CRITICAL REVIEW

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Microplastics removal in wastewater treatment plants: a critical review†

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Wastewater treatment plants (WWTPs) are an important route for microplastics to enter aquatic environments. Microplastics have been recently identified in sewage samples in Russia, Sweden, France, Finland, USA, UK, Netherlands, Germany, Canada, Australia, Italy, Turkey, Denmark, Poland, China and South Korea. The aim of this study was to examine and quantify the removal efficacy of microplastics by WWTPs. Experimental methods employed in sampling, analysis and quantification of microplastics vary widely between studies. Microplastic removal rates in 21 studies were compared. Secondary and tertiary WWTPs removed an average of 88% and 94% of microplastics, respectively. The majority of microplastics, 72% on average, were removed during preliminary and primary treatment. Calculations of the settling/ floating velocities of commonly used polymers indicate that primary sedimentation removes spherical particles >27–149 μm in diameter, depending on the polymer in question. Thus, the majority of microplastics removed during wastewater treatment are likely to be present in sewage sludge. Although the removal of microplastics is high, WWTPs are still an important entry point into aquatic and terrestrial systems, given the high volumes involved and the amount of sludge reused *via* land application. The major concerns are with small particles (especially <∼150 μm) in discharged wastewater effluent and the impact that particles which accumulate in sewage sludge may have on terrestrial ecosystems.

Water impact

Secondary and tertiary wastewater treatment plants removed an average of 88% and 94% of microplastics, respectively; the majority during preliminary and primary treatment. Key impacts from microplastics in wastewater are predicted to be on terrestrial ecosystems, due to their accumulation in sewage sludge, and associated with small particles (especially < approximately 150 μm) in discharged wastewater effluent.

1. Introduction

The accumulation of plastic litter in aquatic ecosystems is now a well-known problem. Microplastics are typically characterised as plastic particles from 1 nm to <5 mm in size.[1] They can either result from the degradation and weathering of larger plastic items (secondary microplastics), or from the direct discharge of materials originally manufactured at that size (primary microplastics), for example, microbeads used in facial scrub cleansers.[2] The durability and resilience of plastics are major reasons why they pose a hazardous threat to the aquatic ecosystems.[3]
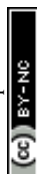
The global occurrence of microplastics in marine and freshwater environments has been affirmed by many research findings. Municipal wastewater treatment plant (WWTP) (or

sewage treatment plant) effluents are an important route for microplastics to enter the aquatic environment[4–20] and can include both primary and secondary microplastics. While WWTPs are not currently designed or optimised for the removal of microplastics, some studies indicate that advanced treatment technologies can improve their removal.[11–13,16,21] It can be inferred that the removal efficacy of microplastics in WWTPs will vary depending on the treatment process and the physiochemical properties of the polymer in question (density, particle size, charge, hydrophobicity *etc.*).[22] WWTPs receive wastewater from domestic, industrial and commercial sources, and sometimes also surface run-off. Depending on the country and location, effluent may be released directly into the ocean or into freshwater environments, typically rivers, from where it may be transported to the marine environment.[23] Microplastics removed during sewage treatment may be retained in the sludge;[5,24] which sometimes is treated and applied to land, such as for agricultural reuse.[7,25]

While there have been a number of recent reviews about plastics in wastewater treatment,[26–29] none of these

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calculated removal efficiencies for microplastics for relevant treatment processes based on all available literature or settling/floating velocities for polymers commonly found in wastewater. The objective of this study was to analyse the fate and behaviour of microplastics during wastewater treatment, which included quantifying their removal efficiency by different processes and concentrations in sewage sludge.

2. Methods

A focussed literature search for papers on microplastics in WWTPs on scientific databases (specifically ScienceDirect, ACS publications and Google Scholar) was carried out for the terms "microplastic" and "wastewater". The search yielded 740, 62 and 284 results respectively, out of which 21 research papers, which were considered relevant to the objectives of this study, were selected after reading abstracts and scanning through the contents of the papers. The removal rates, methods of sampling and analysis, and predominant type of detected microplastics from these 21 papers are summarised in Table 1. The percentage removal of different treatment stages (Table 3) and concentration of microplastics in sewage sludge (Table 4) were calculated. In order to make a preliminary assessment of settling/floating velocities of common polymer types during sewage treatment (Fig. 1), the approach of Dietrich[30] as has been previously applied to calculate theoretical settling/floating velocities for microplastics in water was followed.[31,32] In brief, the first step was to calculate the dimensionless particle diameter (D_*) for spherical plastic particles, using:

$$D_* = \frac{(\rho_s - \rho_w)gD_n{}^3}{\rho_w v^2} \tag{1}$$

where:

p_s = polymer density, kg m^{-3} (see ESI† for values used)p_w = density of water, (998.2 kg^3 m^{-1} at 20 °C)[33]g = acceleration due to gravity (9.81 m s^{-2})D_n = spherical diameter of particle, (a value of 1.6 mm was used, the largest size of plastic fragments found after primary treatment).[34]

v = kinematic viscosity, (1.003 × 10^{-6} m^2 s^{-1} °C)[33]

Subsequently, the dimensionless settling velocity (ω_*) was calculated, using:

$$\omega_* = 1.74 \times 10^{-4}D_*^2 \text{ for } D_* < 0.05 \tag{2}$$

Or:

$$\log(\omega_*) = -3.7615 + 1.92944(\log D_*) - 0.09815(\log D_*)^2 \\ - 0.00575(\log D_*)^3 + 0.00056(\log D_*)^4 \tag{3}$$

For $0.05 \leq D_* \leq 5 \times 10^9$

Finally, the theoretical settling velocity (V_s) was calculated, using:

$$V_s = \left(\left(\frac{\rho_s - \rho_w}{\rho_w}\right)gv\omega_*\right)^{1/3} \tag{4}$$

where the relevant terms are as defined above. Stokes' law can be used to directly calculate the terminal settling velocity of low-density spheres with diameter <200 μm.[31] Rearrangement of the version used by Metcalf and Eddy[33] was used for a preliminary estimation of the critical diameter above which spherical polymer particles are expected to be removed by density-driven clarification during primary sedimentation:

$$D_n = \left(\frac{18\mu V_s}{g(\rho_s - \rho_w)}\right)^{1/2} \tag{5}$$

where μ is the dynamic viscosity (1.003 × 10^{-3} kg m^{-1} s^{-1} at 20 °C,[33]) and the other terms are as defined above. When calculating the critical diameter, a settling/floating velocity of 4.6 × 10^{-4} m s^{-1} was used, which is equivalent to an overflow velocity (or surface loading rate) of 40 m^3 m^{-2} per day, typical of primary sedimentation.[33] The surface loading rate is a key design parameter for clarification processes, as if the actual settling velocity of a particle is less than the overflow velocity then it will not be removed by sedimentation. Here it has been assumed that the noted overflow velocity applies to buoyant polymers (which float),

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Table 1 Techniques utilised and microplastic influent/effluent concentrations in selected studies

