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Chapter

The Effect of Wastewater Treatment Methods on the Retainment of Plastic Microparticles

Rana Zeeshan Habib, Ruwaya al Kindi and Thies Thiemann

Abstract

Microplastics as plastic pieces of ≤5 mm in size, are found in most ecosystems, both terrestrial and aquatic. Many of the microplastics find their way into the environment through the wastewater. For this reason, a knowledge of the microplastic retainment performance of wastewater treatment plants of various design is important. In this regard, several wastewater treatment processes have been studied, including new methods that are still at the development stage. This manuscript reviews the literature on such wastewater treatment methods and their ability to retain microplastics.

Keywords: microplastics, wastewater treatment plants, wastewater treatment method, plastic retainment

1. Introduction

A number of reviews have appeared on the topic of wastewater treatment and microplastic retainment [1–17]. Over the last 10 years, the understanding of microplastics and their impact on the environment has developed as have the analytical techniques to identify and quantify microplastics. In this respect, the focus has shifted to even smaller plastic particles dubbed "nano plastics". The identified sources of micro(nano)plastics have increased to include secondary plastics created by such mundane processes as opening a package [18] or making tea using plastic tea bags [19]. On the other hand, the importance of the individual sources of primary microplastics has shifted, with the ban of plastic microbeads in cosmetics coming into effect in many regions [20–25], changing the attention more to microtyres [26, 27], synthetic fibers [28, 29] and to secondary micro- and nano plastics [30]. There will be a shift of the sources of secondary microplastics as the ban in certain regions of plastic bags [31–33] and single use plastics [34, 35] comes into effect, as both are potential materials for microplastics due to subsequent degradative fragmentation processes. Plastics already existing in the environment degrade very slowly [36]. Furthermore, the examination of food articles and drinking water [37–40] for micro- and nano plastics has increased, as micro- and nano plastics have been found in foods and drink as diverse as table salt [41], soft drinks [42], beer [43, 44], and meat [45].

Microplastics (MPs) are defined as plastic particles of ≤5 mm in size [46–48]. For smaller particles, of size ≤1 μm, the term nanoparticles (NPs) is often used [46, 49]. Some authors define NPs as particles of up to 100 nm in size [50]. Plastic particles include polymeric films and synthetic fibers. Plastic microparticles come from different sources. They can be degraded and fragmented materials from tires (tires and road-wear) [51], clothing [52, 53], plastic bags [30] and packaging [18], where larger pieces of plastic are exposed to wear or weathering [54]. These are secondary MP. Primary MP are materials that are produced industrially at this small size. These include solid micropellets in cosmetic formulations, such as in facial cleaners and body scrubs [20, 55], microspherules in toothpastes (2–5 μm in size) [56], microparticles in washing powder/detergents [57, 58] and scrubbers used for air-blasting surfaces to remove paints and rust [59, 60] in paints and coatings themselves [58], and in drilling fluids in oil and gas exploration [1]. Drug delivery systems have used plastic micro−/nanoparticles, also – these are often biodegradable materials [61]. 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 [62–65] and by the European Community [66]. Often, sediments of water bodies [67], especially oceans [68, 69], and terrestrial soil are some of the places where MPs may end up when released into the environment. There are a number of ways that MPs can enter the world's oceans that include direct run-offs into the oceans or into rivers that lead to oceans. Additionally, atmospheric transfer of MPs [70], which has been largely neglected until relatively recently, has been found to contribute to the accumulation of MPs in rivers, lakes [71] and oceans [72]. Terrestrial acquisition of MPs in soils can also happen in a number of ways that again includes atmospheric transport, but can also occur through fertilizer and even irrigation water [73]. Plastic mulching also contributes [74]. In both the dispersal of MPs to the aquatic and the terrestrial environment wastewater treatment plants (WWTPs) play a major role. On the one hand, WWTPs play a major part in retaining MPs from the sewage water, on the other hand, MPs can enter the soil through the application of sewage sludge [75, 76]. In the treatment of wastewater, WWTPs themselves can become point sources of MPs [77–80], releasing MPs into the receiving water. Thus, many examples have been found where the concentration of MPs downriver of a WWTP was higher than upriver. As the volume of wastewater is bound to increase over the years with an increase of population, new methods of wastewater treatment are being developed that help retain MPs better. This comes against the background of studies that assess the retaining capabilities of different treatment methods in existing WWTPs. Both are topic of the current review.

