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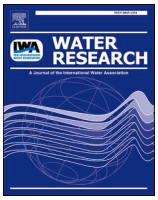

Treating wastewater for microplastics to a level on par with nearby marine waters

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ABSTRACT

Retention of microplastics (MPs) at the third largest wastewater treatment plant (WWTP) in Sweden was investigated. The plant is one of the most modern and advanced of its kind, with rapid sand filter for tertiary treatment in combination with mechanical, biological, and chemical treatment. It achieved a significantly high treatment efficiency, which brought the MP concentration in its discharge on par with concentrations measured in marine waters of the same region. This novel data shows that properly designed modern WWTPs can reduce the MP content of sewage down to background levels measured in the receiving aquatic environment. Opposite to current understanding of the retention of MP by WWTPs, a modern and well-designed WWTP does not have to be a significant point source for MP. MPs were quantified at all major treatment steps, including digester inlet and outlet sludge. MPs sized 10–500 μm were analyzed by a focal plane array based micro-Fourier transform infrared (FPA-μFTIR) microscopy, a hyperspectral imaging technique, while MPs above 500 μm were analyzed by Attenuated Total Reflectance-Fourier transform infrared (ATR-FTIR) spectroscopy. Mass was estimated from the hyperspectral images for MPs <500 μm and from microscope images >500 μm. The overall treatment efficiency was in terms of MP counts 99.98 %, with a daily input of 6.42×10^{10} and output of 1.04×10^{7} particles. The mass removal efficiency was 99.99 %. The mechanical part of the treatment, the pre-treatment, and primary stages, reduced both the MP counts and mass by approximately 71 %. The combined biological treatment, secondary settling, and final polishing with rapid sand filtration removed nearly all the remaining 29 %. MPs became successively smaller as they passed the different treatment steps. The digester inlet received 1.04×10^{11} MPs daily, while it discharged 9.96×10^{10} MPs, causing a small but not significant decrease in MP counts, with a corresponding MP mass reduction of 9.56 %.

1. Introduction

Plastic is a versatile material which enjoys widespread use. Compared to natural materials, plastics are durable, low cost, light weight, and rather strong (Ammala et al., 2011). Plastics take many sizes and shapes, and when manufactured in the size range 1–5000 μm, they are termed primary microplastics (MPs). Some of these are manufactured as additives to cosmetics, personal care products, industrial scrubbers, and cleansers, and some as plastic powders for molding (Talvitie et al., 2017a). MPs are also formed in the environment as breakdown products of larger plastic items (Thompson, 2005), these are termed secondary MPs (Andrady, 2011; Mason et al., 2016; Weinstein et al., 2016). The degradation usually occurs due to harsh mechanical, chemical, and biological processes (Mason et al., 2016; Singh and Sharma, 2008; Sussarellu et al., 2016). The continuous disintegration of larger items lead to accumulation of MPs, for example in marine sediments (Simon-Sánchez et al., 2022). Due to the ubiquity of plastics and their persistency, MPs are present in all environments, such as soil, sediment, water, and air (Duis and Coors, 2016; Gasperi et al., 2018; Simon-Sánchez et al., 2022). The list continues, and MPs have for example been reported in tap water and bottled water (Kirstein et al., 2021; Koelmans et al., 2019), and have been identified in marine

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organisms (Galloway, 2015).

Wastewater is no exception when it comes to MPs, and various sources releasing MPs into it, such as domestic discharge of textile fibers, cleansing beads from personal care products, industrial plastics, and stormwater runoff have been identified (Carr et al., 2016; Iyare et al., 2020; Long et al., 2019). However, wastewater is commonly treated before discharge, and the retention of MPs by wastewater treatment plants (WWTP) has been the focus of many a study (Gies et al., 2018; Lares et al., 2018; Talvitie et al., 2017a). Since the beginning of MP research, there has been significant concern from various stakeholders on the ability of WWTPs to retain MPs, leading to a significant body of research on this topic. Most studies treated WWTPs as black boxes, focusing on particle concentration in the inlet versus the outlet, largely ignoring the huge variation in treatment technologies applied by different plants. Depending on WWTP layout and operation, treatment efficiencies have often been found to be high, commonly at or above 95–99 % (Simon et al., 2018; Talvitie et al., 2017b). The retained MPs are either concentrated in the sludge (Michielssen et al., 2016; Xu et al., 2019) or skimmed off with the grit and grease (Chand et al., 2021; Rasmussen et al., 2021). The larger-sized MPs have been reported residing in grit, grease, and primary sludge, while smaller MPs seem to have a higher probability to reach the outlet (Lusher et al., 2017; Murphy et al., 2016; Ren et al., 2020). Sun et al. (2019) reported that 35–59 % of MPs were removed by pre-treatment and 50–98 % by primary treatment, while Iyare et al. (2020) found that tertiary treatment could further remove 5–20 %. Other studies showed that pre-treatment could remove even more. Ziajahromi et al. (2021) found for example a retention of 79 % by the screening and grit removal. Where the MPs go will depend on the WWTP layout as well as the physical characteristics of the MPs, such as size, shape, density, and chemical characteristics. Moreover, different polymers may follow different hydraulic transport patterns (Molazadeh et al., 2023a) which further might affect their fate in the primary and secondary settling tanks.

Most MP ends up in the sludge, which at larger WWTPs often is further treated in anaerobic digesters (Jiang et al., 2022). The MPs in sludge have been found rather resistant to mesophilic anaerobic digestion, with the exception of some possible fragmentation (Chand et al., 2021; Jiang et al., 2022). An effective degradation of MPs in digester inlet sludge might only be possible at extreme conditions, for instance, the high temperatures of hydrothermal liquefaction (Chand et al., 2022). Fragmentation of large-sized plastics (>5 mm) that can enter the digester via primary sludge and grease (Rasmussen et al., 2021), might also contribute to MPs in its outlet sludge. Some macroplastics and larger MPs probably disintegrated into smaller MPs during anaerobic digestion, which increases the MP numbers. At the same time, the polymer composition by MP counts might be altered, which could affect their fate in the digester (Askham et al., 2022). Where sludge is used as fertilizer, this might increase the risk of MP soil pollution (Iyare et al., 2020; Raju et al., 2018; Wei et al., 2022).

Despite their high efficiency towards MPs removal, WWTPs have been reported to impact the aquatic environment (Carr et al., 2016; Gies et al., 2018; Mintenig et al., 2017). This has been evidenced for example by elevated MP-levels in receiving waters downstream of WWTPs (Molazadeh et al., 2023b; Sönmez et al., 2023). Schmidt et al. (2020) for instance found 4–4.5 $\times$ 10^5 MP m^{-3} in the effluent of 79 German WWTPs. Similarly, Ren et al. (2020) reported a daily discharge of 8.7 $\times$ 10^8 MPs from a Chinese WWTP treating combined municipal sewage and stormwater. Many studies on MP retainment at WWTPs overlook that WWTPs differ widely in the technology they apply. Some are limited to simple screening for gross solids before discharge, others add a gravimetric settling step. More advanced WWTPs make use of biological and chemical treatment processes, ranging from simple treatment in high-loaded activated sludge reactors to complex biological nutrient removal treatment trains. A few added a polishing step of the so-treated water (Funck et al., 2021; Kılıç et al., 2023). They are all called WWTPs, however, their efficiency to retain MPs will depend on the details of the treatment train they apply.

The objective of the present work is to further develop the understanding of how individual treatment steps affect MP retainment within a WWTP, including its anaerobic digester. The study investigates the MPs in successive treatment stages and internal flows at Käppala WWTP in Lidingö, Sweden, as well as the fate of MPs in its mesophilic anaerobic digester.

2. Materials and methods

2.1. Description of the WWTP and sample collection

Käppala WWTP is one of the three largest wastewater treatment facilities in Sweden, receiving sewage from eleven municipalities in the greater Stockholm area. It serves a population over 525,000 and is considered one of the most effective plants in Sweden with quite low nutrients and organic pollutants discharge, partly due to its tertiary treatment based on rapid sand filtration. Approximately 50.5 million m^3 of sewage is being treated annually and the treated water discharges to the Baltic Sea. The sewage sludge (digester inlet) is being treated in two mesophilic anaerobic digesters. The produced sludge (digester outlet) is used as fertilizer on arable land.

Wastewater was collected flow proportionally over 24 h applying the samplers used for routine measurements. Seven consecutive samples were mixed into a weekly sample. Three such weekly samples were collected per sampling point and transported to Aalborg University for MP analysis. The samples were collected at: Influent (inlet before preliminary treatment); Rinse water (water from the washing of screen debris); Screening effluent (inlet after screening and before grit and fat removal); Primary effluent (wastewater after primary sedimentation); Secondary effluent (wastewater after secondary sedimentation); and Backwash water (from the tertiary sand filters). All wastewater samples were collected in aluminium bottles, stored at 4 °C, and trasported to the laboratory.

Three final effluent samples were collected by pumped filtration using a plastic-free filtration device (Universal Filtering Object, UFO) holding 167 mm diameter stainless steel filters. First the water passed a 300 μm pre-screen upon which it was distributed onto two 10 μm screens (Rasmussen et al., 2021). More than 1000 L treated water was filtered per sample. Upon filtration, all three filters were treated together as one sample. Sludge was collected from the inlet and the outlet of the digesters as grab samples. Approx. 1 kg wet weight of each type was collected and stored in metal buckets. The sampling was conducted over a time span of one year (September 2020 to October 2021). The operation of and load on the WWTP was unchanged during this period. An overview of the plant and sampling spots is presented in the graphical abstract.