Study	Location	Method of sampling	Sampled particles size range (μm)	Method of identification	Major type of detected microplastics	Influent microplastic concentration (MP per L)	Effluent microplastic concentration (MP per L)	Daily microplastic discharge (MP d^{-1})
35	Russia	Filtration device	20–300	Visual observation	Fibres, synthetic particles	467, 160	16, 7	NA
5	Sweden	Filtration	>300	Visual observation & FT-IR spectroscopy	Fibres, fragments	15	8.3	3.6×10^4
36	France	Automatic sampler	NA	Visual observation	Fibres	260–320	14–50	8.4×10^9
8	Finland	Filtration device	20–200	Visual observation	Synthetic particle, textile fibres	430, 180	9, 5	3.7×10^9
21	USA	Sieving	20–400	Visual observation & FT-IR spectroscopy	Fibres (polyethylene)	1	~0.0007	9.3×10^5
9	Scotland	Steel bucket and steel sieve	>65	Visual observation & FT-IR spectroscopy	Fibres (polyester, olyamide, acrylic)	16	0.3	6.5×10^7
24	Netherlands	Glass bottle	10–5000	Visual observation & FT-IR spectroscopy	Fibres	68–910	52	7.5×10^8 – 4.3×10^{10}
11	Germany	Filtration device	50–100	Visual observation & FPA-based micro-FT-IR imaging	Fibres (polyethylene)	ND	ND	4.2×10^4 – 1.2×10^7
12	Finland	Filtration device	NA	Visual observation & FT-IR spectroscopy	Fibres	7	0.01	1.3×10^6 – 6.6×10^7
13	Australia	Sampling device	25–500	Visual observation & FT-IR spectroscopy	Fibres (polyethylene)	2[a]	0.3, 0.2	3.6×10^6 – 1.0×10^7
14	Canada	Glass jar	1–65	Visual observation & FT-IR spectroscopy	Fibres, fragments	14	1	10.6×10^9 – 19.9×10^9
15	Turkey	Sampling device	1000–5000	Visual observation & μ-Raman spectroscopy	Fibres (polyester)	4.8×10^6 MP per day; 2.0×10^6 MP per day	1.2×10^6 MP per day; 0.35×10^6 MP per day	2.2×10^5 – 1.5×10^6
16	Finland	Steel bucket	250–5000	Visual observation, FT-IR & μ-Raman spectroscopy	Fibres (polyesters, polyethylene, polyamide)	57.6	1, 0.4	1.0×10^7
37	South Korea	NA	106–300	Visual observation & FT-IR spectroscopy	Fibre, fragments	30, 17, 14[b]	0.4, 0.1, 0.3[b]	NA
38	Denmark	Sampling device	10–500	Visual observation & FPA-based FT-IR imaging	Synthetic particle, fibres	7216	54	NA
39	Poland	Plastic canisters	109–>300	Visual observation	Fibres	19–552	0.028–0.96	NA
17	USA	Glass jar	NA	Visual observation & μFT-IR spectroscopy	Fibres	ND	ND	5.0×10^8 – 1.0×10^9
34	China	Steel bucket	20–4200	Visual observation & μ-Raman spectroscopy	Fibres, fragments	80	28	NA
18	China	Sampling device	43–5000	Visual observation & FT-IR spectroscopy	Fibres	6.6	0.6	6.5×10^8
19	China	Sampling device	25–>500	Visual observation & FT-IR spectroscopy	Fragments, fibres	4	0.1, 0.1	6.5×10^6, 3.5×10^6
20	Italy	Steel bucket and steel sieve	10–5000	Visual observation & μFT-IR spectroscopy	Fibres (polyesters, polyamide)	3	0.4	1.6×10^8

[a] Primary effluent (in lieu of influent) concentration was used for estimation. [b] Concentration from three treatment technologies. NB: concentrations separated by commas represent different microplastic types whereas others separated by dashes represent a concentration range.

as well as non-buoyant polymers (which settle). For calculations of settling/floating velocity it has been assumed that no interactions occur between plastics and other particles present. More experimental research into this topic may be fruitful, in order to validate or improve the approach followed here.

2.1 Observation and identification of microplastics

2.1.1 **Visual identification.** Visual examination uses morphological characteristics such as size, shape and colour to identify and characterise microplastics, often with a dissecting microscope *i.e.* stereomicroscope.[43–47] Particles were often classified into shapes (fibres, fragments, foils,

Table 2 Wastewater treatment processes present in selected studies

Study	Primary treatments	Secondary treatment	Tertiary treatment
35	Screening, grit and grease removal, settling	Activated sludge	
5	Screening, grit and grease removal, settling	Activated sludge	
36	Screening, grit and grease removal, settling	Biofiltration[a]	
8	Screening, grit and grease removal, pre-aeration, settling	Activated sludge	Biological filtration
21	Screening, grit and grease removal, skimming, settling	Activated sludge	Gravity filtration
9	Screening, grit and grease removal, skimming, settling	Activated sludge	
24	Screening, grit and grease removal, settling	Activated sludge	
11	Screening, grit and grease removal, skimming, settling	Activated sludge	Gravity filtration
12	Screening, grit and grease removal, skimming, settling	Activated sludge	DF, RSF, DAF, MBR
13	Screening, grit and grease removal, skimming, settling	Activated sludge	Ultrafiltration, reverse osmosis
14	Screening, grit and grease removal, settling	Trickling filters, solids contact tanks	
15	Screening, grit and grease removal, settling	Activated sludge	
16	Screening, grit and grease removal, settling	Activated sludge	MBR
37	Screening, grit and grease removal, settling	Activated sludge	
38	Screening, grit and grease removal, settling	Activated sludge	
39	Screening, grit and grease removal, settling	Activated sludge	
17	Screening, grit and grease removal, skimming, settling	Activated sludge	
34	Screening, grit and grease removal, settling	Activated sludge	
18	Screening, grit and grease removal, settling	Activated sludge	
19	Screening, grit and grease removal, settling	Activated sludge (oxidation ditch)	MBR
20	Screening, grit and grease removal, settling	Activated sludge	Gravity filtration

[a] No secondary settling involved. MBR: membrane bioreactor; DF: disc-filter, RSF: rapid sand filter; DAF: dissolved air flotation.

Table 3 Removal of microplastics during wastewater treatment in selected studies

Study	Primary	Secondary	Tertiary & advanced	Primary + secondary	Primary + secondary + tertiary
35	87–93	4–9		96–97	
5				99.9	
36	69	19		88	
8	32–92	0.2–52	5–14		97–98
21					99.9
9	78	20		98	
24				72	
11					95.5
12					99.9
13[a]					90
14	92	7		98	
15				76	
16				98.3	99.4
37				98.5	
38				99.3	
39				98.2	
17				91.4	
34	41	24		64	
18				90.5	
19				53.6	82.1
20	NA	NA	20		84
Mean (n)	72% (6)	17% (6)	15% (2)	88% (15)	94% (8)

[a] Primary effluent (in lieu of influent) concentration was used for estimation.

foam, spheres, granules, sheets, pellets, *etc.*), colour (white, black, red, blue, *etc.*), as well as quantities. Results from four studies were based on visual identification only.[8,35,36,39] Others included visual pre-selection of particles prior to spectroscopic identification (FT-IR or Raman spectroscopy). Visual examination prior to analysis represents an important step to isolate plastic particles from materials such as organic debris and other anthropogenic materials (tar, glass, metal paint coatings, *etc.*).[41] However, analysis based on visual observation is prone to bias from overestimation or underestimation by misidentification of particles.[41,48] It was difficult to differentiate between natural and synthetic fibres using visual identification alone.[36,39] Sample preparation, involving digestion, staining and extraction could decrease the tendency for misidentification.[49] The presence of synthetic fibres was reported in all 21 studies investigated. The following criteria was applied by two studies[41,50] to visually distinguish synthetic from biological fibres:

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Table 4 Microplastic concentrations in sewage sludge

