

An evaluation of microplastics fate in the wastewater treatment plants: frequency and removal of microplastics by microfiltration membrane

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Abstract

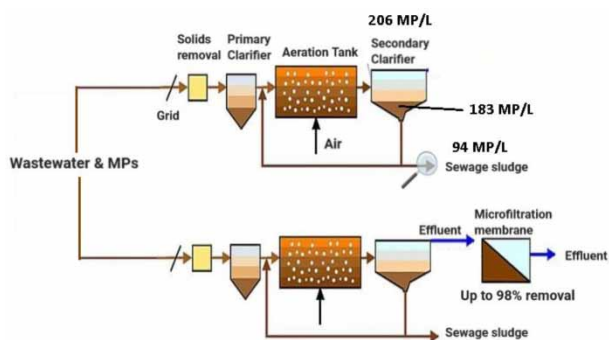
In this research, the fate and removal of microplastics (MPs) entering and leaving a wastewater treatment plant (WWTP) was investigated. Additionally, application of the microfiltration membrane technique for MPs removal was evaluated. In the first phase, the quantity, type and size of the MPs were studied from three different points of the WWTP. The results showed that the average amount of MPs entering into the WWTP, accumulated in the sludge matrix and discharged from the effluent were 206, 183 and 94 MP/L, respectively. The MPs were observed mainly in the forms of fibers, pellets, and fragments, with a proportion of 35%, 39%, 22%, and 34%, 22%, 38%, and 31%, 39%, 37.7% in the influent, effluent and sludge of the WWTP, respectively. It should be noted, a minor amount of foam (2%) and film (0.3%) was also observed. The particle size distribution of the MPs in the effluent of the system was almost identical (6–14% for 1–5,000 μm), while the influent mostly contained particles within 1–100 μm (above 26%) with other particle sizes within 100–5,000 μm (8–17%). Particle size within a range of 500–5,000 μm was abundant in the sludge. In the second phase, a micro-filtration (MF) membrane with a pore size of 0.1 μm was applied to eliminate the MPs from the WWTP effluent. It was observed the application of the MF membrane technique could achieve MPs removal efficiency up to 98% in the effluent of the system.

Key words: microfiltration membrane, microplastics, municipal wastewater treatment plants, sewage sludge

Highlights

- The fate of microplastics (MPs) in the wastewater treatment plant (WWTP) was evaluated.
- The most abundant MPs were fibers, pellets, and fragments in the in/effluent and sludge of WWTP.
- This investigation provides a preliminary insight for management of MPs in WWTPs.
- An advanced tertiary treatment process was applied for MPs removal.
- The MPs particles were reduced up to 98% by application of the microfiltration membrane (MBR).

Graphical Abstract



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INTRODUCTION

Existence of microplastics (MPs) in wastewater streams has become a major concern for many wastewater treatment plants (WWTP) (Brepols *et al.* 2020; Cuartucci 2020). MPs can be classified based on their color, shape, and polymer characteristics (Jung *et al.* 2018). In terms of particle size classification, MPs are defined as particles with an average diameter less than 5 mm (Talvitie *et al.* 2017). In terms of shape, MPs are found to be in the shape of granules, fibers, films, fragments, and pellets (Patel *et al.* 2009; Leslie *et al.* 2017). On the other hand, MPs can be categorized according to their chemical composition such as applied polymers during the manufacturing process of the origin plastics. Common polymers of MPs include polyethylene (PE), polyvinyl-chlorides (PVC), polystyrenes (PS), polyurethanes (PU), polypropylene (PP), polyethylene terephthalate (PET), polyamides (PA), and other polymers (Jung *et al.* 2018). Although the exact point source of MPs generation is unclear, these particles are usually by-products of industrial materials and household wastes like personal care products (i.e. soaps, face cleansers, lotions, etc) as well as food industry products (i.e. food containers, plastic bags, plastic bottles, etc) as primary MPs (Reisser *et al.* 2013; Lares *et al.* 2018; Rolsky *et al.* 2019). The other forms of MPs, known as secondary MPs, can be generated after the breakdown of large plastic wastes or primary MPs into small pieces during a harsh environment (Michielssen *et al.* 1970; Claessens *et al.* 2013). The Environmental Protection Agency (EPA) reported that about 80,000–219,000 tons of MPs are generated in the European countries annually, and consequently they are discarded into the environment (House of Commons Environmental Audit Committee 2016). As a result, the presence of these materials can impose many adverse impacts on the environment.

Considering the impacts of MPs on the aquatic environment, it is fair to say they are responsible for the physical, biological, or chemical distortion of water quality during the MPs fragmentation phase (Lusher *et al.* 2013; Wright *et al.* 2013). It was also reported that MPs can accumulate toxic materials and heavy metals of wastewater and sewage sludge in their texture by adsorption/absorption mechanisms (Bardi & Aminirad 2020; Naqash *et al.* 2020) and provide a suitable environment for attachment of microbial communities, viruses, and pathogens (Browne *et al.* 2011). Previous reports have shown that MPs can sorb endocrine disrupting chemicals, and emit them into the aqueous environment (Chen *et al.* 2019; Naqash *et al.* 2020). Also, MPs can be consumed by different aquatic species and induce biological disorders (Possatta *et al.* 2011; Lusher *et al.* 2013; Magnusson & Wahlberg 2014). Moreover, the association of MPs into the water body can affect the removal efficiency of MPs from water/wastewater treatment plants due to their hard separation feature, and consequently, operational procedures can be impaired (Phillips & Bonner 2015). Therefore, removal of pollutants and toxic substances from MPs is inevitable in the aquatic environment.

