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RESEARCH ARTICLE

Fate of microplastics in a coastal wastewater treatment plant: Microfibers could partially break through the integrated membrane system

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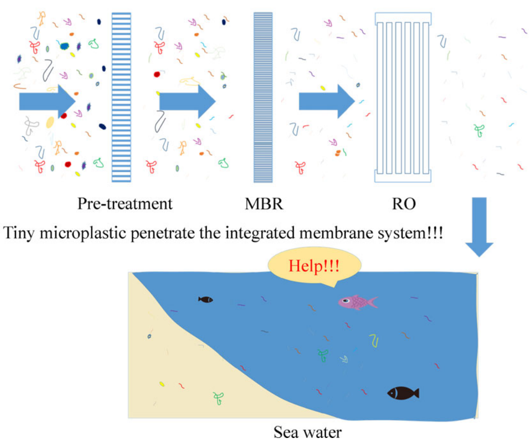
HIGHLIGHTS

- Fate of microplastics in integrated membrane system for water reuse was investigated.
- Integrated membrane system has high removal efficiency (>98%) for microplastics.
- Microplastics (>93%) were mainly removed through membrane bioreactor treatment.
- Small scale fiber plastics (< 200 μm) could break through reverse osmosis (RO) system.
- The flux of microplastics maintained at 2.7 × 10^{11} MPs/d after the RO treatment.

GRAPHIC ABSTRACT



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ABSTRACT

Rare information on the fate of microplastics in the integrated membrane system (IMS) system in full-scale wastewater treatment plant was available. The fate of microplastics in IMS in a coastal reclaimed water plant was investigated. The removal rate of microplastics in the IMS system reached 93.2% after membrane bioreactor (MBR) treatment while that further increased to 98.0% after the reverse osmosis (RO) membrane process. The flux of microplastics in MBR effluent was reduced from 1.5 × 10^{13} MPs/d to 10.2 × 10^{11} MPs/d while that of the RO treatment decreased to 2.7 × 10^{11} MPs/d. Small scale fiber plastics (< 200 μm) could break through RO system according to the size distribution analysis. The application of the IMS system in the reclaimed water plant could prevent most of the microplastics from being discharged in the coastal water. These findings suggested that the IMS system was more efficient than conventional activated sludge system (CAS) for the removal of microplastics, while the discharge of small scale fiber plastics through the IMS system should also not be neglected because small scale fiber plastics (< 200 μm) could break through IMS system equipped with the RO system.

1 Introduction

The majority of marine microplastic waste comes from land. According to statistics, 10% of land plastic waste enters the ocean every year, and most of these micro-plastics enter the ocean through waste water treatment plants. Microplastic (defined to be less than 5 mm) is a

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growing environmental pollution problem (Baldwin et al., 2016; Liu and Wang, 2020). The increase in the amount of plastic waste can cause microplastics to accumulate in various ecosystems and environments include oceans, coastal areas and inland (Xu and Ren, 2021). These microplastics waste may come from the plastic industry (Lares et al., 2018), or from personal care products and the washing of synthetic textiles (Browne et al., 2011). Almost no environment on earth can escape plastic pollution (Taylor et al., 2016). There are many studies on the abundance of microplastics in the ocean (Zhang et al., 2020a; Shi et al., 2021), coastal (Lu et al., 2019; Lu et al., 2020a) and freshwater environment. Microplastics are generally more abundant in densely populated areas (Browne et al., 2011). Microplastics have attracted more and more attention due to their physical hazards and interactions with other pollutants (Wu et al., 2019).

Many animals and plants ingest microplastics from the environment (van Weert et al., 2019; Xue et al., 2021). After being ingested by animals, microplastics can block their digestive tract, destroy the gastric mucosa, reduce food intake, and eventually cause the animal to starve and die (Taylor et al., 2016). Microplastics have been detected in more than 2000 marine organisms (Jabeen et al., 2017). In addition, microplastics release a large number of chemicals, which have an impact on organisms (Koelmans et al., 2013). Microplastics are hydrophobic and more susceptible to being adsorbed on many chemicals such as endocrine disruptors, antibiotics and other organic pollutants in water (Qu et al., 2018). There are many reports on the adsorption of various types of pollutants by microplastics (Chen et al., 2019; Zuo et al., 2019). Liu et al. (2019) studied the adsorption properties and mechanism of UV-aged microplastics. They also studied the effects of salinity and pH on the adsorption capacity of microplastics (Liu et al., 2019). Camacho et al. (2019) investigated the adsorption of 81 compounds on the Canary Islands microplastics (Camacho et al., 2019).

Various studies have shown that waste treatment plant is the most important way for the discharge of various emerging contaminants including microplastics into the environment (Mintenig et al., 2017; Lu et al., 2020b; Zhang et al., 2020b; Lu et al., 2021a). It was difficult to use traditional wastewater treatment methods such as advanced oxidation and biofiltration (Lu et al., 2020b, Lu et al., 2021b) to remove microplastics. How to prevent microplastics from entering the environment is still a challenge. Membrane technology is a prospective treatment method for various pollutants removal in wastewater treatment process (Guo et al., 2019; Ding et al., 2020; Lu et al., 2020b; Wang et al., 2020). In recent years, membrane technology has been widely used in wastewater treatment, and has been growing rapidly (Lu et al., 2020b). Membrane technology has a good removal rate for COD (Sun et al., 2014), NH_4^+-N, bacteria (Chaudhry et al., 2015), organic pollutants, antibiotic resistance genes (Lu et al., 2020b). With the shortage of water resources and water pollution, the IMS technology for reclaimed water reuse has attracted more and more attention (Lu et al., 2020b).

