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Average daily flow of microplastics through a tertiary wastewater treatment plant over a ten-month period

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ABSTRACT

Microplastics (MPs, <5 mm in size) are classified as emerging contaminants but treatment processes are not designed to remove these small particles. Wastewater treatment systems have been proposed as pathways for MPs pollution to receiving waters but quantitative and qualitative data on MP occurrence and transport remains limited, hindering risk assessment and regulation. Here, for the first time, the stepwise abundance and loading of MPs (60–2800 μm) in a tertiary wastewater treatment plant in the UK was assessed by sampling from May 2017 to February 2018. Microplastics were found in all sampling campaigns, with an average inflow of 8.1×10^8 (95% CI, 3.8×10^8 to 1.2×10^9) items day^{-1}. Their prevalence decreased from influent to final effluent. Overall abundances decreased on average by 6%, 68%, 92%, and 96% after the pre-treatment, primary, secondary, and tertiary treatment stages respectively, although considerable variability occurred throughout the year. Sufficient particles remained in the treated effluent to generate an average discharge of 2.2×10^7 (95% CI, 1.2×10^7 to 3.2×10^7]) particles day^{-1} to the recipient river. Secondary MPs were predominant, while primary MP abundances were minimal. Fibres comprised 67% of all items, followed by films (18%) and fragments (15%). Chemical characterisation confirmed the presence of different types of polymers, with polypropylene fibres and fragments most abundant (23%). This research informs understanding of how wastewater effluent may channel MPs to the natural environment and their composition, and helps understand control points for optimising advanced treatment processes.

1. Introduction

Microplastics (MPs; <5 mm) are ubiquitous in the environment and may pose a threat to biota and humans (Anbumani and Kakkar, 2018), thus are classed as emerging contaminants but remain unregulated by water quality standards. This may be largely because they have not been fully assessed due to their heterogeneous nature and high spatio-temporal variations, even within localized environmental compartments. Furthermore, a lack of standardized protocols leads to limited comparability across available surveys and a lack of guidelines to monitor MPs in aquatic systems. Current empirical data is still too limited to fully understand the extent of their pollution and the severity of their threat, making it difficult for regulators to determine what types of MPs need to be prioritised in monitoring programmes and where controls should be implemented. Nevertheless, similar to other anthropogenic contaminants, 80% of MPs are considered to originate from land-based sources (Rochman et al., 2015). Therefore the role of wastewater treatment plants (WWTPs) as potential barriers of MP pollution should be considered, as they are important links between the anthropogenic and natural environments (Ou and Zeng, 2018).

Wastewater treatment systems are designed to remove contaminants from household and trade effluent, so their role in MPs removal has been generating increasing attention, yet they remain largely unexplored (Table 1). The majority of available studies quantify MPs in secondary effluent, with fewer studies considering tertiary treatment plants (Table 1). Here, secondary treatment refers to biological wastewater treatment (e.g. activated sludge) resulting in the separation of decanted effluent and sludge containing microbial biomass (European Environment Agency, 2019). Tertiary or advanced treatment refers to post-secondary polishing

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Table 1
Summarised research from 2011 to 2019 on MPs in WWTPs.

Ref[a]	Location	Treatment Type	Plant size (p.e.)	Sites	Sample Volume (L)	Stages Sampled	Biosolid Samples	Analytical Method	Size Range (μm)	Effluent Concentration (count L^{-1})	Removal (%)
1	Australia	Tertiary		2	0.75	Effluent	None	FTIR	<1000	1	
2	Sweden	Secondary	1.4×10^4	1	2–1000	Influent, final effluent	Sewage sludge	Visual sorting; FTIR	>300	8×10^{-3}	99.9
3	France	Secondary		1	0.05	Influent, primary, final effluent		Visual sorting	100 –5000	14–50	83–95
4	USA	Secondary & Tertiary		7	189,000 –232,000	Influent, primary, secondary, final effluent	Sewage sludge, activated sludge	Visual sorting; FTIR	45 –400	8×10^{-4}	~99.9
5	USA	Secondary & Tertiary	3.5×10^3 –5.6×10^7	17	500 –21,000	Final effluent	None	Visual sorting	>125	5×10^{-2}	
6	USA	Secondary & Tertiary		3	1–38	Influent, pre-treatment, primary, secondary, final effluent	None	Visual sorting	20 –4750	1.4–2.6	95.6 –99.4
7	Scotland	Secondary	6.5×10^5	1	30–50	Influent after screens, pre-treatment, primary, final effluent	Grit and grease, sludge cake from centrifuge	Visual sorting; FTIR	>65	2.5×10^{-1}	98.41
8	USA	Secondary & Tertiary		8	2-hr composite	Final effluent	None	Visual sorting	125 –355	4.7×10^{-2} - 1.9 x $^{-1}$	
9	USA	Secondary	6.8×10^5	1	2–24 h composite	Final effluent	None	Visual sorting; Raman; FTIR	125 –5000	0.3–2.4	
10	Netherlands	Secondary & Tertiary		7	2	Influent, final effluent	Sewage sludge	Visual sorting; FTIR	10 –5000	9–91	
11	Germany	Secondary & Tertiary	7.0×10^3 –2.1×10^5	12	390 –1000	Final effluent	Sewage sludge	Visual sorting; FTIR	20 –5000	1×10^{-3} - 9	~97
12	Finland	Tertiary	5×10^4 –8×10^5	4	0.4–1000	Influent, final effluent	None	Visual sorting; FTIR	20- >300	5×10^{-3} - 3×10^{-1}	40–99.9
13	Finland	Tertiary	8×10^5	1	0.1–1000	Influent, pre-treatment, secondary, final effluent,	Excess sludge, dry sludge	Visual sorting; FTIR	20- >300	7×10^{-1} - 3.5	>99
14	Australia	Primary, Secondary & Tertiary	1.5×10^5 –1.2×10^6	3	3–200	Final effluent	None	Staining and visual sorting; FTIR	25 –500	2.8×10^{-1} - 1.54	90
15	Canada	Secondary	1.3×10^6	1	1–30	Influent, primary, final effluent	Sewage sludge, activated sludge	Visual sorting; FTIR	1–65	5×10^{-1}	99
16	Finland	Secondary		1	4–30	Influent after screens, primary, final effluent	Activated sludge, digested sludge, membrane bioreactor sludge	Visual sorting; FTIR, Raman	0.25 –5000	4×10^{-1} - 1	98.3
17	Denmark	Secondary & Tertiary		10	1–81.5	Influent after screens, final effluent	None	FTIR-FPA	10 –500	54	99.3
18	USA	Secondary	180,000-53000	3	3.6–30	Influent; final effluent	None	Visual sorting; FTIR		1–30	74.8 –98.1
19	Italy	Tertiary	1.2×10^6	1	30	Influent, after settler, outlet	None	Visual sorting; FTIR	63 –5000	4×10^{-1}	84