The collected sample volume depended upon the expected MP content in the matrix, as the goal was to identify a sufficient number of MPs to quantify the content herein. The higher the expected concentration, the smaller the volume, and samples like influent wastewater were hence sampled at much lower volumes than final effluent. Of the collected samples, a subsample of 3 L of influent, screening effluent, primary effluent, and secondary effluent was taken into work to extract MPs, while 2–3 L of backwash water and 1 L of rinse water was taken into work. For sludge, the sample was homogenized by manual mixing, upon which 100 g wet weight of digester inlet sludge and 200 g wet weight of digester outlet sludge was taken into analysis.

2.2. MPs extraction

2.2.1. Removal of natural organic materials

All samples required thorough treatment to remove cellulose, proteins, and other organic materials prior to analysis. For this purpose, all samples except effluent water were first pre-oxidized by adding 50 % hydrogen peroxide (H_2O_2). The amount of H_2O_2 needed depended on

the organic content of the samples. Pre-oxidation took 72–120 h and was considered completed when no more bubbles were seen upon adding H_2O_2. The undigested particles were filtered on a stainless-steel filter of 10 μm mesh and Ø47 mm, upon which the particles were transferred to 250–500 mL of sodium dodecyl sulfate solution (SDS, 5% w/vol). For the effluent samples, the particles collected on the Ø167 mm filters were detached by ultrasonication and transferred into 500 mL of SDS, 5% w/vol solution where they were incubated for 48 h at 50 °C and constantly mixed. All further filtration was done on 10 μm stainless steel filters and particles were detached by ultrasonication.

All samples were then treated enzymatically in two steps. The particles were first transferred to 300 mL of tris buffer (pH 8.2) and 500 μL protease (Protease from Bacillus sp. ®, Sigma-Aldrich) added. The solution was incubated for 48 h at 50 °C. Hereafter the liquid was filtered, and the collected particles transferred into 300 mL of acetate buffer (pH 4.8) to which 500 μL of cellulase (Cellulase enzyme blend®, Sigma-Aldrich) and 500 μL cellulolytic enzyme mixture (Viscozyme®L, Sigma-Aldrich) were added and the solution incubated for 48 h at 50 °C. The liquid was filtered, and particles transferred to 200 mL of milli-Q water. A catalytic oxidation (Fenton) was carried out by adding 145 mL of 50 % H_2O_2, 65 mL of 0.1 M NaOH, and 62 mL of 0.1 M $FeSO_4$. The NaOH was needed to adjust the pH to 2.5–3.0 throughout the process. The temperature was maintained between 15 and 30 °C by keeping the samples in an ice bath.

After Fenton oxidation, the liquid was filtered on a 500 μm stainless steel sieve followed by a 10 μm stainless steel filter. The particles collected on the 500 μm sieve were dried at 50 °C for 7 days and each particle analyzed chemically to identify its material (Section 2.3).

2.2.2. Removal of inorganic materials

The particles on the 10 μm filter were transferred into 250 mL of sodium polytungstate (SPT) solution of 1.78 g cm^{-3} density. The liquid was transferred to a pear-shaped separating funnel, mixed by compressed air from the bottom for 30 min, and left to settle for 24 h. The settled heavy materials were discharged, and the top floating materials were collected and transferred into approximately 25 mL of 50 % ethanol. The ethanol was evaporated, and the particles collected in a 10 mL vial. A final volume of 5 mL of 50 % ethanol was added, making the extract ready for chemical analysis (Section 2.3).

2.3. FTIR (ATR and FPA-μ-FTIR) analysis

Particles >500 μm were manually sorted and imaged with a stereoscopic microscope (ZEISS, SteREO Discovery.V8) with an Axiocam 105 color camera and max. 8x magnification. The dimension of each particle was measured by the software ZenCore (Zen2Core SP1 from ZEISS) coupled to the stereoscopic microscope. The material of each particle was identified by a Cary 630 FTIR (Fourier transform infrared, Agilent Technologies) equipped with a diamond ATR (Attenuated Total Reflectance) accessory. The obtained spectrum was interpreted by the software OMNIC 8.2.0.387 (Thermo Fisher Scientific Inc., version 1) applying a commercial library.

Particles between 500 and 10 μm were analyzed by hyperspectral imaging applying FTIR microspectroscopy (μFTIR). After a thorough mixing of the sample concentrate, a subsample was taken with a disposable glass pipette and deposited on a 13 × 2 mm zinc selenide (ZnSe) window (Crystran, UK) and the window dried on a heating plate at 50 °C. This process was repeated till the window was sufficiently covered for the hyperspectral imaging. The window was analyzed in full by μFTIR using an Agilent Cary 620 FTIR microscope equipped with a 128 × 128-pixel FPA (Focal Plane Array) with Mercury Cadmium Telluride detector and coupled to an Agilent 670 IR spectroscope. Three separate windows were analyzed for each sample. Not all the 5 mL of concentrate could be deposited on the 3 windows. How much depended on the particle content in the concentrate. The full hyperspectral image consisted of 14 × 14 FPA-tiles covering the whole area of the Ø10 mm window. For further details see Chand et al. (2021) and Simon et al. (2018). MPs were identified and characterized by an automated approach using the software siMPle combined with a dedicated reference database (Liu et al., 2019; Primpke et al., 2020b). Besides information on particle number and polymer types, the analysis yielded shape parameters such as particle area, maximum and minimum Feret diameters, and particle mass estimation (Simon et al., 2018).

2.4. Contamination control and recovery test

Proper precautions to avoid contamination during sample preparation and analysis were undertaken as described in Chand et al. (2022a, 2021). Contamination was assessed by three laboratory blanks where 1 L of filtered Milli-Q was treated following the same treatment processes as for the samples. A field air blank was also collected during effluent sampling by keeping a clean glass Petri-dish open near the sampling location. The particles from the Petri-dish were transferred into a vial and concentrated into 5 mL HPLC ethanol 50 % and approx. one third of it was analyzed by μFTIR.

The recovery efficiency was assessed by spiking with known MP particles. Spherical polystyrene (PS) beads (microparticles GmbH, Berlin, Germany) of mean diameter 106 μm and a clearly distinguishable red color were used alongside fragments of polyvinyl chloride (PVC), polyethylene (PE), and polypropylene (PP). These fragments were produced by cry-milling common plastic objects, such as PVC from a PVC pipe, PE from a water bottle, and PP from a plastic sheet, and were obtained in the size range of 20 – 200 μm, 40 – 80 μm, and 20 – 200 μm, respectively. Three different sets of particles were made for the recovery, always mixing the spherical PS beads with one of the fragmented polymer types: i) PS and PVC; ii) PS and PE; and iii) PS and PP. The particles were counted by fluid imaging with a FlowCam 8400 (Yokogawa Fluid Imaging Technologies) at 4x magnification (

Y information Figure S1). The counted particles were then spiked into 250 mL of 5 % SDS. The FlowCam analysis also yielded the size of the added beads and fragments stated above. The spiked particles were then treated following the same sample treatment protocol as the effluent samples and the extracted spiked particles were again counted by the FlowCam. The treatment efficiency was calculated as the proportion of recovered particles to the total spiked particles (Table 2).

2.5. Data analysis

Hyperspectral images acquired by FPA-μFTIR were analyzed by siMPle, an automated software that compares the FTIR-spectrum of each pixel with a library tailored to MP analysis and containing more than 450 spectra (Primpke et al., 2020; Simon-Sánchez et al., 2022). The IR spectra from ATR-FTIR were analyzed by OMNIC (Thermo Fisher Scientific Inc., 8.2.0.387 version 1) for chemical identification. Fibers were distinguished from fragments by their length to width ratio being >3 (Vianello et al., 2019). A Shapiro-Wilk normality test was performed on the distribution of the determined MP major dimensions and estimated masses. Likewise, a pair-wise Wilcoxon rank sum test was conducted to test the significance of data among different sampling spots. In all cases, the significance level was set to 0.05. Data analysis and visualization was performed using R studio (4.2.2) and Microsoft Excel.

3. Results and discussion

3.1. Background contamination and recovery

Almost one third (32 %) of the concentrate from the three laboratory blanks was analyzed by scanning 3 windows of each blank, and 5 MPs were detected: 2 polyester, 1 PVC, 1 PP, and 1 PS. The total estimated mass was 692.5 ng. The contamination per processed blank sample hence was 5.2 MP per sample (721.4 ng per sample). Polyester contributed 40 % of the MP-counts while PS contributed the most mass

(57 %). Comparing the per sample blank concentration to that of the samples, the digester outlet sludge was relatively speaking the least affected by contamination (0.02 %), while the most affected was secondary effluent (7.52 %). The relative contamination measured as mass was lowest in rinse water (0.001 %), and highest in secondary effluent (29.17 %). For the field blank collected during effluent sampling, 2 MPs were found: 1 PVC and 1 polyester. The PVC particle was the largest and contributed most to the estimated mass (83 %). Further details are in supplementary information Table S1 and Table S2.

