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Factors affecting microplastic retention and emission by a wastewater treatment plant on the southern coast of Caspian Sea

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HIGHLIGHTS

- Sari wastewater treatment plant removed 96.7% of microplastics from wastewater.
- Primary settling tank removed more than 70% of plastic fibers in all sizes.
- Primary settling and clarifier units equally contributed to removing microparticles.
- Clarifier unit eliminated particles <500 μm, primary settling removed larger ones.

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ABSTRACT

Understanding how wastewater treatment plants (WWTPs) process microplastics (MPs) will help informing management practices to reduce MP emissions to the environment. We show that composite 24 h samples taken at three replications from the outflow of the grit chamber, primary settling tank and clarifier of the WWTP of Sari City, on the southern coast of the Caspian Sea, contained 12667 ± 668 , 3514 ± 543 and 423 ± 44.9 MP/m³, respectively. Fibers accounted for 94.9%, 89.9% and 77.5% of the total number of MPs, respectively. The MP removal efficiency was 96.7%. MP shape (fiber, particle), size and structure were the most important factors determining their removal in different steps of the wastewater treatment process. The structure of microfibers (polyester, acrylic and nylon) and the consequent higher density than water explained their high removal (72.3%) in the primary settling tank. However, size was more important in microparticle removal with particles ≥ 500 μm being removed in the primary settling tank and <500 μm in the clarifier unit. The smallest particles (37–300 μm) showed the lowest removal efficiency. The predominant types of fibers and particles were polyester and polyethylene, respectively, which are likely to originate from the washing of synthetic textiles and from microbeads in toothpaste and cosmetics. Despite the efficiency of the Sari WWTP in removing MPs, it remains a major emission source of MPs to the Caspian Sea due to its high daily discharge load.

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1. Introduction

The presence of plastic objects under 5 mm in all dimensions in the environment, which are known as microplastics (MPs), has attracted increasing attention (Lambert and Wagner, 2016; Lusher et al., 2017; Mehdinia et al., 2020) since the first reports of plastics in the oceans (Colton and Knapp, 1974; Fowler, 1987). These small plastics debris are found in two different types in the

environment. Primary MPs are plastics produced in microscopic sizes that are used in the manufacture of facial cleansers and cosmetics, such as polyethylene microbeads in toothpaste, or for making larger plastic products (Lassen et al., 2015; Estahbanati and Fahrenfeld, 2016; Bayo et al., 2017). Secondary MPs are generated from degradation of larger plastic wastes released into the environment through chemical, physical or biological processes (Andrady, 2011; Hammer et al., 2012).

MPs are considered a serious environmental concern due to their potential physical and chemical damage. They not only may have direct effects through uptake by organisms but they may also adsorb and transfer pollutants to organisms and facilitate their entry into the food chain, ultimately affecting human health (Chua

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et al., 2014; Rochman et al., 2015; Talvitie et al., 2017a; Magni et al., 2019).

MP pollution has been detected in various aquatic and terrestrial environments (Michielssen et al., 2016; Rillig et al., 2017; Ziajahromi et al., 2017; Wang et al., 2017). Despite the widespread presence of MPs all over the world, identifying the sources that emit them to the environment still is one of the most challenging topics in science. Wastewater treatment plants (WWTPs) have been identified as a source of various types of MPs like particles (including fragments, films, pellets, granules, lines, foams and beads) and fibers. MPs are discharged into the sewage as microbeads used in cosmetics or synthetic fibers from washing clothes, and eventually enter the treatment plant (Liu et al., 2019; Magni et al., 2019). Recent studies investigating the fate and transport of MPs in WWTPs stated that the equipment used in the treatment plant does not completely remove plastic materials from wastewater (Leslie et al., 2017). Consequently, significant amounts of MPs will be emitted to the environment every day due to the vast volumes of effluent discharged by WWTPs (Carr et al., 2016; Michielssen et al., 2016; Mintenig et al., 2017; Talvitie et al., 2017a; Ziajahromi et al., 2017). This triggered researchers to examine the effectiveness of MP removal during various steps of the sewage treatment process in order to have a better perception of its role in reducing the emission of these types of pollutants.

Some studies have examined the removal efficiency of MPs throughout the entire WWTP treatment plant (e.g. Talvitie et al., 2015; Murphy et al., 2016; Lares et al., 2018; Liu et al., 2019; Magni et al., 2019). These studies however, lack information on the simultaneous effect of MP size, shape, and structure on their removal efficiency (Sundt et al., 2015; Murphy et al., 2016). Investigating these factors may provide a better understanding of the performance of various wastewater treatment processes in the separation and removal of MPs.

The aim of this study was to identify the factors determining MP removal efficiency at different treatment stages in the wastewater treatment process in a conventional activated sludge WWTP in the city of Sari, northern Iran on the southern coast of the largest lake in the world, the Caspian Sea. We investigated the efficacy of MP removal according to MP size (37–300, 300–500 and ≥ 500 μm), shape and structure for two general types (fiber and particle) of MPs at each treatment stage.

2. Material and methods

2.1. Description of the Sari WWTP

Sari's WWTP was selected as one of the largest facilities in operation on the southern coast of the Caspian Sea in northern Iran. The WWTP is located in the Mazandaran province and receives and treats the wastewater of the approximately 105,800 inhabitants of Sari city. The treated wastewater is discharged into the Tajan River, one of the most important rivers entering the Caspian Sea.