2. Studies of retaining microplastics in existing wastewater treatment plants

2.1 Standard functional units of WWTPs

WWTPs are of different design and of different sizes (**Figure 1**). In general, most WWTPs start off with passing the wastewater through bar screens (screening) to remove large solids and an oil and grit removal tank as pre-treatment, before the water is left to settle in a clarifier, where solids are removed in form of sludge sunk to the bottom and in form of scum on the water surface as primary wastewater treatment. This primary treatment is followed by activated sludge treatment as secondary wastewater treatment. The treatment uses flocs of microorganisms that decrease the BOD (biological oxygen demand) of the water due to a decrease of
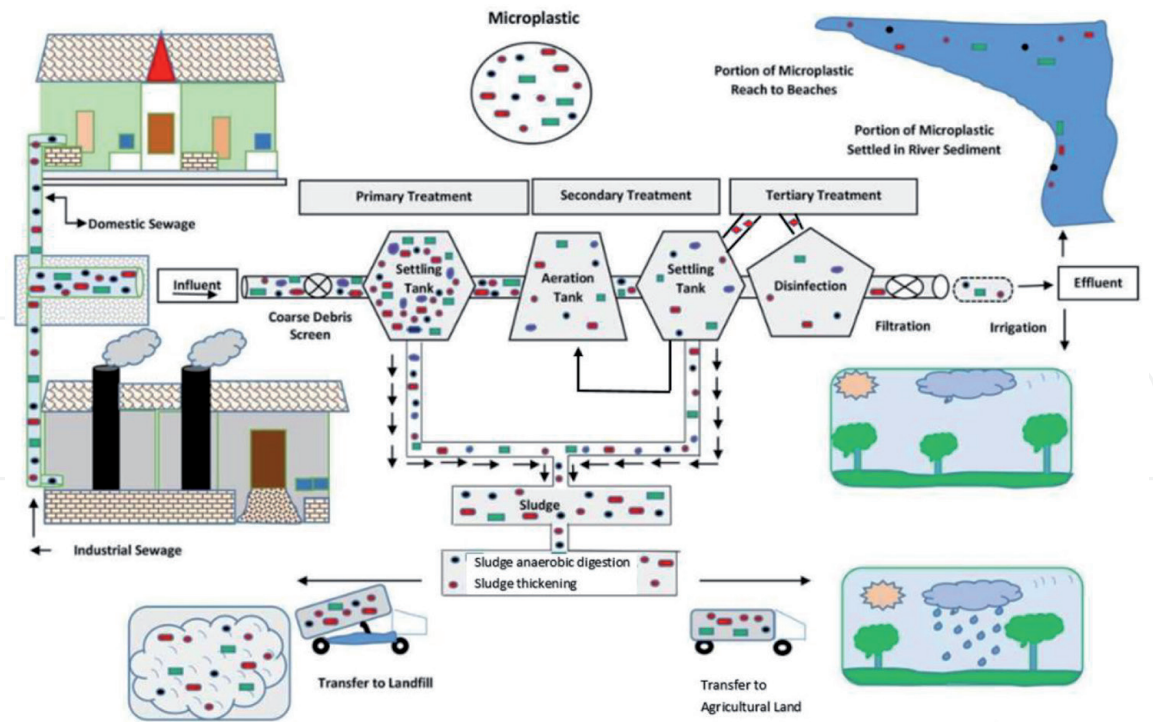
Figure 1.
Schematic flow of microplastic (MP) particles through a wastewater treatment plant (WWTP).

organic components through conversion to CO2 along with a decrease in nitrogen content by conversion of ammonia bound nitrogen to elemental nitrogen through a nitrification process and a subsequent denitrification process, involving both heterotrophic as well as autotrophic microorganisms and both aerobic and anoxic reaction zones. The activated sludge is separated from the treated water in a subsequent clarifier where some of the activated sludge is recycled. Activated sludge treatment can be conducted in a number of different ways. It is also possible to operate this process in a sequential batch reactor (SBR), where bioreactor and clarifier are run in a timed sequence within one vessel. Tertiary treatment methods can be added. They can include rapid sand filtration, membrane filtration, reverse osmosis, advanced oxidation processes and further biological methods [81]. Finally the treated water is disinfected, either by UV irradiation, addition of ozone (O3), of chlorine (Cl2) to produce hypochlorous acid (HOCl) or of chlorine dioxide (ClO2) [82], before it is discharged, mostly into a natural receiving water body such as a lake, river or the ocean. Especially in countries with extreme water scarcity or in large metropolises the water can also be recycled directly for consumption/use [83].