Wastewater treatment plants (WWTPs) are partially able to remove fragments above 300 micrometers, while the rest of the particles that leave the system are mainly in the range of the nano-scale (Carr *et al.* 2016). Although the WWTPs can diminish the amount of these particles from influent, the huge quantity of MPs in the effluent of the system still pour into the aquatic environment (Mason *et al.* 2016; Talvitie *et al.* 2017). It was previously reported that preliminary, primary, secondary, and tertiary treatment processes can reduce the particles up to 6, 68, 90, and 95%, respectively (Blair *et al.* 2019). Furthermore, applying chemical, biological, and physical treatment processes can remove MPs up to 99%, while the rest of the particles are introduced into the environment (Liu *et al.* 2021). A recent investigation on MPs removal showed that the advanced oxidation (UV and O₃), anaerobic-anoxic-oxic, and aeration grit chambers processes can reduce the MPs particles by 71.7%, 54.5%, and 58.5% respectively (Yang *et al.* 2019). Therefore, innovative approaches still need to be proposed in order to reduce the amount of MPs from the effluent of WWTPs (Carr *et al.* 2016; Lay *et al.* 2017; Mintenig *et al.* 2017; Ziajahromi *et al.* 2017).

Recently, MPs removal through a tertiary treatment process as a final treatment by application of membrane technology has received increased attention due to its versatility, and higher MPs removal potential (Lu *et al.* 2016; Salerno *et al.* 2016; Lares *et al.* 2018). Previous studies have shown that

membrane treatment presents a higher MPs removal efficiency (>99%) in comparison with the preliminary, primary, and secondary treatment processes (Sun *et al.* 2019; Iyare *et al.* 2020). However, the robust performance of the membrane process depends on the wastewater, physicochemical features of MPs, as well as the membrane applied (Wang *et al.* 2018). It should be noted that membrane shape, pore size, polymer type, and fouling persistence are the most important factors that affect the removal efficiency of MPs (Ma *et al.* 2018; Poerio *et al.* 2019). Therefore, a primary knowledge about types and fate of MPs in WWTPs is required prior to adopting a suitable strategy for MPs removal from WWTP streams.

The main objectives of this study are: (i) to assess the physicochemical characteristics of MPs, and its quantity in WWTP streams and (ii) to evaluate the removal efficiency of MPs by an advanced treatment technology, the microfiltration membrane. To do this, first, the quantity and quality of the MPs were evaluated from three different points of the WWTP of Sari city in the north part of Iran. Then, the application of a micro-filtration (MF) membrane with a pore size of 0.1 μm was investigated to eliminate the MPs from the effluent of the WWTP.

MATERIALS AND METHODS






Materials and sample collection

In this study, the wastewater treatment plant (WWTP) of Sari city in the north part of Iran was evaluated as a case study. This plant was designed with an average capacity of 18,000 m^3/day . The collected sludge was attained from the influent and effluent of the secondary settling tank in different seasons. Three samples were collected using an electric pump (Godwin-GSL80) from the different levels of the settling tank (i.e. ~ 1 m from the surface, middle, and bottom of the tank), as well as in the in/effluent of the settling tank. Due to the separate system of wastewater collection in the study area, where wastewater sources are mainly from domestic and limited commercial sources and observing low variability in amount of MPS through random sampling, it was decided to take three samples with three replicates, which showed a reasonable consistency. Therefore, three samples were collected per season and labeled as Sample 1, Sample 2, and Sample 3, and collected in autumn, winter, and summer, respectively. The collected samples were taken to the laboratory and kept at 4 °C prior to analyses. Then the collected samples were filtered to giving particles above 1 μm .

MPs extraction and detection assay

In order to extract MPs from the collected samples, 200 mL of hydrogen peroxide (H_2O_2) solution (35%) was added to 500 mL of each sample to eliminate the organic matter. Then they were kept at room temperature for 7 days prior to detection by a microscope analyzer (OPTO-EDU A10.1018) (Foekema *et al.* 2013; Mathalon & Hill 2014; Wesch *et al.* 2016). The extracted samples were filtered by Whatman filter paper (grade 42, 2- μm pore size), and then they were kept at room temperature for 2 days prior to checking with an integrated HD stereomicroscope camera (model AC 230 V 50 Hz; LABEX, UK). According to volatile solids and total solids tests (Water Environment Federation 1999), above 90% of filtered samples were MPs. ATR-FTIR spectroscopy (model WQF510A, China), in the wavelength region of 400–4,000 cm^{-1} with 16 scans, was used to identify the chemical characteristics of MPs. The polymer type of filtered samples was identified based on the absorption frequencies at a specified bound. ImageJ software (2016), was used to present the size of MPs. Table 1 shows the characteristics of MPs, which were observed according to their size and type.

Table 1 | Characteristics of MPs based on their size and type

Abbreviation	Type	Size	Definition	Figure
PT	Pellet	<5 mm–1 μ m	A spherical piece of plastic	
FR	Fragment	<5 mm–1 μ m	An irregular shaped piece of plastic	
FB	Fiber	<5 mm–1 μ m	A strand or filament of plastic	
FI	Film	<5 mm–1 μ m	A thin sheet or membrane-like piece of plastic	
FM	Foam	<5 mm–1 μ m	A piece of sponge, foam, or foam-like plastic	

Operation condition of microfiltration membrane

After the phase of extraction and detection of MPs, an advanced treatment technology was applied to evaluate the MPs removal in the effluent of the WWTP. In this regard, a submerged microfiltration membrane (MFM) with 0.1 μ m pore size was applied to separate the MPs from the effluent of the wastewater. The characteristics of MFM are shown in Table 2. The effluent of the system was manually pumped into the microfiltration tank with a working volume capacity of 50 L (i.e. depth-60 cm, length-30 cm, width- 28 cm). The MFM system was operated semi-continuously to treat 2 m³ of wastewater per day. A digital process controller was installed to monitor the performance of the microfiltration membrane through detecting the displacement of the water level of the tank. In the case of membrane fouling, the process controller orders the influent and effluent of the system to stop and the backwash process begins. Moreover, an aeration system was installed to provide agitation and prevent the membrane fouling as well as the flux declining.