The data were not blank corrected as the level of contamination was low compared to the concentration in most samples. There are furthermore several fundamental issues when blank-correcting MP data. If, for example, a blank holds an MP of a material not found in the related sample, a blank correction is meaningless as negative concentrations cannot exist. A similar issue arises when assessing MP size distributions. Blank correction might also lead to meaningless results when mass is estimated, e.g., having a positive number of MPs but at the same time having a negative mass. Blank correction can hence bias the data instead of improving it. Many studies took the same approach for similar reasons, for example Lares et al. (2018), Liu et al. (2019), Ren et al. (2020), and Simon et al. (2018). Instead, these studies reported background contamination to qualify how trustworthy the data was.

MPs may be lost in the extraction process, and assessing recovery rates during extraction is an essential part of quality assurance. A common way to assess recovery is by spiking with a known amount of MPs and recovering them after extraction. Three sets of recovery tests were prepared with PS beads (106 μm) together with fragments (20–200 μm) of PVC, PE, and PP (Section 2.4), yielding an overall recovery above 68 % (Table 1). PP, PVC, and PE fragments were recovered above 70 % while PS beads were recovered at about 61 %. In comparision, Simon et al. (2018) recovered 77.7 ± 11.6 % PS beads of 100 μm and 57.6 ± 25.1 % HDPE fragments of 80–150 μm following a similar protocol of MP extraction. Long et al. (2019) obtained recovery rates of 99.0 % and 89.3 % of 1000 μm and 90 μm PS, respectively, where the extraction was done by wet oxidation without enzymatic treatment. Various other researchers have presented recovery results applying spiked material (Ruggero et al., 2020). However, it is difficult to be sure that the recovery of artificially spiked material will reflect the true recovery of MPs naturally present in complex environmental samples as some few polymer types and standard sized particles can never fully reflect the variety of MPs present in such samples. It is on the other hand we used four different polymer types with two different shapes and rather small sized MPs which makes this recovery test fairly representative compared to other examples.

Table 1
Composite samples over the course of week, and sample amount.

Sample Matrices	Daily collected sample volume	Total volume/mass collected per sample
Influent	0.5 – 1 L	5 L (flow proportionally collected weekly sample)
Screening effluent	0.5 – 1 L	5 L (flow proportionally collected weekly sample)
Rinse water (from rinsing screen debris)	1 L	5 L (combined spot sample)
Primary effluent	0.5 – 1 L	5 L (flow proportionally collected weekly sample)
Secondary effluent	0.5 – 1 L	5 L (flow proportionally collected weekly sample)
Backwash effluent (from sand filters)	0.5 – 1 L	5 L (combined grab sample/spot sample)
Effluent	–	1042.7 L, 1269.9 L, and 1274 L (spot sample via collecting particles on filter)
Digester inlet sludge	–	1 kg (grab sample/spot sample)
Digester outlet sludge	–	1 kg (grab sample/spot sample)

Table 2
Recovery of MP particles.

Recovery sample sets	Spiked particles		Recovered particles			Overall average recovery
i) PS and PVC	PS = 1188	PVC = 1234	PS = 741 (62.37 %)	PS_{avg} = 61.45 ± 0.91 %	PVC = 922 (74.71 %)	68.16 ± 7.29 %
ii) PS and PE	PS = 450	PE = 397	PS = 271 (60.22 %)		PE = 278 (70.02 %)	
iii) PS and PP	PS = 364	PP = 213	PS = 225 (61.81 %)		PP = 170 (79.81 %)	

3.2. MP in the waters of the WWTP

A total of 3315 MPs were identified in the water samples (raw wastewater and various effluents) based on scanning on average 29 % of the 5 mL of microplastic concentrate from the sample preparation (supplementary information Table S8). Here of, 822 were in samples from the sand filter backwash, 766 in rinse water, 540 in influent, 513 in screening effluent, 375 in primary effluent, 170 in final effluent, and 129 in secondary effluent (Table S3 gives further details). The highest MP counts, and mass concentration were in the rinse water, while effluent held the lowest MP concentration (Table 3). For details see supplementary information Table S4 and Table S5.

Table 3
MP particle and mass concentration in various sample matrices. Sd stands for standard deviation.

Samples	Particle conc. [counts L^{-1} ±Sd]		Mass conc. [ng L^{-1} ±Sd]	
	MP < 500 μm	MP > 500μm	MP < 500 μm	MP > 500μm
Influent	$4.52 \times 10^2 \pm 2.24 \times 10^2$	$3.07 \times 10^{\circ} \pm 2.95 \times 10^{\circ}$	$4.00 \times 10^4 \pm 2.28 \times 10^4$	$2.37 \times 10^4 \pm 3.15 \times 10^4$
Screening effluent	$3.91 \times 10^2 \pm 1.08 \times 10^2$	$9.56 \times 10^{\circ} \pm 9.66 \times 10^{\circ}$	$4.08 \times 10^4 \pm 2.50 \times 10^3$	$2.21 \times 10^5 \pm 1.08 \times 10^5$
Rinse water (from screened debris)	$5.78 \times 10^3 \pm 2.33 \times 10^3$	$6.23 \times 10^1 \pm 7.02 \times 10^{\circ}$	$8.04 \times 10^5 \pm 4.41 \times 10^5$	$4.28 \times 10^7 \pm 3.76 \times 10^7$
Primary effluent	$1.14 \times 10^2 \pm 2.89 \times 10^1$	$8.64 \times 10^{-1} \pm 4.78 \times 10^{-1}$	$1.27 \times 10^4 \pm 7.40 \times 10^3$	$3.37 \times 10^3 \pm 1.36 \times 10^3$
Secondary effluent	$2.82 \times 10^1 \pm 5.22 \times 10^{\circ}$	$2.14 \times 10^{-1} \pm 3.70 \times 10^{-1}$	$1.39 \times 10^3 \pm 7.89 \times 10^2$	$2.86 \times 10^1 \pm 4.95 \times 10^1$
Back wash (from sand filters)	$5.84 \times 10^2 \pm 4.81 \times 10^2$	$5.05 \times 10^{\circ} \pm 7.48 \times 10^{\circ}$	$3.81 \times 10^4 \pm 3.24 \times 10^4$	$5.69 \times 10^4 \pm 4.95 \times 10^4$
Effluent	$8.64 \times 10^{-2} \pm 2.18 \times 10^{-2}$	Not detected	$3.80 \times 00 \pm 7.24 \times 10^{-1}$	Not detected

Samples	Particle conc. [counts (kg wet weight)$^{-1}$ ±Sd]		Mass conc. [ng (kg wet weight)$^{-1}$ ±Sd]	
	MP<500 μm	MP>500μm	MP<500 μm	MP>500μm
Digester inlet sludge	$1.10 \times 10^5 \pm 8.21 \times 10^3$	$5.67 \times 10^2 \pm 5.00 \times 10^2$	$7.32 \times 10^6 \pm 1.60 \times 10^6$	$3.66 \times 10^6 \pm 2.86 \times 10^6$
Digester outlet sludge	$1.16 \times 10^5 \pm 2.86 \times 10^4$	$5.11 \times 10^2 \pm 3.35 \times 10^2$	$8.61 \times 10^6 \pm 3.19 \times 10^6$	$2.28 \times 10^6 \pm 2.25 \times 10^6$

MP levels decreased continuously between influent, primary effluent, secondary effluent, and final effluent (Table 3), with the caveat that the internal stream of rinse water held the highest concentrations of the water samples. The MPs likely were trapped on the bar screen (opening: 3 mm) and its filter cake. During washing of the screenings, the MPs smaller than the bar screen opening were detached and carried on with the rinse water. Thus, the input of rinse water before the grit chamber represents an internal load which increased the MP concentration in its inflow. The rinse water MPs were larger than those of other sampled matrices (Fig. 3).

Comparing to other studies can be challenging as wastewater treatment technologies differ, as do catchment characteristics (Mason et al., 2016). Further complicating comparison is that studies have used different sampling approaches, sample preparation and MP isolation protocols, and analytical instruments (Y. Y. Liu et al., 2023). They have consequently achieved different size quantification limits (Liu et al., 2021; Simon et al., 2018; Talvitie et al., 2017a). Reported numbers on influent and effluent consequently vary substantially. For instance, Üstün et al. (2022) reported 135.3 ± 28 counts L^{-1} in influent and 8.5 ± 4.7 counts L^{-1} in effluent of a Turkish WWTP. Liu et al. (2021) reported 0.28–3.14 × 10^4 counts L^{-1} in the influent and 0.01–2.97 × 10^2 counts L^{-1} in the effluent of 38 WWTPs of 11 countries. Magni et al. (2019) reported effluent concentrations of 0.90 counts L^{-1}, Gies et al. (2018) found 0.53 counts L^{-1}, and Talvitie et al. (2017b) found 0.5–2.5 counts L^{-1}. Mason et al. (2016) reported an average of 0.05 counts L^{-1} in the effluent from WWTPs in the USA. Taking the last reference as an example, Mason et al. (2016) applied visual identification of MPs, and it is reasonable to assume that such analytical approach will yield lower concentrations than the μFTIR analysis of the present study.