The WWTP is a conventional activated sludge plant; its main units are a bar screen (distance between the bars in the bar screen was 20 mm), an aerated grit chamber, a flow metering, a primary settling tank, an anoxic tank, an aeration basin, a clarifier, and a disinfection unit. In addition, the produced sludge is treated through sludge thickening, aerobic digestion and mechanical dewatering.

2.2. Sampling method and MP extraction

Twenty-four hour composite sampling, with intervals of 1 h, was carried out from the outflow of the grit chamber as the first available sampling point, primary settling tank, and clarifier unit

(final effluent) during a week in April 2018, in sunny conditions (Fig. 1). The amount of sample taken by the pump per hour was proportional to the wastewater flow announced by the treatment plant operator.

In order to increase the accuracy of the estimate of the number of MPs, three replicate samples were taken on three consecutive days with the same weather conditions. The final results are based on the average of three repetitive samplings. The sampling time interval between the treatment units was determined based on their mean hydraulic retention time. Sample volumes based on the amount of organic and inorganic matter at each stage were 30, 100 and 270 L from the outflows of the grit chamber, primary settling tank and clarifier unit, respectively. Each sample was passed over sieves with mesh sizes of 500, 300, and 37 μm (Damavand Sieve, ASTM-E11, mesh numbers 35, 50 and 400, respectively). Sieving was done on site.

The high organic load in the outflows of the grit chamber and the primary settling tank led to rapid clogging of the 37 μm sieve by organic and inorganic materials. To solve this problem, the sieve was washed several times with distilled water on the site and the washed matter was poured into a clean glass bottle. After passing the samples through the sieves, the sieves were washed with about 1 L of distilled water, and the water and the matter on the sieve were poured into clean glass bottles. To prevent algal and microbial growth, the samples were kept in the dark at 4 °C until they were transferred to the laboratory (Mintenig et al., 2017).

In the laboratory, the bottles were emptied into clean beakers, and the samples dried at 70 °C to reduce the volume to 100 ml. The beakers were placed on magnetic heater stirrers at 60 °C and hydrogen peroxide (H_2O_2) solution (30%) was added to digest the decomposable organic matter in the samples, such as algae and bacteria (Ziajahromi et al., 2017). After digesting the organic matter and evaporating hydrogen peroxide, 15 ml of sodium iodide (NaI) solution with a density of 1.7–1.75 g/ml were added to the dried sample for density separation of the MPs. The floated MPs were collected by centrifuging and following filtering the supernatant over a 37 μm screen (Nuelle et al., 2014; Rocha-Santos and Duarte, 2015; Carr et al., 2016; Ziajahromi et al., 2017).

2.3. Coloring method and MPs characterization

To avoid over-estimating the number of MPs, natural particles and fibers were stained by adding 5 ml of 0.2 mg/ml Bengal Rose solution to each screen (Liebezeit and Liebezeit, 2014; Ziajahromi et al., 2017). After 5 min at room temperature, the solution was washed off with ultrapure water. Then the samples were dried at 60 °C for 15 min and their morphological characteristics visually analyzed using a stereomicroscope (KERN, OZL-45). The pink stained particles and fibers, suspected to be natural matter, were removed from the samples after confirming their natural origin by Micro-Raman spectrometry.

In order to investigate any potential microplastic contamination caused by the sampling equipment and the laboratory environment, equipment and laboratory blanks were taken before and after sampling. No MPs were found in the blanks taken through all steps of processing the samples, indicating that our samples were not contaminated during the transportation and analyses. The thorough rinsing of the filtering device and all other equipment with pure water prior sampling apparently was efficient in minimizing microplastic contamination.

Using a stereomicroscope (KERN, OZL-45) all suspected MPs were extracted from each sample (except the pink fibers and particles), examined morphologically and then categorized and counted according to their shape (fiber, particle). Finally, a micro-Raman spectrometer at laser wavelengths of 785 nm and 633 nm

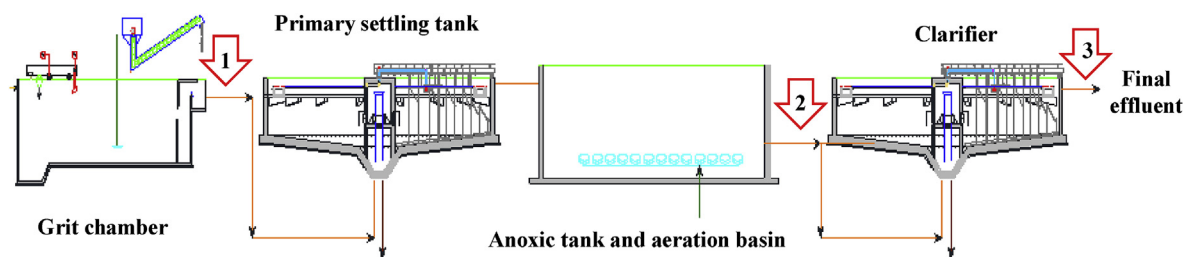


Fig. 1. Wastewater treatment process at Sari City. Sampling points are marked with arrows: after the Grit chamber (1), the primary settling tank (2), and the clarifier (final effluent) (3).

(Confocal Raman microscope, LabRAM HR Evolution—HORIBA) was used to determine the type of MPs. The instrument was controlled using the LabSpec6 software.

