

Understanding Microplastic Presence in Different Wastewater Treatment Processes: Removal Efficiency and Source Identification

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Abstract

Municipal effluents discharged from wastewater treatment plants (WWTPs) are a considerable source of microplastics in the environment. The dynamic profiles of microplastics in treatment units in WWTPs with different treatment processes remain unclear. This study quantitatively analyzed microplastics in wastewater samples collected from different treatment units in two tertiary treatment plants with distinct processes. The influents contained an average of 15.5 ± 3.5 particles/L and 38.5 ± 2.5 particles/L in the two WWTPs with in the oxidation ditch process and the integrated fixed-film activated sludge process, respectively. Interestingly, microplastic concentrations in the influent were more influenced by the population density in the served area than sewage volume or served population equivalent. Throughout the treatment process, concentrations were reduced to 1.5 ± 0.5 particles/L and 1.0 ± 1.0 particles/L in the final effluents, representing an overall decrease of 90% and 97%, in WWTPs with the oxidation ditch process and integrated fixed-film activated sludge process, respectively. A significant proportion of the microplastics were removed during the primary treatment stage in both WWTPs, with better performance for foam, film, line-shaped and large-sized microplastics. Most microplastics were accumulated in activated sludge, indicating its key role as the primary sink in WWTPs. The multiple correspondence analysis identified laundry washing and daily necessities such as packaging and containers as the major contributors to microplastics in WWTPs. The study proposed recommendations for upgrading WWTPs, modifying designs, and implementing strategies to reduce microplastic sources, aiming to minimize the release of microplastics into the environment. These findings can shed lights on the sources of microplastics in WWTPs, and advance our understanding of the mechanisms for more effective microplastic removals in wastewater treatment technologies in future applications.

Keywords: Microplastics; Population density; Removal; Source apportionment; WWTP upgrade

1. Introduction

Microplastics (MPs), defined as plastic particles measuring less than 5 mm, have emerged as a growing concern due to their potential threat to ecosystems and to human health (Arthur et al., 2009; Thompson et al., 2004; Eerkes-Medrano et al., 2015; Daziroglu and Bilici, 2023). The significant demand and production of plastic products in our modern era have progressively contributed to the issue of plastic pollution. In 2020, a total of 6.51 million tons of plastics for daily necessities were produced by large-scale plastic industrial enterprises (designated as those with an income greater than \$2.78 million per year) in China (CPPIA, 2021). However, due to the fact that the trash separation systems were poorly developed, the plastic recycling rate in China is much lower (below 25%) than those in Japan and Germany (CPPIA, 2021), resulting in the unmanaged discharge of MPs to the environment.

Several studies have reported that wastewater treatment plants (WWTPs) are major sources of MPs in freshwater streams (Dyachenko et al., 2017; Estahbanati and Fahrenfeld, 2016; Murphy et al., 2016; Prata et al., 2021). Liu et al. reported that an average of 7.2 billion MPs discharged into the aquatic environment each day due to the high daily treatment capacity of WWTPs (Liu et al., 2021). Therefore, there is an urgent need to understand the occurrence and removal of MPs in different wastewater treatment processes, and to propose effective strategies for minimizing their source input and enhancing WWTP design. There have been studies examining MPs occurrence, distribution, and removal efficiency in specific WWTPs from cities like Hong Kong (Cao et al. 2020), Wuhan city (Liu et al. 2019), Wuxi city (Lv et al. 2019), and Harbin city (Jiang et al. 2020), significant gaps persist in comprehensively comparing MPs across various treatment processes.

Researchers emphasized that the presence and removal of MPs were impacted by the characteristics of MPs in different treatment processes, to which more attention should be paid (Hamidian et al., 2021; Liu et al., 2021). However, recent research on the transport and fate of MPs across diverse treatment processes has primarily focused on influents and effluents, lacking the detailed analysis of wastewater characteristics in the different treatment units (Conley et al., 2019; Hidayaturrahman and Lee, 2019; Long et al., 2019; Tadsuwan and Babel, 2022), and the varying MP removal efficiencies among different WWTP technologies. The difficulty in comparing the MP removal efficiency among different WWTP technologies lies in the lack of standard methodology for MP characterization, highlighting the need for further systematic investigations to achieve the comparative results (Lv et al., 2019; Sadia et al., 2022).