Table 2 | Characteristics of MFM

Specification	Membrane material	PVDF + PET
	Nominal pore size	0.1 μ m
	Inside/outside diameter	0.8/1.9 mm
	Dimension (mm)	485*46*134
Operation conditions	Filtering type	Intermittent filtration/intermittent aeration
	Temperature	5–40 °C
	Backwash flux	50–85 LMH
	pH range	1–11

The sludge at the bottom of the filtration tank was pumped out as a collected sample. Three samples were taken at different levels (bottom, middle and top) of the treated effluent tank, and then the samples were mixed as a unique collected sample. The collected samples were taken to the laboratory and kept at 4 °C prior to analyses. The detection and separation of MPs from aqueous solution were similar to the previous section (see MPs extraction and detection assay). The collection, separation and detection of samples was done in triplicate. Figure 1 shows the schematic microfiltration membrane set-up.

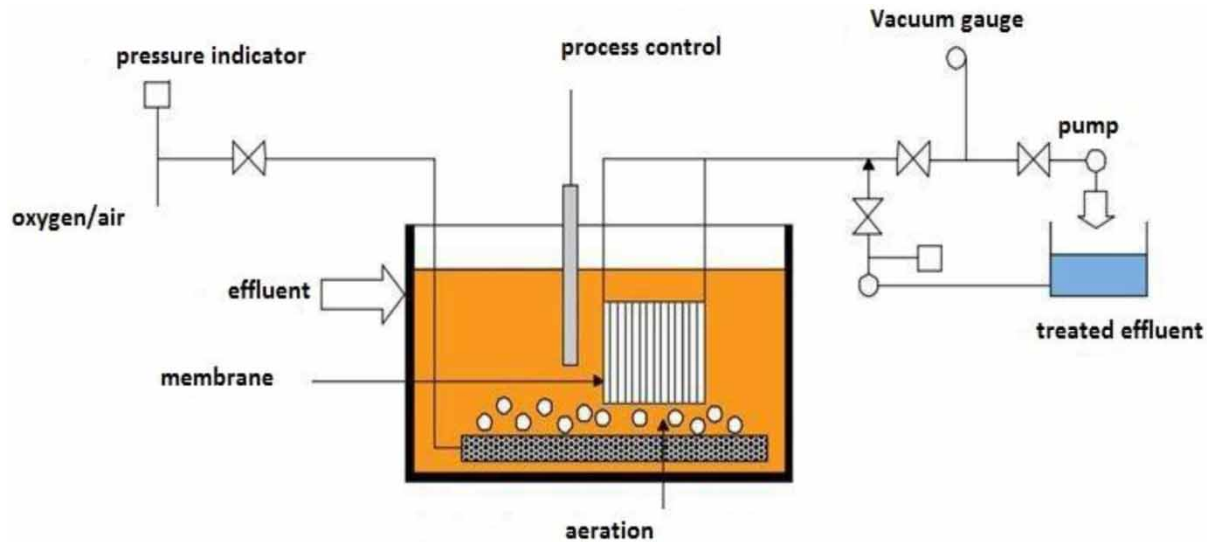


Figure 1 | The schematic microfiltration membrane set-up.

RESULTS AND DISCUSSION

Fate of MPs in the WWTP

Table 3 describes the MPs particles in the in/effluent and sludge of the secondary settling tank. As can be seen from Table 3, the highest amount of MPs was observed in the influent of the system with an average of 206 MP/L. The results of analyses showed that MPs were accumulated in the sludge at 183 MP/L. As previously reported, the accumulation of MPs in the sludge could have been due to the adsorption of these materials by the sludge matrix (Li *et al.* 2019). Furthermore, from Table 3, it can be found that the quantity of MPs in the influent and effluent of the WWTP varied corresponding to the season. It can be noticed that the number of MPs increased in the influent of samples 1, and 2 rather than sample 3 (Table 3). This phenomenon could have been due to climate change, by which rainfalls wash out the MPs into WWTPs (Xia *et al.* 2020). However, the settleability of MPs increased in sample 1 with an efficiency of 90.91%. The settleability of samples 1 and 2 was 88.9%, and 86.4% respectively (Table 3). From Table 3, the amount of MPs in the sludge samples of 1–3 was 200, 192, and 159 MP/L respectively. It was reported that the temperature and operational conditions (i.e. pH, dissolved oxygen, etc) have an indispensable role in sludge flocculation (Nouha *et al.* 2018). Thus, at the higher temperature, the sludge floc disintegrates and consequently results in low settleability of flocs. As a result of this phenomenon, the amount of MPs in the sludge matrix was lower in the summer (sample 3) due to sludge disintegration than that of autumn and winter (i.e. samples 1 and 2). From Table 3, it can be estimated that about 1,123.5 billion MPs enter into the WWTP annually. However, about 617.6 billion MPs are discharged into the aquatic environment annually from the effluent of the WWTP. Therefore, the post-treatments need to be applied after effluent of the system before the land application or direct discharge. Moreover, the incorporation of MPs into

Table 3 | The MPs particles in the in/effluent and sludge of secondary settling tank

Location	Sample 1 (MPs/L)	Sample 2 (MPs/L)	Sample 3 (MPs/L)	Average (MPs/L)
Influent	220 ± 23	216 ± 19	184 ± 26	206 ± 31
Sludge	200 ± 41	192 ± 24	159 ± 23	183 ± 21
Effluent	106 ± 31	95 ± 17	81 ± 21	94 ± 28

the sewage sludge is above 1,202 billion MPs annually, hence the application of sewage sludge must be banned from the land application or should be treated prior to use.

Taking the types of MPs into the account, from Figure 2 and Table 4, it can be seen that all types of MPs are found in the in/effluent and sludge of the secondary settling tank. The most abundant MPs is fibers, fragments, and pellets followed by a minor amount of foam and film in the sludge. The amount of film in the in/effluent of the system was negligible (i.e. almost 0.00%). Also, in the sludge of the system, a minor percentage of film (i.e. 0.3%) was observed. The same was also observed for foam with 2%. Thus, it can be concluded that the film and foam are either removed in the previous unit prior to the secondary settling tank or the existence of these MPs in the influent of WWTP is trivial.