Whether the waste water treatment plant can prevent the microplastics from entering the marine environment is a crucial problem. There is still rare information on the fate of microplastics in the IMS system used for water reclamation. In this paper, the fate of microplastics in CAS and IMS in a coastal reclaimed water plant was investigated. The removal behaviors of microplastics in traditional wastewater treatment process and membrane technology were compared. In addition, the type, shape and particle size of microplastics in water samples were also studied.

2 Materials and methods

2.1 Description of the coastal wastewater treatment plant

The coastal reclaimed water plant is located in Yantai City, Shandong Province, China. It is divided into phase I and phase II projects. The phase I project is the CAS, including grille, aerated sand sink, primary sedimentation tank, biological tank, secondary sedimentation tank, UV disinfection etc. Water treatment capacity is 2.5×10^8 m^3/d. The effluent is discharged into the tail water discharge area of the surrounding sea. The phase II reclaimed water project is semi-underground, uses IMS on the basis of CAS, treatment capacity is 1.5×10^8 m^3/d. The CAS consists of grille, aerated sand sink, primary sedimentation tank, biological tank, MBR tank, ultrafiltration (UF) (pretreatment of RO) and RO system. The MBR system composed of PVDF hollow fiber membrane with pore size of 0.4 μm, and the water treatment capacity is 1.50×10^8 m^3/d. The RO system composed of flat membrane with pore size of 0.0001 μm. Water treatment capacity of RO treatment is 4.0×10^7 m^3/d. The MBR effluent is discharged into the tail water discharge area. The recycled water produced by RO process could be reused as industrial and greening water.

2.2 Sampling collection

Water samples were taken from the coastal waste water treatment plant (WWTP). Samples obtained from the CAS were collected from the influent of CAS (A1), primary sedimentation treatment of CAS (A2), and secondary treatment of CAS (A3). Samples obtained from the IMS system were collected from the influent of IMS (M1), primary sedimentation treatment of IMS (M2), MBR outlet pond (M3), and from RO outlet pond (M4). Surface water samples in area of the surrounding sea were collected from the discharge area (S1) and 2.0 km away from discharge area (S2) (Fig. 1). S1 and S2 are far away from the coast while S2 is far away from the coast than S1. There is no wastewater discharge from around so that they are not polluted by other microplastics.

2.3 Sampling processing

The samples were poured to four stacked stainless sieves with mesh sizes of 5 mm, 1.25 mm, 0.375 mm and 0.075 mm. Total 5-L waste water and 100-L for seawater were collected at each sampling point. Wash the sieves with as little deionized water (8–20 μs/cm, Wahaha, China) as possible and then transfer all the particles to glass beakers. The particles were dried in a drying oven at 80°C until completely dry. Digest the particles using 30% H_2O_2 (GR, Sinopharm Chemical Reagent Co., Beijing, China) in the presence of Fe^{2+} catalyst (wet peroxide oxidation, WPO) (Lares et al., 2018). The samples were treated by vacuum filtration use a glass microfiber filters (GE Whatman 1825-047, UK) and dried at room temperature.

To reduce contamination, all utensils are made of stainless steel or glass. They are cleaned with ultra-pure water and covered with aluminum foil before using. In addition, all participants in the sampling and experiment were uniformly dressed in cotton clothes. Ultra-pure water was used as a blank sample in the laboratory. There was no microplastics were detected in the blank sample, showing that the analysis is reliable.

2.4 Characterization and analysis of microplastics

All samples were examined and taken photos under a stereo microscopes (Leica S9i stereo zoom, Switzerland) with dedicated image software (LAS version, Leica). All possible microplastics were collected and detected via a FTIR spectrometer (Thermo Scientific Nicolet iN10 Infrared Microscope, USA). The FTIR spectra was tested from 500 to 4000 cm^{-1} at room temperature.

3 Results and discussion

3.1 Morphology of microplastics in the IMS system

Figures 2 and 3 showed the morphology and distribution of microplastics at each sampling point in the reclaimed water plant. Figure 2 shows the photos of appearance microplastics obtained in this study. According to the physical shape of microplastics, the particles can be divided into four main morphologies, includes fragments (Figs. 2(a)–2(c)), film (Figs. 2(d) and 2(e)), pellet (Fig. 2(f)) and fibers (Figs. 2(g)–2(i)). Among them, the morphology of microplastics in influent water was mainly fibers and fragments, which accounted for 80%–90% of the total amount, and the proportion of fragments and fibers is about the same. The shape distribution of microplastics in the effluent of traditional activated sludge process is similar to that in the influent (Lares et al., 2018). The distribution of microplastics in the nearby seawater is similar. Different from the CAS process, the proportion of fiber microplastics in the effluent of membrane technology system gradually increased, especially after RO membrane treatment, the proportion of fiber microplastics accounted for 94% (Fig. 3). This could be explained by that large-piece microplastics can not penetrate the membrane after