Furthermore, the influences of different factors such as served equivalent population, commercial and industrial activities, catchment sizes, and adjacent land use on the occurrence, distribution, and variability of MPs in WWTPs require more comprehensive assessment (Conley et al., 2019; Sadia et al., 2022; Sun et al., 2019). MPs in the environment usually come from industrial and domestic sources, including textiles, personal care products, city dust, plastic bags and bottles, fishing wastes, and farming films, which are closely related to industrial productions and human activities (An et al., 2020, IUCN 2017, Lin et al., 2023). Previous studies have reported that there is no correlation between MP abundance and population equivalents (Long et al., 2019), but they overlooked other possible factors such as the types of commerce, human activities, and service area, which could impact MP variability in different WWTPs (Conley et al., 2019; Sun et al., 2019). Therefore, the data of population density in Changchun city is obtained from WorldPop (<https://www.worldpop.org/>) in the current

study to explore the relationship between the existence of MPs and human activities. In addition, previous studies highlighted that source reduction is the most efficient method to reduce MP contamination (Prata, 2018). However, limited studies have tracked the sources of MPs in WWTPs, due to the lack of sophisticated methods to investigate the sources of MPs in WWTPs. For example, potential sources of MPs can be elucidated by highly distinctive and recognizable profiles (such as shape, color, size and polymer) of MP particles (de Carvalho et al., 2021; Fahrenfeld et al., 2019), yet only one or two profiles were used to explore the MP characteristics and sources (Klein et al., 2015; Su et al., 2016). The link between MP characteristics to their specific sources indicated that further analysis on their correlation using approaches such as multiple correspondence analysis (MCA) can shed lights on the source identification of MPs (Fahrenfeld et al., 2019). Therefore, it is imperative to address the sources of MPs, explore control strategies, and enhance existing WWTPs to incorporate MP removal capabilities into treatment processes, given the significant number of MPs that would be released into the environment due to the large discharge volumes of effluent and sludge (Picó et al., 2020).

In this study, the dynamic profiles of MPs in two WWTPs employing two different yet broadly applied core processes, oxidation ditch (OD) and integrated fixed film activated sludge (IFAS), were explored. The OD process in WWTPs facilitates a circular flow of wastewater and activated sludge whereas the IFAS process is a combination of both the attached and suspended biofilm growth systems with the benefits of space efficiency, improved nutrient removal, and enhanced removal of anthropogenic compounds (Waqas et al., 2020; Zhang et al., 2023). The main objectives of this study were (1) to conduct a comprehensive analysis and comparison of MPs characteristics and removal

efficiency across diverse treatment processes, (2) to explore correlations between treatment capacity, catchment area, served population equivalent of a WWTP, and MP concentrations, (3) to trace MP sources using multiple categorical variables, and (4) to offer recommendations for source reduction and retrofitting of WWTPs to prevent MP release.

2. Materials and methods

2.1. WWTP introduction and MP sample collection

This study focused on the assessment of two representative WWTPs in China, including a WWTP-OD and a WWTP-IFAS, both located in Changchun City, Northeast China. Changchun is densely populated and characterized by high industrial and commercial activities as the capital of Jilin Province. The WWTP-OD has a treatment capacity of $1 \times 10^5 \text{ m}^3$ per day, while the WWTP-IFAS has a treatment capacity of $2 \times 10^4 \text{ m}^3$ per day. Detailed information regarding these two WWTPs can be found in Table S1.

Wastewater sampling was carried out across the various treatment stages of each WWTP, including influent (S1), effluent from primary treatment (S2), activated sludge-wastewater mixture in biological treatment (S3), secondary effluent (S4), and final effluent (S5) (Fig. 1). Sampling days were deliberately selected on a sunny day to ensure a rain free period in the preceding week. For each sampling location, 1 L of wastewater was collected following a modified methodology described by Tagg et al. (2020). Briefly, the wastewater was collected using a stainless-steel barrel, and then stored in a cleaned glass bottle after passing through a 5 mm mesh sized stainless-steel sieve. All samples were taken in duplicate. The samples were shipped for further processing to the laboratory. To avoid cross contamination, the sampling equipment

was rinsed three times with deionized water between sampling operations.