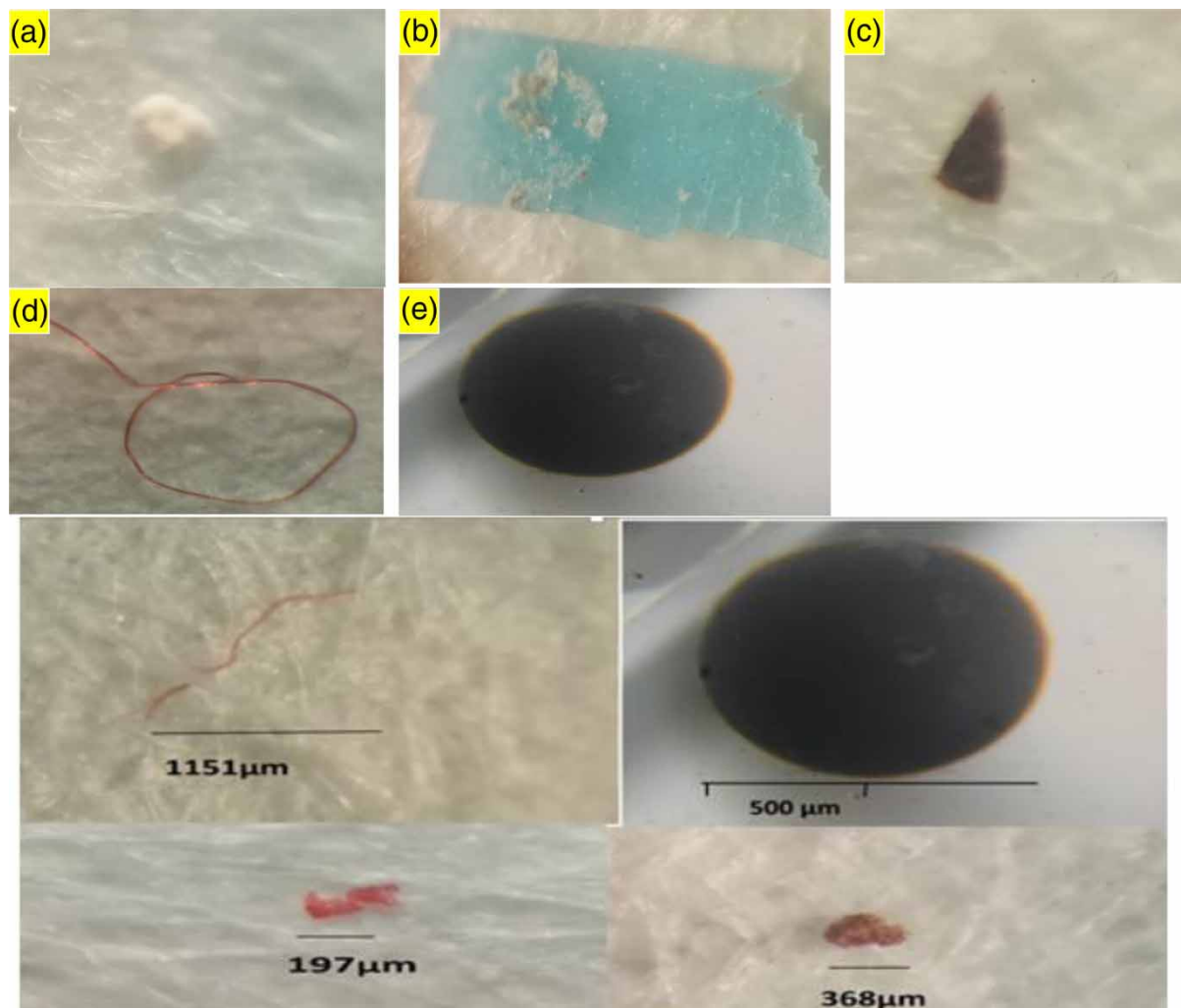


Figure 2 | The magnified picture of MPs in the system (Foam (a), Film (b), Fragment (c), Fiber (d), Pellet (e)).

Table 4 | The quantity of MPs in the in/effluent and sludge of secondary settling tank

Location	Film	Fragment	Pellet	Foam	Fiber
Influent	ND	31 ± 5.5%	34 ± 6%	ND	35 ± 6%
Sludge	0.3 ± 0.2%	37.7 ± 9%	38 ± 5%	2 ± 1%	22 ± 8%
Effluent	ND	39 ± 7%	22 ± 8%	ND	39 ± 7%

ND, not detectable.

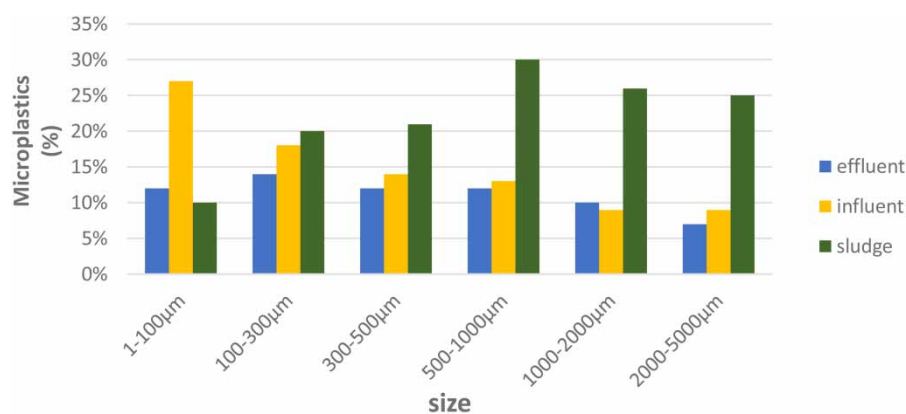
Table 5 | The results of MFM performance on MPs removal

Effluent	Before (MPs/L)	After (MPs/L)	Removal (%)
Sample 1	106 ± 16	2 ± 1	98.1%
Sample 2	81 ± 12	ND	100%
Sample 3	95 ± 7	1 ± 2	98.9%

On the other hand, the percentage of fragments, pellets, and fibers in all collecting stations (e.g. in/effluent and sludge), was partially the same, ranging from 31–39%, 22–38%, and 22–39%, respectively (Table 5). Since these materials are heavier than film and foam, they tend to settle or stick to the sludge matrix. Consequently, the presence of these materials impairs the subsequent operational units such as anaerobic digestion, when the digesters are fed with MPs-concentrated sludge (Li *et al.* 2020; Aminzadeh *et al.* 2021). Therefore, an appropriate post-treatment should also be adopted to eliminate the MPs from sewage sludge (Noguchi *et al.* 2019).