Käppala WWTP achieved a high MP removal efficiency. It decreased an MP influent load of 6.42 × 10^{10} counts day^{-1} and 8.99 kg day^{-1} to an effluent load of 1.04 × 10^7 counts day^{-1} and 0.00046 kg day^{-1}, yielding an overall efficiency of 99.98 % and 99.99 % for respectively MP counts (Fig. 1A and Fig. 1B) and mass (Fig. 1C and Fig. 1D). The mechanical part of the treatment, the preliminary, and the primary stages, reduced most of MPs by approximately 71 %, which corresponds well with the 35–59 % and 57–98 % reported by respectively Michielssen et al. (2016) and Xu et al. (2021) for these stages. The efficiency of the combined biological treatment, secondary settling, and final polishing with sand filtration removed nearly all the remaining 29 % not retained by the previous stages. The individual treatment process all achieved good individual efficiencies, were especially that of the sand filtration was very high, namely 99 % (Fig. 2).

The overall removal efficiency was comparable to data reported for Ryaverket (MP count efficiency 99.25 % and mass efficiency 99.60 %), another Swedish WWTP, which also applies tertiary treatment (disc filters) (Rasmussen et al., 2021). It was also comparable to Viikinmaki WWTP in Finland, which uses biologically active filters as tertiary treatment (overall retention >99 %) (Talvitie et al., 2017b). Despite very high removal rates, a complete MP elimination cannot be achieved (Yaseen et al., 2022), and the daily discharge of MPs through WWTPs is a product of the treated water volume and the effluent concentration

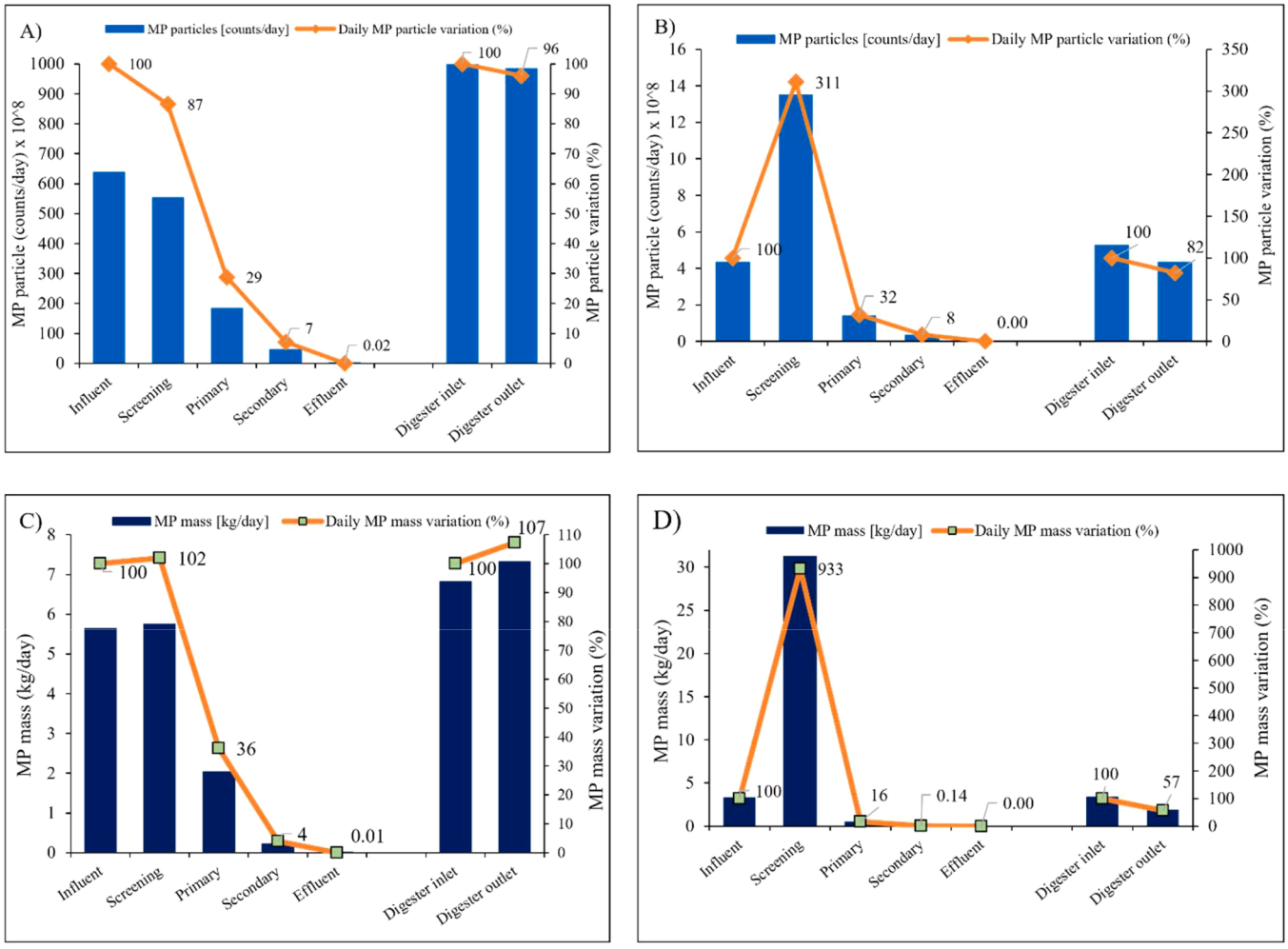
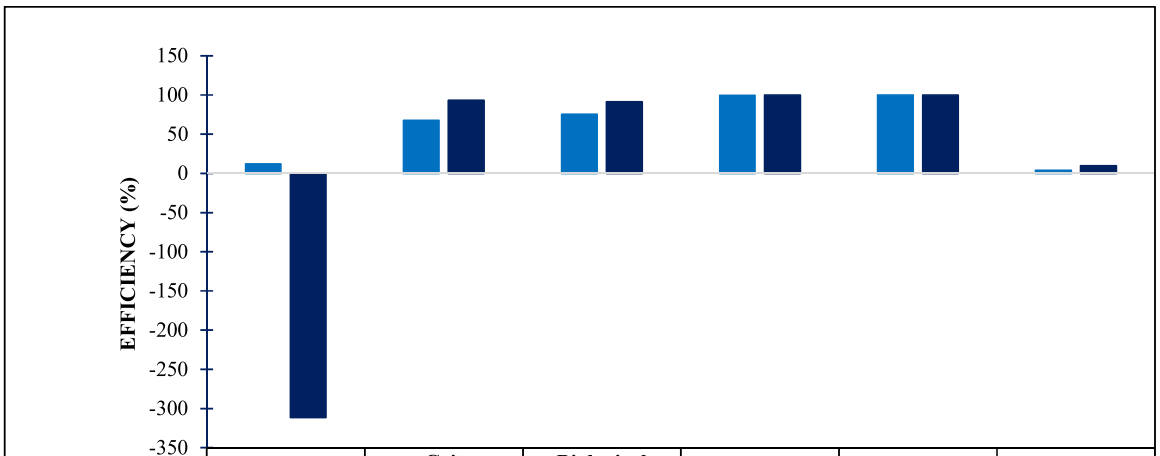


Fig. 1. Estimated daily variation of MPs in various treatment points based on flow rate of wastewater; A) particle variation for MPs < 500 μm, B) particle variation for MPs> 500 μm, C) mass variation for MPs < 500 μm, and D) mass variation for MPs > 500 μm.



	Pre-treatment	Grit chamber+Primary sedimentation	Biological treatment + Secondary sedimentation	Sand filtration	Overall	Daily variation in anaerobic digester
MP particle(%)	11.92	67.34	75.25	99.77	99.98	3.95
MP mass(%)	-311.43	93.02	91.14	99.80	99.99	9.56

Fig. 2. Overall MP removal efficiency at different treatment stages.

(Ziajahromi et al., 2017). In the present study, the effluent held very low concentrations (86.4 × 10–3 counts L^{-1}), but due to the large volume this still means that 1.04 × 107 counts day^{-1} were discharged to the nearby receiving waters. With the caveat that data are not really comparable, Magni et al. (2019) and Xu et al. (2019) found quite comparable numbers released into receiving waters, namely 1.6 × 108 and 3.63 × 108 MP counts day^{-1}, respectively.

Another way of evaluating the environmental impact of wastewater effluent is by comparing it to concentrations in the aquatic environment. Käppala effluent was on par with levels in the nearby marine waters between Sweden and Denmark where Y. Liu et al. (2023) and Gunaalan et al. (2023) found 17–28.6 × 10^{-3} counts L^{-1} and 11–87 × 10^{-3} counts L^{-1}, respectively. Both studies were done in the same laboratory as the present one and applied the same sampling tools, analytical protocols, and equipment. While the absolute numbers of an analytical pipeline always can be discussed, the fact that the same approach was used gives confidence in the comparability of concentrations between these studies.

3.3. Sludge digestion over mesophilic anaerobic digester

The MPs removed from the wastewater after the bar screens will largely be retained in the WWTP's sludge, which then undergoes anaerobic digestion. Here 3126 MPs were found of which 1007 were in digester inlet sludge and 2119 in digested sludge (supplementary information Table S3). The MP particle and mass concentrations in the inlet and outlet were rather similar (Table 3). The purpose of a digester is to convert sludge into methane and carbon dioxide gas, and a third or more of the sludge organic matter can be expected to be converted to gas (Appels et al., 2008; Zhang et al., 2018). To be able to compare inlet and outlet, the concentrations were hence stated per wet weight and not per dry weight of sludge. In comparison, Ryaverket WWTP, also in Sweden, showed a reduction of 29 % MP counts and 33.3 % mass (Chand et al., 2021).