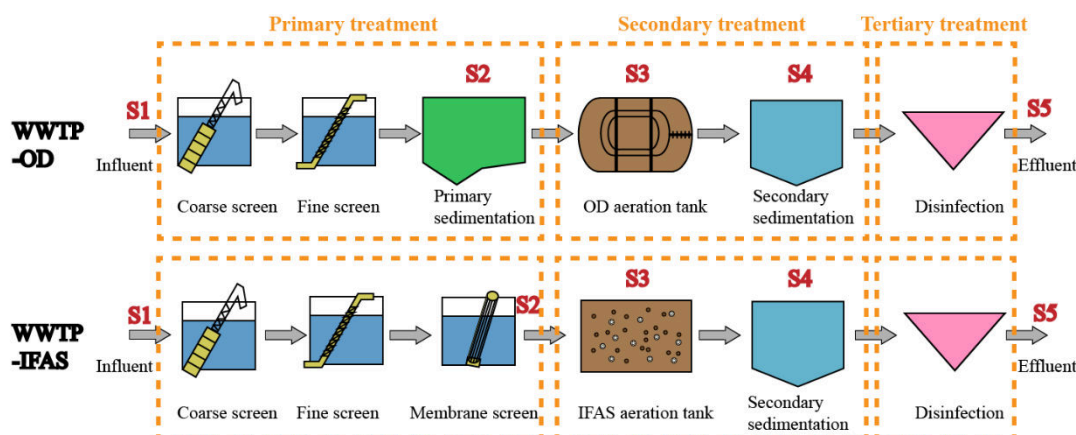


Fig. 1. Sampling locations of MPs in the WWTPs, where S1 represents the influent before the primary treatment, S2 represents the effluent after primary treatment, S3 represents the activated sludge and wastewater mixture in the aeration tank, S4 represents the effluent after the secondary treatment, and S5 represents the final effluent after tertiary treatment.

2.2. Sample pretreatment

Wastewater samples in glass bottles were filtered on a 50 μm stainless-steel membrane, using vacuum pump (GM-0.5B, 30 L/min, Jinteng, China), and a suction filter bottle (Jinteng, China). The materials captured on the stainless-steel membrane were then digested with 100 mL of 30% hydrogen peroxide (H_2O_2 , Fuchen Chemical Reagent, Tianjin, China) for at least 2 hours at 75°C until the mixture was transparent. The mixture was then filtered again through an 8 μm pore-sized polycarbonate filter paper (47 mm diameter, Merck Millipore Ltd., Ireland) as described by Barrett et al. (2020). The filtrate was transferred to a separatory funnel containing a saturated sodium iodide (NaI, Sinopharm Chemical Reagent, Shanghai, China) solution (density $\approx 1.8 \text{ g/cm}^3$). The lid of the funnel was tightly closed and vigorously shaken and then left overnight for density-based separation. Any floating particles in the resultant supernatant were filtered, and the funnel was rinsed with deionized water several times after discarding

the sediment. The filtered pieces of paper were kept in glass petri dishes so they could be examined later. For the activated sludge samples, the digestion process was carried out first, and then the extraction process was the same as the wastewater samples.

2.3. MP Characterization

The MPs were visually examined using a stereomicroscope with a magnification range of $6.7\times - 45\times$ (SZ45, Sunny, China). The shapes were categorized according to the classification standard of MPs morphology established (GESAMP, 2019). The colors were recognized by the naked eye under the stereomicroscope. The sizes of the particles were measured using MvImage software (Sunny V1.0, China). The stereomicroscope coupled with a digital camera (Sunnyoptical OD500M, China) was applied to take images of the suspected MPs. The suspected MPs were counted and recorded based on their shape, color, and size.