Distribution of MPs according to the classified size in the WWTP

Figure 2(b) shows the different sizes and types of MPs particles in the WWTP. From the figure, it was observed that the current size of pellets, fibers, fragments, and foam was 500 µm, 1,151 µm, 368 µm, and 197 µm, respectively. Also, the typical MPs size was classified into six groups, namely 1–100 µm, 100–300 µm, 300–500 µm, 500–1,000 µm, 1,000–2,000 µm, and 2,000–5,000 µm (Figure 3). Figure 3 shows that the distribution of MPs according to the classified size is almost the same in the effluent. Therefore, MPs particles such as fragments, pellets, and fibers, which are abundant in the effluent of the WWTP, can be found in a wide range of sizes (i.e. 1–5,000 µm) (Table 4 and Figure 3). However, in the influent, the abundant particles are within the range of 1–100 µm (above 26%). It can be noticed from Figure 3, as the particle size of MPs increases, the distribution of MPs decreases in the influent of the system. This effect could have been due to the large MPs being removed in the previous WWTP units (Blair *et al.* 2019). At higher particle size (within the range of 100–5,000 µm), MPs are mostly accumulated in the sewage sludge due to their higher density and specific weight (Carr *et al.* 2016; Lares *et al.* 2018). The most abundant particle in the sludge was within a range of 500–5,000 µm with a proportion of 25–29%. Thus, it can be assumed that the WWTPs do not have an intrinsic potential to eliminate particles greater than 1 µm; therefore, a new approach for removal of MPs particles greater than 1 µm is proposed through incorporating a microfiltration membrane system, which will be discussed further.

**Figure 3** | The distribution of MPs in the in/effluent and sludge according to the classified size.

FTIR spectroscopy of MPs

The polymer features of collected MPs from in/effluent and sludge of settling tank were analyzed using an FTIR spectrometer with 16 scans and the spectra from $4,000\text{ cm}^{-1}$ to 450 cm^{-1} (Figure 4). A minimum of three adsorption bands was detected within $1,300\text{--}1,690\text{ cm}^{-1}$ in the collected MPs from the influent (Figure 4(a)). These functional broad bands were found to be abundant in methyl group in branched low-density polyethylene, and polyvinyl chloride (Nishikida & Coates 2003; Noda *et al.* 2007; Asensio *et al.* 2009; Brandon *et al.* 2016). The broad band within the region of $1,330\text{--}1,400\text{ cm}^{-1}$ represents C-H stretch, and CH_2 bend and was found to be polypropylene (Asensio *et al.* 2009). The functional region at $2,260\text{ cm}^{-1}$ shows the CN stretch group and can be assigned to nitrile (Verleye *et al.* 2001). Also, from Figure 4, broadband within $2,970\text{--}3,400\text{ cm}^{-1}$, with a maximum of $3,434$, is presented. This band can be attributed to aromatic C-H stretch, implies the presence of polystyrene (Verleye *et al.* 2001; Noda *et al.* 2007; Asensio *et al.* 2009).

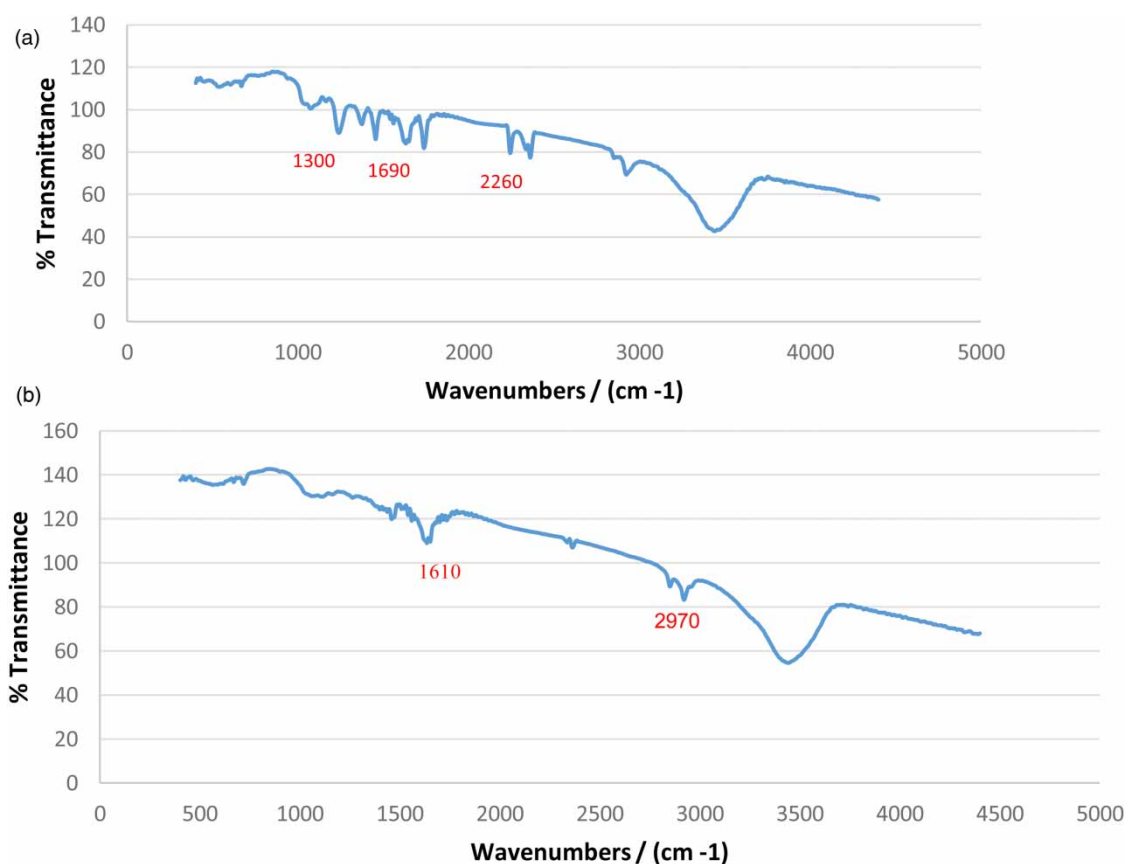


Figure 4 | FTIR image for MPs ((a) influent, (b) effluent and sludge).