2.2 Early MP retainment studies in WWTPs

More than 70 WWTPs have been studied to date as to their MP retaining capability [1]. Some of these studies were devoted to the assessment of microplastic accumulation in the sludge of the WWTPs [8]. While early studies were carried out in North America [80, 84–90], Europe [78, 90–106], and Australia [79, 107], a larger number of recent studies emanate from East Asia [108–123]. It must be noted that some WWTPs were handling sewerage and storm run-off separately, others were not. In some of the cases where sewerage and storm run-off were handled together, specific note was made of black fragments in the influent that derived from tire abrasion in form of microtires [124]. **Table 1** gives an overview of many of the published studies on MP retention efficiency of WWTPs around the world [77–80, 84–88, 90, 92–94, 97–100, 103–144].

Ref.	MP conc. in influents	MP conc. in effluent	WWTP type	Overall retention/ efficiency	Country
Conley et al., 2019 [77]	147, 126, 146 MP/L	3.7, 17.6 and 17.2 MP/L	Primary and secondary	97.6, 85.2, 85.5%	USA
Talvitie et al., 2015 [78]	610 MP/L	13.5 MP/L (incl. all textile fibers)	Primary, secondary and tertiary	97.6%	Finland Viikinmäki
Browne et al., 2011 [79]	n.a	90 MP/L	Primary, secondary and tertiary	n.a.	Australia
Carr et al., 2016 [80]	1 MP/L	1 MP/L	Primary, secondary and tertiary	95–99%	USA
Carr et al., 2016 [80]	1.10×10^9 MP/day (681 million L./day)	0.88 MP/m^3	Secondary and Tertiary	99.9%	USA
Mason et al., 2016 [84]	n.a	0.05 MP/L	17 WWTPs, Tertiary	n.a.	USA
Michielssen et al., 2016 [85]	367 MP/L	0.5 MP/L	Tertiary (AnMBR)	99.4%	USA (Northfield)
Michielssen et al., 2016 [85]	133.0 MP/L	5.9 MP/L	Primary and secondary	93.8%	USA (Detroit)
Michielssen et al., 2016 [85]	367 MP/L	2.6 MP/L	Primary, secondary and tertiary	97.2%	USA (Northfield)
Estahbanadi and Fahrenfeld 2016 [86]	n.a	0.028 to 0.44 MP/L	Primary and secondary	n.a.	USA
Sutton et al., 2016 [87]	n.a	0.086 MP/L	Primary and secondary	n.a.	USA
Dyachenko et al., 2017 [88]	n.a	0.02 MP/L	Primary, secondary and tertiary	n.a.	USA
Gies et al., 2018 [90]	31.1 MP/L	0.5 MP/L	Primary and secondary	98.3%	Canada
Lares et al., 2018 [92]	57.6 MP/L	1.05 MP/L	Primary and secondary	98.3%	Finland (Mikkeli)
Mintenig et al., 2014 and 2017 [93, 94]	n.a	0.1 to 10.1 MP/L	12 WWTPs: mostly secondary and tertiary	97%	Germany (Oldenburg)
Talvitie and Heinonen 2014 [97]	627 MP/L in addition to 3160 black particles/L	23 MP/L In addition to 125 black particles/L	Secondary	96.1%	Russia (St. Petersburg)

Ref.	MP conc. in influents	MP conc. in effluent	WWTP type	Overall retention/ efficiency	Country
Dris et al., 2015 [98]	n.a	14 to 50 MP/L	Secondary	83–95%	France
Magnusson and Norén 2014 [99]	1.5×10^4 MP/m^3 3.2×10^6 MP/h	1.77×10^3 MP/h	Secondary	>99%	Lysekil (Sweden)
Murphy et al., 2016 [100]	15.70 MP/L	0.25 MP/L	Secondary	98.4%	UK
Leslie et al., 2017 [103]	73 MP/L	9 to 91 MP/L	7 WWTPs	72%	Netherlands
Kalčiková et al., 2017 [104]	n.a	0.021 MP/L	Primary (Mechanical and Biological)	87%	Slovenia
Simon et al., 2018 [105]	7216 MP/L	54 MP/L	—	98.3%	Denmark
Wisniowska et al., 2018 [106]	$19.4 \cdot 10^3$ to $552.2 \cdot 10^3$ MP/1 m^3	0.028 to 0.96 MP/L	n.a.	95–99%	Poland
Ziajahromi et al., 2017 [107]	n.a	0.28 MP/L	Primary, secondary and tertiary	92–99%	Australia
Yang et al., 2019 [108]	12.03MP/L	0.59 MP/L	Primary and secondary	95%	China (Beijing)
Long et al., 2019 [109]	1.57–13.69 MP/L	0.20–1.73 MP/L	Primary and secondary	97.8%	China
Xu et al., 2019 [110]	196.00 MP/L	9.04 MP/L	Primary and secondary	97.2%	China (Changzhu)
Lv et al., 2019 [111]	0.28 mp/L	0.13 and 0.05 MP/L	n.a.	MBR 99.5%,	China
Liu et al., 2019 [112]	80 MP/L	28.4 MP/L	Primary and secondary	64.4%	China
Ren et al., 2020 [113]	16.0 MP/L	2.9 MP/L	Primary, secondary and tertiary	81.9%	China (Zhengzhou)
Wei et al., 2020 [114]	430–2154 MP/m^3	430–2154 MP/m^3	RD-WWTFs	84%	China (Hangzhou)
Tang et al., 2021 [115]	23.3 MP/L and 80.5 MP/L	23.3 to 7.9 MP/L	Primary and secondary	66.1 and 62.7%,	China (Wuhan City)
Nguyen et al., 2021 [116]	n.a	n.a	Primary, Secondary and tertiary	80%	Korea (Seoul)