2.4. Statistical analysis

One-way ANOVA and Chi-Square Tests based on the normalized data were used to compare total numbers of released MPs among the different wastewater treatment stages. All statistical analyses were performed using the SPSS16 software.

3. Results

3.1. MPs in the wastewater water treatment plant

On average 12667, 3514 and 423 MP/m³ were found at the outflows of the grit chamber, primary settling tank and clarifier unit, respectively (Table 1). The MPs consisted of microfibers (MFi) and microparticles (MPa). MFi constituted 94.9%, 89.9% and 77.5% of the MPs at the outflows of the grit chamber, primary settling tank and clarifier unit, respectively. Numbers of total MPs (Chi-square: 7.26, $p < 0.05$), and specifically MFi and MPa (Chi-square: 7.2, $F: 43.2$, $p < 0.05$), differed significantly among stages. The total number of MFi decreased from 12022 MFi/m³ at the outflow of the grit chamber to 3157 MFi/m³ at primary settling tank and 328 MFi/m³ at the clarifier unit. There were 645, 357 and 95.1 MPa/m³, respectively (Table 1).

3.2. Size of MPs

At all stages in the wastewater treatment process, the dominant size of MFi and MPa was 37–300 μm , representing 63.0%, 73.6% and 62.7% of the MPs at the outflows of the grit chamber, primary settling tank and clarifier unit, respectively (Fig. 2).

3.3. Removal efficiency

The Sari WWTP, from the outflow of grit chamber to the final effluent, removed 96.7% of the MPs, with much of this removal being realized in the primary settling unit (72.3%) and the

remainder by the clarifier (24.4%) (Table 2). Removal was most efficient for the fibers (97.3%). MPa removal was slightly less efficient (85.2%) with equal contributions of the primary settling tank and the clarifier.

The bigger the fibers, the more efficient removal was in the primary settling tanks (Fig. 3). Removal of the particles in the settling tank however, was only efficient for the largest size classes; the clarifier unit removed the larger proportion of the smaller MPa. MP removal efficiencies in the WWTP were similar at 96.7%, 94.8% and 98.0% for the sizes of 37–300, 300–500 and ≥ 500 μm , respectively (Fig. 3).

3.4. Types of MPs

Micro-Raman analysis of MFi and MPa from the outflows of the grit chamber, primary settling tank, and clarifier unit indicated the presence of polyethylene terephthalate or polyester (PET/PES), acrylic, polyamide or nylon (PA), polyethylene (PE), and polypropylene (PP). The proportion of different polymers varied depending on the treatment unit.

Polyester was the most abundant type of fiber in the outflows from the grit chamber (57.0%), primary settling tank (70.0%) and clarifier unit (40.0%), followed by polyamide and acrylic fibers and a small amount of polypropylene were observed in the primary settling and clarifier.

Most of the particles were made of polyethylene, with 91.0%, 85.0% and 73.0% in the outflows from the grit chamber, primary settling tank and clarifier unit, respectively. A small amount of polypropylene MPa was observed in all phases and also some of the PET was seen in the clarifier (Fig. 4, Fig. SI-1 in the supplementary files).

4. Discussion

4.1. MPs in WWTPs

The outflow of the grit chamber contained on average some 13000 MP/m³, 95.0% of which was MFi and 5.0% MPa. Comparing these data with other studies is difficult because previous studies investigated MPs before and after the bar screen as influent wastewater, while in this study we investigated the number of MPs after the grit removal process as influent. Also the use of different processes in the WWTP makes it hard to compare data obtained in different studies. Table 3 gives an overview of the results of different studies reported in the literature.

The influent of the Sari treatment plant contained less MPs than WWTPs in Finland, Canada and China (Talvitie et al., 2017b; Gies et al., 2018; Lares et al., 2018; Liu et al., 2019). The very high number of MPs in the influent in these countries may be related to the population, the level of urbanization, and industrial activities (Li et al., 2018; Raju et al., 2020). The level of MPs in Sari influent

Table 1

The total number of microplastics (MPs), microfibers (MFi) and microparticles (MPa) per m³ (\pm SE, $n = 3$) at the outflow of units of the wastewater treatment plant in Sari, northern Iran.

Treatment unit	MFi/m ³	MPa/m ³	MP/m ³
grit chamber	12022 \pm 656 ^a	645 \pm 58.8 ^a	12667 \pm 668 ^a
primary settling	3157 \pm 548 ^b	357 \pm 40.6 ^b	3514 \pm 543 ^b
Clarifier	328 \pm 33.4 ^c	95.1 \pm 12.2 ^c	423 \pm 44.9 ^c

a,b,c: Different letters indicate significant differences in the number of microplastics, microfibers and microparticles between the treatment units ($P < 0.05$).

