confocal Raman microscopy	Sujathan *et al.*, 2017 [164]
Secondary	UK (Glasgow)	1.34 - 1.62 mm	About 2000[b]	FT-IR	Murphy *et al.*, 2016 [90]
Primary and secondary	Finland (Mikkeli)	<1mm	8.2 - 301.4[b]	FT-IR and Raman Microscope	Lares *et al.*, 2018 [95]
Primary and secondary	Canada (Vancouver)	64 µm	4400[b]	Microscopy and FT-IR	Gies *et al.*, 2018 [119]
	Norway (10 WWTPs)	54 µm to 5mm	6 077[a]	µ-FT-IR	Lusher *et al.*, 2018 [163]
n.a	China (Jiangsu)	25 to >500 µm	1.6 and 0.7[b]	FT-IR	Lv *et al.*, 2019 [125]
Primary and secondary	China (Wuhan)	100 to 800 µm	24,030[a]	Visual inspection and FT-IR	Liu *et al.*, 2019 [126]

Continued

Primary and secondary	China (Beijing)	681.46 µm	95.16	Microscopy and FT-IR	Yang *et al.*, 2019 [121]
Primary, secondary and tertiary	Italy (Italy)	0.5 - 0.1 mm	113,000[a]	FT-IR	Magni *et al.*, 2019 [115]
Secondary	Australia (New South Wales, Hunter Region)	>1.5 µm to >1 mm	7.91 ± 0.44 MP/L (in activated sludge	Visual sorting and ATR-FT-IR	Raju *et al.*, 2020 [144]
Primary and secondary	Italy	<1 mm	1.67 MP/g TS in the Sludge, 5.3 MP/g TS in the AerWAS and 4.74 MP/g TS dewatered sludge	Stereomicroscope and µ FT-IR	Pittura *et al.*, 2021 [142]
Primary and secondary	Spain (Valencia)	<1 mm	280 MP/Lof activated sludge	μ-ATR-FT-IR/ATR-FT-IR	Alvim *et al.*, 2020 [140]
Primary and secondary	Iran (Bandar Abbas City)	>250 µm	328.50 (±56.42, SD) MP/50 g dw and 278.4 (±26.87, SD) MP/50 g dw	SEM and X-ray EDX	Naji *et al.*, 2021 [133]
Primary and secondary	Spain	25 to 104 µm	183 ± 84 MP/g (mixed sludge), 165 ± 37 MP/g (dried sludge)	Visual inspection and FT-IR	Edo et al, 2020 [109]

The site map of **Figure 3** shows the locations of the studies on a world map. [a]DM, dry matter. [b]The unit is particles kg^{-1} wet weight and the dry weights of the wet sewage sludge samples were all below 1%.

Oftentimes, sludge after thickening, is treated in a thermal hydrolysis reactor and then goes into an anaerobic digester. Afterwards, the sludge is dried. It must be noted that while sewage sludge is added to soil, smaller amounts of sludge can also be incinerated. The latter might also be added to a landfill. Also, an appreciable quantity [90] [114] [167] of microplastics, especially those of low density such as LDPE and PP, can be separated off in the primary steps of the wastewater treatment, in grit traps and during grease skimming. These separated wastes are usually put into landfills or are incinerated [99] [163].

The application of sludge to agricultural soils and municipal green areas as well as its use by soil producers raises the concentration of microplastics in soils significantly [168] [169] [170]. In addition, microplastics are entering soil via plastic mulching, irrigation with grey water, and through run-offs. Also, air movement contributes to the dissemination of microplastics in farming areas [91] [171]. The occurrence and fate of microplastics in soil are less studied [172] [173] [174] [175] [176] than of microplastics in the marine environment. It has been calculated that in Norway alone, 500 billion (5×10^{11}) pieces of microplastic find their way into the soil via sewage sludge applied to agricultural soils [163]. This compares with an estimated 1.56×10^{14} plastic particles per year entering Chinese soil [166], and 300,000 and 430,000 tons/year of plastic distributed over European and North American agricultural land [73] [172]. While plastics structurally weather under the influence of humidity, temperature, UV-radiation and wind [173], they remain chemically intact in the soil for long periods of time [177], with slow chemical oxidation and UV driven bond scis-

sion being the two main degradation mechanisms reported [178].