MPs removal by microfiltration membrane (MFM)

MFM technique is a versatile technology for water and wastewater treatment, and it is estimated that the application of different membranes for MPs removal will increase in the following years (Li *et al.* 2018; Poerio *et al.* 2019; Chuanwen *et al.* 2020; Lorain *et al.* 2020). Since the selection of MFM is based on the composition of treated wastewater, and the particle size of MPs, deep knowledge about types and size distribution of MPs are required (i.e. it was evaluated in the previous section) (Cerón-Vivas & Noyola 2017; Li *et al.* 2018). From Table 4, it can be seen that higher MPs are entered into the MFM tank (sample 1) before discharge with 106 MP/L. However, only 2 MP/L was observed

in the effluent of the system. Thus, MFM could remove the MPs by up to 98%. Next to that, sample 3 received 95 MP/L and achieved MPs removal efficiency of 98.9%. However, in Sample 2, removal efficiency of 100% was observed. From Table 4, it can be concluded that the number of MPs affects the efficiency of MFM since, at a higher amount of MPs, the removal efficiency of MFM declined. Also, the detected particles after the MFM system were below 0.1 μm , which implies that the particles were within nano-scale. It should be noted that our findings are fitted by other investigations (Carr *et al.* 2016). Mintenig *et al.* (2017) reported that the application of the post-filtration unit (e.g. pile cloth media filtration) could completely remove the MPs particles greater than 500 μm , and particles within the range of 20–500 μm with a removal efficiency of 95%. Also, Ziajahromi *et al.* (2017) found that the ultrafiltration membrane can significantly reduce the MPs to 0.28 MP/L in the effluent of the system.

CONCLUSIONS

The quantity of MPs was evaluated in the in/effluent and sludge of WWTP. The results showed that about 1,123.5 billion MPs are entered into the WWTP, and about 55% leave the WWTP, annually. Also, the results showed that 1,202 billion MPs accumulate in the sludge, annually. Most of the MPs were fibers, fragments, and pellets, and only a minor percentage of foam and film was observed. The heavier particles are trapped in the sludge matrix, although the smaller particles within the range of 1–300 μm are observed in the influent of the system. The trapped particles in the sludge were in the range of 500–5,000 μm , while in the effluent of system the distribution of particle size is identical with a proportion of 6–14%. In this study, the advanced MFM could achieve MPs removal up to 98% in all samples. Therefore, this study provides knowledge for evaluating the fate of MPs in the WWTPs and possibly eliminates MPs in the effluent of WWTP prior to discharge.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

REFERENCES

- Aminzadeh, M., Bardi, M. J. & Aminirad, H. 2021 A new approach to enhance the conventional two-phase anaerobic co-digestion of food waste and sewage sludge. *International Journal of Environmental Health Research*. <https://doi.org/10.1007/s40201-020-00603-8>
- Asensio, R. C., Moya, M. S. A., de la Roja, J. M. & Gómez, M. 2009 Analytical characterization of polymers used in conservation and restoration by ATR-FTIR spectroscopy. *Analytical and Bioanalytical Chemistry* **395**(7), 2081–2096.
- Bardi, M. J. & Aminirad, H. 2020 Synergistic effects of co-trace elements on anaerobic co-digestion of food waste and sewage sludge at high organic load. *Environmental Science and Pollution Research International* **27**(15), 18129–18144. <http://dx.doi.org/10.1007/s11356-020-08252-y>
- Blair, R. M., Waldron, S. & Gauchotte-Lindsay, C. 2019 Average daily flow of microplastics through a tertiary wastewater treatment plant over a ten-month period. UNSP 114909 *Water Research* **163**, 114909. <https://doi.org/10.1016/j.watres.2019.114909>
- Brandon, J., Goldstein, M. & Ohman, M. D. 2016 Long-term aging and degradation of microplastic particles: comparing in situ oceanic and experimental weathering patterns. *Marine Pollution Bulletin* **110**(1), 299–308.
- Brepols, C., Drensla, K., Janot, A. & Schäfer, H. 2020 Energetic refurbishment of an existing large-scale MBR. *Water Practice and Technology* **15**(1), 182–187. doi:10.2166/wpt.2020.009.
- 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. *Environmental Science & Technology* **45**(21), 9175–9179.
- Carr, S. A., Liu, J. & Tesoro, A. G. 2016 Transport and fate of microplastic particles in wastewater treatment plants. *Water Research*. Available from: <https://www.sciencedirect.com/science/article/abs/pii/S0043135416300021>