[a] **1**, Browne et al., (2011); **2**, Magnusson and Norén, 2014; **3**, Dris et al., (2015); **4**, Carr et al., (2016); **5**, Mason et al., (2016); **6**, Michielssen et al., (2016); **7**, Murphy et al., (2016); **8**, Sutton et al., (2016); **9**, Dyachenko et al., (2017); **10**, Leslie et al., (2017); **11**, Mintenig et al. 2017; **12**, Talvitie et al., (2017a); **13**, Talvitie et al., (2017b); **14**, Ziajahromi et al., (2017); **15**, Gies et al., (2018); **16**, Lares et al., (2018); **17**, Simon et al., (2018); **18**, Conley et al., (2019); **19**, Magni et al., (2019)..

steps (e.g. chemical removal, advanced filtration) to eliminate pollutants not removed by secondary treatment (European Environment Agency, 2019). Current understanding suggests that a mixture of primary and secondary MPs may be entering the treatment facilities daily, at varying levels of pollution (Sun et al., 2019). Microplastic concentrations in raw wastewater are reported so far to range from <1 particle L^{-1} as observed by multiple studies (Table 1), to 18,285 particles L^{-1} reported in a secondary treatment site in Denmark (Simon et al., 2018). Conversely, effluent concentrations between 8×10^{-4} (Magnusson and Norén, 2014) and 447 (Simon et al., 2018) particles L^{-1} have been observed in secondary WWTPs, and between 0 (Carr et al., 2016) and 51 particles L^{-1} (membrane bioreactor, MBR; Leslie et al., 2017) after advanced treatment (Sun et al., 2019), with larger facilities likely discharging higher loads (Mason et al., 2016). While the WWTP literature has grown over the past two years, each study differs in methodologies (e.g. sampling volumes, detection limits), plant capacity, and type of treatment technologies and stages examined. Therefore, it is difficult to determine what variation across studies is due to site differences or analytical bias, limiting comparability of findings and comprehensive understanding of the occurrence and fate of MPs in these systems.

Comparison of influent vs effluent concentrations is a common approach to estimate removal efficiencies, which range between 40% and 99.9% (Table 1). While absolute values may be difficult to compare, reporting of removal percentages may improve intra-study comparisons, but not all studies report this. Despite high retention efficiencies, low concentrations in final or treated effluent may represent daily releases of millions of MPs when scaled for the discharge volumes (Mason et al., 2016; Murphy et al., 2016). For instance, concentrations of 2.5×10^{-1} and 4×10^{-3} particles L^{-1} in final effluent, equated to discharges of 6.5×10^7 and 5×10^4 MPs day^{-1}, respectively in secondary treatment plants in Scotland, UK (Murphy et al., 2016) and San Francisco, USA (Mason et al., 2016). Microplastic discharges from WWTPs appear highly variable, and treatment procedure employed at the facility is presumed to be crucial in their retention.

The role of different treatment processes in removing contaminants from these systems can be assessed by a stage-wise inspection of MPs abundances during their passage through a single facility. Owing to challenges of sample collection and processing times, only a few studies have done this (Table 1), and stages

sampled vary across studies. It appears that between ~63 and 98% of the removal can occur by the primary stage (Sun et al., 2019). Secondary treatment may reduce an additional 7–20% of MPs not captured by preliminary and primary treatment (Talvitie et al., 2017b; Ziajahromi et al., 2017; Gies et al., 2018). The observation of MPs in different types of biosolids suggest that their removal during earlier stages is through their capture in various sludge fractions including grit and grease skimmings (Murphy et al., 2016), sewage sludge (Bayo et al., 2016; Murphy et al., 2016; Leslie et al., 2017; Mintenig et al. 2017; Li et al., 2018), and returned activated or excess sludge (Carr et al., 2016; Talvitie et al., 2017a; Lares et al., 2018).