As stated above, in WWTPs, sludge undergoes prior steps before being released such as thickening and final dewatering, which can involve a centrifugation step. Oftentimes, digestion steps are incorporated in the treatment. These can be anaerobic digestion or aerobic digestion/composting. Here, L. Pittura *et al.* have shown that 86% of MPs (which included 100% of the particles and 87% of the fibers) can be retained with a pilot scale upflow granular anaerobic sludge blanket (UASB) in combination with an anaerobic membrane bioreactor (AnMBr) [142]. However, MPs can have potential adverse effects on anaerobic processes, which normally yield biogas as a useful side-product. Thus, L. Pittura *et al.* [142] have shown that in their case the presence 50 polypropylene MPs per g of solid led to a decrease of the methanogenic activity by 58%. This "poisoning" of the anaerobic microorganisms may be due to leachates from the MPs, which contain small organic molecules but also heavy metals. The organic molecules can stem from plastic additives to MPs such as added plasticizers, antioxidants or UV absorbents or they themselves could have been adsorbed to the MPs at some point in their history and are now released. Also, other research groups have commented on this inhibition of methanogenic activity by MPs (and NPs) in lab scale anaerobic digestion processes of wastewater or simulated wastewater and in lab scale UASB processes [179] [180] [181] [182] [183]. PE-MPs [180], PEST-MPs [179], PET-MPs [182], PS-MPs [183], and PVC-MPs [181] all showed this inhibition.

It has been shown that a fraction of up to 20% the microplastic can be recycled back into the reject water during the sludge dewatering step [114] and thus will cycle through the system on more time. The fate of microplastics in thermal treatment processes such as in the Cambi process (*i.e.*, at 160˚C) or incineration processes has not been studied in detail. Mahon *et al.* have described a potential shredding of MPs in lime stabilization processes, leading to smaller sized MPs. MPs having gone through the sludge thermal drying process showed signs of melting and blistering [161]. We have noted that some plastic microbeads contained in personal care products undergo fragmentation when cycled on silica gel from room temperature to 100˚C and back to rt [17]. Synthetic fibers, which are more ubiquitous, are usually more indestructible. Rom *et al.* have looked at polylactide microfibers and have found that they were not biodegraded by treatment with activated sludge under mesophilic (36˚C) or thermophilic (56˚C) conditions, even after 4 weeks [184].

3. Laboratory-Scale Development of New Methods for the Removal of Micro- and Nanoplastics from Aqueous Systems Suitable for Implementation in Wastewater Treatment

There are a number of new methods under development for the removal of micro- and nanoparticles that can be deemed suitable for implementation at a future time. Mainly, they are based on adsorption [185] [186] [187], coagulation

techniques [157] [158] [188]-[194] and membrane filtration [156]. This becomes important also in regard to eliminating microplastics (and potentially nanoplastics) from drinking water derived from drinking water treatment plants (DWTPs) [195] [197] [198], where MP have been found not only in the ground water used by DWTPs, but also in tap water [199] and bottled mineral water [200] [201] [202]. Kosuth *et al.* have found MP in 81% of 156 drinking water samples collected from 14 countries [203].