Ref.	MP conc. in influents	MP conc. in effluent	WWTP type	Overall retention/ efficiency	Country
Yuan et al., 2020 [117]	n.a	10.30 MP/L, 6.10 MP/L	Primary, Secondary and tertiary	97.67% and 98.46%	China (Nanjing)
Park et al., 2020 [118]	10 to 470 MP/L	10 to 470 MP/L	Primary, Secondary and tertiary	98.7–99.99%	Korea
Zhou et al., 2020 [119]	54,100 MFs/L	537.5 MFs/L (MF)	Primary, Secondary and tertiary	85%	China (Keqiao industrial park)
Mak et al., 2020 [120]	n.a	10,816 MP/m^3	Primary, Secondary	86.4%	Hong Kong (Victoria Harbor)
Zou et al., 2020 [121]	n.a	1.719 ± 1.035 MP/L	n.a	n.a	China (Guangzhou)
Hidayaturrahman et al., 2020 [122]	13813 MP/L	132 MP/L	Primary, Secondary and tertiary	> 98%	South Korea (Daegu)
Talvitie et al., 2017 [125]	6.9	0.005 MP/L	4 tertiary WWTPs	99.9%	Finland
Magni et al., 2019 [126]	2.5 MP/L	0.4 MP/L	Primary, secondary and tertiary	84%	Italy
Bayo et al., 2020 [127]	15.70 MP/L	13.04 MP/L	Primary	90.3%	Spain (Cartagena)
Gűndoğdu et al., 2018 [128]	4,825,697/day	7.02 MP/L	Secondary	73%	Turkey (Seyhan)
Gűndoğdu et al., 2018 [128]	2,040,639/day	4.11 MP/L	Secondary	79%	Turkey (Yűreğir)
Bayo et al., 2019 [129]	15.70 MP L^{-1}	0.25 MP/L	Primary	90.3%	Spain (Cartagena)
Blair et al., 2019 [130]	3 and 10 MP L^{-1}	<1 and 3 MP/L	Tertiary	96%	UK
Wolff et al., 2019 [131]	n.a	59 and 30 MP/L	Primary and secondary	n.a	Germany
Ziajahromi et al., 2021 [132]	n.a	22.1×10^6 to 133×10^6 per day	n.a	99.8–98.2%	Australia
Petroody et al., 2020 [133]	12667 MP/m^3	12667 ± 668, 3514 ± 543 and 423 ± 44.9 MP/m^3	n.a	96.7%	Iran (Sari)
Edo et al., 2020 [134]	n.a	12.8 ± 6.3 MP/L	Primary and secondary	>90%	Spain (Madrid)

Ref.	MP conc. in influents	MP conc. in effluent	WWTP type	Overall retention/ efficiency	Country
Ben-David et al., 2021 [135]	28.28 MP/L	1.97 MP/L	Primary, Secondary and tertiary	97%	Israel (Karmiel)
Tagg et al., 2020 [136]	n.a	1.5 MP/L	Primary, Secondary and tertiary	76.9%	UK (East Midlands)
Akarsu et al., 2020 [137]	1.1 and 3.6 MP/L	0.9 MP/L	Primary, Secondary and tertiary	55–97%	Turkey Mersin Bay
Naji et al., 2021 [138]	74 (±11.01, SD) and 67 (±18.35, SD) MP 35/L	70.66 MP/L	Primary and secondary	n.a	Iran (Bandar Abbas City)
Rajala et al., 2020 [139]	n.a .	0.1 mg/L, 6.7 mg/L (used)	Secondary	99.4%	Finland
Alvim et al., 2020 [140]	n.a	11.1 MP/L	Primary, Secondary	n.a	Spain (Valencia)
Pittura et al., 2021 [141]	(12,170,000 MP/h) 3.6 MP/L	1,730,000 MP/h	Primary, Secondary	94%	Italy
Raju et al., 2020 [142]	11.80 ± 1.10 MP/L	2.76 ± 0.11 MP/L	Secondary	76.61%	Australia (New south wales Hunter Region)
Ferreira et al., 2020 [143]	n.a	0.24 ± 0.07 MP/m^3 (Laucala Bay) and 0.09 ± 0.02 MP/m^3 (Suva Harbour)	79 WWTPs'	N.a	Fiji (Suva)
Schmidt et al., 2020 [144]	n.a	$4 * 10^0$ and $4.5 * 10^5$ MP/m^3	Secondary	n.a	Germany