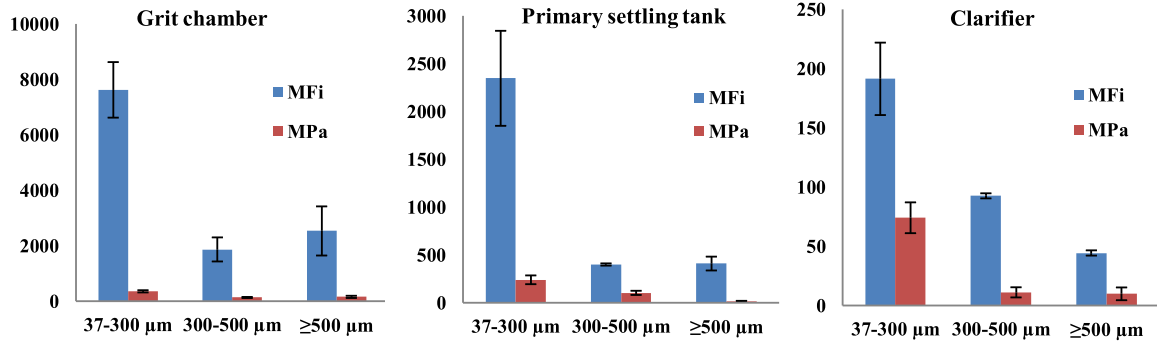


Fig. 2. The number of microfibers (MFi) and microparticles (MPa) in different size classes in the outflows of different steps in the wastewater treatment plant of Sari, northern Iran.

Table 2
The removal efficiency of the microplastics (MPs) and of microfibers (MFi) and microparticles (MPa) in different units of the wastewater treatment plant in Sari, northern Iran.

	Primary settling tank	Clarifier	Total
MPs (T)	72.3%	24.4%	96.7%
MFi (T)	73.7%	23.5%	97.3%
MPa (T)	44.6%	40.7%	85.2%

was also slightly lower than in raw wastewater and higher than in the de-gritted wastewater of a Scottish WWTP (Murphy et al., 2016). Although the numbers in the influent of both treatment plants were approximately the same, the type of MPs was very different. While most of the MPs entering the Scottish WWTP were MPa with only 18.5% being MFi (Murphy et al., 2016), more than 90.0% of the MPs in the influent of Sari's WWTP were MFi.

The number of MPs in the grit chamber outflow in this study

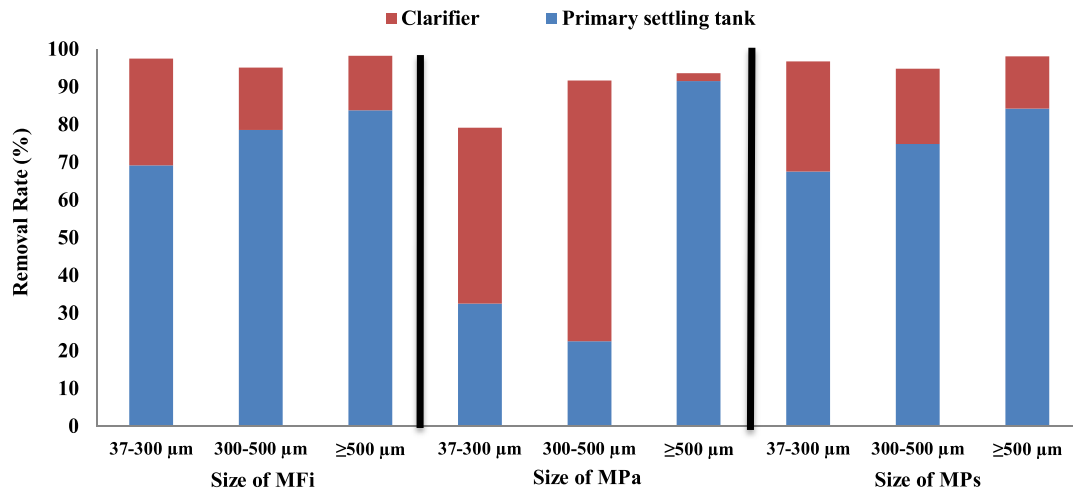


Fig. 3. The removal efficiency of different size classes of microplastics (MPs), microfibers (MFi) and microparticles (MPa) in different steps of the wastewater treatment plant of Sari, northern Iran.

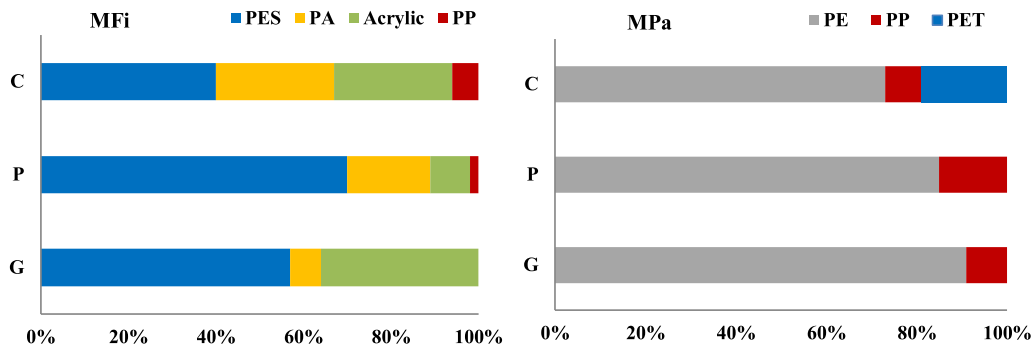


Fig. 4. The relative proportion (%) of the type of microfibers (MFi) and microparticles (MPa) in the outflows from different treatment steps in the wastewater treatment plant (G: Grit chamber, P: Primary settling tank, C: Clarifier) of Sari, northern Iran. PES= Polyester, PA= Polyamide or nylon, Acrylic, PE= Polyethylene, PP= Polypropylene, PET= Polyethylene terephthalate.