First, we turn to adsorption or other types of retention of MPs on different materials leading to different filtrations. Z. Wang *et al.* have looked at replacing sand filters with biochar filters [187]. The filters were either comprised of corn straw biochar produced at 500 ˚C or of commercially available hardwood biochar. Leachate column tests were performed with spherical 10 μm polystyrene beads at a concentration of 1.6 × 108 suspended beads/L distilled water. The removal efficiency of such filters was found to be more than 95% [187]. C. Sun *et al.* have developed artificial sponges made from chitin and graphene oxide cross-linked with the help of epichlorohydrin, which were found to take up polystyrene, carboxylate modified polystyrene and amine-modified polystyrene microbeads of 1 μm size with maximum adsorption capacities of 5.99. 7.53 and 8.46 mg/g, respectively [186]. The sponges could be reused, although the regeneration of the sponges involves a complex process of rinsing with larger amounts of ethanol, cooling them down to −80˚C for 4h and freeze-drying [186].

Looking at chemical coagulation-flocculation processes, G. Zhou *et al.* [190] used polyaluminium chloride (PAC) and ferric chloride ($FeCl_3$) as coagulants for PE and PS microplastics, finding 77.8% (PS) and 29.7% (PE) removal efficiencies with PAC and 64% (PS) and 17.4% (PE removal efficiencies with $FeCl_3$. Interestingly, with these systems the smaller sized PE microplastics could be removed more efficiently, while for PS it was just the reverse [190]. N. Shahi *et al.* have turned to using alum coagulant alone or in combination with polyamine-coated (PC) sand. Here, a combination of PC sand (500 $mg \cdot L^{-1}$) with alum (20 $mg \cdot L^{-1}$) gave an MP removal of 97% for PE-MPs, where it was noted that size, shape and surface morphology plays a significant role in the coagulation and flocculation processes [192]. K. Rajala *et al.* [138] have studied the effectiveness of PAC and $FeCl_3$ in MP removal by artificially adding PS microspheres, 1 and 6.3 μm in size, respectively, to a WW matrix stemming from the effluent of a secondary WWTP in Finland. M. Lapointe *et al.* [194] looked at the difference in coagulation and flocculation behaviour of pristine and weathered microplastics, using polyethene microspheres (15 μm and 140 μm in size), polystyrene microspheres (140 μm in size) and polyester fibers (12 - 16 μm wide, 105 - 1325 μm long). Alum (0.45 - 3.64 mg Al/L) and polyacrylamide (PAM, 0.05 - 0.30 mg/L) were used as coagulant/flocculant. It was seen that polyester fibers were removed more efficiently (99%) than the microbeads (82% - 84%). Again, smaller sized microbeads were removed more facilely than larger ones. Weathered (aged) PE microbeads were more easily removed than pristine beads. The authors rationalized this behaviour

with the slow oxidation of the PE bead surface which allows such functionalities as –OH, -CO_2H, and –C=C- to form. With a quartz crystal microbalance, the deposition of coagulant and flocculant was investigated on a plastic surface. Here, it was seen that cationic PAM showed the highest deposition rate. Z. Chen *et al.* [204] have demonstrated an interesting flocculation method for the removal of NPs with different salt-based flocculants, which not only included aluminium but also calcium. Calcium ions show an excellent sedimentation performance for impurities at high pH. It has been described that flocculation occurring between composite metal calcium-aluminium (Ca/Al) ions and nanoplastics showed the best performance, again at high pH [204]. As many synthetic polymers used as flocculants exhibit some toxicity, especially due to small concentrations of monomers in the materials, bio-based polymeric flocculants have been suggested [205]. Here, the problem is their poor water solubility and their low charge density [206] [207]. Nevertheless, S. Magalhães *et al.* have performed initial experiments using a cationic hydrophobically modified cellulose derivative as a bio-flocculant for the flocculation PET microplastics [206].

Electrocoagulation uses sacrificial iron or aluminium electrodes and usually forms iron- and aluminium hydroxides as coagulant material. As with the addition of chemical coagulants, these destabilize the surface charges on the surface of the suspended solids. The suspension is broken up, and the particles, approaching each other more closely, interact by van der Waals forces to form a conglomerate of particles. The hydrogen liberated during the electrolysis process lifts these larger conglomerates together with the blanketing coagulant to the water surface. In their set-up, W. Perren *et al.* [189] used seven metal electrodes, one as cathode, one as working anode and the five remaining as sacrificial anodes. The added spherical polyethene beads of 300 - 355 μm size were found to aggregate on the positive electrode faces. The microplastic particles underwent simultaneous charge neutralisation and flocculation to create a stable floc on the water's surface. A minimum of 2 g/L NaCl salt concentration was employed. The microbead removal efficiency increased with increasing salt concentration [189].