Table 1.
Published studies on the microplastic (MP) retention efficiency of different WWTPs.

In the following, we describe the outcome of some of the earlier studies (2012–2016) in differently-sized WWTPs in various parts of the world. In 2014, Magnusson and Norén [99] studied the MP retention in a smaller WWTP in Lysekil, Sweden (Långeviksverket, serving 14.000 inhabitants, flow rate of 5160 m^3/day). During the study time, the WWTP received per m^3 15.1 ± 0.89·10^3 MP (10.7 ± 0.39·X 10^3 plastic fibers; 2.67 ± 0.77 X 10^3 plastic fragments; 1.78 ± 0.80·X 10^3 plastic flakes). Of these, only 8.25 ± 0.85 MP m^{-3} (4.00 ± 0.58 plastic fibers, 3.75 ± 1.25 plastic fragments, 0.50 ± 0.50 plastic flakes) could be found in the effluent, which was released into the sea. This amounted to 99.96% retention for plastic fibers. While MPs in the effluent were still appreciably higher than in the receiving water, Magnusson and Norén [99] found a steady decrease in fiber concentrations with increasing distance from the discharge point, from 1.82 ± 0.45 fibers/m^3 at 20 m from the discharge point to 1.14 ± 0.38 fibers/m^3 at 200 m from the discharge point. A larger sized WWTP was studied in 2012 by J. Talvite et al. [78] at Viikinmäki, Finland (serving 800.000 inhabitants in the Helsinki metropolitan area, flow rate 270.000 m^3/day). The influent carried 180 textile fibres L^{-1} and 430 synthetic particles L^{-1}. After the primary sedimentation, the wastewater contained an average of 14.2 (±0.7) fibres and 290.7 (±28.2) synthetic particles L^{-1}. After the secondary sedimentation, 13.8 (±1.6) fibres and 68.6 (±6.3) synthetic particles were still present. The remainder of the fibers and particles had settled in the sludge. Thus, most of the fibers were eliminated in the primary sedimentation process; most of the other synthetic particles were removed in the second sedimentation. As a tertiary stage, the WWTP also included a biological filtration, which removed further particles from the treated water. Removal of fibers in the second and third treatment stages was insignificant. The final effluent carried 4.9 (±1.4) fibres and 8.6 (±2.5) synthetic particles L^{-1}. This means that 3.73 x 10^9 fibers were released daily, with the effluent [78] into the Gulf of Finland, Baltic Sea. In 2014, Talvitie and Heinonen [97] published a study on the Central WWTP of Vodokanal in St. Petersburg, Russia (serving about 4 million people, flow rate of 959.000 m^3/day), carried out in collaboration with HSY, Vodokanal of St. Petersburg and Water Research and Control Center. The influent was found to carry 467 fibers L^{-1}, 160 synthetic particles L^{-1}, in addition to 3160 less identified black particles L^{-1}, most likely of synthetic nature. After pre- and primary treatment, these values decreased to 33 fibers L^{-1}; 21 synthetic particles L^{-1}, and 302 black particles L^{-1}. After, secondary treatment, these values decreased further to 16 fibers L^{-1}; 7 synthetic particles L^{-1}, and 125 black particles L^{-1}. Nevertheless, still 153.4 X 10^9 fibers were released daily with the effluent, some of which lastly will reach the Baltic Sea. Murphy et al. studied the MP retaining capability of a secondary WWTP in UK. Here, the influent contained on average 15.70 (±5.23) MP·L^{-1}. This was reduced to 0.25 (±0.04) MP·L^{-1} in the final effluent, which is a decrease of 98.4% [100]. The team reported that about 45% of microplastics were removed in the grease and grit tank, while primary sedimentation in the first clarifier accounted for 34% removal [97]. 20% of the microplastics were removed in the secondary stage [100]. In 2016, M.R. Michielssen et al., investigated the retainment efficiency towards small anthropogenic litter (SAL) of a Detroit WWTP (Great Lakes Water Authority, serving 2.36 million people, flow rate 2.5 million m^3/day). SAL includes both plastic based and cellulose derived materials. Pretreatment at the Detroit WWTP removed 58.6% SAL. Primary treatment retained an additional 25.5%, secondary treatment (with activated sludge) an additional 9.7% SAL for a total of 93.8% SAL removal overall [85]. Nevertheless, the effluent was found to release about 8.94 billion fibers a day [85]. M.R. Michielssen et al. also looked at the much smaller Northfield WWTP (Michigan, serving 100.000 people, flow rate 1700 m^3 day^{-1}), which features sand filtration as a tertiary treatment [85]. Here, pretreatment was found to retain