Table 3
Comparison of the presence of microplastics (MPs) in wastewater and effluents and removal efficiencies at different stages in the process of wastewater treatment in the present study and other studies. Also indicated are numbers of microfibers (MFi) and microparticles (MPa).

Country	Sampling location	Discharge (MP/m ³)	Removal (%)	Smallest mesh (μm)	Population	Reference
Iran	Influent (After grit chamber)	12667 ± 668 (MFi: 12022 ± 656, MPa: 645 ± 58.8)	–	37	105800	This study
	After primary settling tank	3514 ± 543 (MFi: 3157 ± 548, MPa: 357 ± 40.6)	72.3 (MFi: 73.7, MPa: 44.6)			
	Final effluent (After clarifier)	423 ± 44.9 (MFi: 328 ± 33.4, MPa: 95.1 ± 12.2)	24.4 (MFi: 23.5, MPa: 40.7)			
Sweden	Influent (Raw wastewater)	15100 ± 890	–	300	12000	Magnusson and Norén (2014)
Scotland	Final effluent (Treated wastewater)	8.25 ± 0.85	99.9			
	Influent (after 19 mm coarse screening)	15700 ± 5230	–	65	650000	Murphy et al. (2016)
	Grit and grease effluent	8700 ± 1560	44.6			
	Primary effluent	3400 ± 280	33.7			
	Final effluent	250 ± 40	20.1			
			Total removal:98.4			
Finland	Influent	567800	–	–	800000	Talvitie et al. (2017b)
Finland	Final Effluent (Secondary effluent)	1430	99.7			
	Influent (After 6 mm screen, in beginning of grit separation basin)	57600 ± 12400	–	250	–	Lares et al. (2018)
	After primary clarification	600 ± 200	–			
Canada	Final effluents (After disinfection)	1050 ± 400	98			
	Influent	31100 ± 6700	–	1.63	1300000	Gies et al. (2018)
	Primary effluent	2600 ± 1400	91.6			
	Final effluent (Secondary effluent)	500 ± 200	6.7			
			Total removal:98.3			
Italy	Influent	2500 ± 300	–	63	1200000	Magni et al. (2019)
	Secondary effluent	900 ± 300	64			
	Final effluent (sand filter treatment and disinfection)	400 ± 100	20			
			Total removal:84			
China (7WWTPs)	Influent	1570-13690 (ave. = 6550)	–	43	–	Long et al. (2019)
	Final Effluent (Secondary effluent)	200-1730 (ave. = 590)	79.3–97.8 (ave. = 90.5)			
China	Influent (Inlet of coarse grid)	79900 ± 9300	–	47	–	Liu et al. (2019)
	Primary effluent	47400 ± 7000	40.7			
	Secondary effluent	34100 ± 9400	16.6			
	Final effluent	8400 ± 7000	7.1			
			Total removal:64.4			
Spain	Influent (grit and grease removal)	3200 ± 670	–	–	210000	Bayo et al. (2020)
	primary clarifier	2590 ± 850	19.06			
	Final effluent (After clarifier)	10 ± 60	71.25			
			Total removal:90.3			

was about 4 times higher than in the outflow of grit and grease removal unit of a Spanish WWTP, while the population covered by the Spanish WWTP was twice that of Sari (Bayo et al., 2020). This difference may be due differences in the lifestyle of people in these regions (Table 3). The number of MPs in the influent in different countries may be affected by habits, weather and seasonal conditions (Magni et al., 2019). In addition to the difference in the finest mesh size used for sampling, the population covered by the wastewater network and the sampling location have significant impacts on the differences in the number of MPs recovered from the wastewater.

About 3500 and 420 MPs were found at the outflows of the primary settling tank and the clarifier unit of the Sari's WWTP, respectively. These numbers are much lower compared to the values found at the outflows of the primary settling and the clarifier unit of treatment plants in Finland (Talvitie et al., 2017b; Lares et al., 2018) and almost equal to the number of MPs in a WWTP in Scotland (Murphy et al., 2016). Compared to the Sari's WWTP, in Swedish WWTPs the MP numbers were slightly higher in the influent, but much lower in the effluent (Magnusson and Norén, 2014). Despite the differences in the MP numbers in the influent and effluent of treatment plants, the removal efficiencies of MPs in all treatment plants were approximately similar. This high removal efficiency indicates the good performance of the treatment plant for the separation of MPs (Table 3).

Results of studies on wastewater treatment plants can differ because of different sampling methods (simple or composite, sampling time, mesh size of sieves, etc.), extraction methods (digestion method and salt used for density separation), identification methods, and social, economic and climate conditions. This makes it difficult to compare the results of different studies (Avio et al., 2015; Raju et al., 2020), and emphasizes the importance of developing standard protocols for the measurement and reporting of MPs to facilitate the comparison of data.

4.2. Removal efficiency

Primary settling had the greatest effect on the removal of MPs, leading to a 72.3% decrease, while the studied processes of the Sari wastewater treatment plant together removed about 96.7% of all MPs (Table 1). This reduction of MPs by primary settling agrees with the work of Murphy et al. (2016) who found 78.3% MP removal by pre-treatment and primary treatment. Most of the MFi (in all three size classes) were eliminated in the primary settling (73.7%), while only 23.5% was removed in the clarifier unit. This result is in line with Talvitie et al. (2015). For the MPa, however, removal rates were similar for both processes.