It was seen that mesoparticles (*i.e.*, 20 μm - 0.5 μm, falling within the size of many microplastics) can play a role in irreversible membrane fouling and that a prior coagulation process is of utmost importance [208]. An alternative membrane separation technology is that using dynamic membranes (DMs). DMs operate with a layer produced on a supporting membrane by particles in the influent. So, these particles in the influent form a filtration layer that can be supported by a larger pore-sized mesh or by low-cost porous materials. DMs have been run successfully with particles that are of a similar size to microplastics [209]. Also membranes made from electrospun lignin-zeolite composite nanofibers, with 1 wt% zeolite, were found to retain polystyrene microbeads of size > 1 μm (R = 94.7%) after five cycles of filtration [210]. Zirconium based metal organic frameworks (MOFs) in form of foams have been utilized as filters. They are recyclable and operate at a MP removal efficiency of up to 95.5%. A large

scale filtration with these materials as filters was carried out [211].

The following shows some further intriguing approaches for the removal of MPs from wastewater, but they may be less suitable for a high through-put WWTPs and more suitable for batch treatments. One such approach centres on centres on the adsorption of nanoparticles on MPs that induce a property change in the MPs such as a change in density or giving the composite magnetic properties. Also, the use of hydrophobic magnetic nanoparticles has been forwarded. One such method uses magnetic polyoxometalate supported ionic liquid phases (magPOM-SILPs), in which magnetic iron oxide/silica core-shell precursor particles, with an average size of 16 nm, are treated with a polyoxometalate ionic liquid $(n\text{-}C_7H_{15})_4N^+)_8$ $[\alpha\text{-}SiW_{11}O_{39}]_8$-in acetone, giving a viscous coating to the particles. This makes the magPOM-SILPs attach themselves to the MPs. The mag-POM-SILPs with the attached MPs are then removed from the water with a permanent magnet [212]. The use of a coating of magnetic iron nanoparticles by reaction of the nanoparticles with hexadecyltrimethoxysilane (HDTMS) has also been reported to be suitable for the adsorption of MPs. Similar to the above, the nanoparticles with the attached MPS are removed from the water by a permanent magnet [213]. Lastly, an interesting approach to the removal of microparticles, incl. microplastics, in water is through their interaction with magnetic, self-moving microswimmers of the type $Au@Ni@TiO_2$, linked together in form of a chain. The microswimmers moving in a magnetic field have been used to clear water of the river Warnow in Germany from microparticles by "shoveling" or "pushing" interactions [214]. The microswimmers still lack selectivity to identify microplastics among other microparticles [214].

4. Conclusion

The emission of microplastics into the environment will be an ongoing problem for the foreseeable future. The phase-out of plastic microbeads in personal care products and of single use plastic in the food industry in certain regions is laudable, but will only have a limited impact on the continuous generation of polymeric microparticles that especially include synthetic fibers, microtires and secondary microplastics, incl. from plastic packaging. Therefore, WWTPs will play an ever-increasing role in frustrating the dispersal of these materials, specifically in the marine ecosystem. In this context, further research in four broader areas continues to be needed: 1) the creation of new techniques to better retain microparticles; 2) a more complete understanding of possible physiological effects of the materials in humans and animals; 3) the development of a more complete inventory of microplastic emissions; 4) a better knowledge of the fate and lifetime of microplastics in aquatic ecosystems and soil. This should go hand-in-hand with the development of new methods for the analysis of micro- and nanoplastics, also and especially in drinking water and food.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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