Study	Location	Sampling point	Microplastic size range (μm)	Concentration (particle per kg)
5	Sweden	Slightly dewatered sludge	300–5000	16 700 720[a]
24	Netherlands	Sewage sludge	10–5000	510–760[a]
11	Germany	Sewage sludge	<500	1000–24 000
14	Canada	Primary sludge Secondary sludge	1–65	14 900[a] 4400[a]
16	Finland	Activated sludge Digested sludge MBR sludge	250–1000	23 000 170 900 27 300
37	South Korea	Anaerobic–anoxic–aerobic Sequence batch reactor Media process	106–300 >300 106–300 >300 106–300 >300	13 275 1620 7340 2315 10 615 2585
39	Poland	Sewage sludge	109–5000	6700–62 600
34	China	Dewatered sludge	20–000	240 300
20	Italy	Recycled activated sludge	10–5000	113 000

[a] Wet weight. Note: concentrations based on dry weight of sludge.
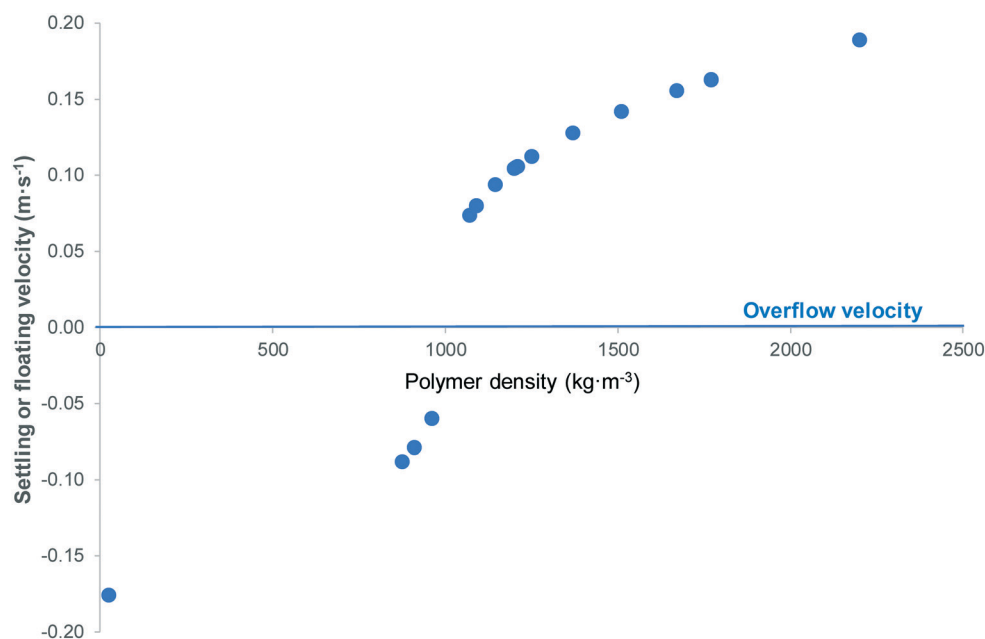

Fig. 1 Theoretical settling or floating velocities of 1.6 mm diameter spherical particles of polymers commonly found in WWTPs (using density values based on Andrady,[40] Hidalgo-Ruz et al.,[41] and Duis and Coors[42]). Polymers with density ≤960 kg m^{-3} (expanded polystyrene, polypropylene, low-density and high-density polyethylene) are predicted to float; remaining polymers, all with density ≥1070 kg m^{-3}, are predicted to be removed by sedimentation.

i. The fibres had to be equally thick through their entire length.

ii. Fibres should not be entirely straight, which indicates a biological origin.

iii. No cellular or organic structures should be visible in microplastic fibres.

iv. Transparent fibres were examined with higher magnification to confirm their nature. Green fibres were also carefully observed because this colour is very widespread in natural particles.

Another problem associated with using visual identification only is size limitation, as smaller particles (especially 10–30 μm particle sizes) could not be sorted and identified easily even under microscopes and were referred to as "potential" microplastics.[20,51] Thus, visual examination may include error rates ranging from 20%[52] to 70%,[41] which increased with decreasing size of particles.

2.1.2 Polymer identification. Spectroscopic methods are required for reliable polymer identification in microplastic samples.[11,53] FT-IR spectroscopy was the most frequently

used technique in analysing microplastics from WWTPs: 11 studies applied FT-IR spectroscopy, 2 focal plane array (FPA)-based FT-IR imaging, 2 micro-FT-IR spectroscopy and 3 micro-Raman spectroscopy (Table 1).

FT-IR identification involves comparing characteristic peaks with those present in a reference spectrum. However, there are often discrepancies between samples and those in reference spectra, as commercially available spectral libraries represent ideal samples, which may not be representative of environmental samples.[9] Comparison and identification of microplastics from environmental matrices can be improved by creating a more representative library of weathered/degraded plastics.[9,11] Applying visual pre-selection steps prior to FT-IR analysis, using a set of qualitative characteristics, was considered as labour intensive and prone to error.[54–58] In some cases, FT-IR was only used for fragments or sub-samples considered ambiguous (on the basis of visual identification).[9,21,24,27] Despite comprehensive sample processing (including organic matter digestion, density separation and staining), 22–90% of suspected microplastics were identified as non-plastic particles, following FT-IR analysis.[13] Fourier transform infrared microscopy (μFT-IR), a tool that combines FT-IR spectroscopy and microscopy, have been used by recent studies.[17,20] μFT-IR gives improved spatial resolution[59] relative to more standard types of FT-IR and requires little sample preparation, so can be used to directly identify microplastics which have been separated on membrane filters.[58] The need for visual sorting of microplastics was eliminated by introducing a chemical mapping technique using reflectance micro-FT-IR spectroscopy for detecting microplastics in marine sediments.[58] They also reported that focal plane array (FPA) detectors reduced the measurement time for complete filter analyses. Further, FPA-based micro-FT-IR has been suggested as a method for unbiased microplastic analysis in environmental samples, including wastewater.[11,51,60] By simultaneously obtaining thousands of spectra within minutes, FPA detectors can provide fast identification of even smaller microplastic fragments, without compromising spatial resolution and eliminating the need for the visual pre-selection.[51,60,61] This gives a clear advantage compared to analytical methods based on visual pre-selection. Attenuated total reflection FT-IR (ATR-FT-IR) and FPA-based transmission μFT-IR imaging, as well as an enzymatic–oxidative sample preparation were applied to identify polymers of all microplastics down to a size of 20 μm.[11] However, the whole-filter analysis was time consuming, up to 10 hours per sample of treated wastewater. This may further lead to compromises like performing partial analysis of the samples in order to process the analysis in a reasonable amount of time.[11]

Raman spectroscopy has also been frequently used for identifying microplastics.[15,16,34] Raman spectroscopy has a better size resolution than FT-IR (down to 1 μm – by automated single-particle exploration coupled to μ-Raman spectroscopy), but fluorescence often impairs the quality of Raman spectra of environmental microplastics. In contrast, FT-IR has a less precise size resolution (down to 10–20 μm – by μ-FTIR spectroscopy), and its spectral quality is not influenced by fluorescence but by the presence of water.[62] FT-IR spectroscopy was reported to be faster than Raman spectroscopy,[63] while the latter may suffer from interferences resulting from additives (*e.g.* colorants) or contaminants (including microalgae).[63,64] Raman spectroscopy is a valuable alternative to FT-IR spectroscopy when the analysis of wet samples is required or when complementary spectral profiles are desired.[60] Micro-Raman spectroscopy has been demonstrated as a reliable tool for polymer identification.[65–71]

3. Results and discussion

3.1 Overall removal of microplastics in wastewater treatment plants

While sewage treatment plants are not designed to remove microplastics,[72] an average removal value of 88% for WWTPs applying preliminary/primary plus secondary treatment, and 94% for WWTPs applying preliminary/primary plus secondary plus tertiary treatment was calculated from the 21 studies summarised in Table 3. The bulk of the removal, an average of 72%, comes during preliminary and primary treatment. Although overall removal is high, the residual amount in treated effluent (~10% of the microplastics in influent wastewater) nonetheless represents an important release of microplastics to the aquatic environment, given the large volumes of effluent involved. Data from Table 1 indicates that from 0.0–54 particles per litre (MP per L) microplastic particles were discharged into the receiving water body daily from treated wastewater. Corresponding influent concentrations of microplastics ranged from 1 to 7216 MP per L.