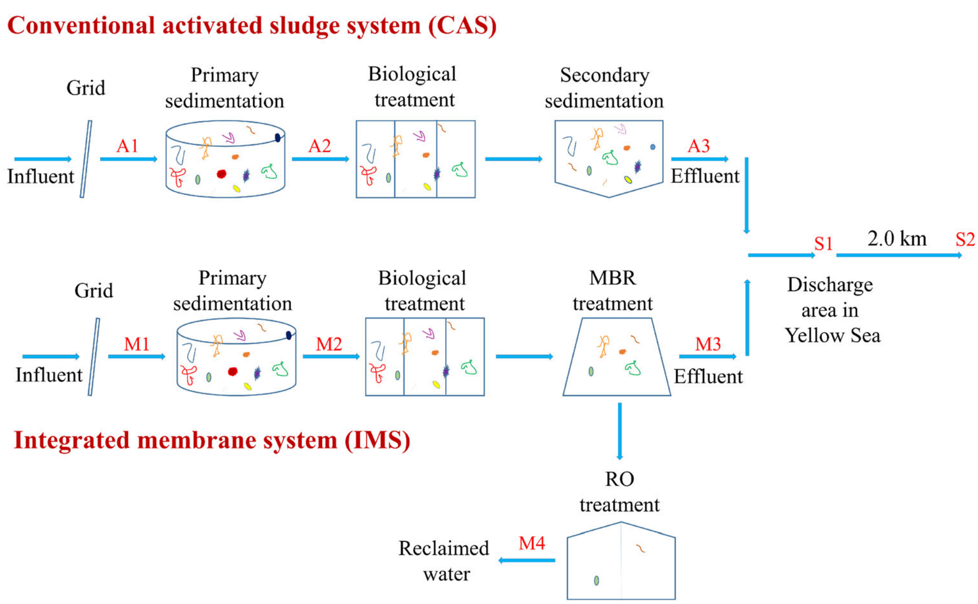

Fig. 1 Wastewater treatment process at the coastal reclaimed water plant. M1: Influent of IMS; M2: Primary treatment of IMS; M3: MBR treatment of IMS; M4: RO treatment of IMS; A1: Influent of CAS; A2: Primary treatment of CAS; A3: Secondary treatment of CAS; S1: discharge area of the Yellow Sea; S2: 2.0 km away from discharge area.

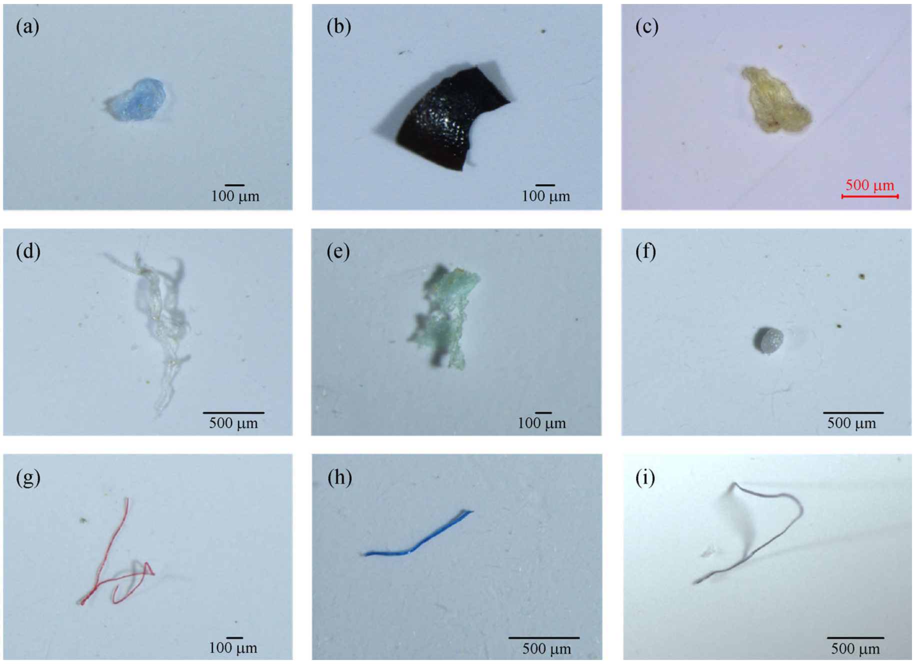

Fig. 2 Photos of appearance microplastics found in different sampling point at the coastal reclaimed water plant. (a–c) fragments; (d and e) films; (f) pellet; (g–i) fibers.