While no statistically significant change in MP concentration over the digester was seen in the present study. A potential decrease could be caused by degradation of the polymers and by fragmentation below the size quantification limit of the analytical method, which was seen in earlier studies that have observed morphological changes on MP surfaces during anaerobic digestion (Akbay et al., 2022; Mahon et al., 2017). A study by Chand et al. (2021) reported a higher MP count concentration in digester outlet sludge, pointing in the same direction. However, detailed studies on anaerobic digesters are scarce, which makes it challenging to compare results. This is further complicated by different studies using different MP extraction protocols, analytical techniques, and data presentation. For example regarding the latter, Mahon et al. (2017) reported for seven Irish WWTPs an MP abundance in dry weight of sludge ranging from 4.2 × 10^3 to 1.5 × 10^5 counts kg^{-1} while Yuan et al. (2022) presented results from five Chinese WWTPs ranging from 0.02 × 10^3 to 5.81 × 10^3 counts kg^{-1} based on wet weight.

The amount of MP entering the treatment plant and the amount entering the digesters did not agree well, as about 38 % more MP by counts and 12 % more by mass entered the digesters than the treatment plant. Most likely this difference was caused by sampling uncertainty. Sludge samples were taken as grab samples during working hours, while wastewater was collected flow proportionally over 24 h. As MP concentrations vary over the day and between days, this means that the time window sampled was not the same. Furthermore, while the sludge retention time in the primary settlers is counted in hours, the sludge retention time in the biological processes is in the order of a month. Both these waste streams become mixed before the inlet to the digester, hence representing by themselves different time windows of influent. These different time scales of sources contributing to the digester inlet sludge and the WWTP inlet illustrates that comparison between MP load on the WWTP and the digesters must be treated carefully.

3.4. MP size and mass

Of the 6441 identified MPs from wastewater and effluents and sludge (supplementary information Table S3), 6326 had a major dimension <500 μm while 115 had a major dimension >500 μm. Of the latter, 44 were in rinse water, 18 in digester outlet, 9 in digester inlet, 17 in screening effluent, 15 in backwash, 6 in primary effluent, 5 in influent, 1 in secondary effluent and no large MPs were found in final effluent. Of the 6441 MPs, 1388 (22 %) had a length to width ratio >3 and could hence be characterized as fibers. The largest MP was 4775 μm and the smallest was 11 μm (corresponding to the lower size limit of detection). The median size of all particles was 56.9 μm with first and third quartiles of 37.6 and 95.6 μm, respectively (for detail of each sample type see supplementary information Figure S3). The corresponding MP mass of the identified particles varied by 9 orders of magnitude, between 0.14 ng and 1.11 × 10^7 ng with a median of 14 ng (first and third quartiles: 4 ng

and 49 ng, respectively. For the detail of each sample type see supplementary information Figure S4). A Shapiro-Wilk normality test showed that the MP major dimensions and estimated masses were non-normally distributed ($p < 2.2 \times 10^{-16}$).

The major dimension of the MPs from different treatment steps were compared by a pair-wise Wilcoxon rank sum test, showing a significant difference ($p < 0.05$) between the mean size of influent and screen effluent, between rinse water and primary effluent, and between influent and effluent water (Fig. 3A). The major dimensions of digester inlet and outlet sludge were also significantly different. The MP masses also showed a significant difference between influent and rinse water and between screening effluent and primary effluent. Likewise, the particle mass between digester inlet and outlet sludge was significantly different (Fig. 3B). The analysis furthermore showed that the presence of a few large MPs influenced the mass variation much, for instance, less than 2 % large MPs (>500 μm) contributed more than 99.5 % of all mass.

Overall, particle sizes decreased through the treatment plant (Fig. 3), leading to a complete retainment of the very large particles > 500 μm. Size, shape, and mass of the MPs play a significant role in their removal. Especially fibers have gained much attention in microplastics research and is also one of the shapes more readily defined (F. F. Liu et al., 2023). Fibers have by several studies been reported to dominate in wastewater and retained to a higher degree in sewage sludge, while other studies could not confirm this (Harley-Nyang et al., 2023). In the current study, there were less fibers in the effluent from the rapid sand filter for tertiary treatment compared to the influent to the treatment plant (10 % in the

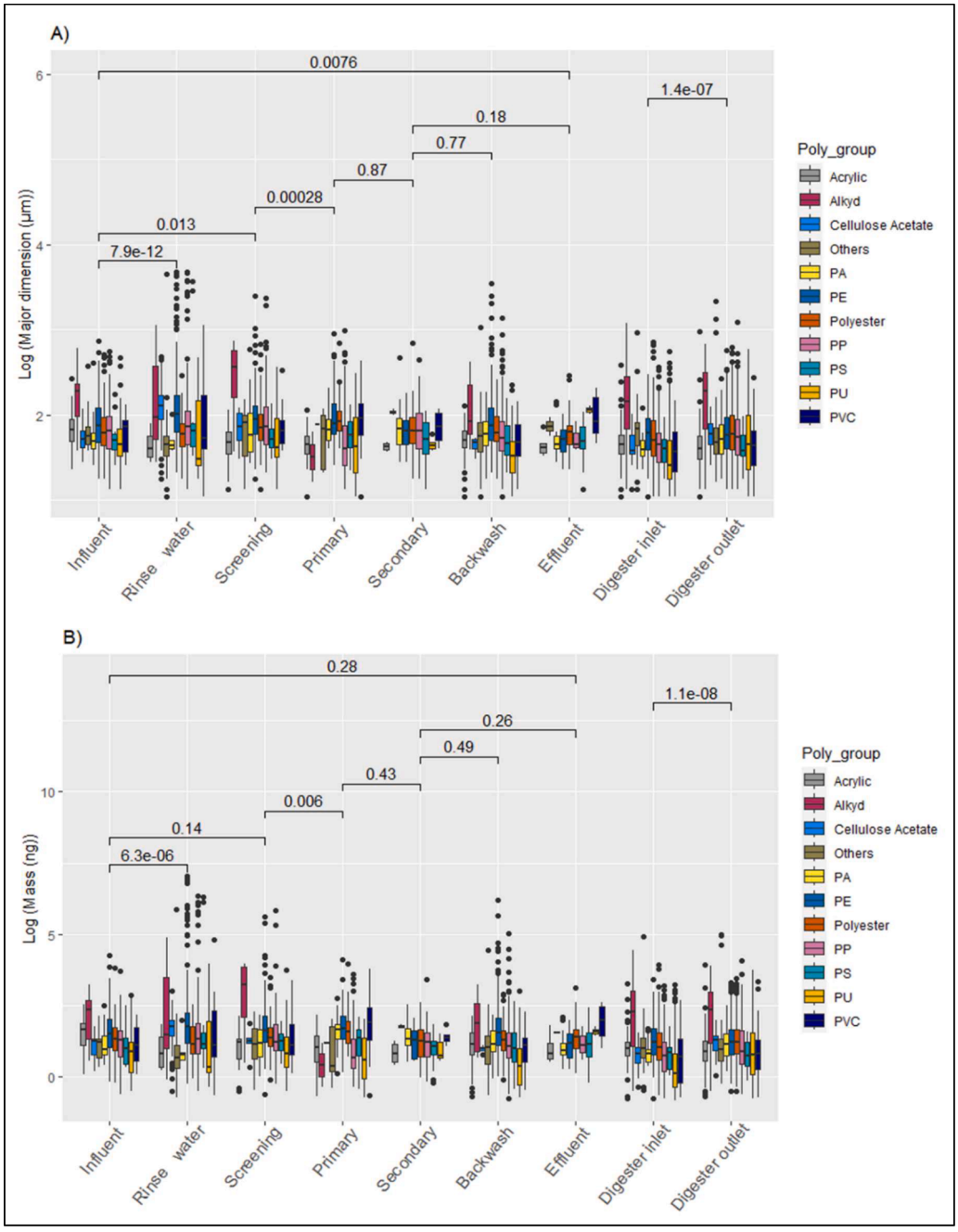


Fig. 3. Box plot for identified MP polymer types and the comparison (significance level of $P = 0.05$) between analyzed sample matrices at various treatment steps; A) major dimension of the identified particles, and B) estimated mass of the identified particles.'Others' indicates polymer types: aramid, acrylonitrile butadiene styrene (ABS), epoxy, polyvinyl acetate (PVaC), pan acrylic (PAN), poly(oxymethylene) (POM), vinyl copolymer, poly(ethyl acrylate), poly butylene terephthalate, poly (lauryl acrylate), and polyvinyl acetate (PVA).

outlet versus 20 % in the inlet) (supplementary information Table S7 and Figure S4). The inlet to the sand filters, on the other hand, held more fibers (26 %) than the inlet to the treatment plant. This implies that the tertiary treatment retained fibers better than less elongated MPs, while the other treatment step did not show such preference.

Large and buoyant particles of low density can remain at the surface and be removed by skimming in the primary treatment steps (Alavian Petroody et al., 2020; Bilgin et al., 2020). Larger and dense MPs can settle in the grit chamber and the primary sedimentation tank (Iyare et al., 2020; Ngo et al., 2019). Small particles, on the other hand, can pass the primary and secondary treatment. Talvitie et al. (2017a) found for example that approximately 80 % of the MPs in secondary effluent were 20–100 μm while 100–300 μm MPs were almost absent.