Note the high variability in both influent and effluent concentrations of microplastics. A large proportion of this can be explained by the variability in polymer sampling, isolation and detection methods, as discussed above. This is illustrated by the lower end of the size range of plastic particles sampled, isolated and identified in selected studies, which ranged from 1 to 1000 μm (Table 1). Due to the variability in the experimental methods deployed, comparing removal values for microplastics across multiple studies should be viewed as approximate. Nonetheless, the data presented in Table 3 does provide an important indication of the efficacy of WWTPs for the removal of microplastics.

Based on data provided by Talvitie *et al.*,[8] Murphy *et al.*[9] and Magni *et al.*,[20] intra-study experimental variability in the number of microplastic particles, as quantified by the standard deviation of replicate samples, ranged from 12–33%, with a mean value of 25%. The authors of the current study are unaware of comparisons of variability in experimental microplastic measurements between multiple studies. However, they are presumably somewhat higher than the quoted intra-study values, given the current lack of

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standardised methods for sampling, sample processing and polymer identification. In the current study, percentage microplastic removal was selected as the principal means of quantifying the performance of wastewater treatment processes, but it should be kept in mind that direct comparison between different studies is problematic and that the removal values presented here are approximate.

3.1.1 Preliminary and primary treatment. Preliminary treatment encompasses the removal of materials such as rags, sticks, floatables, grit and grease that may impair or inhibit the operation of downstream processes.[33] Preliminary and primary treatment processes in the selected studies included coarse and fine screening, grit and grease removal, skimming and primary settlement (sedimentation) (Table 2). Screens of varying sizes – typically coarse (6 to 150 mm) and fine (less than 6 mm) – retain suspended and floating solids,[33] so plastic particles larger than this are expected to be removed during preliminary treatment. This is consistent with data from Liu et al.,[34] in which a screen mesh size of 6 mm was used, who reported that the largest size of plastic fragments after primary treatment was 1.6 mm.

From Table 3, an average of 72% (range 32–93%) of microplastic particles were removed during the preliminary and primary wastewater treatment. Data from WWTPs in Russia,[35] Finland[8] and Canada[14] showed that preliminary and primary wastewater treatment removed 92–93% of fibres. Talvitie et al.[8] found that fibres were largely removed during primary sedimentation, while removal during secondary sedimentation and biological filtration was insignificant in comparison. Further, Michielssen et al.[73] reported that screening and primary sedimentation removed 84–88% of small anthropogenic litter. To illustrate this point further, analysis of microplastics retained by a secondary WWTP showed that the largest reduction in microplastics was during grit and grease removal treatment stage, 45%, followed by primary sedimentation, 34%.[9] These data are consistent with the results of the settling/floating velocity calculations displayed in Fig. 1 and Table S1.† For polymers which have been commonly reported as components of the microplastics found in WWTPs, spherical particles of diameter 1.6 mm are predicted either to be completed removed by floatation (polymers of density ≤960 kg m^{-3}, i.e. expanded polystyrene, polypropylene, low-density and high-density polyethylene) or sedimentation; (remaining polymers, all with density ≥1070 kg m^{-3}). Based on an overflow velocity of 40 m^3 m^{-2} per day, spherical polymer particles of diameter above 27–149 μm, depending on the particular polymer, are predicted to be removed during primary sedimentation (Table S1†). Note that these values are estimations, as they do not account for deviation from non-ideal behaviour of, for example, highly irregular particles or those fouled with biofilm, or of alternative removal mechanisms such as sorption. Nonetheless, they do emphasise the general point that larger particles, which in this context can be defined as those >~150 μm, are well-removed by density-driven separation in preliminary and primary wastewater treatment.

It is smaller particles which will comprise the bulk of microplastics entering subsequent treatment stages. A complicating factor is that smaller particles are also more difficult to isolate and analyse, so many will be missed during experimental surveys, even using state-of-the-art techniques which have a spatial resolution as low as 10–20 μm (see section 2.1.2).

3.1.2 Secondary treatment. Secondary treatment operations are typically biological processes which further reduce the residual suspended and dissolved solids remaining in wastewater after primary treatment.[74] In 19 of the 21 studies, secondary treatment comprised variations of the activated sludge process (Table 2), whereas Dris et al.[36] applied biofiltration and Gies et al.[14] utilised trickling filters and solids contact tanks. Secondary treatment removes additional microplastics through entrapment in solid flocs,[21] sedimentation in secondary clarifiers, or even by ingestion by microorganisms, e.g. protozoa or metazoan[75,76] present. Ferric sulfate or other flocculants used in secondary sedimentation have been suspected to aid the removal of microplastics through aggregation of particulate matter into flocs.[9] However, the mechanism of the interaction between microplastics and flocs, and the degree to which they aid microplastic removal has not, as far as we are aware, been determined. It is probable that microplastics trapped in unstable flocs may not be effectively removed, resulting in a dynamic redistribution of the particles and their subsequent escape during clarification.[21] Lee and Kim[37] suggested that smaller microplastics (106–300 μm) have a higher removal efficacy compared to larger microplastics (>300 μm), because they are not only retained in grit and grease removal stage, but also easily adsorbed to sticky media such as biofilm or floc. Liu et al.[34] suggested that turbidity removal, from secondary treatment as well as primary sedimentation, had a closer relationship with microplastic removal than other parameters.

Overall, secondary treatment removed a further 16% of microplastics, on average, (0.2–52%) relative to preliminary and primary treatment (Table 3). The activated sludge process recorded an average removal rate of 16% (0.2–52%). Biofiltration was more effective for microplastic removal (19%) compared to trickling filters and solids contact tanks (7%). Talvitie et al.[8] found that the removal of fibres was insignificant compared to the removal of synthetic particles during secondary sedimentation and the reverse was recorded during primary sedimentation. For example, 92% and 32% of textile fibres and synthetic particles were respectively removed by primary treatment, with equivalent values 0.2% and 52% during secondary treatment. HELCOM[35] also recorded a higher removal of particles compared with textile fibres during secondary treatment.