35.1% SAL. Primary and secondary treatment held back a further 53.3% and 1.4% SAL, respectively. Sand filtration as a tertiary treatment method further reduced SAL by 7.4% of the total. Overall, the Northfield WWTP reduced the SAL load by 97.2%, with 8.9 million fibers released daily with the effluent [85]. In addition, M.R. Michielssen et al. studied the effect of an anaerobic membrane (AnMBR) test reactor, situated at the Northfield WWTP, as a stage directly after the pretreatment of the influent. Here, 99.4% of the inflowing MP were retained – this made for 64.5% of the total MP in the influent. Thus, pretreatment and AnMBR retained 99.6% of the MP in total [85]. Finally, in 2014–2015, Carr et al. [80] studied 7 tertiary WWTPs and one secondary WWTP in Los Angeles County, Southern California. The studied WWTPs had a gravity filtration as the tertiary stage. Bench studies of the group with water spiked with microplastics showed that all MPs could be retained by such filtration processes. This, however, did not hold up in the" real-life" scenario of the WWTPs. Nevertheless, in this study, Carr et al. showed that gravity filtration as a tertiary stage in a WWTP can give up to 99.9% MP retention, calculated over all stages [80].

2.3 Retention of MPs in preliminary and primary treatment stages of WWTPs

P.U. Iyare et al. [5] showed that the bulk of the removal of MPs, at an average of 72%, comes during pre- and primary treatment. Dris et al. reported 69% MP retention in the pre- and primary stages of a WWTP in Paris [98]. Gies et al. saw that the MP concentration decreased from 31.1 ± 6.7 MP L^{-1} in the influent to 2.6 ± 1.4 MP L^{-1} (91.7% MP retention efficiency) in the primary effluent of a major secondary WWTP near Vancouver, Canada [90]. Michielssen et al. reported that screening and primary sedimentation removed 84–88% SAL ([85], see above) in studied US WWTPs. From WWTPs in Russia [97], Finland [78] and Canada [87] it was found that pre- and primary wastewater treatment removed 92–93% of fibres. In 2015, Ziajahromi et al. performed one of few studies on a WWTP with solely a pre- and primary treatment stage, receiving wastewater from over 1 million inhabitants in the Sydney area [107]. The pre- and primary stages were standard screening (mesh size of 5 mm), grit removal and sedimentation, with the effluent discharged in the deep ocean. Here, 1.5 MP L^{-1} were detected in the effluent. With a through-put of 300.000 m^3 day^{-1}, 460 million MP day^{-1} were being discharged from the WWTP into the ocean [107].

2.4 Retention of MPs in the secondary treatment stage of WWTPs

Different studies have looked at the MP retention in the secondary treatment stages of WWTPs, where the MP is then collected in the accumulating sludge. In most cases, the secondary treatment in a WWTP involves an activated sludge process, where different bacteria lower both the organic content as well as the nitrogen content of the water. Due to the different requirements of the bacteria, some of which are autotrophs and some of which are heterotrophs, some operating under aerobic conditions, some under anoxic conditions, the process involves different stages, where the water passes through aerated zones to anoxic zones and back. Different set-ups for such processes have been developed, involving different reaction chambers or a single batch reactor, where the different stages of the process run sequentially in time. After the process, the water needs to be separated from the sludge. This may be through passing the mixture to a settling tank or through a membrane in form of microfiltration or ultrafiltration, where the activated sludge is passed back to the bioreactor. This combination then is called a membrane bioreactor (MBR). MBRs have also be seen as a separate entity as a tertiary treatment method [125].