In the current study, 82 % of the MPs in the secondary effluent were smaller than 100 μm, which increased to 90.4 % in the final effluent. The tertiary treatment step hence not only reduced the MP concentration but also preferentially removed the larger particles which can also be seen in the data on the mass estimate (Table 3). Mintenig et al. (2017) similarly found few large MPs (>500 μm) in final effluent after the post-filtration, and Carr et al. (2016) showed that a tertiary treatment system with sand

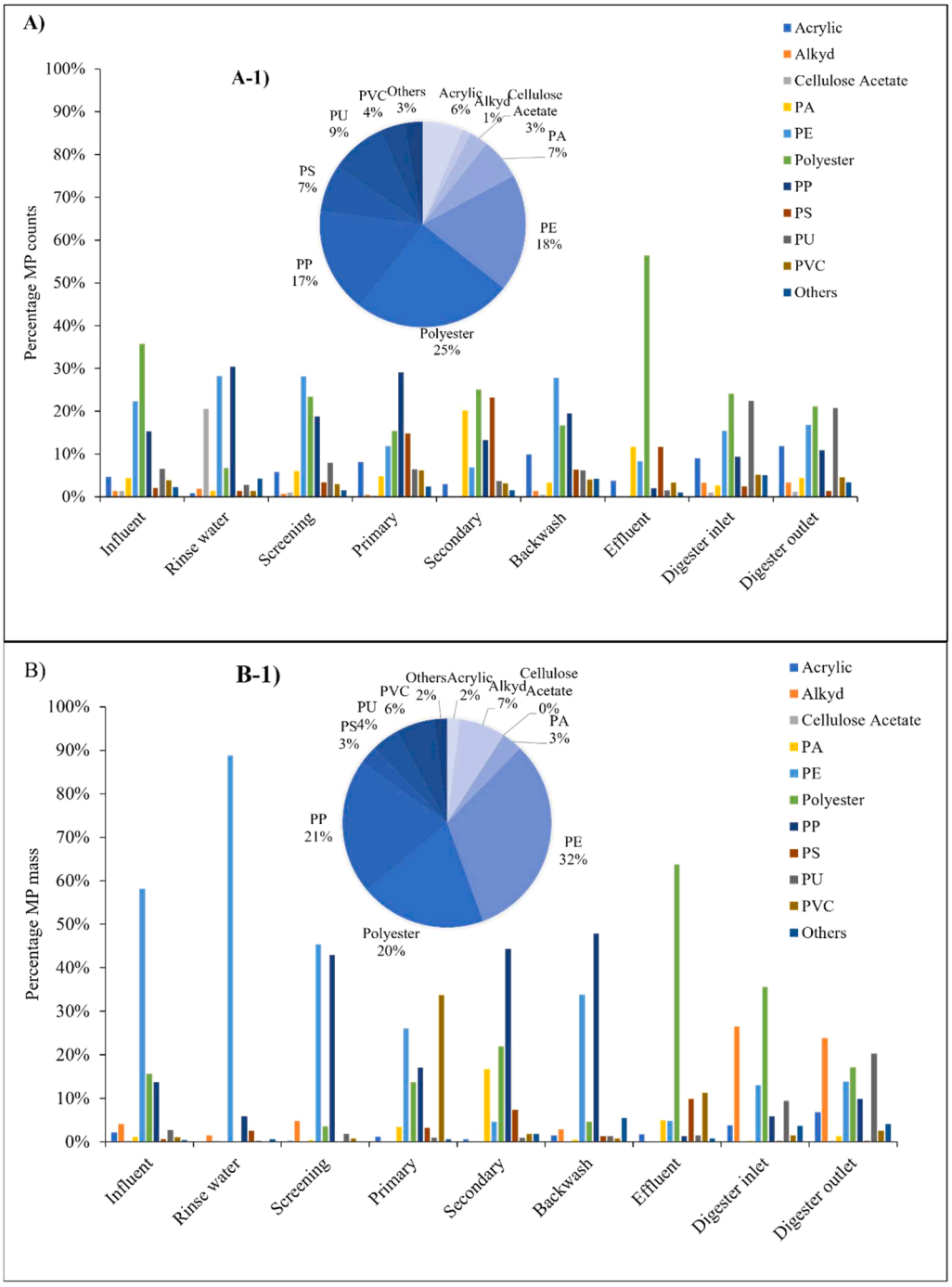


Fig. 4. Distribution of MPs among different polymer types as percentage of total; A) particle counts where A-A-1) shows the overall polymer distribution by particle counts, and B) particle mass where B-B-1) shows the overall mass distribution of the estimated mass from all sample matrices. 'Others' indicates polymer types: aramid, acrylonitrile butadiene styrene (ABS), epoxy, polyvinyl acetate (PVaC), pan acrylic (PAN), poly(oxymethylene) (POM), vinyl copolymer, poly(ethyl acrylate), poly butylene terephthalate, poly(lauryl acrylate), and polyvinyl acetate (PVA).

filtration removed most MPs >45 μm and that MPs <20 μm dominated the final effluent. Likewise, Lares et al. (2018) reported MPs between 0.5 and 1 mm as a major particle size in the final effluent. However, these studies address simpler treatment plants without tertiary polishing. This indicates that the treatment technique is important for which particle sizes are found in the effluent (Wu et al., 2021).

3.5. Polymeric composition

A total of 21 synthetic polymer types were identified (Fig. 4): acrylic, alkyd, aramid, acrylonitrile butadiene styrene (ABS), cellulose acetate (CA), epoxy, polyamide (PA), PE, PP, polyester (PEsT), PVC, polyvinyl acetate (PVaC), polyurethane (PU), PS, pan acrylic (PAN), poly(oxymethylene) (POM), vinyl copolymer, poly(ethyl acrylate), poly(butylene terephthalate), poly(lauryl acrylate), and polyvinyl acetate (PVA)). Polyester, PE, PP, PS, PU, PA, acrylic, alkyd, PVC were the major types of polymers, which have also commonly been found in wastewater treatment systems (Azizi et al., 2022; Sun et al., 2019). These polymers accounted for 80–95 % in the water samples. The rest of the polymers, grouped as "Others", contributed less than 1–5 %. The polymer composition was not systematically affected by the individual treatment steps, and neither was the size or mass of MP. Like for the water samples, there was little variation between digester inlet and outlet sludge. No systematic removal of specific polymer types could hence be identified, i.e., none of the treatment steps preferentially removed specific polymer types.

4. Conclusion

Käppala WWTP with its advanced treatment processes and tertiary polishing step by sand filtration technique was highly efficient in retaining MPs. It achieved an overall reduction of 99.98 % measured by MP counts and 99.99 % by MP mass, bringing the discharged water to a level on par with which was reported for marine environments in its vicinity. All steps contributed to the removal, with the final tertiary step, the sand filter, achieving percentually the highest efficiency. The MPs became continuously smaller as the water passed the mechanical treatment, the biological processes, and finally the sand filtration. The polymer composition, on the other hand, was not affected, indicating that none of the steps had a preference towards specific polymer types. Käppala WWTPs digesters similarly did not affect the polymer composition, that is, no polymer types were preferentially removed. Neither did it significantly affect the MP concentration measured as counts or mass. Overall, the study led to a novel realization, namely that a properly designed, advanced and modern WWTP can reduce MP down to background levels in the receiving aquatic environment. Such modern, advanced, and well-designed WWTP is hence not a significant point source for MP.

CRediT authorship contribution statement

Rupa Chand: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Writing – original draft, Writing – review & editing. **Lucian Iordachescu:** Writing – review & editing. **Frida Bäckbom:** Writing – review & editing. **Angelica Andreasson:** Writing – review & editing. **Cecilia Bertholds:** Writing – review & editing. **Emelie Pollack:** Writing – review & editing. **Marziye Molazadeh:** Writing – review & editing. **Claudia Lorenz:** Methodology, Writing – review & editing. **Asbjørn Haaning Nielsen:** Writing – review & editing. **Jes Vollertsen:** Funding acquisition, Supervision, Writing – review & editing.

Declaration of competing interest

The authors declare no competing interest.

Data availability

Data will be made available on request.

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.watres.2024.121647.

References

Akbay, H.E.G., Akarsu, C., Isik, Z., Belibagli, P., Dizge, N., 2022. Investigation of degradation potential of polyethylene microplastics in anaerobic digestion process using cosmetics industry wastewater. Biochem. Eng. J. 187, 108619 https://doi.org/10.1016/j.bej.2022.108619.

Alavian Petroody, S.S., Hashemi, S.H., van Gestel, C.A.M., 2020. Factors affecting microplastic retention and emission by a wastewater treatment plant on the southern coast of Caspian Sea. Chemosphere 261, 128179. https://doi.org/10.1016/j.chemosphere.2020.128179.

Ammala, A., Bateman, S., Dean, K., Petinakis, E., Sangwan, P., Wong, S., Yuan, Q., Yu, L., Patrick, C., Leong, K.H., 2011. An overview of degradable and biodegradable polyolefins. Prog. Polym. Sci. https://doi.org/10.1016/j.progpolymsci.2010.12.002.

Andrady, A.L., 2011. Microplastics in the marine environment. Mar. Pollut. Bull. https://doi.org/10.1016/j.marpolbul.2011.05.030.