Overall, preliminary/primary and secondary wastewater treatment removed an average of 88% of microplastics (Table 3), although far higher removals have been reported by individual WWTPs. Magnusson and Norén[5] recorded a 99.9% removal rate for a relatively small (12 000 population

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equivalents) Swedish WWTP, based on one sampling event. Lee and Kim[37] compared the microplastic removal efficacies of three different configurations of the activated sludge process: the anaerobic–anoxic–aerobic (A2O), sequence batch reactor (SBR), and media process (an anaerobic, anoxic, and aerobic basin that uses a filled carrier). All processes were found to have removal efficacies above 98%, with the SBR process recording the highest rate of 99.2%. Although, experiments from a lab-scale sequencing batch biological WWTP reported a low removal rate of 52%, and smaller particles (up to 60–70 μm) were retained in the activated sludge, while the larger particles were found in the effluent.[77] However, the study focused on the fate of polyethylene microbeads and not a complex mixture of microplastics as found in authentic wastewater.
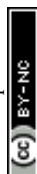
3.1.3 Tertiary and advanced treatment. Tertiary treatment technologies are used to remove specific inorganic and organic pollutants beyond levels attainable by conventional secondary treatment processes, typically as required by a particular discharge consent or reuse requirement.[33] The technologies used in the selected tertiary WWTPs included variations of filtration processes including depth, surface, biological and membrane filtration, as well as dissolved air flotation (DAF; Table 2). Tertiary treatment reduced the concentration of microplastics by a further 5–20%, relative to the level of removal achieved by secondary treatment. On average, tertiary WWTPs removed 94% (range 82–99%) of microplastics in sewage influent (Table 3). Talvitie et al.[12] investigated the removal efficacies of microplastics (>20 μm) from tertiary treatment technologies commonly used in Finland: disc-filter (DF), rapid sand filter (RSF) and DAF treating secondary effluent, and MBR treating primary effluent. Samples were collected before and after the tertiary treatment steps. MBR produced the highest percentage removal (99.9%) whereas RSF, DAF and DF removed 97%, 95% and 40–98.5% respectively. Although the MBR was treating primary clarified effluent with a much higher microplastic concentration than the other processes, which received secondary effluent, this technology still gave the lowest concentration of microplastics in final effluent. The MBR filters had the finest pore size (0.4 μm) of all the filters in the study. Furthermore, Lares et al.[16] used a pilot-scale MBR and reported 99.4% removal of microplastics. The plant consisted of an anaerobic tank, aerobic tank, as well as a membrane filtration tank with a submerged MBR unit with a pore size of 0.4 μm. Other studies have reported lower removal using MBRs as a tertiary treatment: Lv et al.[19] reported a 99.5% removal based on plastic mass but 82.1% based on the number of microplastics. Removal efficacies based on microplastic particle numbers were negatively impacted by certain unit operation, e.g. grit removal, which resulted in microplastic fragmentation and an increase in particle numbers. Thus, these authors suggested that estimated that microplastic removal should be based on mass, as well as particle number. Ultrafiltration and reverse osmosis (RO) completely removed particles larger than 190

μm, whereas the smallest size fractions (<190 μm) were reported as the most abundant after tertiary treatment.[13] Also, a post-filtration system of 12 rolling filters of pile fabric removed MP >500 μm completely but could only remove 93% of microplastics <500 μm in a German sewage treatment works.[11]

Conversely, while, Carr et al.[21] recorded 99.9% removal rate across the whole WWTP, microplastics were mainly removed in the primary treatment stage, especially through solids skimming and sludge settling processes, and the effect of gravity filters was minimal. Biofiltration systems involving upstream denitrification (UD), downstream denitrification (DD) and combined upstream–downstream denitrification (U–DD) further removed 5% of textile fibres and 14% of synthetic particles, relative to the removal achieved by secondary treatment (Table 3).[8] Liu et al.[78] assessed the efficiency of a pilot-scale biofilter to remove microplastics from WWTP effluent. 79–89% of microplastics were removed and all microplastic particles larger than 100 μm were retained. These data are consistent with results from full-scale wastewater treatment plants (Table 3). Sand filtration decreased the microplastic concentration of secondary treated effluent by over 50%, from 0.9 ± 0.3 MPs per L to 0.4 ± 0.1 MPs per L.[20] Compared to membrane filtration, this is a simple and cost-effective treatment method. Reliable estimation of microplastics removal efficacy by tertiary treatment processes require larger volumes of samples than for pre-treatment and secondary treatment processes.[27] This is because microplastic concentrations from tertiary treatment units may be very low (<1 particle per L), leading to false zero results from limited sample volumes.

3.2 Microplastic concentrations in sewage sludge

The residual solids separated during various stages of wastewater treatment are known as sewage sludge. More than 10 million tonnes of sewage sludge were generated in WWTPs in the EU in 2010.[79] The sludge produced is pumped to sludge treatment facilities such as thickening, dewatering and anaerobic digestion. Fat, oil and grease (FOG), typically separated by skimming during preliminary treatment, makes sludge more viscous and waxy, thus reducing its dewatering efficiency.[80] Sludge which contains FOG is often incinerated or landfilled.[81] As discussed above, it is likely that the majority of microplastics removed during wastewater treatment accumulate in sludge. This is illustrated by Magnusson and Norén[5] who reported that the amount of microplastic particles in the wastewater effluent entering a Swedish WWTP with a load of 12 000 population equivalent (PE) was 15 000 microplastic particles per m^3. More than 99% of this amount was retained in the sewage sludge while the concentration in the treated effluent was only 8 microplastics per m^3. Li et al.[82] suggested that laundry and plastic-industry wastewater may be the principal microplastic sources in sewage sludge. Consistent with this, plastic microfibres have been reported to dominate the composition of microplastics

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in sludge. Fibres comprised 90% of the total plastics reported by Corradini *et al.*,[83] 76% by Mahon *et al.*,[84] 70% by Magnusson and Norén[5] and 63% by Li *et al.*[82] Other items found included shafts, fragments, films, flakes and spheres. From Table 4, microplastic counts of 720–14 900 particles per kg (wet weight) have been reported from sewage sludge samples in various studies. Based on these numbers, an approximate estimation for the number of plastic particles present in the 10 million tonnes of sewage sludge generated each year in the EU is in the region of $7.2 \times 10^{12} – 1.49 \times 10^{14}$ per year. Similarly, Li *et al.*[82] estimated that the average number of microplastic particles entering the natural environment in sewage sludge in China as 1.56×10^{14} particles per year.

On a dry weight basis, a higher microplastic sludge concentration range 1000–170 900 particles per kg has been reported in literature (Table 4). The sizes of microplastics retained in the sludge of a large secondary WWTP in Glasgow, with a load of 650 000 PE, were considerably larger than those obtained from various sampling points in the WWTP.[9] This again emphasises that larger polymer particles are more easily removed through settlement operations in WWTPs. In investigating the effects of sludge treatment processes, Mahon *et al.*[84] found that microplastics in smaller size classes were high in lime stabilisation samples (*e.g.* 0.2 g kg^{-1} in the <45 μm size fraction), suggesting that the treatment process shears microplastic particles. However, a lesser abundance of microplastics observed in anaerobic digestion samples (*e.g.* 0 g kg^{-1} in the <45 μm size fraction) suggested that the process may decrease microplastic abundances. Corradini *et al.*[83] evaluated 31 agricultural fields in Chile with different sludge application records, covering a period of 10 years, and reported that successive sludge application on agricultural fields resulted in microplastic accumulation over time. Microplastic content in sludge of the agricultural fields ranged from 18 000 to 41 000 particle per kg. It is expected that microplastics in soil enter the aquatic environment through surface runoff, irrigation or wind.[85,86]

4. Conclusions

More accurate comparison of data across multiple studies would be facilitated by at least some standardisation of experimental methods for the extraction, isolation, and identification of microplastics in complex environmental matrices. Nonetheless, based on critical analysis of scientific literature on microplastics in WWTPs and sewage sludge, the following conclusions can be drawn:

• Preliminary and primary treatment, especially sedimentation and solids skimming, removed 72%, on average, of plastic particles present in sewage influent.

• Calculations of the settling/floating velocities of spherical polymer particles predict that those of diameter >27–149 μm, depending on the particular polymer, are removed during primary sedimentation. Thus, the majority

of microplastics in sewage effluent discharged to the environment are likely to be smaller particles.