membrane filtration. Only fibrous microplastics with very small cross section can penetrate the membrane and enter the outlet so that the relative content of fiber microplastics is increased. The influent (M1) of IMS contained film (2%), pellet (8%), fragment (32%) and fiber (58%). After MBR treatment, the effluent (M3) contained film (3%), pellet (18%), fragment (33%) and fiber (46%). The reclaimed water after RO treatment (M4) contained film (2%), pellet (0%), fragment (4%) and fiber (94%). After the multi-stage membrane treatment, almost all of the spherical microplastics were removed, the film and fragment microplastics are also greatly reduced. The microplastics remained in the effluent were mainly fibers. For CAS, the content of microplastics of fragments shape was relatively high, followed by fibrous shape, microplastics with film shape were the least (A1 and A3). There was no obvious change in the morphology of microplastics in the CAS. The content of fibrous microplastics in the treated water does not change significantly, the proportion of other three kinds of granular microplastics changed slightly (A3). Among them, the proportion of fiber in CAS effluent has little change, still 40%. The proportion of fibrous microplastics in MBR effluent accounts for 46% of the total. After further using RO technology, the proportion of fiber increased to 94%. Compare with traditional technology, the membrane technology has a good removal effect on both pellets, films and fragments microplastics. Fibrous microplastics are difficult to be removed. The fiber microplastics are microfibers like needles that might penetrate through the membrane easily. The shape of microplastics in the discharge area (S1) was film (10%), pellet (6%), fragment (48%) and fiber (36%). IT could not distinguish the MPs from CAS and IMS because S1 possessed the same discharge area for both CAS and IMS. However, the distribution proportion of fiber microplastics in water samples (M3, M4) after membrane treatment was greatly improved, illustrating that membrane technology, especially reverse osmosis technology, has a good removal effect on other shapes of microplastics. The shape of microplastics in the 2.0 km away from discharge area (S2) is as follows, film (13%), pellet (2%), fragment (53%) and fiber (32%). This result is similar to that of A3, which demonstrated that the wastewater discharge of traditional wastewater treatment plant does have a great impact on the surrounding environment (Fig. 3).

3.2 Type and abundance of microplastics in the IMS system

A total of five types of common microplastics were detected and identified in this study, including polypropylene (PP), polyethylene (PE), polyamide (PA), polyethylene terephthalate (PET), polyvinyl chloride (PVC). The infrared spectras of five microplastics are shown in Fig. 4. Type distribution of microplastics was shown in Fig. 5. The total abundance in influent was 96.6 and 81.8 MPs/L at sampling points M1 and A1 respectively. In the IMS, MBR membrane technology replaced the traditional secondary sedimentation treatment, and the concentration of microplastics in effluent dropped significantly to 6.6 MPs/L, the removal rate reached to 93.2% (M3). For RO membrane process (M4), the removal rate of microplastics was further increased to 98.0%, the effluent is directly

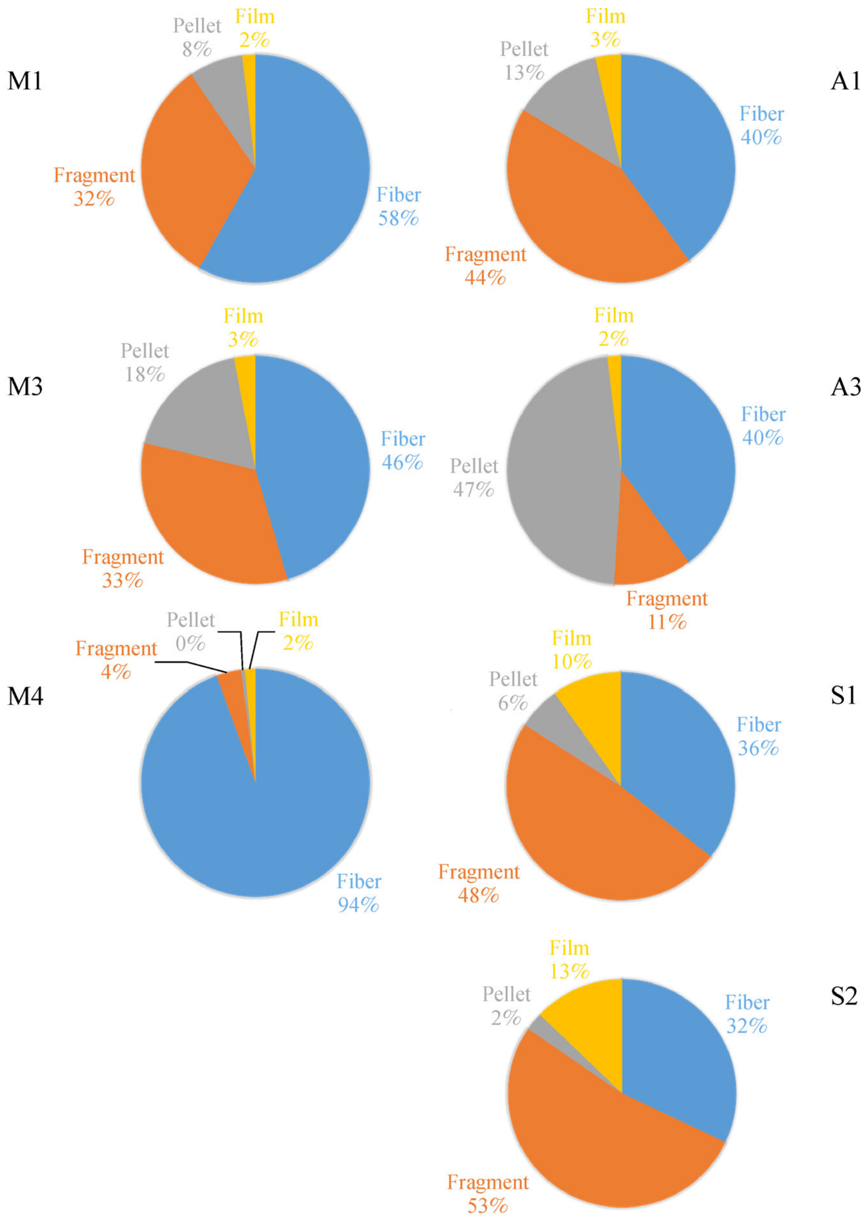

Fig. 3 Shape fraction of microplastics at the coastal reclaimed water plant. M1: Influent of IMS; M3: MBR treatment of IMS; M4: RO treatment of IMS; A1: Influent of CAS; A3: Secondary treatment of CAS; S1: discharge area of the Yellow Sea; S2: 2.0 km away from discharge area.