While the nature of primary and secondary treatment is mostly consistent across studies, there is an array of advanced treatment techniques. Studies comparing MPs in tertiary vs. secondary effluent found that different advanced treatment technologies can further decrease MPs before discharge (Michielssen et al., 2016; Mintenig et al. 2017; Talvitie et al., 2017a,b; Ziajahromi et al., 2017; Lares et al., 2018; Magni et al., 2019). Overall, MBR (Lares et al., 2018; Talvitie et al., 2017a) and advanced filtration technologies (Michielssen et al., 2016; Mintenig et al. 2017; Talvitie et al., 2017a,b; Ziajahromi et al., 2017; Magni et al., 2019) have been reported as effective means in reducing MPs from final effluent. Dissolved air flotation in Finland (Talvitie et al., 2017a) and reverse osmosis and decarbonation in Australia (Ziajahromi et al., 2017) also showed high performance. However, in other studies, advanced treatment by gravity sand filtration (Carr et al., 2016) and MBR (Leslie et al., 2017) did not promote further reduction in particle concentrations. These different findings in advanced WWTP studies support the need for further research on a range of treatment technologies to produce a representative assessment of their role in removing MPs from wastewater. This information could help identify control points within these systems, and what development or modification of operational procedures may decrease MPs discharge to the recipient waters.

Further research of WWTPs is crucial in MPs research because wastewater is a complex and heterogeneous matrix, and pollution levels and removal efficiencies appear to exhibit high inter- and intra-site variability (Mason et al., 2016). Especially, empirical data are needed for multiple stages other than final effluent and to explore factors driving spatio-temporal variabilities. Here, a study was conducted in a WWTP in the UK (Scotland) to: (1) understand the inflow and outflow loading of MPs (quantity and composition) in a tertiary treatment plant, accommodating temporal variability, and (2) assess the stepwise effect of treatment stage on the distribution and fate of MPs sized between 60 and 2800 μm. To our knowledge, this is the first study to evaluate MPs in advanced treatment systems in the UK by long-term (i.e. 10 months) spatial sampling in a single facility.

2. Materials and methods

2.1. Study site and sampling

The study site was a tertiary wastewater treatment plant in Scotland, UK, with 184,500 population equivalents (p.e.) and receiving a mix of trade and domestic sewage. The plant consists of preliminary treatment of wastewater by coarse screening (12 mm) and grit removal, primary settling tanks (phases 1 and 2), activated sludge treatment and clarification in final settling tanks (phases 1 and 2), and nitrification on plastic media trickling filters (Fig. 1), with final discharge of treated effluent into a freshwater river. Phases 1 and 2 were created due to an expansion of the treatment plant. This splits the stream into parallel channels for primary and secondary stages but there is no difference in treatment between the two.

Sampling was conducted five times between May 2017 and February 2018: 19 May 2017 (sampling event, SE1), 13 July 2017 (SE2), 20 October 2017 (SE3), 11 January 2018 (SE4), and 16 February 2018 (SE5). The flow range covered by the sampling events was 111,496 to 184,703 m^3 day^{-1}, representing low to medium flow (Qmean = 166,422 m^3 day^{-1}; Fig. S1). During each sampling event, a 5-L wastewater sample was collected from each of eight sample collection points (P): influent before screens (P1), preliminary effluent after coarse screening and grit removal (P2), primary effluent phase 1 (P3a) and phase 2 (P3b), secondary effluent phase 1 (P4a) and phase 2 (P4b), secondary effluent mixed liquor (P5), and final effluent after tertiary treatment (P6) (Fig. 1). Samples were collected in the morning, with two additional afternoon samples on the same day during SE5 from the influent (P1, pm) and effluent (P6, pm), to explore daily fluctuations. A bulk sample, taken by lowering a metal bucket into the stream, was filtered through a 2.8 mm metal sieve, and collected in plastic bottles for transport to the laboratory. Bottles were kept in black plastic bags at 3 °C until processing within a maximum of 8 weeks after collection.

2.2. MP extraction

The methodology for extraction and characterisation is broadly adapted from wet peroxide oxidation (WPO) protocols (Nuelle et al., 2014). As sewage can contain pathogens, all samples were processed in a Category 2 biological safety cabinet (Cat 2 BSC) and room, which also helped minimise potential background contamination of samples. Samples were transferred to glass Erlenmeyer flasks and spiked with 50 standard polyethylene (PE) beads each (0.71–0.85 mm diameter, $\rho = 0.96\ g\ cm^{-3}$; Cospheric LLC, Santa Barbara, California), to determine recovery rates. The spiked samples were treated with 30% hydrogen peroxide (H_2O_2; 1:1, v/v) for digestion of labile organics, heated in a water bath to 75 °C for 30 min to accelerate the reaction, stirred using a magnetic stirrer for 10 min, and digested at room temperature for three days. After the digestion period, samples were treated with UV light for 30 min to ensure they were sufficiently sterile to be removed from the Cat 2 BSC room for filtration under vacuum through Whatman 1.2-μm glass fibre filters (47 mm diameter). This processing stage was very time-consuming, indeed samples still contained some level of suspended solids and therefore filtration of 5-L samples was slow and required several filters. It was the step that limited the volume of samples that could be processed between sampling events. However, the entire sample was processed and filtered in this fashion to minimise the potential loss of smaller MPs by on-site filtration.

2.3. MP characterisation

Particle characterisation followed a two-step process starting with visual sorting of suspected MPs into four categories based on morphology: pellets, fibres, fragments, and films. Each entire filter area was examined using a Leica MX_{75} stereo microscope with magnification between 10× and 32× to identify and quantify particles of size range between 60 and 2800 μm (Blair et al., 2019).