• Secondary and tertiary WWTPs removed respectively 88% and 94%, on average, of the number of microplastic particles present in sewage influent. Even though these values are high, the amount of sewage effluent discharged means that WWTPs still contribute significant amounts of microplastic pollution to the environment.

• Compared with other advanced (tertiary) treatment technologies, MBRs demonstrated a higher potential for more effective microplastic removal, from 82.1–99.9%.

• The majority of plastics removed during sewage treatment are retained in sewage sludge. A wide range of concentrations for microplastics in sewage sludge have been reported in literature: from 720–14 900 particles per kg (wet weight) and from 1000–170 900 particles per kg (dry weight). The majority, from 63–90%, of microplastics in sludge were fibres. Large numbers of plastic particles enter the terrestrial environment where sludge is reused for agriculture. More research is needed on the environmental fate and impact of plastics in sludge-amended soils, in particular where agricultural reuse of sewage sludge is common practice.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

References

1 GESAMP, *Sources, fate and effects of microplastics in the marine environment: A global assessment*, (IMO/FAO/UNESCO-IOC/UNIDO/WMO/IAEA/UN/UNEP/UNDP Joint Group of Experts on the Scientific Aspects of Marine Environmental Protection), Rep. Stud. GESAMP, London, 90, 2015.

2 M. Chang, Reducing microplastics from facial exfoliating cleansers in wastewater through treatment versus consumer product decisions, *Mar. Pollut. Bull.*, 2015, **101**(1), 330–333.

3 N. A. C. Welden, Microplastic pollution in the Clyde sea area: a study using the indicator species Nephrops norvegicus, *PhD. Thesis*, University of Glasgow, 2015.

4 M. Cole, P. Lindeque, C. Halsband and T. S. Galloway, Microplastics as contaminants in the marine environment: A review, *Mar. Pollut. Bull.*, 2011, **62**(12), 2588–2597.

5 K. Magnusson and F. Norén, *Screening of microplastic particles in and down-stream a wastewater treatment plant*, IVL Swedish Environmental Research Institute, Stockholm, 2014.

6 M. Wagner, C. Scherer, D. Alvarez-Muñoz, N. Brennholt, X. Bourrain, S. Buchinger, E. Fries, C. Grosbois, J. Klasmeier

2672 | *Environ. Sci.: Water Res. Technol.*, 2020, **6**, 2664–2675

This journal is © The Royal Society of Chemistry 2020

and T. Marti, Microplastics in freshwater ecosystems: what we know and what we need to know, *Environ. Sci. Eur.*, 2014, **26**(1), 12.

7   D. Eerkes-Medrano, R. C. Thompson and D. C. Aldridge, Microplastics in freshwater systems: a review of the emerging threats, identification of knowledge gaps and prioritisation of research needs, *Water Res.*, 2015, **75**, 63–82.

8   J. Talvitie, M. Heinonen, J. P. Paakkonen, E. Vahtera, A. Mikola, O. Setala and R. Vahala, Do wastewater treatment plants act as a potential point source of microplastics? Preliminary study in the coastal Gulf of Finland, Baltic Sea, *Water Sci. Technol.*, 2015, **72**(9), 1495–1504.

9   F. Murphy, C. Ewins, F. Carbonnier and B. Quinn, Wastewater Treatment Works (WwTW) as a Source of Microplastics in the Aquatic Environment, *Environ. Sci. Technol.*, 2016, **50**(11), 5800–5888.

10   S. Ziajahromi, P. A. Neale and F. D. Leusch, Wastewater treatment plant effluent as a source of microplastics: review of the fate, chemical interactions and potential risks to aquatic organisms, *Water Sci. Technol.*, 2016, **74**(10), 2253–2269.

11   S. M. Mintenig, I. Int-Veen, M. G. J. Löder, S. Primpke and G. Gerdts, Identification of microplastic in effluents of waste water treatment plants using focal plane array-based micro-Fourier-transform infrared imaging, *Water Res.*, 2017, **108**, 365–372.

12   J. Talvitie, A. Mikola, A. Koistinen and O. Setälä, Solutions to microplastic pollution–Removal of microplastics from wastewater effluent with advanced wastewater treatment technologies, *Water Res.*, 2017, **123**, 401–407.

13   S. Ziajahromi, P. A. Neale, L. Rintoul and F. D. L. Leusch, Wastewater treatment plants as a pathway for microplastics: Development of a new approach to sample wastewater-based microplastics, *Water Res.*, 2017, **112**, 93–99.

14   E. A. Gies, J. L. LeNoble, M. Noël, A. Etemadifar, F. Bishay, E. R. Hall and P. S. Ross, Retention of microplastics in a major secondary wastewater treatment plant in Vancouver, Canada, *Mar. Pollut. Bull.*, 2018, **133**, 553–561.

15   S. Gündoğdu, C. Çevik, E. Güzel and S. Kilercioğlu, Microplastics in municipal wastewater treatment plants in Turkey: a comparison of the influent and secondary effluent concentrations, *Environ. Monit. Assess.*, 2018, **190**(11), 626.

16   M. Lares, M. C. Ncibi, M. Sillanpää and M. Sillanpää, Occurrence, identification and removal of microplastic particles and fibers in conventional activated sludge process and advanced MBR technology, *Water Res.*, 2018, **133**, 236–246.

17   K. Conley, A. Clum, J. Deepe, H. Lane and B. Beckingham, Wastewater treatment plants as a source of microplastics to an urban estuary: Removal efficiencies and loading per capita over one year, *Water Research X*, 2019, **3**, 100030.

18   Z. Long, Z. Pan, W. Wang, J. Ren, X. Yu, L. Lin, H. Lin, H. Chen and X. Jin, Microplastic abundance, characteristics, and removal in wastewater treatment plants in a coastal city of China, *Water Res.*, 2019, **155**, 255–265.

19   X. Lv, Q. Dong, Z. Zuo, Y. Liu, X. Huang and W.-M. Wu, Microplastics in a municipal wastewater treatment plant:

Fate, dynamic distribution, removal efficiencies, and control strategies, *J. Cleaner Prod.*, 2019, **225**, 579–586.

20   S. Magni, A. Binelli, L. Pittura, C. G. Avio, C. Della Torre, C. C. Parenti, S. Gorbi and F. Regoli, The fate of microplastics in an Italian Wastewater Treatment Plant, *Sci. Total Environ.*, 2019, **652**, 602–610.

21   S. A. Carr, J. Liu and A. G. Tesoro, Transport and fate of microplastic particles in wastewater treatment plants, *Water Res.*, 2016, **91**, 174–182.

22   T. Bond, V. Ferrandiz-Mas, M. Felipe-Sotelo and E. van Sebille, The occurrence and degradation of aquatic plastic litter based on polymer physicochemical properties: A review, *Crit. Rev. Environ. Sci. Technol.*, 2018, **48**(7–9), 685–722.

23   A. McCormick, T. J. Hoellein, S. A. Mason, J. Schluep and J. J. Kelly, Microplastic is an Abundant and Distinct Microbial Habitat in an Urban River, *Environ. Sci. Technol.*, 2014, **48**(20), 11863–11871.

24   H. A. Leslie, S. H. Brandsma, M. J. van Velzen and A. D. Vethaak, Microplastics en route: Field measurements in the Dutch river delta and Amsterdam canals, wastewater treatment plants, North Sea sediments and biota, *Environ. Int.*, 2017, **101**, 133–142.

25   M. C. Rillig, Microplastic in terrestrial ecosystems and the soil?, *Environ. Sci. Technol.*, 2012, **46**(12), 6453–6454.