transported to the factory for reuse. For CAS, after primary and secondary treatment, the concentration of microplastics in effluent of CAS was reduced to 30.6 MPs/L, and the removal rate is only 62.6%. The abundance of microplastics in effluent is still high, far below the IMS. The more advanced the water treatment process, the higher the removal rate of microplastics. This study shows that the overall removal rate of IMS is much higher than that of the CAS. The membrane process has a higher removal rate of microplastics in wastewater than the traditional process (Lares et al., 2018). IMS greatly improves the removal rate of microplastics in WWTPs. At the same time, IMS can prevent microplastics from entering the environment and convert wastewater into usable reclaimed water. To investigate the influence of WWTPs on the surrounding sea area, the microplastics in the discharge area and 2.0 km away from discharge area were examined. The abundance of microplastics in discharge area of is 4.30 MPs/L, which is only 0.78 MPs/L at 2.0 km away from discharge area. It can be seen that the microplastic pollutants in seawater mainly come from the discharge of waste treatment plant. Considering that the water treatment process with IMS has a good removal efficiency on the microplastics, especially the fibrous microplastics which are difficult to remove in the traditional process, the membrane process is recommended as the main process of the sewage treatment plant

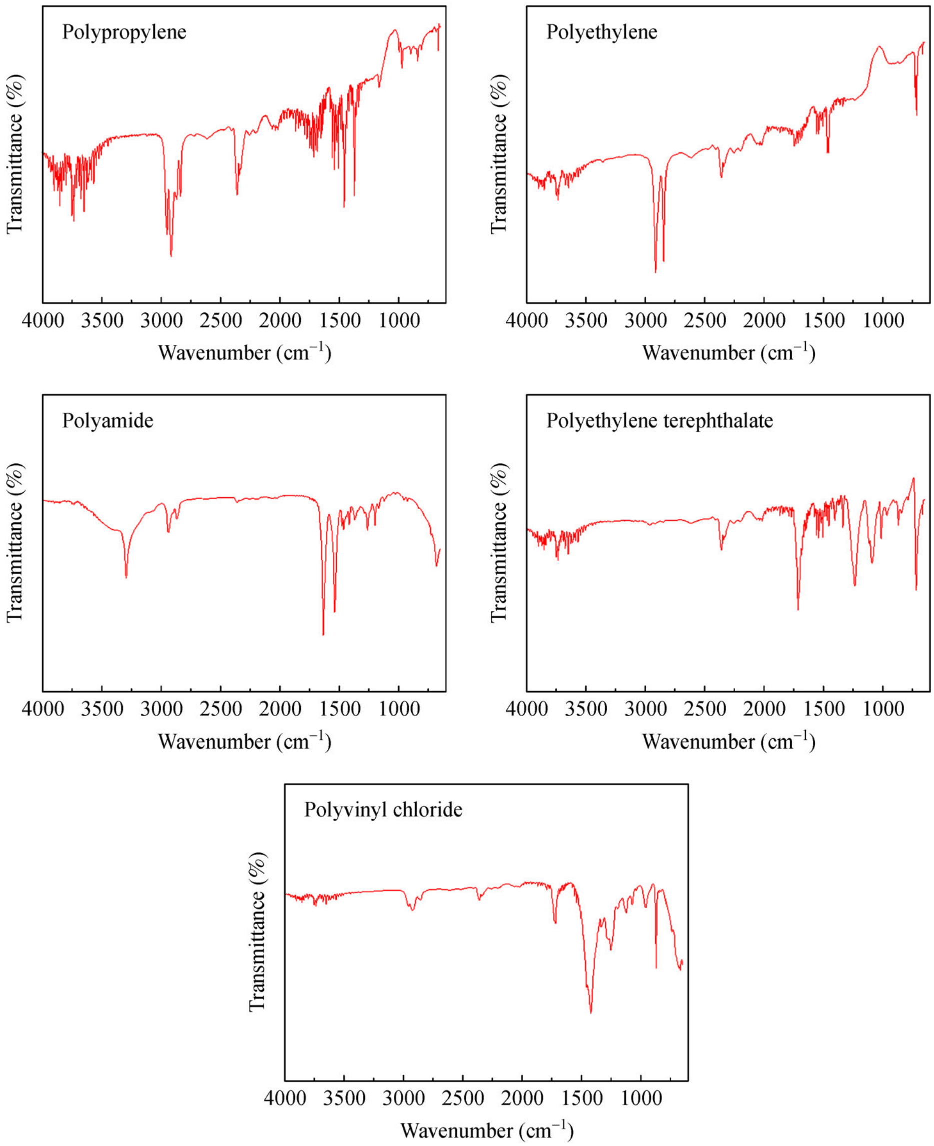

Fig. 4 Infrared spectras of detected microplastics: polypropylene (PP), polyethylene (PE), polyamide (PA), polyethylene terephthalate (PET), polyvinyl chloride (PVC).

in the coastal area. It can be seen that wastewater discharge from CAS does affect the abundance of microplastics in the surrounding sea area. Discharges from wastewater treatment plants do have impact on the surrounding environment. Therefore, membrane technology has a good application prospect in wastewater treatment.