A subsample of 70 pieces, equivalent to 5% of total particles identified during visual inspection, was selected for chemical confirmation of plastics by Fourier-transform infrared-attenuated total reflectance spectroscopy (FTIR-ATR), using a Shimadzu IRAffinity-1S FTIR with diamond crystal and 20 scans. Manipulation of small particles was difficult, thus chemical analysis was only possible for fibres (n = 19), fragments (n = 10) and films (n = 41) larger than 300 μm. Pellets could not be analysed as they were lost

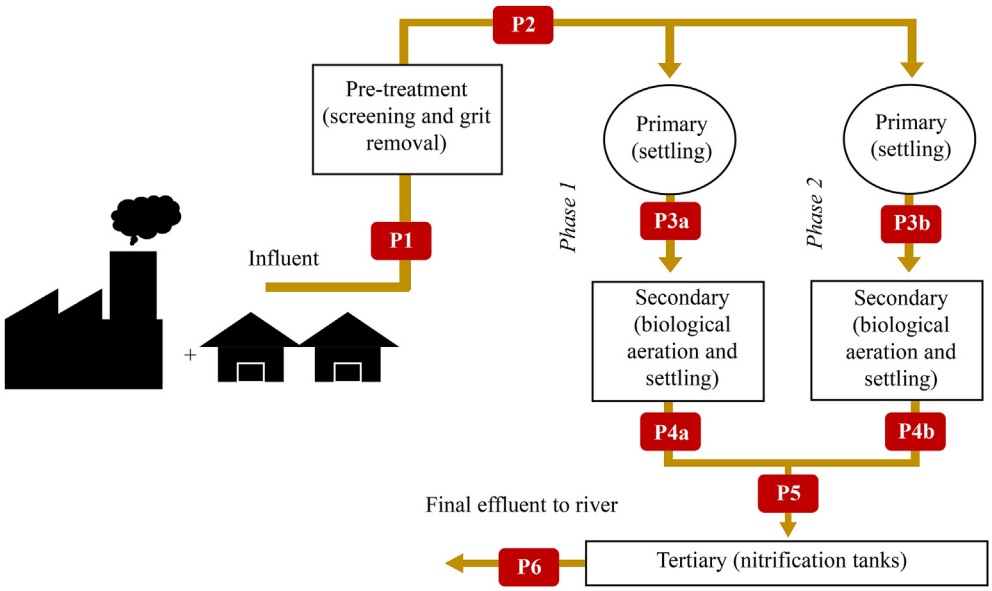

Fig. 1. Generalised diagram of the tertiary sewage treatment process in the selected study site, illustrating eight sample collection points (P1–P6).

during transfer due to their small sizes and smooth surfaces. Materials were identified by comparing the unknown spectra to those in the Shimadzu LabSolutions IR libraries, which contain approximately 12,000 reference spectra. For each particle, the top three automated matches were compared visually to assess closeness of match, and except for four pieces, the highest score was considered acceptable and reported (Table S3). The counts for confirmed plastics were used to estimate percentages for each category, subsequently extrapolated to correct all visual counts, including the 60–300 μm fraction. Further details of the FTIR-ATR characterisation process are in the Supplementary Material.

2.4. Quality control

A procedural blank was created for each SE by running 5 L of DI water through the same sample equipment used to collect samples, and processed the same way as wastewater. The purpose of the procedural blanks was to evaluate possible cross-contamination from generation of particles from plastic equipment used during sampling – these include plastic bottles, synthetic ropes, and a plastic funnel. Laboratory blanks were created in triplicates by placing 1 L of DI water in the same glass containers used for sample processing and leaving uncovered on lab benches during the extraction process, and filtering in parallel with each run of field samples. The purpose of the lab blanks was to capture cross-contamination from deposition of airborne particles in the general environment. Procedural and lab blanks, respectively, contained 4–14 and 0–3 coloured fibres by count (Supplementary Material), while no other type of particles were observed. It was not possible to analyse fibres in the blanks chemically, but their presence is considered evidence of cross-contamination from the environment and the use of synthetic sampling ropes.

Fragmentation tests using MP-spiked DI water were carried out to assess if the extraction process could generate secondary MPs at various stages. This is reported in the Supplementary Material. It was found fragmentation could occur, but the MPs used to assess this (microbeads) were rare in the samples, and so this understanding could not be used to refine MP estimates.

2.5. MP estimation

For each category, visual counts were corrected by subtracting the corresponding procedural blank. To ensure MPs were quantified correctly, blank-correct data were multiplied by the percentage of FTIR-confirmed plastics in each category. Such FTIR correction was employed for conservative estimates of daily discharge from a secondary WWTP in Vancouver, although blank correction was not incorporated in their calculation (Gies et al., 2018). The FTIR-corrected counts were summed to estimate total MP abundance (items L^{-1}), for each stage and each sampling campaign. Daily flow data for the WWTP were used to estimate incoming and outgoing MP loads in items day^{-1} and stage-wise removal efficiencies.

3. Results and discussion

3.1. Chemical confirmation of MPs

During visual characterisation, a total of 1308 items across all samples were considered potential MPs: 871 fibres, 191 fragments, 239 films, and 7 pellets (n = 7) (Fig. 2). Chemical characterisation confirmed that MPs were present and comprised 39% of the total pieces measured by FTIR-ATR (Fig. 3). Within each category of suspected MPs, plastics comprised 63%, 80%, and 17% of fibres, fragments, and films respectively. In absence of chemical confirmation and thus based on appearance, all micropellets (the lowest abundance of particle) recovered from wastewater samples were counted as primary MPs. Thus, based on FTIR-corrected data, a total of 749 MPs were observed across all wastewater samples, consisting of 549 fibres, 153 fragments, 41 films, and 7 pellets.