Appels, L., Baeyens, J., Degrè Ve, J., Dewil, R., 2008. Principles and potential of the anaerobic digestion of waste-activated sludge ARTICLE IN PRESS. Prog. Energy Combust. Sci. 34, 755–781. https://doi.org/10.1016/j.pecs.2008.06.002.

Askham, C., Pauna, V.H., Boulay, A.-M., Fantke, P., Jolliet, O., Lavoie, J., Booth, A.M., Coutris, C., Verones, F., Weber, M., Vijver, M.G., Lusher, A., Hajjar, C., 2022. Generating environmental sampling and testing data for micro- and nanoplastics for use in life cycle impact assessment. Sci. Total Environ. 160038 https://doi.org/10.1016/J.SCITOTENV.2022.160038.

Azizi, N., Nasseri, S., Nodehi, R.N., Jaafarzadeh, N., Pirsaheb, M., 2022. Evaluation of conventional wastewater treatment plants efficiency to remove microplastics in terms of abundance, size, shape, and type: a systematic review and Meta-analysis. Mar. Pollut. Bull. 177, 113462 https://doi.org/10.1016/j.marpolbul.2022.113462.

Bilgin, M., Yurtsever, M., Karadagli, F., 2020. Microplastic removal by aerated grit chambers versus settling tanks of a municipal wastewater treatment plant. J. Water Process Eng. 38, 101604 https://doi.org/10.1016/j.jwpe.2020.101604.

Chand, R., Kohansal, K., Toor, S., Pedersen, T.H., Vollertsen, J., 2022. Microplastics degradation through hydrothermal liquefaction of wastewater treatment sludge. J. Clean. Prod. 130383 https://doi.org/10.1016/J.JCLEPRO.2022.130383.

Chand, R., Rasmussen, L.A., Tumlin, S., Vollertsen, J., 2021. The occurrence and fate of microplastics in a mesophilic anaerobic digester receiving sewage sludge, grease, and fatty slurries. Sci. Total Environ. 798 https://doi.org/10.1016/j.scitotenv.2021.149287.

Duis, K., Coors, A., 2016. Microplastics in the aquatic and terrestrial environment: sources (with a specific focus on personal care products), fate and effects. Environ. Sci. Eur. https://doi.org/10.1186/s12302-015-0069-y.

Funck, M., Al-Azzawi, M.M.S., Yildirim, A., Knoop, O., Schmidt, T.C., Drewes, J.E., Tuerk, J., 2021. Release of microplastic particles to the aquatic environment via wastewater treatment plants: the impact of sand filters as tertiary treatment. Chem. Eng. J. 426 https://doi.org/10.1016/j.cej.2021.130933.

Galloway, T.S., 2015. Micro- and nano-plastics and human health. Mar. Anthropog. Litter 343–366. https://doi.org/10.1007/978-3-319-16510-3_13/FIGURES/3.

Gasperi, J., Wright, S.L., Dris, R., Collard, F., Mandin, C., Guerrouache, M., Langlois, V., Kelly, F.J., Tassin, B., 2018. Microplastics in air: are we breathing it in? Curr. Opin. Environ. Sci. Heal. 1, 1–5. https://doi.org/10.1016/j.coesh.2017.10.002.

Gies, E.A., LeNoble, J.L., Noël, M., Etemadifar, A., Bishay, F., Hall, E.R., Ross, P.S., 2018. Retention of microplastics in a major secondary wastewater treatment plant in Vancouver, Canada. Mar. Pollut. Bull. 133, 553–561. https://doi.org/10.1016/j.marpolbul.2018.06.006.

Gunaalan, K., Almeda, R., Lorenz, C., Vianello, A., Iordachescu, L., Papacharalampos, K., Rohde Kiær, C.M., Vollertsen, J., Nielsen, T.G., 2023. bundance and distribution of microplastics in surface waters of the Kattegat/Skagerrak (Denmark). Environ. Pollut. 318, 120853 https://doi.org/10.1016/j.envpol.2022.120853.

Harley-Nyang, D., Memon, F.A., Baquero, A.O., Galloway, T., 2023. Variation in microplastic concentration, characteristics and distribution in sewage sludge & biosolids around the world. Sci. Total Environ. 891, 164068 https://doi.org/10.1016/j.scitotenv.2023.164068.

Iyare, P.U., Ouki, S.K., Bond, T., 2020. Microplastics removal in wastewater treatment plants: a critical review. Environ. Sci. Water Res. Technol. 6, 2664–2675. https://doi.org/10.1039/D0EW00397B.

Jiang, C., Ni, B.-J., Zheng, X., Lu, B., Chen, Z., Gao, Y., Zhang, Y., Zhang, S., Luo, G., 2022. The changes of microplastics' behavior in adsorption and anaerobic digestion of waste activated sludge induced by hydrothermal pretreatment. Water Res. 118744 https://doi.org/10.1016/J.WATRES.2022.118744.

Kirstein, I.V., Hensel, F., Gomiero, A., Iordachescu, L., Vianello, A., Wittgren, H.B., Vollertsen, J., 2021. Drinking plastics? – Quantification and qualification of microplastics in drinking water distribution systems by μFTIR and Py-GCMS. Water Res. 188, 116519 https://doi.org/10.1016/j.watres.2020.116519.