Investigating the effect of size and shape on the removal of MPs during wastewater treatment is very important to provide appropriate solutions for their optimal separation. Unlike MFi, most of the particles <500 µm were best removed from the wastewater by the clarifier, followed by primary settling. For removing particles ≥500 µm, however, primary settling played the most important and effective role, with only 2% removal by the clarifier unit (Fig. 3).

The removal of MFi ≥37 µm and MPa ≥500 µm in primary settling and the role of the clarifier unit in the removal of particles <500 µm may be attributed to their density, size, and shape. A way to approach this is by looking at the settling velocities of the MFi and MPa, which may follow Stokes' law (eq. (1)):

Equation (1):

$$\omega_f = \frac{\left(\frac{2}{9}\right) \cdot g \cdot \left(\frac{\rho_s}{\rho_f} - 1\right) \cdot r^2}{\eta_f} \quad (1)$$

where ω_f is the settling velocity [L/T], g the acceleration due to gravity [L/T²], ρ_s the density of the spherical particle [M/L³], ρ_f the density of the fluid [M/L³], r the radius of the particle [L], and η_f the kinematic viscosity of the fluid [L²/T].

Most of the extracted MFi were polyester, acrylic and nylon with densities of 1.24–2.3, 1.18 and 1.02–1.16 g/ml, respectively. Based on the Stokes' equation, one of the possible reasons for the separation of these MPs in the primary settling may be that their density is higher than that of water. Consistent with these results, Zhang et al. (2020) state that MPs with a higher density than water are separated from the sewage during the primary sedimentation processes.

Most of the MPa were polyethylene, which have a lower density than water (0.89–0.98). According to the Stokes' law, differences in the size of this type of MPs play a decisive role in their removal, as particles <500 µm require more time to be separated from the wastewater.

Lower-density MPs can be removed from the wastewater during the primary treatment by surface skimming, because they float on the surface of the wastewater due to their lightness (Carr et al., 2016; Talvitie et al., 2017a; Lares et al., 2018). Murphy et al. (2016) found that microbeads are effectively eliminated by the skimming process, likely because most microbeads are made of polyethylene and probably associate with the fat, oil, and grease that float on the wastewater surface. According to our results, it can be concluded that the large size of low-density particles (MPa ≥500 µm) causes them to be removed in the primary treatment by surface skimming.

Although some particles less than 500 µm, which were mainly light MPs (with density less than water, <1 g/ml), were removed in the primary treatment, the important role of the clarifier unit in their removal can be related to the adsorption of these particles by biological flocs in the secondary treatment process. Flocs are made of extracellular polymeric substances (EPS) secreted by microorganisms, and their viscous properties enable catching remaining MPs from the primary treatment process, especially low-density MPs, separating them from the wastewater (Carr et al., 2016; Zhang et al., 2020).

The reason for the removal of most particles smaller than 500 µm and the small amount of fibers (which were not removed during primary settling) in the clarifier tank may also be due to the formation of biofilms, which increased their density (Carr et al., 2016; Raju et al., 2020), or their adhering to the bioflocs formed in the aeration tank which settled in the clarifier (Turovskiy and Mathai, 2005; Zhang et al., 2020).

4.3. MPs size and morphology

Most MFi and MPa were in size of 37–300 µm. The smaller the MPs, the larger their surface-to-volume ratio making them more reactive and also more likely to bind persistent organic pollutants (POPs), heavy metals as well as hydrophobic organic pollutants (HOCs) (Rios et al., 2010). As a consequence, the small MPs released by WWTPs can pose significant environmental hazards.

The dominance of MFi (polyester, nylon, and acrylic) in Sari's wastewater can be due to washing clothes and other textiles such

as carpets. Browne et al. (2011) found on average > 1900 MFi released per wash of one piece of clothing. One fleece clothing (Almroth et al., 2018) and 5 kg of polyester textile (Falco et al., 2018) can release approximately 110,000 and 6,000,000 MFi, respectively. Aalipour et al. (2020) showed that washing one square meter of machine-woven carpet can, on average, release between 1825 and 3098 MFi.

Among the MPa, spherical and irregular blue MPa were often in the size of 37–300 µm and made of polyethylene, so similar to the polyethylene MPa used in personal care and cosmetic products (Carr et al., 2016; Lares et al., 2018). Thus, the most likely source of these particles are personal care products such as toothpaste and face scrub that enter the wastewater during washing (Chua et al., 2014; Wright et al., 2013). In addition to polyethylene particles, which are the majority of MPa in all stages, a few polypropylene particles were observed in the samples which are also rarely used in cosmetics (Hammer et al., 2012). Also, the origin of other particles in the effluents, such as PET and some of the PE and PP, can be the degradation or erosion of plastic objects.

5. Conclusion

The abundance, characteristics, and removal of MPs in different stages of wastewater treatment as well as the role of factors affecting the removal efficiency in WWTPs were studied by analyzing samples from the Sari WWTP.

Most of the MFi in all three sizes were removed from the wastewater at the primary settling process, with a smaller but significant role also of the clarifier unit. This may be due to the high density of fibers compared to water. Initial settling was most important for removing particles ≥500 µm, the clarifier for particles <500 µm.

In spite of a 96.7% reduction of MPs, the Sari WWTP can be considered one of the main sources of MPs release into the Tajan River and the Caspian Sea. The high discharge rate of treated effluent (22000 m³/d) results in a total emission of more than 9.3 million MPs/d to the environment of which 77.5% as MFi especially with size of 37–300 µm. So, despite the high removal efficiency of the primary settling tank and clarifier unit, complementary treatment processes such as filtration are necessary to further reduce the emission of MPs from the Sari WWTP.