2.4. MP Identification

MPs were further identified via Fourier Transform Infrared (FTIR) spectrometry and Raman spectrometry. Prior studies have shown that employing a combination of multiple analytical techniques enhances the precision in identifying MPs (Ding et al., 2019). Particles $> 1,000\ \mu\text{m}$ were identified using an attenuated total reflection (ATR) accessory in conjunction with an FTIR (Spectrum Two, PerkinElmer, USA). Particles between 50 and $1,000\ \mu\text{m}$ were identified using a micro-FTIR (μ -FTIR, Spectrum Spotlight 400, PerkinElmer, USA). A germanium (Ge) crystal-based ATR attachment was equipped with the μ -FTIR to run the ATR imaging mode. The spectrum range was $4000\text{-}400\ \text{cm}^{-1}$, the pixel size was $6.25\ \mu\text{m}$, and the spectral resolution was $16\ \text{cm}^{-1}$. The type of polymers was confirmed by utilizing the Spectrum IMAGE-Spotlight 400 spectral database. A confocal Raman micro spectrometer (P300, HOOKE, China) fitted

with a -70°C cooled CCD detector and a 532 nm solid-state laser was used to perform the Raman measurements. A 50× objective was employed to focus the laser beam, offering a lateral resolution of less than 1 μm. The laser power was 3 mW. The spectral resolution of the Raman measurements was $\sim 2\text{ cm}^{-1}$. The exposure time was set at 2 s to obtain the spectra. Finally, recalculated and recorded the confirmed MPs.

2.5. Quality control

Appropriate measures were taken to prevent MP contamination during the sampling and in the laboratory. Throughout the entire experimental procedure, nitrile gloves and cotton laboratory coats were worn. Prior to and during the experiment, 70% ethanol (Sinopharm Chemical Reagent, Shanghai, China) was used to scrub the laboratory benches. All tools and containers were rinsed with deionized water prior to their use. A sample of deionized water was prepared and used as the blank control, and it was verified that no MP contamination occurred throughout the experiment procedure. Furthermore, a recovery test was conducted to verify the reliability of the method. The detailed experimental procedures are provided in Text S1. The results demonstrated that the recovery rate was 97%.

3. Results and Discussion

3.1. MP characterization

A total of 12 types of polymer MPs were identified in the samples collected from both WWTPs. Representative FTIR spectra and Raman spectra are shown in Fig. 2 and Fig. S1. The most frequently detected type of polymer was polyethylene (PE), accounting for 30% of the total MPs in number, followed by polyethylene terephthalate (PET), polypropylene (PP), and polystyrene (PS), accounting for 29%, 11% and 7%,

respectively. PE and PET were found to be the most common polymers encountered in WWTPs, consistent with the findings in previous studies (Mason et al., 2016; Xu et al., 2021; Ziajahromi et al., 2017, Cheng et al., 2021). In addition, other polymers, such as polyamide (PA), polybutyl acrylate (PBA), polybutyl methacrylate (PBMA), polycaprolactam (PCT), ethylene-acrylic acid copolymer (PEAA), polyurethane (PU), polyvinyl acetate (PVAc), and polyvinyl fluoride (PVF), were detected in small amounts. The comparison among different polymers can be challenging due to the utilization of random sampling methods for the identification and validation of questionable samples across various studies (Cheng et al., 2021). It was assumed that the use of different density flotation techniques during the sample pretreatment will result in the extraction of different MPs in this study. The identification of high-density MPs, such as PVF with a density $\approx 1.7 \text{ g/cm}^3$, was attributed to the use of high-density flotation reagent NaI, which has a density of 1.8 g/cm^3 .