Different types of polymers identified (Fig. 3) included commonly-used plastics like polypropylene (PP, 23%) and PE (4%), and some less common, such as polyvinyl stearate (PVS, 7%) and polyoxymethylene (POM, 1%). The remaining MPs identified here were grouped as copolymers and included an ethylene-ethyl acrylate film and a PE-PP fragment. Polypropylene and PE are often reported in relatively high abundances across available surveys (Sun et al., 2019), as they are used in a wide number of applications including personal care and packaging products. The second-most detected polymer was PVS, a material not yet

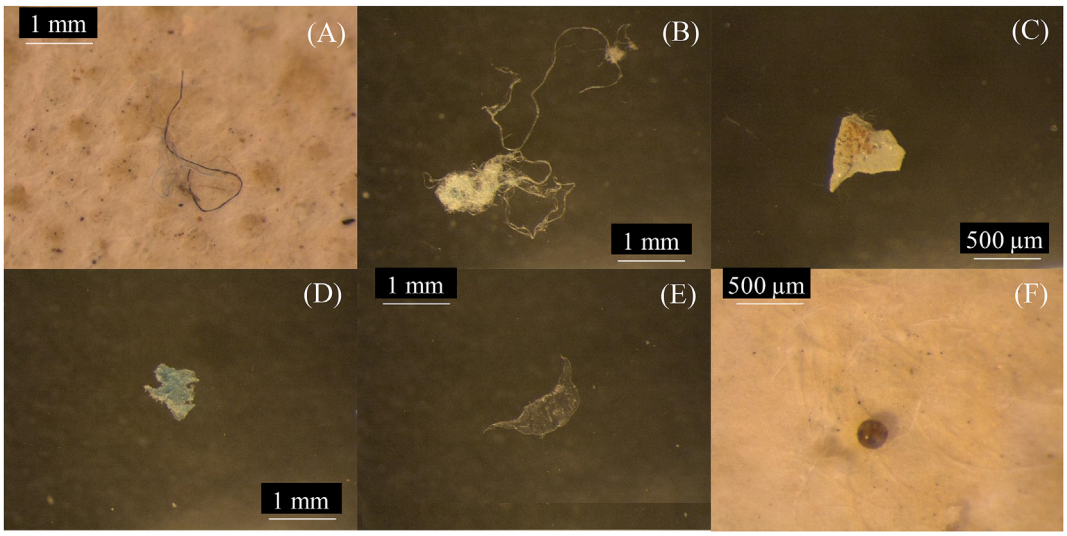

Fig. 2. . Examples of secondary and primary types of MPs extracted from wastewater samples and identified visually: fibres (A–B), fragments (C–D), film (E), and pellet (F).

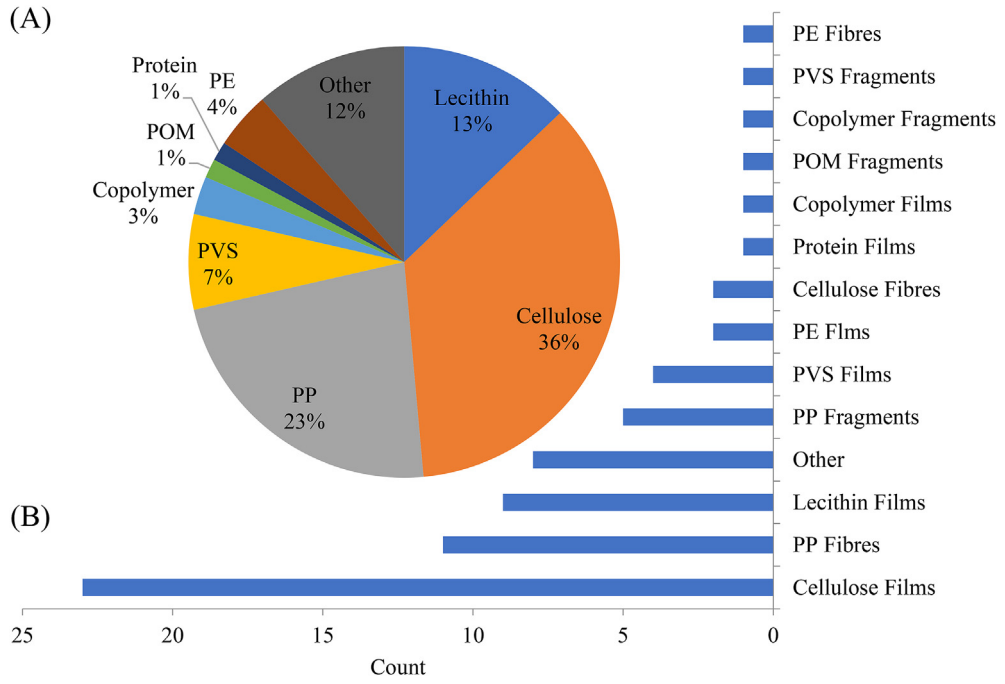

Fig. 3. (A) Pie chart showing the chemical distribution in percentages of different types of materials identified in a subsample of suspected secondary MPs (n = 70); (B) Bar graph showing the repartition by count for the chemical and categorical data combined.

reported in other studies to date, and of limited use in the plastics industry (Gooch, 2011). Polyvinyl stearate can be co-polymerised with polyvinyl chloride, PVC (Gooch, 2011) so may indicate construction applications. The POM particles also may not be common, only reported to date from a Danish secondary WWTP. The same study found PE-PP copolymers in raw and treated wastewater (Simon et al., 2018), but in higher abundance than this study.

Non-plastic materials were also present in the subsample (Fig. 3): cellulose (36%), lecithin (13%), and protein (1%). While these are not the focus of this paper, their presence should still be noted as depending on sample purification process, they may not be entirely removed from samples and thus mistaken as MPs. The remaining pieces classed as "Other" included 5 fibres, 2 fragments, and 1 film. These particles could not be identified as they showed no distinguishable peaks to allow for manual annotation or to produce any hits during the library search (Fig. S3).