Credit author statement

Somayye Sadat Alavian Petroody: Methodology, Validation, Formal analysis, Investigation, Writing - original draft, Visualization. Seyed Hossein Hashemi: Conceptualization, Methodology, Resources, Formal analysis, Data curation, Supervision. Cornelis A.M. van Gestel: Resources, Formal analysis, Writing - review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.chemosphere.2020.128179>.

References

- Aalipour, S., Hashemi, S.H., Alavian Petroody, S.S., 2020. Release of microplastic fibers from carpet-washing workshops wastewater. *Journal of Water and Wastewater; Ab va Fazilab* (in persian). (accepted).
- Almroth, B.M.C., Åström, L., Roslund, S., Petersson, H., Johansson, M., Persson, N.K., 2018. Quantifying shedding of synthetic fibers from textiles; a source of microplastics released into the environment. *Environ. Sci. Pollut. Res.* 25, 1191–1199. <https://doi.org/10.1007/s11356-017-0528-7>.
- Avio, C.G., Gorbi, S., Milan, M., Benedetti, M., Fattorini, D., d'Errico, G., Paoletto, M., Bargelloni, L., Regoli, F., 2015. Pollutants bioavailability and toxicological risk from microplastics to marine mussels. *Environ. Pollut.* 198, 211–222. <https://doi.org/10.1016/j.envpol.2014.12.021>.
- Bayo, J., Martínez, A., Guillen, M., Olmos, S., Roca, M.J., Alcolea, A., 2017. Microbeads in commercial facial cleansers: threatening the environment. *Clean* 45 (7), 1600683. <https://doi.org/10.1002/cle.201600683>.
- Bayo, J., Olmos, S., Lopez-Castellanos, J., 2020. Microplastics in an urban wastewater treatment plant: the influence of physicochemical parameters and environmental factors. *Chemosphere* 238, 124593. <https://doi.org/10.1016/j.chemosphere.2019.124593>.
- Browne, M.A., Crump, P., Niven, S.J., Teuten, E., Tonkin, A., Galloway, T., Thompson, R., 2011. Accumulation of microplastic on shorelines worldwide: sources and sinks. *Environ. Sci. Technol.* 45, 9175–9179. <https://doi.org/10.1021/es201811s>.
- Carr, S.A., Liu, J., Tesoro, A.G., 2016. Transport and fate of microplastic particles in wastewater treatment plants. *Water Res.* 91, 174–182. <https://doi.org/10.1016/j.watres.2016.01.002>.
- Chua, E.M., Shimeta, J., Nuggeoda, D., Morrison, P.D., Clarke, B.O., 2014. Assimilation of polybrominated diphenyl ethers from microplastics by the marine amphipod, *Allorchestes compressa*. *Environ. Sci. Technol.* 48 (14), 8127–8134. <https://doi.org/10.1021/es405717z>.
- Colton, J.B., Knapp, F.D., 1974. Plastic particles in surface waters of the Northwestern Atlantic. *Science* 185, 491–497. <https://doi.org/10.1126/science.185.4150.491>.
- Estahbanati, S., Fahrenfeld, N.L., 2016. Influence of wastewater treatment plant discharges on microplastic concentrations in surface water. *Chemosphere* 162, 277–284. <https://doi.org/10.1016/j.chemosphere.2016.07.083>.
- Falco, F., De Pia, M., Gentile, G., Di, E., Escudero, R., Villalba, R., Mossotti, R., Montarolo, A., Gavignano, S., Tonin, C., Avella, M., 2018. Evaluation of microplastic release caused by textile washing processes of synthetic fabrics. *Environ. Pollut.* 236, 916–925. <https://doi.org/10.1016/j.envpol.2017.10.057>.
- Fowler, C.W., 1987. Marine debris and Northern Fur seals: a case study. *Mar. Pollut. Bull.* 18, 326–335. [https://doi.org/10.1016/S0025-326X\(87\)80020-6](https://doi.org/10.1016/S0025-326X(87)80020-6).
- 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>.
- Hammer, J., Kraak, M.H.S., Parsons, J.R., 2012. Plastics in the marine environment: the dark side of a modern gift. *Rev. Environ. Contam. Toxicol.* 220, 1–44. https://doi.org/10.1007/978-1-4614-3414-6_1.
- Lambert, S., Wagner, M., 2016. Characterisation of nanoplastics during the degradation of polystyrene. *Chemosphere* 145, 265–268. <https://doi.org/10.1016/j.chemosphere.2015.11.078>.
- Lares, M., Ncibi, M.C., Sillanpää, M., Sillanpää, M., 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>.
- Leslie, H.A., Brandsma, S.H., van Velzen, M.J., Vethaak, A.D., 2017. Microplastics en route: field measurements in the Dutch river delta and Amsterdam canals, wastewater treatment plants, North Sea sediments and biota. *Environ. Int.* 101, 133–142. <https://doi.org/10.1016/j.envint.2017.01.018>.
- Li, X., Chen, L., Mei, Q., Dong, B., Dai, X., Ding, G., Zeng, E.Y., 2018. Microplastics in sewage sludge from the wastewater treatment plants in China. *Water Res.* 142, 75–85. <https://doi.org/10.1016/j.watres.2018.05.034>.
- Liebezeit, G., Liebezeit, E., 2014. Synthetic particles as contaminants in German beers. *Food Addit. Contam.* 31 (9), 1574–1578. <https://doi.org/10.1080/19440049.2014.945099>.
- Liu, X., Yuan, W., Di, M., Li, Z., Wang, J., 2019. Transfer and fate of microplastics during the conventional activated sludge process in one wastewater treatment plant of China. *Chem. Eng. J.* 362, 176–182. <https://doi.org/10.1016/j.cej.2019.01.033>.
- 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>.
- Lusher, A., Hollman, P., Mendoza-Hill, J., 2017. Microplastics in Fisheries and Aquaculture: Status of Knowledge on Their Occurrence and Implications for Aquatic Organisms and Food Safety. *FAO Fisheries and Aquaculture Technical*, p. 615.