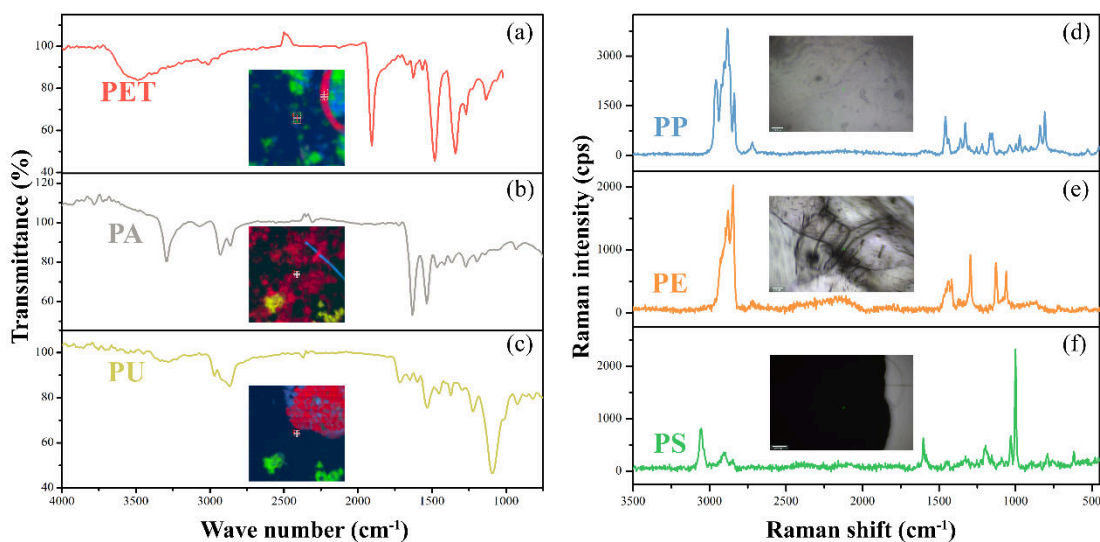


Fig. 2. Examples of MPs identified: μ -FTIR image and spectra (a, b, c), and Raman image and spectra (d, e, f).

To the best of our knowledge, there is no standard method for morphological

characterization of MPs, hence, general shape categories (e.g., fragment, foam, film, fiber, and bead) are used (Salam et al., 2023; Vitali et al., 2022; Wu et al., 2022). Scientists recommended that the shape categories should be subdivided into finer portions, which would shed lights on specific source identification (GESAMP, 2019). Therefore, in this study, the shapes of MPs were divided into granule, line, fiber, flake, foam, and film. And the shapes of “line” and “fiber” were distinguished on the basis of our previous study that the “fiber” is thought to possess length and thickness in two-dimension, while the “line” encompasses length, thickness, and width in three-dimension (Ma et al., 2022). “Flake” and “film” were differentiated by their hardness, with hard particles being categorized as “flake” while soft particles being categorized as “film”. Fig. 3 demonstrated the photographs and composition of MPs by types of shape, color, and size.

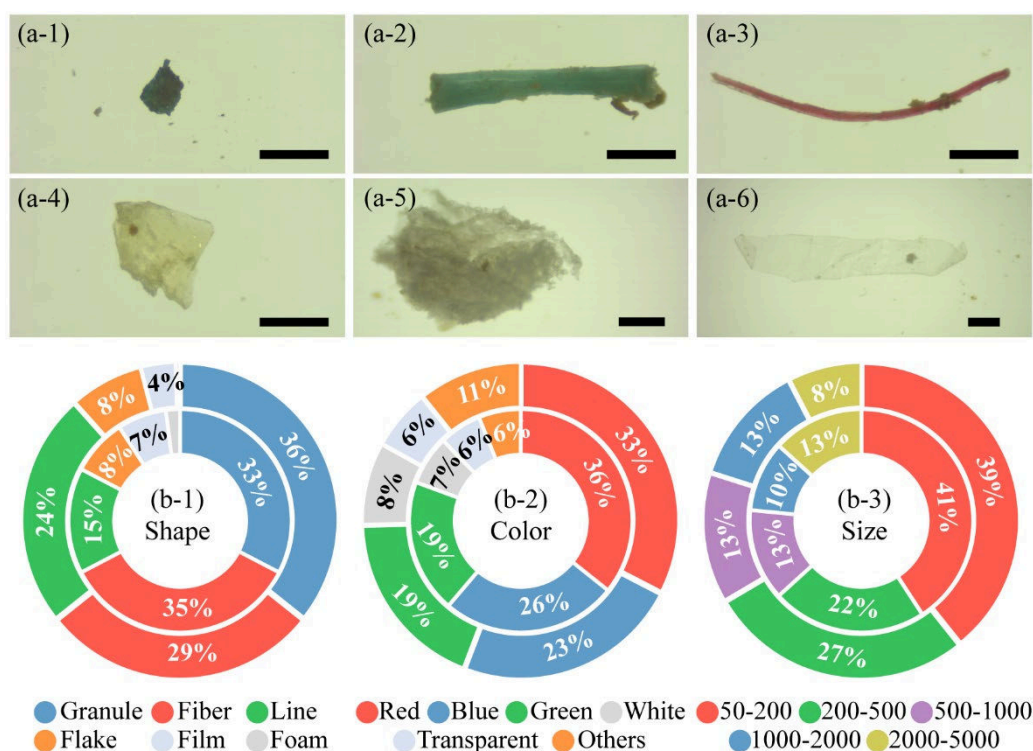


Fig. 3. Photographs of MPs taken under a stereomicroscope with an image software representing each shape, color, and size (all scale bars are 500 μm): blue granule (a-1), green line (a-2), red fiber (a-3), yellow flake (a-4), white foam (a-5), and transparent