3.2. MP morphology

Secondary MPs were predominant in the wastewater samples, comprising 99.5% of total pieces. Fibres were the most common type of MPs, followed by fragments and films. The predominance of fibres here is consistent with previous wastewater surveys (e.g. Sutton et al., 2016; Gies et al., 2018; Lares et al., 2018; Conley et al., 2019). Fibre abundance is expected to be higher in densely-populated areas as they can be carried by washing machine effluent. For example, clothes washing can release between 1.9×10^3 (Browne et al., 2011) and 6×10^6 fibres per wash (De Falco

et al., 2018). The highest releases have been observed from polyester (Pest) and polyamide (PA) garments, but these materials were not identified by FTIR-ATR here. This may be as Pest and PA fibres were settling out of suspension due to higher densities. Therefore, their concentrations in the liquid fractions would be lower than the detection limit allowed by a 5-L sample. Alternatively, they may have been smaller than 300 μm and thus were not subsampled for chemical identification. However, PP fibres may highlight the importance of other sources like sanitary products, thermal clothing, medical applications, and construction materials (Mandal, 2019), but the discussion on these alternative sources of fibres to WWTPs is limited in the literature. Fibre count was highly-variable across sampling events, and while generally decreased after each treatment stage (Fig. 4), some fibres persisted through the process and were observed in final effluent.

Fragments were present throughout all treatment stages and at least one particle was observed in final effluent (Fig. 4). Most fragment removal seemed to occur after the primary stage (when settling of solids takes place) and again after tertiary treatment. Films were mostly removed during pre-treatment, which may indicate they are more likely to be captured in the grit and grease biosolids as observed in a similar study in a Scottish secondary WWTP (Murphy et al., 2016). Different types of fragmented pieces have also been observed across multiple WWTPs (Sun et al., 2019) and generally refer to uneven or irregular pieces. As observed here, fragments were the second most-abundant MPs after fibres in a Swedish secondary WWTP (Magnusson and Norén, 2014), in secondary and tertiary WWTPs in the USA (Mason et al., 2016; Sutton et al., 2016), and in an Italian tertiary treatment plant (Magni et al., 2019). Here, fragmented pieces were categorised as either films or fragments to distinguish between two-dimensional thin particles and three-dimensional pieces with broken edges, respectively. However, the terms used to categorise these particles may vary across surveys (Hidalgo-Ruz et al., 2012), thus it is necessary to unify classifications for adequate consideration.

Fragments can be produced from a wide variety of sources and enter the wastewater stream via household and industrial effluent, but fragments generated during the treatment process cannot be excluded, supported by evidence of fragmentation of larger MPs beads (>700 μm) in controlled tests here. This needs to be validated for other particle types and sizes. Furthermore, the WWTP may have plastic equipment that if degrades over time could release MPs, but to our knowledge this has not been explored. The mechanical generation of MP fragments, particularly in sizes that may be evading detection, presents an important research gap in these systems that warrants further investigation as without it WWTP loading and MP redistribution cannot be fully understood.

Lastly, microbeads were only observed before secondary treatment (Fig. 4). This is consistent with previous observations in Swedish secondary WWTPs where 95–99% of microbeads were considered to settle out in sludge (Magnusson and Norén, 2014), and in the UK where microbeads were only found in grease fractions removed during pre-treatment (Murphy et al., 2016). These observations are for particles >65 μm. Therefore, entrapment in sludge may explain why these particles were only observed in the early treatment stages in this study also. Primary MPs (i.e. microbeads) can be introduced to WWTPs via household sewage, but primary MPs represent only a small portion of the plastic load in this catchment. This discussion is relevant to current considerations on MP control measures of MPs, especially as current actions such as regulatory bans are mainly aimed at reducing primary MPs inputs, and few focus on secondary sources.

3.3. MP abundances

Microplastics were present throughout the system. Concentrations ranged from ~1 to 13 MP L^{-1}, with highest abundances in pre-treatment effluent during SE1 (Fig. 5). Total concentrations of MPs were highly-variable across sampling dates and time, consistent with other reports of high variability (Sun et al., 2019). Influent concentrations were between 3 and 10 MP L^{-1}, with maximum abundances observed in January and minimum in February and July. In effluent, concentrations were between <1 and 3 MP L^{-1}. The lowest concentrations were mostly observed after tertiary treatment (final effluent), except during SE2, when concentrations reached their minimum after the mixed secondary liquor. Both influent and effluent abundances observed here are comparable to those in a secondary WWTP in Glasgow, Scotland (Murphy et al., 2016) but considerably lower than in three secondary WWTPs in South Carolina, USA (Conley et al., 2019). Nevertheless, current methods may not be suited to detect small MPs (e.g. <300 μm) so it is probable that MP concentrations are underestimated, especially as small MPs have been observed in greater abundances than larger pieces (Carr et al., 2016; Mintenig et al. 2017). Moreover, small MPs (e.g. 20–190 μm) may be more common in final effluent as they are more likely to pass through filtration barriers if not retained in