26   P. L. Ngo, B. K. Pramanik, K. Shah and R. Roychand, Pathway, classification and removal efficiency of microplastics in wastewater treatment plants, *Environ. Pollut.*, 2019, **255**, 113326.

27   J. Sun, X. Dai, Q. Wang, M. C. van Loosdrecht and B.-J. Ni, Microplastics in wastewater treatment plants: Detection, occurrence and removal, *Water Res.*, 2019, **152**, 21–37.

28   X. Zhang, J. Chen and J. Li, The removal of microplastics in the wastewater treatment process and their potential impact on anaerobic digestion due to pollutants association, *Chemosphere*, 2020, 126360.

29   Z. Zhang and Y. Chen, Effects of microplastics on wastewater and sewage sludge treatment and their removal: a review, *Chem. Eng. J.*, 2020, **382**, 122955.

30   W. E. Dietrich, Settling velocity of natural particles, *Water Resour. Res.*, 1982, **18**(6), 1615–1626.

31   N. Kowalski, A. M. Reichardt and J. J. Waniek, Sinking rates of microplastics and potential implications of their alteration by physical, biological, and chemical factors, *Mar. Pollut. Bull.*, 2016, **109**(1), 310–319.

32   M. Kooi, E. H. V. Nes, M. Scheffer and A. A. Koelmans, Ups and downs in the ocean: effects of biofouling on vertical transport of microplastics, *Environ. Sci. Technol.*, 2017, **51**(14), 7963–7971.

33   Metcalf and Eddy Inc., G. Tchobanoglous, H. D. Stensel, R. Tsuchihashi and F. L. Burton, *Wastewater engineering: treatment and resource recovery*, McGraw-Hill Higher Education, New York, 2014.

34   X. Liu, W. Yuan, M. Di, Z. Li and J. Wang, Transfer and fate of microplastics during the conventional activated sludge process in one wastewater treatment plant of China, *Chem. Eng. J.*, 2019, **362**, 176–182.

This journal is © The Royal Society of Chemistry 2020

Environ. Sci.: Water Res. Technol., 2020, **6**, 2664–2675 | **2673**

35 J. Talvitie and M. Heinonen, *HELCOM BASE project 2012–2014: Preliminary study on synthetic microfibers and particles at a municipal waste water treatment plant*, Baltic Marine Environment Protection Commission (HELCOM), Helsinki, 2014.

36 R. Dris, J. Gasperi, V. Rocher, M. Saad, N. Renault and B. Tassin, Microplastic contamination in an urban area: a case study in Greater Paris, *Environ. Chem.*, 2015, **12**(5), 592–599.

37 H. Lee and Y. Kim, Treatment characteristics of microplastics at biological sewage treatment facilities in Korea, *Mar. Pollut. Bull.*, 2018, **137**, 1–8.

38 M. Simon, N. van Alst and J. Vollertsen, Quantification of microplastic mass and removal rates at wastewater treatment plants applying Focal Plane Array (FPA)-based Fourier Transform Infrared (FT-IR) imaging, *Water Res.*, 2018, **142**, 1–9.

39 E. Wiśniowskaa, K. Moraczewska-Majkutb and W. Nocońb, Efficiency of microplastics removal in selected wastewater treatment plants–preliminary studies, *Desalin. Water Treat.*, 2018, **134**, 316–323.

40 A. L. Andrady, Microplastics in the marine environment, *Mar. Pollut. Bull.*, 2011, **62**(8), 1596–1605.

41 V. Hidalgo-Ruz, L. Gutow, R. C. Thompson and M. Thiel, Microplastics in the marine environment: a review of the methods used for identification and quantification, *Environ. Sci. Technol.*, 2012, **46**(6), 3060–3075.

42 K. Duis and A. Coors, Microplastics in the aquatic and terrestrial environment: sources (with a specific focus on personal care products), fate and effects, *Environ. Sci. Eur.*, 2016, **28**(1), 2.

43 D. G. Shaw and R. H. Day, Colour-and form-dependent loss of plastic micro-debris from the North Pacific Ocean, *Mar. Pollut. Bull.*, 1994, **28**(1), 39–43.

44 C. J. Moore, S. L. Moore, S. B. Weisberg, G. L. Lattin and A. F. Zellers, A comparison of neustonic plastic and zooplankton abundance in southern California's coastal waters, *Mar. Pollut. Bull.*, 2002, **44**(10), 1035–1038.

45 S. Morét-Ferguson, K. L. Law, G. Proskurowski, E. K. Murphy, E. E. Peacock and C. M. Reddy, The size, mass, and composition of plastic debris in the western North Atlantic Ocean, *Mar. Pollut. Bull.*, 2010, **60**(10), 1873–1878.

46 L. M. Rios, P. R. Jones, C. Moore and U. V. Narayan, Quantitation of persistent organic pollutants adsorbed on plastic debris from the Northern Pacific Gyre's "eastern garbage patch", *J. Environ. Monit.*, 2010, **12**(12), 2226–2236.

47 M. J. Doyle, W. Watson, N. M. Bowlin and S. B. Sheavly, Plastic particles in coastal pelagic ecosystems of the Northeast Pacific ocean, *Mar. Environ. Res.*, 2011, **71**(1), 41–52.

48 M. G. Löder and G. Gerdts, in *Marine anthropogenic litter*, Springer, 2015, pp. 201–227.

49 S. A. Mason, D. Garneau, R. Sutton, Y. Chu, K. Ehmann, J. Barnes, P. Fink, D. Papazissimos and D. L. Rogers, Microplastic pollution is widely detected in US municipal wastewater treatment plant effluent, *Environ. Pollut.*, 2016, **218**, 1045–1054.

50 Sweden KIMO, *Small plastic particles in Coastal Swedish waters, N-Research report*, commissioned by KIMO Sweden (Submitted to BDC), Sweden, 2007.

51 M. G. J. Löder, M. Kuczera, S. Mintenig, C. Lorenz and G. Gerdts, Focal plane array detector-based micro-Fourier-transform infrared imaging for the analysis of microplastics in environmental samples, *Environ. Chem.*, 2015, **12**(5), 563–581.

52 M. Eriksen, S. Mason, S. Wilson, C. Box, A. Zellers, W. Edwards, H. Farley and S. Amato, Microplastic pollution in the surface waters of the Laurentian Great Lakes, *Mar. Pollut. Bull.*, 2013, **77**(1–2), 177–182.

53 T. Rocha-Santos and A. C. Duarte, A critical overview of the analytical approaches to the occurrence, the fate and the behavior of microplastics in the environment, *TrAC, Trends Anal. Chem.*, 2015, **65**, 47–53.

54 R. C. Thompson, Y. Olsen, R. P. Mitchell, A. Davis, S. J. Rowland, A. W. John, D. McGonigle and A. E. Russell, Lost at sea: where is all the plastic?, *Science*, 2004, **304**(5672), 838.

55 K. Ng and J. Obbard, Prevalence of microplastics in Singapore's coastal marine environment, *Mar. Pollut. Bull.*, 2006, **52**(7), 761–767.

56 M. S. Reddy, S. Basha, S. Adimurthy and G. Ramachandraiah, Description of the small plastics fragments in marine sediments along the Alang-Sosiya ship-breaking yard, India, *Estuarine, Coastal Shelf Sci.*, 2006, **68**(3–4), 656–660.

57 P. L. Corcoran, M. C. Biesinger and M. Grifi, Plastics and beaches: a degrading relationship, *Mar. Pollut. Bull.*, 2009, **58**(1), 80–84.