Kılıç, E., Yücel, N., Şahutoğlu, S.M., 2023. Microplastic composition, load and removal efficiency from wastewater treatment plants discharging into Orontes river. Int. J. Environ. Res. 17, 1–11. https://doi.org/10.1007/s41742-023-00514-0.
Koelmans, A.A., Mohamed Nor, N.H., Hermsen, E., Kooi, M., Mintenig, S.M., De France, J., 2019. Microplastics in freshwaters and drinking water: critical review and assessment of data quality. Water Res. https://doi.org/10.1016/j.watres.2019.02.054.
Lares, M., Ncibi, M.C., Sillanpää, Markus, Sillanpää, Mika, 2018. Occurrence, identification and removal of microplastic particles and fibers in conventional activated sludge process and advanced MBR technology. Water Res. 133, 236–246. https://doi.org/10.1016/j.watres.2018.01.049.
Liu, F., Olesen, K.B., Borregaard, A.R., Vollertsen, J., 2019. Microplastics in urban and highway stormwater retention ponds. Sci. Total Environ. https://doi.org/10.1016/j.scitotenv.2019.03.416.
Liu, F., Rasmussen, L.A., Klemmensen, N.D.R., Zhao, G., Nielsen, R., Vianello, A., Rist, S., Vollertsen, J., 2023a. Shapes of hyperspectral imaged microplastics. Environ. Sci. Technol. 57 (33), 12431–12441. https://pubs.acs.org/doi/10.1021/acs.est.3c03517.
Liu, W., Zhang, J., Liu, H., Guo, X., Zhang, X., Yao, X., Cao, Z., Zhang, T., 2021. A review of the removal of microplastics in global wastewater treatment plants: characteristics and mechanisms. Environ. Int. 146 https://doi.org/10.1016/J.ENVINT.2020.106277.
Liu, Y., Lorenz, C., Vianello, A., Syberg, K., Nielsen, A.H., Nielsen, T.G., Vollertsen, J., 2023b. Exploration of occurrence and sources of microplastics (>10μm) in Danish marine waters. Sci. Total Environ. 865 https://doi.org/10.1016/j.scitotenv.2022.161255.
Long, Z., Pan, Z., Wang, W., Ren, J., Yu, X., Lin, L., Lin, H., Chen, H., Jin, X., 2019. Microplastic abundance, characteristics, and removal in wastewater treatment plants in a coastal city of China. Water Res. 155, 255–265. https://doi.org/10.1016/j.watres.2019.02.028.
Liu, Y., Lorenz, C., Vollertsen, J., Prikler, B., 2023. Science of the Total Environment Does microplastic analysis method affect our understanding of microplastics in the environment ? 902. https://doi.org/10.1016/j.scitotenv.2023.166513.
Lusher, A.L., Hurley, R.R., Vogelsang, C., Nizzetto, L., Olsen, M., 2017. Mapping Microplastics in Sludge. Norweigan Institute for Water Research. https://doi.org/10.13140/RG.2.2.25277.56804.
Magni, S., Binelli, A., Pittura, L., Avio, C.G., Della Torre, C., Parenti, C.C., Gorbi, S., Regoli, F., 2019. The fate of microplastics in an italian wastewater treatment plant. Sci. Total Environ. 652, 602–610. https://doi.org/10.1016/j.scitotenv.2018.10.269.
Mahon, A.M., O'Connell, B., Healy, M.G., O'Connor, I., Officer, R., Nash, R., Morrison, L., 2017. Microplastics in sewage sludge: effects of treatment. Environ. Sci. Technol. 51, 810–818. https://doi.org/10.1021/acs.est.6b04048.
Mason, S.A., Garneau, D., Sutton, R., Chu, Y., Ehmann, K., Barnes, J., Fink, P., Papazissimos, D., Rogers, D.L., 2016. Microplastic pollution is widely detected in US municipal wastewater treatment plant effluent. Environ. Pollut. 218, 1045–1054. https://doi.org/10.1016/j.envpol.2016.08.056.
Michielssen, M.R., Michielssen, E.R., Ni, J., Duhaime, M.B., 2016. Fate of microplastics and other small anthropogenic litter (SAL) in wastewater treatment plants depends on unit processes employed. Environ. Sci. Water Res. Technol. 2, 1064–1073. https://doi.org/10.1039/c6ew00207b.
Mintenig, S.M., Int-Veen, I., Löder, M.G.J., Primpke, S., Gerdts, G., 2017. Identification of microplastic in effluents of waste water treatment plants using focal plane array-based micro-Fourier-transform infrared imaging. Water Res 108, 365–372. https://doi.org/10.1016/j.watres.2016.11.015.
Molazadeh, M., Liu, F., Simon-Sánchez, L., Vollersten, J., 2023a. Buoyant microplastics in freshwater sediments – How do they get there? Sci. Total Environ. 860, 160489 https://doi.org/10.1016/j.scitotenv.2022.160489.
Molazadeh, M.S., Liu, F., Lykkemark, J., Iordachescu, L., Nielsen, A.H., Vollertsen, J., 2023b. What is hiding below the surface – MPs including TWP in an urban lake. Environ Int 182, 108282. https://doi.org/10.1016/j.envint.2023.108282.
Murphy, F., Ewins, C., Carbonnier, F., Quinn, B., 2016. Wastewater Treatment Works (WwTW) as a source of microplastics in the aquatic environment. Environ. Sci. Technol. 50, 5800–5808. https://doi.org/10.1021/acs.est.5b05416.
Ngo, P.L., Pramanik, B.K., Shah, K., Roychand, R., 2019. Pathway, classification and removal efficiency of microplastics in wastewater treatment plants. Environ. Pollut. https://doi.org/10.1016/j.envpol.2019.113326.
Primpke, S., Cross, R.K., Mintenig, S.M., Simon, M., Vianello, A., Gerdts, G., Vollertsen, J., 2020. EXPRESS: toward the systematic identification of microplastics in the environment: evaluation of a new independent software tool (siMPle) for spectroscopic analysis. Appl. Spectrosc. 000370282091776. https://doi.org/10.1177/0003702820917760.
Raju, S., Carbery, M., Kuttykattil, A., Senathirajah, K., Subashchandrabose, S.R., Evans, G., Thavamani, P., 2018. Transport and fate of microplastics in wastewater treatment plants: implications to environmental health. Rev. Environ. Sci. Biotechnol. 17, 637–653. https://doi.org/10.1007/S11157-018-9480-3/FIGURES/2.
Rasmussen, L.A., Iordachescu, L., Tumlin, S., Vollertsen, J., 2021. A complete mass balance for plastics in a wastewater treatment plant - macroplastics contributes more than microplastics. Water Res 201, 117307. https://doi.org/10.1016/J.WATRES.2021.117307.
Ren, P.J., Dou, M., Wang, C., Li, G.Q., Jia, R., 2020. Abundance and removal characteristics of microplastics at a wastewater treatment plant in Zhengzhou. Environ. Sci. Pollut. Res. 27, 36295–36305. https://doi.org/10.1007/S11356-020-09611-5.
Ruggero, F., Gori, R., Lubello, C., 2020. Methodologies for Microplastics Recovery and Identification in Heterogeneous Solid Matrices: a Review 28, 739–748. https://doi.org/10.1007/s10924-019-01644-3.
Schmidt, C., Kumar, R., Yang, S., Büttner, O., 2020. Microplastic particle emission from wastewater treatment plant effluents into river networks in Germany: loads, spatial patterns of concentrations and potential toxicity. Sci. Total Environ. 737, 139544 https://doi.org/10.1016/J.SCITOTENV.2020.139544.
Simon-Sánchez, L., Grelaud, M., Lorenz, C., Garcia-Orellana, J., Vianello, A., Liu, F., Vollertsen, J., Ziveri, P., 2022. Can a sediment core reveal the plastic age? microplastic preservation in a coastal sedimentary record. Environ. Sci. Technol. https://doi.org/10.1021/acs.est.2c04264.
Simon, M., van Alst, N., Vollertsen, J., 2018. 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. 142, 1–9. https://doi.org/10.1016/j.watres.2018.05.019.
Singh, B., Sharma, N., 2008. Mechanistic implications of plastic degradation. Polym. Degrad. Stab. https://doi.org/10.1016/j.polymdegradstab.2007.11.008. Environ. Pollut. 318, 120922 https://doi.org/10.1016/j.envpol.2022.120922.
Sönmez, V.Z., Akarsu, C., Sivri, N., 2023. Impact of coastal wastewater treatment plants on microplastic pollution in surface seawater and ecological risk assessment.
Sun, J., Dai, X., Wang, Q., van Loosdrecht, M.C.M., Ni, B.J., 2019. Microplastics in wastewater treatment plants: detection, occurrence and removal. Water Res. https://doi.org/10.1016/j.watres.2018.12.050.
Sussarellu, R., Suquet, M., Thomas, Y., Lambert, C., Fabioux, C., Pernet, M.E.J., Goïc, N. Le, Quillien, V., Mingant, C., Epelboin, Y., Corporeau, C., Guyomarch, J., Robbens, J., Paul-Pont, I., Soudant, P., Huvet, A., 2016. Oyster reproduction is affected by exposure to polystyrene microplastics. Proc. Natl. Acad. Sci. USA 113, 2430–2435. https://doi.org/10.1073/pnas.1519019113.
Talvitie, J., Mikola, A., Koistinen, A., Setälä, O., 2017a. Solutions to microplastic pollution – Removal of microplastics from wastewater effluent with advanced wastewater treatment technologies. Water Res. 123, 401–407. https://doi.org/10.1016/j.watres.2017.07.005.
Talvitie, J., Mikola, A., Setälä, O., Heinonen, M., Koistinen, A., 2017b. How well is microlitter purified from wastewater? – A detailed study on the stepwise removal of microlitter in a tertiary level wastewater treatment plant. Water Res. 109, 164–172. https://doi.org/10.1016/j.watres.2016.11.046.
Thompson, R., 2005. New directions in plastic debris. Science (80-.) 310. https://doi.org/10.1126/science.310.5751.1117b, 1117b-1117b.
Üstün, G.E., Bozdaş, K., Can, T., 2022. Abundance and characteristics of microplastics in an urban wastewater treatment plant in Turkey. Environ. Pollut. 310 https://doi.org/10.1016/j.envpol.2022.119890.
Vianello, A., Jensen, R.L., Liu, L., Vollertsen, J., 2019. Simulating human exposure to indoor airborne microplastics using a Breathing Thermal Manikin. Sci Rep 9, 8670. https://doi.org/10.1038/s41598-019-45054-w.
Wei, F., Xu, C., Chen, C., Wang, Y., Lan, Y., Long, L., Xu, M., Wu, J., Shen, F., Zhang, Y., Xiao, Y., Yang, G., 2022. Distribution of microplastics in the sludge of wastewater treatment plants in Chengdu, China. Chemosphere 287, 132357. https://doi.org/10.1016/J.CHEMOSPHERE.2021.132357.
Weinstein, J.E., Crocker, B.K., Gray, A.D., 2016. From macroplastic to microplastic: degradation of high-density polyethylene, polypropylene, and polystyrene in a salt marsh habitat. Environ. Toxicol. Chem. 35, 1632–1640. https://doi.org/10.1002/etc.3432.
Wu, M., Tang, W., Wu, S., Liu, H., Yang, C., 2021. Fate and effects of microplastics in wastewater treatment processes. Sci. Total Environ. 757, 143902 https://doi.org/10.1016/j.scitotenv.2020.143902.
Xu, X., Jian, Y., Xue, Y., Hou, Q., Wang, L.P., 2019. Microplastics in the wastewater treatment plants (WWTPs): occurrence and removal. Chemosphere 235, 1089–1096. https://doi.org/10.1016/j.chemosphere.2019.06.197.
Xu, Z., Bai, X., Ye, Z., 2021. Removal and generation of microplastics in wastewater treatment plants: a review. J. Clean. Prod. 291, 125982 https://doi.org/10.1016/j.jclepro.2021.125982.
Yaseen, A., Assad, I., Sofi, M.S., Hashmi, M.Z., Bhat, S.U., 2022. A global review of microplastics in wastewater treatment plants: understanding their occurrence, fate and impact. Environ. Res. 212 https://doi.org/10.1016/J.ENVRES.2022.113258.
Yuan, F., Zhao, H., Sun, H., Sun, Y., Zhao, J., Xia, T., 2022. Investigation of microplastics in sludge from five wastewater treatment plants in Nanjing, China. J. Environ. Manage. 301, 113793 https://doi.org/10.1016/j.jenvman.2021.113793.
Zhang, Y., Zhang, L., Liu, He, Gong, L., Jiang, Q., Liu, Hongbo, Fu, B., 2018. Carbon dioxide sequestration and methane production promotion by wollastonite in sludge anaerobic digestion. Bioresource Technol. https://doi.org/10.1016/j.biortech.2018.10.004.
Ziajahromi, S., Neale, P.A., Rintoul, L., Leusch, F.D.L., 2017. Wastewater treatment plants as a pathway for microplastics: development of a new approach to sample wastewater-based microplastics. Water Res. 112, 93–99. https://doi.org/10.1016/J.WATRES.2017.01.042.
Ziajahromi, S., Neale, P.A., Silveira, I.T., Chua, A., Leusch, F.D.L., 2021. An audit of microplastic abundance throughout three Australian wastewater treatment plants. Chemosphere 263, 128294. https://doi.org/10.1016/j.chemosphere.2020.128294.