- Magni, S., Binelli, A., Pittura, L., Avio, C.G., Torre, C.D., 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>.
- Magnusson, K., Norén, F., 2014. Screening of Microplastic Particles in and Down-Stream a Wastewater Treatment Plant. Report C55. Swedish Environmental Research Institute, Stockholm.
- Mehdinia, A., Dehbandi, R., Hamzehpour, A., Rahnema, R., 2020. Identification of microplastics in the sediments of southern coasts of the Caspian Sea, north of Iran. *Environ. Pollut.* 258, 113738. <https://doi.org/10.1016/j.envpol.2019.113738>.
- Michielssen, M.R., Michielssen, E.R., Niac, 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>.
- Mintinig, S.M., Int-Veen, I., Löder, M.G.J., Primpke, S., Gerdt, G., 2017. Identification of microplastic in effluents of wastewater 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>.
- 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>.
- Nuelle, M.T., Dekiff, J.H., Remy, D., Fries, E., 2014. A new analytical approach for monitoring microplastics in marine sediments. *Environ. Pollut.* 184, 161–169. <https://doi.org/10.1016/j.envpol.2013.07.027>.
- Raju, S., Carbery, M., Kuttykattil, A., Senthirajah, K., Lundmark, A., Rogers, Z., Scb, S., Evans, G., Palanisami, T., 2020. Improved methodology to determine the fate and transport of microplastics in a secondary wastewater treatment plant. *Water Res.* 173, 115549. <https://doi.org/10.1016/j.watres.2020.115549>.
- Rillig, M.C., Ingrassia, R., De Souza Machado, A., 2017. Microplastic incorporation into soil in agroecosystems. *Front. Plant Sci.* 8, 1–4. <https://doi.org/10.3389/fpls.2017.01805>.
- Rios, L.M., Jones, P.R., Moore, C., Narayan, U.V., 2010. Quantitation of persistent organic pollutants adsorbed on plastic debris from the Northern Pacific Gyre's "eastern garbage patch". *J. Environ. Monit.* 12 (12), 2226–2236. <https://doi.org/10.1039/c0em00239a>.
- Rocha-Santos, T., Duarte, A.C., 2015. A critical overview of the analytical approaches to the occurrence, the fate and the behavior of microplastics in the environment. *Trac. Trends Anal. Chem.* 65, 47–53. <https://doi.org/10.1016/j.trac.2014.10.011>.
- Rochman, C.M., Tahir, A., Williams, S., Baxa, D.V., Lam, L., Miller, J., The, C., Werorilangi, S., The, J., 2015. Anthropogenic debris in seafood: plastic debris and fibers from textiles in fish and bivalves sold for human consumption. *Sci. Rep.* 5, 14340. <https://doi.org/10.1038/srep14340>.
- Sundt, P., Schulze, P.E., Syversen, F., 2015. Sources of Microplastic Pollution to the Marine Environment. Report M-321|2015. Norwegian Environment Agency.
- Talvitie, J., Heinonen, M., Pääkkönen, J.P., Vahtera, E., Mikola, A., Setälä, O., Vahala, R., 2015. Do wastewater treatment plants act as a potential point source of microplastics? Preliminary study in the coastal Gulf of Finland. *Baltic Sea. Water Sci. Technol.* 72, 1495–1504. <https://doi.org/10.2166/wst.2015.360>.
- 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>.
- Turovskiy, I.S., Mathai, P.K., 2005. *Wastewater Sludge Processing*. John Wiley & Sons, Inc, pp. 30–59. <https://doi.org/10.1002/047179161X>.
- Wang, J., Peng, J., Tan, Z., Gao, Y., Zhan, Z., Chen, Q., Cai, L., 2017. Microplastics in the surface sediments from the Beijiang River littoral zone: composition, abundance, surface textures and interaction with heavy metals. *Chemosphere* 171, 248–258. <https://doi.org/10.1016/j.chemosphere.2016.12.074>.
- Wright, S.L., Thompson, R.C., Galloway, T.S., 2013. The physical impacts of microplastics on marine organisms: a review. *Environ. Pollut.* 178, 483–492. <https://doi.org/10.1016/j.envpol.2013.02.031>.
- Zhang, X., Chen, J., Li, J., 2020. The removal of microplastics in the wastewater treatment process and their potential impact on anaerobic digestion due to pollutants association. *Chemosphere* 251, 126360. <https://doi.org/10.1016/j.chemosphere.2020.126360>.
- 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>.