The waste treatment plant mainly deals with the domestic waste water of Yantai and its surrounding areas. In all water samples, the highest content of microplastics was PE and PET while the lowest content was PVC (Fig. 5). PE is the most produced plastic polymer in the world, and it is also is the main part of plastic used in our daily life. PE in wastewater mainly exists in the form of fragments, spheres and films, which may come from product packaging, daily necessities and personal care products (Browne et al., 2011). PET mainly exists in the form of fibers, and these may come from washing wastewater (Carr et al., 2016; Ziajahromi et al., 2017). This is because PET is the first choice material for clothing in synthetic fiber, it provides strength and durability to clothes (Gundogdu et al., 2018). Domestic washing machines can release thousands of fiber microplastics in each washing cycle. It is estimated that about 160 million fiber microplastics will be discharged into the ocean (Lares et al., 2018). The abundance of PE in M1 was 32.4 MPs/L, which is reduced to 2.6 MPs/L (M3) after MBR treatment, after RO treatment it reduced to 0.1 MPs/L (M4), the removal rate is up to 99.7%. PET decreased from 37.8 to 1.7 MPs/L (M4), the removal rate reached to 95.5%. For CAS, the removal rates of PE and PET are only 51.8% and

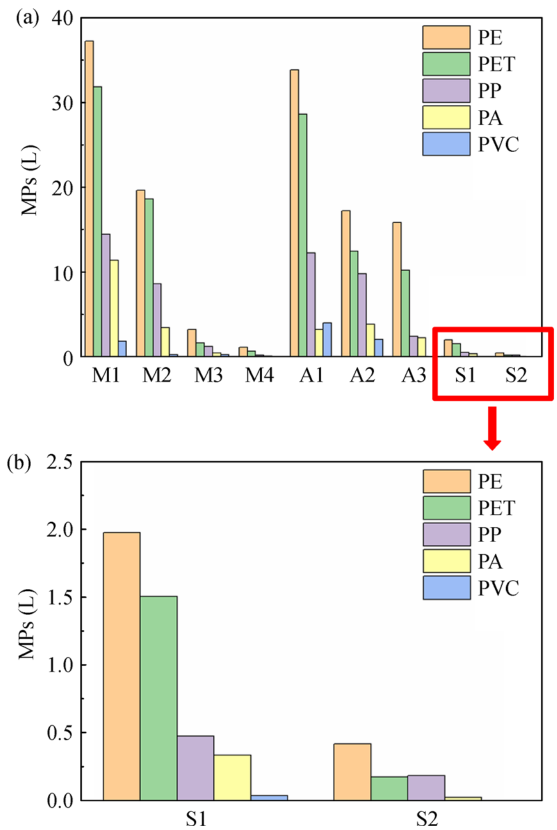

Fig. 5 Type distribution of microplastics at the coastal reclaimed water plant. M1: Influent of IMS; M2: Primary treatment of IMS; M3: MBR treatment of IMS; M4: RO treatment of IMS; A1: Influent of CAS; A2: Primary treatment of CAS; A3: Secondary treatment of CAS; S1: discharge area of the Yellow Sea; S2: 2.0 km away from discharge area. (a) Type distribution of microplastics at all sampling points; (b) Type distribution of microplastics in sea water.

65.5%, respectively. The abundance of microplastics in effluent is still high. There are still 15.8 MPs/L PE and 10.2 MPs/L PET in its effluent, there effluent will be discharged into the sea. In IMS, the removal rate of PE and PET is very high, and much higher than that of CAS. This shows that IMS system has more advantages than traditional technology in dealing with all types of microplastics. In addition, the content of PE and PET in the effluent of CAS process is relatively high, which after the IMS treatment, PE and other particle microplastics are almost removed, the abundance of PET fiber is the most. The abundance of PE and PET is 2.0 MPs/L and 1.5 MPs/L in S1, while the abundance of PE and PET in S2 is reduced to 0.4 MPs/L and 0.2 MPs/L. The concentration of PE and PET in the wastewater discharge area is much higher than that 2.0 km away from the discharge area, which indicates that the abundance of microplastics in the surrounding waters of the discharge area is affected by the discharge of the wastewater treatment plant.