- Cerón-Vivas, A. & Noyola, A. 2017 Fouling membrane in an anaerobic membrane bioreactor treating municipal wastewater. *Water Practice and Technology* **12**(2), 314–321. doi:10.2166/wpt.2017.036.
- Chen, Q., Allgeier, A., Yin, D. & Hollert, H. 2019 Leaching of endocrine disrupting chemicals from marine microplastics and mesoplastics under common life stress conditions. *Environment International* **130**, 104938. Available from: <https://www.sciencedirect.com/science/article/pii/S0160412019303137>
- Chuanwen, S., Haiqiao, W., Qi, Y., Shiqiang, C., Xun, L. & Hanyang, W. 2020 Experimental study of the flux law of flat ceramic membranes under different pressures. *Water Practice and Technology* **15**(2), 416–425. doi:10.2166/wpt.2020.028.
- Claessens, M., Cauwenberghe, L. V., Vandegehuchte, M. B. & Janssen, C. R. 2013 New techniques for the detection of microplastics in sediments and field collected organisms. *Marine Pollution Bulletin* **70**, 227–233.
- Cuartucci, M. 2020 Ultrafiltration, a cost-effective solution for treating surface water to potable standard. *Water Practice and Technology* **15**(2), 426–436. doi:10.2166/wpt.2020.039.
- Foekema, E. M., De Grijter, C., Mergia, M. T., Van Franeker, J. A., Murk, A. J. & Koelmans, A. A. 2013 Plastic in North Sea fish. *Environmental Science & Technology* **47**, 8818–8824.
- House of Commons Environmental Audit Committee, Environmental impact of microplastics, Fourth Report of Session 2016–17. House of Commons, London, UK.
- Iyare, P. U., Ouki, S. K. & Bond, T. 2020 Microplastics removal in wastewater treatment plants: a critical review. *Environmental Science: Water Research & Technology* **6**, 2664–2675. Available from: <https://pubs.rsc.org/en/content/articlelanding/2020/ew/d0ew00397b#divAbstract>
- Jung, M. R., Horgen, F. D., Orski, S. V., Rodriguez, V., Beers, K. L., Balazs, G. H., Jones, T. T., Work, T. M., Brignac, K. C., Royer, S. J. & Hyrenbach, K. D. 2018 Validation of ATR FT-IR to identify polymers of plastic marine debris, including those ingested by marine organisms. *Marine Pollution Bulletin* **127**, 704–716. Available from: <https://www.sciencedirect.com/science/article/abs/pii/S0025326X17310949>
- 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 Research* **133**, 236–246. Available from: <https://www.sciencedirect.com/science/article/pii/S0043135418300630>
- Lay, W. C., Lim, C., Lee, Y., Kwok, B. H., Tao, G., Lee, K. S. & Seah, H. 2017 From R&D to application: membrane bioreactor technology for water reclamation. *Water Practice and Technology* **12**(1), 12–24. Available from: <https://iwaponline.com/wpt/article-abstract/12/1/12/20764/From-R-and-D-to-application-membrane-bioreactor?redirectedFrom=fulltext>
- Leslie, H., Brandsma, S., Velzen, M. V. & Vethaak, A. 2017 Microplastics en route: field measurements in the Dutch river delta and Amsterdam canals, wastewater treatment plants, North Sea sediments and biota. *Environment International* **101**, 133–142. Available from: <https://www.sciencedirect.com/science/article/pii/S0160412017301654>
- Li, L., Xu, G., Yu, H. & Xing, J. 2018 Dynamic membrane for micro-particle removal in wastewater treatment: performance and influencing factors. *Science of The Total Environment* **627**, 332–340. doi:10.1016/j.scitotenv.2018.01.239.
- Li, X., Mei, Q., Chen, L., Zhang, H., Dong, B., Dai, X. & Zhou, J. 2019 Enhancement in adsorption potential of microplastics in sewage sludge for metal pollutants after the wastewater treatment process. *Water Research* **157**, 228–237. doi:10.1016/j.watres.2019.03.069.
- Li, L., Geng, S., Li, Z. & Song, K. 2020 Effect of microplastic on anaerobic digestion of wasted activated sludge. *Chemosphere* **247**, 125874. doi:10.1016/j.chemosphere.2020.125874.
- 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. *Environment International* **146**, 106277. Available from: <https://pubmed.ncbi.nlm.nih.gov/33227584/>
- Lorain, O., Marcellino, S., Deratani, A., Gassara, S., Duchemin, I. & Espenan, J. 2020 New ultrafiltration (UF) membrane made with a new polymer material for long-lasting rejection performance. *Neophil®. Water Practice and Technology* **15**(2), 356–364. doi:10.2166/wpt.2020.022.
- Lu, D., Cheng, W., Zhang, T., Lu, X., Liu, Q., Jiang, J. & Ma, J. 2016 Hydrophilic Fe₂O₃ dynamic membrane mitigating fouling of support ceramic membrane in ultrafiltration of oil/water emulsion. *Separation and Purification Technology* **165**, 1–9. Available from: <https://www.sciencedirect.com/science/article/abs/pii/S1383586616301393>
- Lusher, A. L., McHugh, M. & Thompson, R. C. 2013 Occurrence of microplastics in the gastrointestinal tract of pelagic and demersal fish from the English Channel. *Marine Pollution Bulletin* **67**, 94–99.
- Ma, B., Xue, W., Hu, C., Liu, H., Qu, J. & Li, L. 2018 Characteristics of microplastic removal via coagulation and ultrafiltration during drinking water treatment. *Chemical Engineering Journal* **359**, 159–167. Available from: <https://www.sciencedirect.com/science/article/abs/pii/S1385894718323908>.
- Magnusson, K. & Wahlberg, C. 2014 *Screening of Microplastic Particles in and Downstream of A Wastewater Treatment Plant*. Swedish Environmental Research Institute, Stockholm, Sweden. Technical Report published for IVL Swedish Environmental Research Institute, August 2014.
- Mason, S., Garneau, D., Sutton, R., Chu, Y. & Ehmann, K. 2016 Microplastic pollution is widely detected in US municipal wastewater treatment plant effluent. *Environmental Pollution* **218**, 1045–1054.
- Mathalon, A. & Hill, P. 2014 Microplastic fibers in the intertidal ecosystem surrounding Halifax Harbour, Nova Scotia. *Marine Pollution Bulletin* **81**, 69–79.
- Michielssen, M. R., Michielssen, E. R., Ni, J. & Duhaime, M. 1970 Fate of microplastics and other small anthropogenic litter (SAL) in wastewater treatment plants depends on unit processes employed: Semantic Scholar. *Environmental Science:*