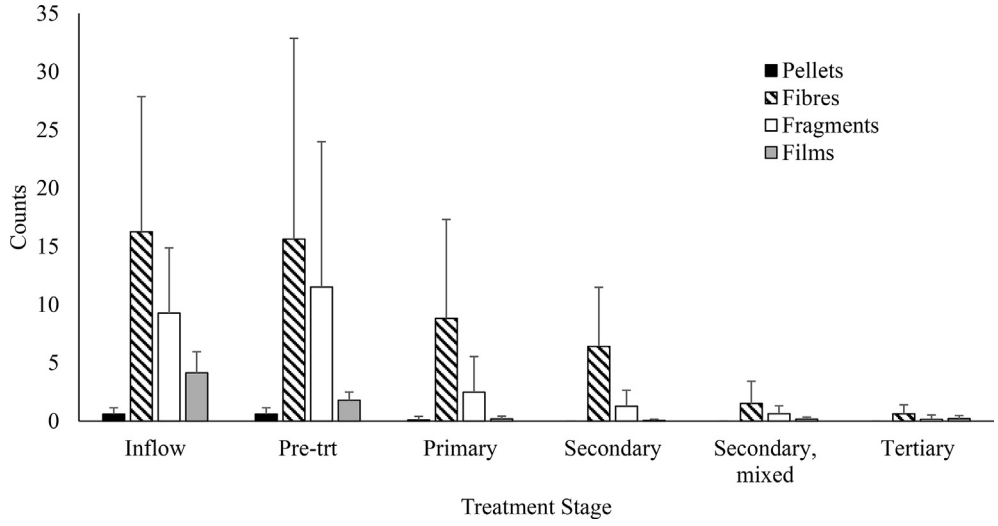

Fig. 4. Mean counts of MPs at different stages using FTIR- and blank-corrected data calculated averaging all sampling campaigns. Error bars represent standard deviation.

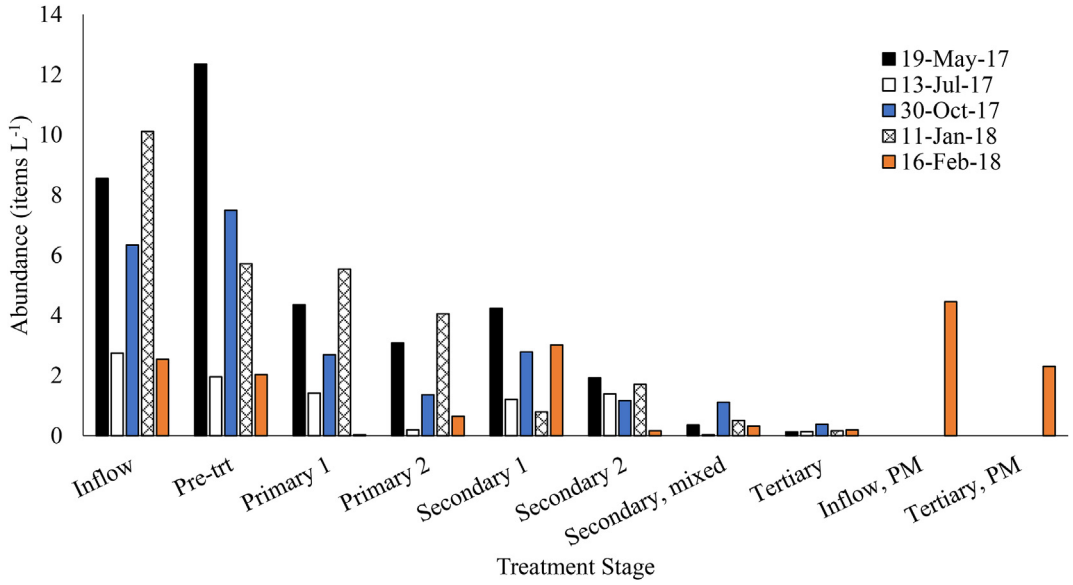

Fig. 5. FTIR-corrected MP abundances across all treatment stages and events in a tertiary sewage treatment plant.

biosolid fractions and smaller than the pore size (Ziajahromi et al., 2017; Sun et al., 2019).

Abundances were highly variable across sampling events and between the morning and afternoon samples collected on the same day, despite similar flow conditions. A survey of three USA WWTPs observed concentrations to vary by a factor of 2.5 and 4.8 in influent and effluent respectively, and long-term variations were greater than in short-term (Conley et al., 2019). However, the absence of replicates in the present study limited this assessment of short-term variation, and future work should explore this to support considerations of regulating inflow concentrations of different types of MPs to the system.

3.4. MP removal and loadings

Average MP inflow to the treatment plant over one year was 8.1×10^8, 95% CI [3.8×10^8, 1.2×10^9] particles day^{-1}. Influent loads based on incoming concentrations and plant flows are only reported by a few studies (Magnusson and Norén, 2014; Murphy et al., 2016; Lares et al., 2018; Conley et al., 2019), but their findings suggest these loads may be partially dependent on the size of population served. For example, among three WWTPs in South Carolina, a WWTP serving 1.8×10^5 p. e. received considerably higher MP loading than a treatment plant serving a smaller population (Conley et al., 2019). In an earlier survey in the same catchment of this study in Scotland, a larger secondary treatment plant serving 6.5×10^5 p. e. received an average daily load of 4×10^9 MPs >65 μm. Incoming loads in the present study were mostly comparable to those of a Finnish secondary treatment plant (p.e. not specified) in Finland with a reported daily inflow of 6.2×10^8 MPs >0.25 μm (Lares et al., 2018).