3.3 Abundance and size distribution of microplastics in the IMS system

Abundance and size distribution of microplastics in IMS and CAS at the wastewater treatment plant was shown in Fig. 6. To better illustrate the advantages of IMS, the size distributions of fibers and non-fibers were analyzed separately. Fiber-shaped microplastics have very small cross-sectional size relative to the other three types, which can easily penetrate through the membrane holes and enter the water outlet. In Fig. 6, M1, M3 and M4 showed the size distribution of microplastics in IMS. The size of non-fiber microplastics in the influent of IMS is mainly 0–400 μm. The removal of large particles is better than that of small particles, the size of microplastics in effluent tends to decrease. Among them, 50–100 μm microplastic was 28 MPs/L in influent (M1). After MBR treatment, the abundance of 50–100 μm microplastic was reduced to 2.4 MPs/L, the removal rate was 91% (M3). After RO treatment, there are less than 0.06 MPs/L of 50–100 μm in reclaimed water, the removal rate increased to 99.8% (M4), the main size of microplastics in effluent is between 0 and 450 μm. The removal rate of non-fiber microplastics is higher than that of fiber microplastics. The size of fiber microplastics in the influent of IMS is widely distributed, ranging from 0 to 5000 μm. The fibers with the largest content of 500–1000 μm decreased from 17 MPs/L (M1) to 0.8 MPs/L (M4), the removal rate was as high as 95%. Figure 6 (A1 and A3) shows the size distribution of microplastics in CAS. Compared with the influent (A1), the microplastics abundance in effluent of various sizes was decreased (A3). Among them, fibers with size between 500 to 1000 μm decreased from 10 to 4 MPs/L, the removal rate reached to 60%. After the first, second and even membrane treatment, the large-scale microplastics are relatively reduced, and the proportion of small-scale microplastics is increased, indicating that the removal rate of large-scale microplastics by water treatment process is better than that of small-scale microplastics. In the study of Lars et al. (Lares et al., 2018), 64% of microplastics are less than 1 mm, half of which are less than 0.5 mm. Microplastics are mainly between 0.5 and 1 mm in the final effluent. However, in this study, we divide microplastics into fiber and non-fiber, and studied separately. It is found that the large particles (greater than 0.5 mm) in the non-fiber microplastics have been completely removed from the wastewater treated by membrane technology (MBR and RO). The large-scale microplastic particles are mainly from fibers microplastics. Although the size of fibers is large, their diameter is very small, usually about 10 μm, which is easy to pass through the membrane and stay in the final effluent. Also in CAS, the removal rate of non-fiber particles is better than that of large size fiber microplastics. Compared with CAS, the removal of all sizes of microplastics especially large size has been greatly

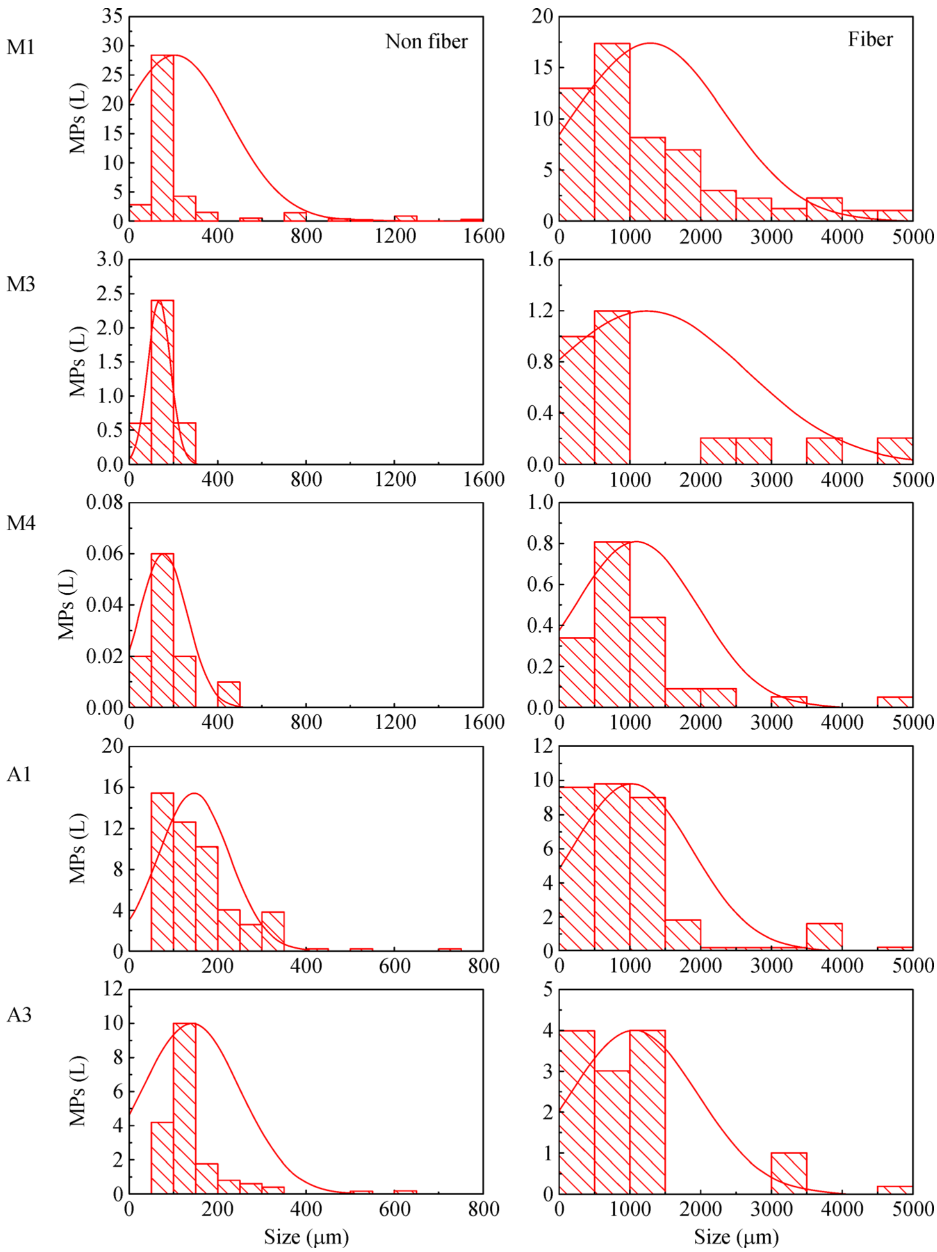

Fig. 6 Size distribution of microplastics in IMS and CAS at the coastal reclaimed water plant. M1: Influent of IMS; M3: MBR treatment of IMS; M4: RO treatment of IMS. A1: Influent of CAS; A3: Secondary treatment of CAS.