H. Lee and Y. Kim [124] looked at the efficiency of three different types of activated sludge processes, the A2O (anaerobic-anoxic-aerobic), the sequence batch reactor (SBR) and the Media process. As of 2013, these were the main processes used at public WWTPs in South Koreas with a capacity of over 500m^3/day [A$_2$O 23.7%, SBR 34.8%, Media 22.8%] with membrane bioreactor (MBR), long term aeration, and special microbial processes accounting for the remainder of the processes [124]. The WWTP running the A2O process, with a throughput of 35.000 m^3 day^{-1} (serving 67.700 inhabitants) reduced 29.85 MP L^{-1} found in the influent (taken before the pre-treatment) to 0.435 MP L^{-1} in the effluent after disinfection (98.5 overall retention efficiency), with 14.9 MP g^{-1} found in the ensuing sludge. The WWTP running the SBR process, with a throughput of 110.000 m^3 day^{-1} (serving 235.700 inhabitants) reduced 16.45 MP L^{-1} in the original influent to 0.14 MP L^{-1} in the final effluent (99.1% overall retention efficiency), with 9.65 MP g^{-1} noted in the sludge. Finally, the WWTP running the Media process, with a throughput of 130.000 m^3 day^{-1} (serving 245.000 inhabitants), reduced 13.86 MP L^{-1} in the original influent to 0.29 MP L^{-1} in the final effluent (98% overall retention efficiency), with 13.2 MP g^{-1} found in the sludge [124]. Based on the MP found in the sludge, the retention efficiencies of the secondary treatment alone were 49.3%, 44.7%, and 49.0% for the A2O process, the SBR process, and the Media process, respectively [121]. It is not clear, if the primary settling tank is included in these numbers as the fate of the sludge from the settling tank has not been discussed. If included, the numbers would compare well with the numbers given by Murphy et al., who reported a 53.8% MP removal in the primary settler and the secondary stage. Still, in the South Korean WWTPs 3 billion, 4 billion, and 11 billion MPs were discharged annually with the final effluent in the A2O process, in the SBR process, and in the Media process, respectively.

2.5 Retention of MPs in the tertiary treatment stage of a WWTP

Tertiary treatment methods have been studied extensively in regard to MP retention. In this regard, sand and gravel filtration is a common tertiary treatment, and quite a few of the early papers looking at the retaining capability of tertiary WWTPs also investigated the performance of such filters. In 2015, New York State had authorized a study of its WWTPs and found that some WWTPs using filtration processes in their tertiary stage still released MPs. The study focused on plastic microbeads, and here it can be said that certain WWTPs using membrane microfiltration, continuous backwash up flow dual sand (CBUDS) microfiltration or rapid sand filtration indeed did not show any plastic microbeads in the effluent at the time. Data on the retention of synthetic fibers was not released, however [120]. Two other studies came from New York State at that time, both citing release of microplastics downstream from WWTPs [84, 145]. One of the studies looked at a WWTP in Western New York State (Lake Erie) (12.000 people served, flow rate 13.000 m^3/day) using granular filtration (sand/anthracite coal) as the tertiary stage, with 0.009 MP/L found in the effluent, leading to a release of 101.000 MP/day, 68% of which were fibers [84]. The effluents of three tertiary WWTPs in the San Francisco Bay area with sand filtration or sand/anthracite coal filtration were found to have higher loadings with 0.064, 0.092, and 0.127 MP/L [84], leading the largest of the WWTPs to release more than 9.6 million MP/day [84]. Also, a later study from a WWTP in Northern Italy [126] showed that sand filtration as the tertiary stage with an overall MP retention rate of 84% can still lead to significant releases in the order of 160 million MPs day^{-1}. For a WWTP in the Murcia region, Spain (serving 29.800 people; flow rate: 12.000 m^3/day) J. Bayo et al. gave a 75.5% MP retention rate for the gravity rapid sand filtration, with 3 sand filters installed in parallel [127]. Here, it was noted that RSF could retain plastic microparticulates (95.5%) better than synthetic fibers

(53.8%) [126]. In contrast, in cases. Rapid sand filtration has also been shown to lead to very significant reduction of MP concentration in the effluent, e.g., from 0.7 (±0.1) to 0.02 MP (±0.007) MP L^{-1} (97% MP retention) at the Kakolanmäki WWTP (Turku Region Waste Water Treatment Plant) in Turku, Southern Finland [122]. On the downside, there has been a report of fragmentation of MP material during sand filtration [146].