58 J. P. Harrison, J. J. Ojeda and M. E. Romero-González, The applicability of reflectance micro-Fourier-transform infrared spectroscopy for the detection of synthetic microplastics in marine sediments, *Sci. Total Environ.*, 2012, **416**, 455–463.

59 J. L. Koenig, S.-Q. Wang and R. Bhargava, FTIR images, *Anal. Chem.*, 2001, **73**(13), 361A–369A.

60 A. S. Tagg, M. Sapp, J. P. Harrison and J. J. Ojeda, Identification and Quantification of Microplastics in Wastewater Using Focal Plane Array-Based Reflectance Micro-FT-IR Imaging, *Anal. Chem.*, 2015, **87**(12), 6032–6040.

61 R. Bhargava, S.-Q. Wang and J. L. Koenig, *Liquid Chromatography/FTIR Microspectroscopy/Microwave Assisted Synthesis*, Springer, Berlin, Heidelberg, 2003, pp. 137–191.

62 L. Cabernard, L. Roscher, C. Lorenz, G. Gerdts and S. Primpke, Comparison of Raman and Fourier transform infrared spectroscopy for the quantification of microplastics in the aquatic environment, *Environ. Sci. Technol.*, 2018, **52**(22), 13279–13288.

63 M. Cole, H. Webb, P. K. Lindeque, E. S. Fileman, C. Halsband and T. S. Galloway, Isolation of microplastics in biota-rich seawater samples and marine organisms, *Sci. Rep.*, 2014, **4**, 4528.

64 L. Van Cauwenberghe, A. Vanreusel, J. Mees and C. R. Janssen, Microplastic pollution in deep-sea sediments, *Environ. Pollut.*, 2013, **182**, 495–499.

65 F. Collard, B. Gilbert, G. Eppe, E. Parmentier and K. Das, Detection of anthropogenic particles in fish stomachs: an

2674 | *Environ. Sci.: Water Res. Technol.*, 2020, **6**, 2664–2675

This journal is © The Royal Society of Chemistry 2020

isolation method adapted to identification by Raman spectroscopy, *Arch. Environ. Contam. Toxicol.*, 2015, **69**(3), 331–339.

66 K. Enders, R. Lenz, C. A. Stedmon and T. G. Nielsen, Abundance, size and polymer composition of marine microplastics ≥ 10 μm in the Atlantic Ocean and their modelled vertical distribution, *Mar. Pollut. Bull.*, 2015, **100**(1), 70–81.

67 L. Frère, I. Paul-Pont, J. Moreau, P. Soudant, C. Lambert, A. Huvet and E. Rinnert, A semi-automated Raman microspectroscopy method for morphological and chemical characterizations of microplastic litter, *Mar. Pollut. Bull.*, 2016, **113**(1–2), 461–468.

68 A. Käppler, F. Windrich, M. G. Löder, M. Malanin, D. Fischer, M. Labrenz, K.-J. Eichhorn and B. Voit, Identification of microplastics by FTIR and Raman microscopy: a novel silicon filter substrate opens the important spectral range below 1300 cm⁻ 1 for FTIR transmission measurements, *Anal. Bioanal. Chem.*, 2015, **407**(22), 6791–6801.

69 B. E. Oßmann, G. Sarau, S. W. Schmitt, H. Holtmannspötter, S. H. Christiansen and W. Dicke, Development of an optimal filter substrate for the identification of small microplastic particles in food by micro-Raman spectroscopy, *Anal. Bioanal. Chem.*, 2017, **409**(16), 4099–4109.

70 R. Lenz, K. Enders, C. A. Stedmon, D. M. Mackenzie and T. G. Nielsen, A critical assessment of visual identification of marine microplastic using Raman spectroscopy for analysis improvement, *Mar. Pollut. Bull.*, 2015, **100**(1), 82–91.

71 S. Zhao, M. Danley, J. E. Ward, D. Li and T. J. Mincer, An approach for extraction, characterization and quantitation of microplastic in natural marine snow using Raman microscopy, *Anal. Methods*, 2017, **9**(9), 1470–1478.

72 E. Schneiderman, *Discharging Microbeads to Our Waters: An Examination of Wastewater Treatment Plants in New York*, 2015.

73 M. R. Michielssen, E. R. Michielssen, J. Ni and M. B. Duhaime, Fate of microplastics and other small anthropogenic litter (SAL) in wastewater treatment plants depends on unit processes employed, *Environ. Sci.: Water Res. Technol.*, 2016, **2**(6), 1064–1073.

74 H. Patel and R. T. Vashi, in *Characterization and Treatment of Textile Wastewater*, ed. H. Patel and R. T. Vashi, Elsevier, Boston, 2015, pp. 1–20.

75 C.-B. Jeong, E.-J. Won, H.-M. Kang, M.-C. Lee, D.-S. Hwang, U.-K. Hwang, B. Zhou, S. Souissi, S.-J. Lee and J.-S. Lee, Microplastic Size-Dependent Toxicity, Oxidative Stress Induction, and p-JNK and p-p38 Activation in the Monogonont Rotifer (Brachionus koreanus), *Environ. Sci. Technol.*, 2016, **50**(16), 8849–8857.

76 C. Scherer, A. Weber, S. Lambert and M. Wagner, in *Freshwater Microplastics*, Springer, Cham, 2018, pp. 153–180.

77 G. Kalčíková, B. Alič, T. Skalar, M. Bundschuh and A. Ž. Gotvajn, Wastewater treatment plant effluents as source of cosmetic polyethylene microbeads to freshwater, *Chemosphere*, 2017, **188**, 25–31.

78 F. Liu, N. B. Nord, K. Bester and J. Vollertsen, Microplastics Removal from Treated Wastewater by a Biofilter, *Water*, 2020, **12**(4), 1085.

79 Eurostat, Sewage sludge production and disposal, 2014 [cited 2019 June]; Available from: http://appsso.eurostat.ec. europa.eu/nui/show.do?dataset=env_ww_spd&lang=en.

80 A. T. Jameel, S. A. Muyubi, M. I. A. Karim and M. Z. Alam, Removal of oil and grease as emerging pollutants of concern (EPC) in wastewater stream, *IIUM Engineering Journal*, 2011, **12**(4), 161–169.

81 D. H. Liu and B. G. Liptak, *Environmental Engineers' Handbook Second Edition*, CRC Press LLC, Florida, 1999.

82 X. Li, L. Chen, Q. Mei, B. Dong, X. Dai, G. Ding and E. Y. Zeng, Microplastics in sewage sludge from the wastewater treatment plants in China, *Water Res.*, 2018, **142**, 75–85.

83 F. Corradini, P. Meza, R. Eguiluz, F. Casado, E. Huerta-Lwanga and V. Geissen, Evidence of microplastic accumulation in agricultural soils from sewage sludge disposal, *Sci. Total Environ.*, 2019, **671**, 411–420.

84 A. M. Mahon, B. O'Connell, M. G. Healy, I. O'Connor, R. Officer, R. Nash and L. Morrison, Microplastics in Sewage Sludge: Effects of Treatment, *Environ. Sci. Technol.*, 2017, **51**(2), 810–818.

85 J. Hammer, M. H. Kraak and J. R. Parsons, in *Reviews of environmental contamination and toxicology*, Springer, 2012, pp. 1–44.

86 C. M. Rochman, S. M. Kross, J. B. Armstrong, M. T. Bogan, E. S. Darling, S. J. Green, A. R. Smyth and D. Veríssimo, Scientific evidence supports a ban on microbeads, *Environ. Sci. Technol.*, 2015, **49**(18), 10759–10761.

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