improved in IMS, both fibers and non-fiber, especially the removal efficiency of IMS for non-fiber. With the introduction of membrane technology, the concentration of microplastics through MBR decreased rapidly, especially for the large particle microplastics and RO effluent, the microplastics were almost completely removed. It can be seen that membrane technology plays an important role in the removal of microplastics in wastewater, and the removal rate increases with the decrease of average membrane pore size and type. This result is consistent with the previous research results on the removal of other emerging particle pollutant such as the antibiotic resistance genes (ARGs) from waster by membrane technology (Lu et al., 2020b).

3.4 Daily flux and removal efficiency of microplatics in the IMS system

According to the water treatment capacity of the wastewater treatment plant, daily flux and removal efficiency of microplatics in IMS (M1, M2, M3 and M4) and CAS (A1, A2 and A3) and was shown in Fig. 7. Results showed that the flux of microplastics in influent of the WWTP was 1.5 $\times$ 10^{13} MPs/d in IMS (M1) while that in MBR effluent

(M3) was reduced to 10.2×10^{11} MPs/d (Fig. 7(a)) with the removal rate of 93.2% (Fig. 7(b)). As to RO treatment (M4), the daily flux decreased to 2.7×10^{11} MPs/d (Fig. 7 (b)). Compared with the IMS, the flux of microplastics in influent of the WWTP were 2.0×10^{13}/d at A1. The daily flux in effluent of CAS reduced to 7.5×10^{12} (A3). The daily flux of microplastics in the effluent varied with the water treatment capacity, influent concentration, treatment technique, and so on. In this work, the IMS is more effective than the CSA in removing microplastics in wastewater treatment process. Most of the microplastics are discharged with the sludge, and only a few permeate RO treatment to enter the reclaimed water. IMS has the same effect on the removal of antibiotic resistance genes (ARGs). The IMS has been proved to have high removal efficiency for the removal of ARGs. The IMS has a good removal of total daily flux of ARGs, which were 1–3 orders of magnitude lower than that in the CAS system (Lu et al., 2020b). Microplastics are small-size particles to be more susceptible to adsorbing antibiotic resistance genes (Lu et al., 2019, Lu et al., 2022). For these reasons, the removal behaviors of microplastics using membrane process were similar. Considering the water shortage in coastal areas, the IMS water reuse system has certain advantages for the removal of similar emerging particulate pollutants.

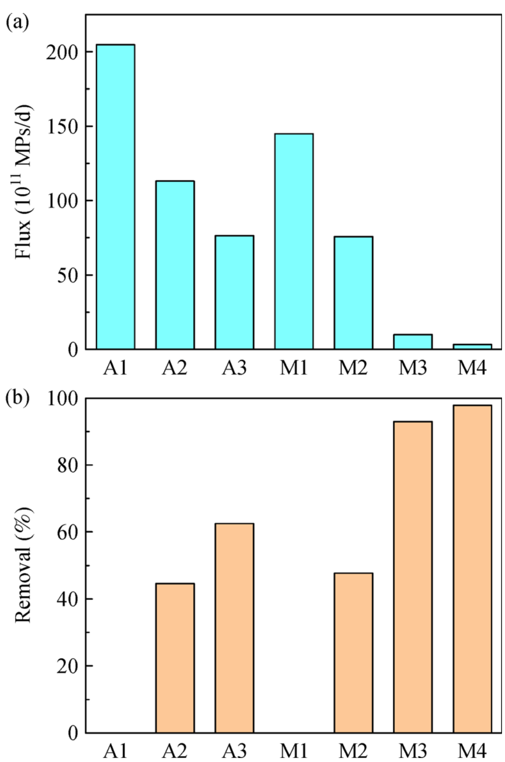

Fig. 7 Daily flux (a) and removal efficiency (b) of microplatics in IMS (M1, M2, M3 and M4) and CAS (A1, A2 and A3) at the reclaimed water plant.

4 Conclusions

The fate of microplastics in traditional water treatment process and in the membrane technology of typical reclaimed water plant in coastal zone were studied. The results show that the removal rate of microplastics by IMS is much higher than that by traditional wastewater treatment process. Membrane treatment has been a good removal rate for microplastics with different type, size and shape. The introduction of IMS into coastal wastewater treatment plants has advantages. Because IMS system could prevent the re-entry of most of the microplastics into the marine environment and convert the wastewater into renewable water, which can subsequently reduce pollution to the ocean and solve the shortage of water resources. Since small scale fiber plastics (<200 μm) could break through RO system with a flux maintained at 2.7×10^{11} MPs/d, the discharge of small scale fiber plastics should not be neglected. Novel separation membrane with low cost and high efficiency for the removal of microplastics is in urge need.

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