film (a-6). Composition of MPs by types of shape (b-1), color (b-2), and size (unit: μm) (b-3) in WWTP-OD (inner ring) and WWTP-IFAS (outer ring). The sum may not equal 100% due to rounding being performed separately on individual values.

As shown in Fig. 3(b-1), granule and fiber MPs were predominant in all samples, followed by line, flake, film, and foam in both WWTPs. Granule MPs accounted for 33% and 36% in WWTP-OD and WWTP-IFAS, respectively. Scientists suggested that the subdivisions can be combined to facilitate the comparison of data (GESAMP, 2019). In this study, line and fiber MPs accounted for 50% and 53% in WWTP-OD and WWTP-IFAS, respectively, which is consistent with previous studies (Cai et al., 2020; Xu et al., 2021). MPs in regular shapes, such as bead and pellet, typically came from the release of primary MPs, whereas the irregular shapes, including fiber and fragment, mainly came from the secondary MPs that were formed from the degradation of large plastics (Li et al., 2018).

It has been reported that MP color may serve as a valuable indicator for the consumptions by aquatic organisms when wastewater is discharged into the environment. Fish can recognize and demonstrate preferences towards MP colors relying on their color vision (Okamoto et al., 2022), and predators preferentially consume MPs that share similar colors to their prey (Boerger et al., 2010; Shaw et al., 1994; Wright et al., 2013). As shown in Fig. 3(b-2), multicolored MPs were found in this study. The dominating MP colors in the two WWTPs were similar, with the following order: red > blue > green > white > transparent, with others (including yellow, purple, black, and gray) accounting for a small proportion. The common usage of colorful plastic products used by household residents in daily life, such as clothing, packaging, and toys, could potentially lead to the emergence of colored MPs. For

example, Watt et al. reported that polymer fiber, a common raw material used in clothes/textiles, is polychromatic colors (Watt et al., 2005). In addition, discoloration may occur due to prolonged UV exposure, biodegradation and physical breakage, resulting in white foam MPs (Galgani et al., 2013; Yin et al., 2021).

As shown in Fig. 3(b-3), small MP particles less than 1 mm in size were the most abundant, accounting for 76% and 79% in WWTP-OD and WWTP-IFAS, respectively. This might be due to MPs with a large particle size could be removed easily and be broken into smaller MPs under the action of external force (Zhang et al., 2015). However, most of the particles smaller than 50 μm couldn't be retained as the stainless-steel sieve used in sampling has an aperture of 50 μm , which would cause the concentration of MPs to be underestimated.

3.2. Removal of MPs by different wastewater treatment processes

3.2.1. Concentration of MPs in different treatment units

The MP concentrations in particles/L in samples collected at different processes were shown in Fig. 4. The results showed that the MP concentration in the influent of WWTP-IFAS was 38.5 ± 2.5 particles/L, much higher than that in the influent collected at WWTP-OD (15.5 ± 3.5 particles/L). In S2 samples, MP concentrations from both WWTPs were lower than those of the influent. However, the MP concentrations were detected at much higher concentrations in S3 significantly in both samples, indicating the activated sludge can be a sink for the MPs. MPs in S4 and S5 were significantly reduced, resulting in the single digit of MP concentrations in both WWTPs. As shown in Table S2, the overall removal efficiencies of MPs by WWTP-OD and WWTP-IFAS were 90% and 97%, respectively, which are consistent with the previous studies where the average removal efficiency of MPs in sewage treatment plants were 97.7%

(Mintenig et al., 2017; Murphy et al., 2016). We carried out a review on the MP removal efficiency in WWTPs (Table S3), which showed that there are significant variations in MP removal efficiency, ranging from 59.8% to 98.4%. The overall removal efficiency of MPs in WWTP-IFAS was higher than that of WWTP-OD, indicating that the IFAS process had better performance in removing MPs than OD process.