Another filtering technique uses disc filters (DF) as a final polishing step, removing particles from biologically treated wastewater. Disc filters are made of a stack of round filter meshes in a closed tank, where the filter mesh is a woven material, made of polypropylene, polyester, or polyamide with a pore size of 10–40 µm. The sludge cake formed from the retained particles is periodically removed by high-pressure back-flushing. M. Simon et al. looked [147] at the efficiency of DF in a WWTP in Grindsted, Denmark (flow rate 10.040 m^3/day). The effluent from the secondary clarifier was noted to carry 20 mg L^{-1} suspended solid. This was reduced by DF to 3–8 mg L^{-1}. When passed through DF of a pore size of 18 µm, the MP content could be reduced from 29 MP L^{-1} to 3 MP L^{-1} (89.7% removal efficiency). Talvitie et al. looked at the filtration of the secondary effluent through a pilot-scale disc filter (Hydrotech HSF 1702-1F) consisting of two discs each composed of 24 filter panels at the Viikinmäki WWTP, located in Helsinki, Finland. Here, DF-10 (10 µm pore size) decreased the MP concentration from 0.5 (± 0.2) to 0.3 (± 0.1) (40% removal efficiency) and DF-20 (20 µm pore size) from 2.0 (± 1.3) to 0.03 (± 0.01) (98% removal efficiency). The results were noted to fluctuate from trial to trial [125].

There are a number of membrane filtration techniques. However, MP removal through micro- and ultrafiltration (UF) has been studied less frequently. Often, UF is used in combination with coagulation and can be used as a secondary or tertiary treatment method. Polymeric or ceramic membranes with a pore size between 1 and 100 nm are used, 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 nano-plastics. 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 has been reported to increase the removal efficiency of small-sized polyethene particles (d < 0.5 mm) significantly from 13 to 91% [148, 149]. UF can also be used as a pretreatment for a reverse osmosis (RO) separation (see below) to protect the RO membrane. Nevertheless, fouling of membranes due to meso-particles, where MP have the same size, continues to be a problem [150].

An alternative membrane separation technique is that using dynamic membranes (DMs). DMs operate with a layer formed on a supporting membrane by particles in the influent. So, these particles in the influent create 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 [151].

Reverse osmosis (RO) is the process filtering water from a region of high solute concentration through a semipermeable membrane to a region of low-solute concentration by applying a pressure larger than the osmotic pressure. RO units are used in desalination plants but are also used in drinking water treatment plants and in some WWTPs. Ziajahromi et al. [107] have looked at a WWTP in the Sydney area operating with a reverse osmosis (RO) unit (13.000 m^3 day^{-1}) as a tertiary treatment. Here, the MP concentration decreased from 2.2 MP L^{-1} in the primary effluent to 0.21 MP L^{-1}, after the reverse osmosis (RO) process. This still leads to a discharge of 10 million MP day^{-1} into the tributary of a major urban river in Australia. It is thought that the occurrence of larger sized pores on the membrane, the membrane material and other membrane imperfections may contribute to the passage of the MP through the membrane [107].

Finally, dissolved air flotation (DAF) as a flocculation process can be used as a tertiary treatment method. It was found to remove 95% of MP remaining from the secondary treatment [125]. In this case, dissolved air flotation (DAF) was studied as a full-scale tertiary treatment at Paroinen WWTP (Hameenlinna Region Water Supply and Sewerage Ltd) located in city of Hameenlinna, Southern Finland. In DAF, water is saturated with air at high pressure and then pumped to a flotation tank at 1 atm, forming dispersed water. The formed air bubbles (typically 20–70 μm in size) in the dispersed water adhere to the suspended solids causing them to float to the surface, from which they are removed by skimming. The process necessitates only a small retention time of the treated water. At the Paroinen WWTP, before the flotation, flocculation chemical polyaluminum chloride was added to the wastewater with a dosage of 40 mg L^{-1} to enhance flocculation [125]. Y. Wang et al. studied DAF with three common types of MP in freshwater and found the hydrophilic-hydrophobic interaction not to be ideal for an efficient separation of MPs without additives, citing a removal of 32–38% of MPs, only. The efficiency could be increased by 13.6–33.7%, however, with two additives that modified the surface of the air bubbles [152].

3. Conclusion

Microplastic is a serious pollutant in our aquatic and terrestrial ecosystems. WWTPs play a major role in limiting the dispersal of MP in the environment. Nevertheless, as waste streams flow through WWTPs, these in turn become point sources of MPs, where MPs are released in the millions into rivers, lakes and lastly into the sea. Studies on different WWTPs around the world have given a good indication of the retaining efficiency of different wastewater treatment stages and methods. Usually, a large part of MP is retained in the preliminary and primary treatment stages. However, the amount of MP released in the final effluent is often a function of the tertiary treatment method used. In this regard, a further development of membranes and techniques used in combination with membranes for the filtration of MP seems of interest.

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