Particles concentration decreased between influent and final outflow with each treatment stage removing different proportions of MPs (Fig. 6). Mean concentrations decreased by 6% (standard error 16) after pre-treatment. Preliminary treatment has only been assessed by two studies, and removal efficiencies in this research are lower than those reported, ~35–58% (Michielssen et al., 2016; Murphy et al., 2016). Primary treatment removed between 60 (P3a, standard error 10) and 76% (P3b, standard error 6) of overall MP counts and is consistent with other surveys (63–81%, Dris et al., 2015; 84–88%, Michielssen et al., 2016; 78%, Murphy et al., 2016; 97.4–98.4%, Talvitie et al., 2017b; ~68%, Ziajahromi et al., 2017). There was indication of further removal after secondary treatment, but this was only evident at the secondary mixed liquor stage after the channels are joined back together (P5). As there is no remediation between P4 and P5 stages, this reduction suggests that engineering parameters and infrastructure may play a role in MP retention, especially if a large portion of removal is attributed to settling. After secondary treatment (P5), removal reached 92% (standard error 3), comparable to a Finnish secondary treatment plant where 7–20% of MPs were removed by activated sludge treatment (Talvitie et al., 2017b). A similar study in a larger UK secondary treatment plant had a retention efficiency of 98% and discharged 6.5×10^7 particles day^{-1} (Murphy et al., 2016). Although the data come from different WWTPs, both studies are located in the same catchment, serve a similar population demographic, and observed a similar profile of MPs. Therefore, the differences between the two plants emphasise that removal of MPs will depend on site-specific engineering parameters besides loading and general treatment process.

Tertiary treatment produced an average 4% (standard error 1) decrease in MPs in secondary effluent, bringing the total retention efficiency to ~96% (Fig. 6). The plant discharges on average 2.2×10^7, 95% CI [1.2×10^7, 3.2×10^7] MPs day^{-1} under low-to medium-flow conditions. The removal ranges and discharges here are within those observed elsewhere (Table 1), noting cross study comparisons are difficult as different sampling volumes and size ranges can introduce uncertainty to MP measurements reported across sites. No other sites of the same type of treatment considered here (i.e. use of plastic media in nitrifying trickling filters) have been documented, but removal percentages in this WWTP were higher than those observed by advanced sand filters (Magni et al., 2019) and lower than MBR (Michielssen et al., 2016; Talvitie et al., 2017b). The differences among these treatment technologies may be expected because of differences in the porosity of the filters they use, and so may indicate a way in which performance of tertiary treatment may be predicted. Nevertheless, the diversity of advanced systems and the contrasting results reported for different facilities, mean more research in WWTPs is needed to help identify which technologies optimise removal of MPs pollution in and from

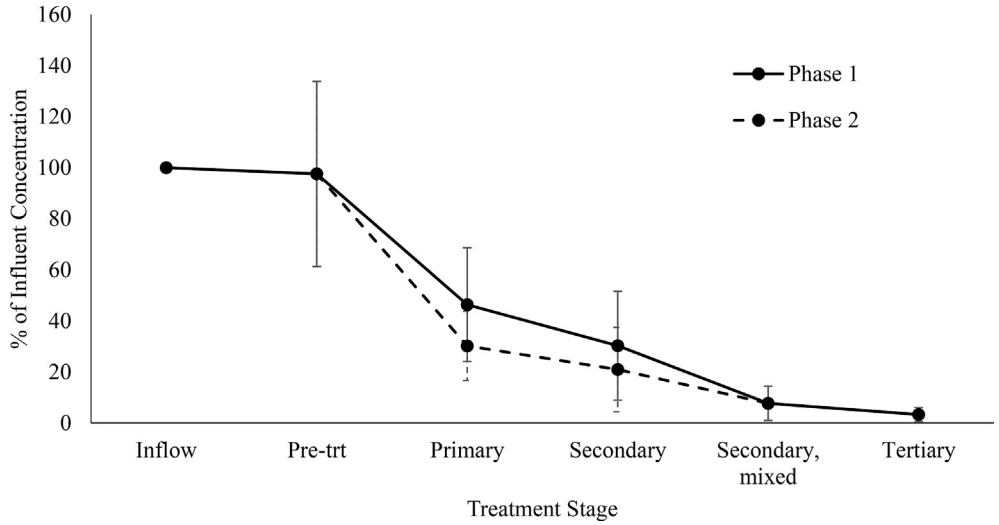

Fig. 6. Percent change relative to influent microplastic concentrations after each treatment stage, averaged across five sampling campaigns. Concentrations are FTIR- and field blank-corrected, then averaged across the five sampling events.

these systems.

4. Conclusions

Here, the occurrence, distribution, and fate of MPs in an advanced WWTP were assessed. A continuous input of MPs and other microdebris to the treatment site was observed over the course of ten months. The presence of MPs was confirmed by FTIR-ATR analysis, with PP identified as the most abundant type and present as fibres and fragments. Microplastics were mainly observed as secondary types, and while a few pellets were present, their chemical composition could not be determined due to size limitations of the FTIR-ATR approach employed here. Fibres were dominant. Their high abundance is expected as they are often associated with washing machine effluent, but their presence in blanks suggests that some may be entering the system via atmospheric, possible as the wastewater is treated in open channels. The system investigated here had apparent removal efficiencies at the higher end of that observed elsewhere, but MPs were not entirely removed and at least 1.2×10^7 particles may be discharged daily from this site even during low flow. These estimates are limited to particles sized 60–2800 μm but there will be smaller MPs in the system that need to be investigated further. As observed by other studies, the largest concentration reduction was observed in early treatment stages. Generally, this is linked to retention of microplastics in the sludge and so the concentration and fate of MPs in sludge needs further attention because rather than providing a solution, it may be displacing delivery of MPs to the environment. This research generates new understanding of MPs in WWTPs by its consideration of multiple stages, including tertiary treatment, not yet considered elsewhere and by employing a longer sampling period in a single facility to generate spatio-temporal understanding. Further research could use larger sample volumes to reduce the blank sensitivity and incorporate greater sampling frequency to assess short-term variation and thus contextualise seasonal observations. As wastewater treatment plants are expected to play an increasingly important role in regulating the delivery of MPs coming from land-based sources, this and similar studies can help to inform regulators about what needs to be prioritised in monitoring programmes and where controls should be implemented, thus guiding fundamental action.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.watres.2019.114909.

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