- Water Research & Technology* 2, 1064–1073. Available from: <https://www.semanticscholar.org/paper/Fate-of-microplastics-and-other-small-anthropogenic-Michielsen-Michielsen/3a09944551ba00580f2e7810581a7ddc292ee475>
- Mintenig, S. M., Int-Veen, I., Loder, M. G., Pimpke, S. & Gerdt, G. 2017 Identification of microplastic in effluents of waste water treatment plants using focal plane array-based micro-Fourier-transform infrared imaging. *Water Research* 108, 365–372.
- Naqash, N., Prakash, S. & Kapoor, D. 2020 Interaction of freshwater microplastics with biota and heavy metals: a review. *Environmental Chemistry Letters* 18, 1813–1824. <https://doi.org/10.1007/s10311-020-01044-3>
- Nishikida, K. & Coates, J. 2003 Infrared and Raman analysis of polymers. In: *Handbook of Plastics Analysis* (Lobo, H. & Bonilla, J. V., eds). Marcel Dekker, Inc., New York, NY, pp. 186–316.
- Noda, I., Dowrey, A. E., Haynes, J. L. & Marcott, C. 2007 Group frequency assignments for major infrared bands observed in common synthetic polymers. In: *Physical Properties of Polymers Handbook* (Mark, J. E., ed.). Springer Science+Business Media, New York, NY, pp. 395–406.
- Noguchi, H., Oo, M. H., Niwa, T., Fong, E., Yin, R. & Supaat, N. 2019 Applications of flat sheet ceramic membrane for surface water and seawater treatments – introduction of performance in large-scale drinking water plant and seawater pretreatment pilot system in Singapore. *Water Practice and Technology* 14(2), 289–296. doi:10.2166/wpt.2019.013.
- Nouha, K., Kumar, R. S., Balasubramanian, S. & Tyagi, R. D. 2018 Critical review of EPS production, synthesis and composition for sludge flocculation. *Journal of Environmental Sciences* 66, 225–245. doi:10.1016/j.jes.2017.05.020.
- Patel, M. M., Goyal, B. R., Bhadada, S. V., Bhatt, J. S. & Amin, A. F. 2009 Getting into the brain: approaches to enhance brain drug delivery. *CNS Drugs* 23, 35–58. doi:10.2165/0023210-200923010-00003.
- Phillips, M. B. & Bonner, T. H. 2015 Occurrence and amount of microplastic ingested by fishes in watersheds of the Gulf of Mexico. *Marine Pollution Bulletin* 100(1), 264–269.
- Poerio, T., Piacentini, E. & Mazzei, R. 2019 Membrane processes for microplastic removal. *Molecules* 24(22), 4148. doi:10.3390/molecules24224148.
- Possatta, F. E., Barletta, M., Costa, M. F., do Sul, J. A. I. & Dantas, D. V. 2011 Plastic debris ingestion by marine catfish: an unexpected fisheries impact. *Marine Pollution Bulletin* 62, 1098–1102.
- Reisser, J., Shaw, J., Wilcox, C., Hardesty, B. D., Proietti, M. & Thums, M. 2013 Marine plastic pollution in waters around Australia: characteristics, concentrations, and pathways. *PLoS One* 8(11), e80466. <https://dx.doi.org/10.1371/journal.pone.0080466>.
- Rolsky, C., Kelkar, V., Driver, E. & Halden, R. U. 2019 Municipal sewage sludge as a source of microplastics in the environment. *Current Opinion in Environmental Science & Health* 14, 16–22. Available from: <https://www.sciencedirect.com/science/article/abs/pii/S2468584419300832>
- Salerno, C., Vergine, P., Berardi, G. & Pollice, A. 2016 Influence of air scouring on the performance of a Self Forming Dynamic Membrane BioReactor (SFD MBR) for municipal wastewater treatment. *Bioresour Technol* 223, 301–306. Available from: <https://www.sciencedirect.com/science/article/abs/pii/S0960852416314602>
- Sun, J., Dai, X., Wang, Q., van Loosdrecht, M. C. & Ni, B. 2019 Microplastics in wastewater treatment plants: detection, occurrence and removal. *Water Research* 152, 21–37. Available from: <https://www.sciencedirect.com/science/article/abs/pii/S0043135418310686>
- Talvitie, J., Mikola, A., Setälä, O., Heinonen, M. & Koistinen, A. 2017 How well is microlitter purified from wastewater? – A detailed study on the stepwise removal of microlitter in a tertiary level wastewater treatment plant. *Water Research* 109, 164–172. Available from: <https://www.sciencedirect.com/science/article/abs/pii/S0043135417305687>
- Verleye, G. A., Roeges, N.P. & De Moor, M. O., 2001 *Easy Identification of Plastics and Rubbers*. Rapra Technology Limited, Shropshire, UK, pp. 174.
- Wang, F., Wong, C. S., Chen, D., Lu, X., Wang, F. & Zeng, E. Y. 2018 Interaction of toxic chemicals with microplastics: a critical review. *Water Research* 139, 208–219. Available from: <https://www.sciencedirect.com/science/article/abs/pii/S0043135418302835>
- Water Environment Federation 1999 *Standard Methods for the Examination of Water and Wastewater: 6430*. APHA/AWWA/WEF, Washington, DC. Available from: <http://scholar.google.com/scholar?hl=en&btnG=Search&q=intitle:Standard+methods+for+the+examination+of+water+&+wastewater#0>
- Wesch, C., Barthel, A.-K., Braun, U., Klein, R. & Paulus, M. 2016 No microplastics in benthic eelpout (*Zoarces viviparus*): an urgent need for spectroscopic analyses in microplastic detection. *Environmental Research* 148, 36–38.
- Wright, S. L., Thompson, R. C. & Galloway, T. S. 2013 The physical impacts of microplastics on marine organisms: a review. *Environmental Pollution* 178, 483–492.
- Xia, W., Rao, Q., Deng, X., Chen, J. & Xie, P. 2020 Rainfall is a significant environmental factor of microplastic pollution in inland waters. *Science of The Total Environment* 732, 139065. doi:10.1016/j.scitotenv.2020.139065.
- Yang, L. B., Li, K. X., Cui, S., Kang, Y., An, L. H. & Lei, K. 2019 Removal of microplastics in municipal sewage from China's largest water reclamation plant. *Water Research* 155, 175–181. <https://doi.org/10.1016/j.watres.2019.02.046>
- Ziajahromi, S., Neale, P. A., Rintoul, L. & Leusch, F. D. 2017 Wastewater treatment plants as a pathway for microplastics: development of a new approach to sample wastewater-based microplastics. *Water Research* 112, 93–99. Available from: <https://www.sciencedirect.com/science/article/abs/pii/S0043135417300489?via=ihub>

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