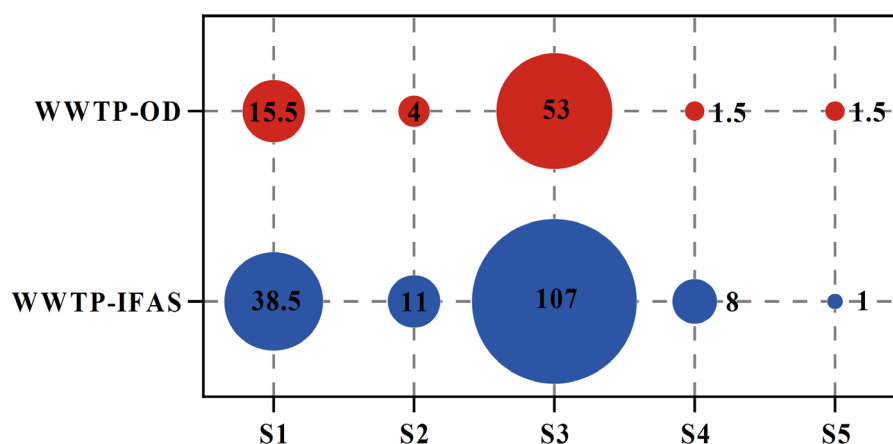


Fig. 4. MP concentrations (particles/L) in all sampling locations in the WWTPs, and the designation S1-S5 can be found in the title of Fig. 1.

At present, the wastewater treatment system is mainly aimed for the removal of conventional pollutants (such as nitrogen and phosphorus) in wastewater, and there is no specific design to remove MPs (Leslie et al., 2017). The data in this study suggested that MP removal efficiencies of 74% and 71% were achieved by the primary treatment processes in WWTP-OD and WWTP-IFAS, respectively, consistent with previous findings that 76.9% MPs can be removed efficiently by the primary treatment process (Tagg et al., 2020). The primary treatment usually consists of filtration, sedimentation, and aeration, to remove suspended solids, with gravity settling and physical entrapment serving as the removal mechanisms. In WWTP-OD, the primary sedimentation provides suitable conditions, such as low turbulence and long hydraulic retention times, which are beneficial to the sedimentation of most solids, including MPs (Bilgin et al.,

2020). In WWTP-IFAS, the membrane screen process contributed to the removal of MPs, except for MPs in the shape of fiber, which may easily pass through the screen (Ding et al., 2020; Sun et al., 2019).

During the secondary treatment process, the removal efficiency of MPs in WWTP-OD and WWTP-IFAS were 63% and 27%, respectively, as shown in Table S2, which were lower than those in the primary stage. In both WWTPs, the anaerobic-anoxic-oxic-secondary sedimentation process was applied. It was found that the concentration of MPs in IFAS activated sludge of WWTP-IFAS was twice more than that in OD activated sludge of WWTP-OD. This may be because the biofilm filler of WWTP-IFAS was made of high-density polyethylene (HDPE), and the addition of filler increases the risk of introducing MP particles. As seen in Fig. 4, the concentrations of MPs in S3 were higher than those in other process units in both WWTP-OD and WWTP-IFAS, which were 3.42 and 2.78 times higher than those in S1, respectively. This might be due to the accumulation of MPs in the tank as sewage sludge can act as a sink for MPs. For example, Chand et al. reported that MPs in wastewater from various treatment steps could be transferred and accumulated in the sewage sludge (Chand et al., 2022). In addition, Nguyen et al. (2022) reviewed that 75%-99% of the MPs are trapped in the sludge, confirming the role of sewage sludge as a sink for MPs. In this study, the high overall removal efficiency of MPs in WWTP-IFAS can be attributed to the enrichment of MPs in activated sludge in the IFAS process. Previous studies reported that conventional activated sludge reactors can only degrade MPs at a rate of less than 10% (Liu et al., 2021). Although plastic polymers such as PP and PET have been reported to be degraded by specific bacteria (Liu et al., 2021), the long degradation time (more than 40 days) limited the microbial degradation of MPs due to the constraints of hydraulic

retention time (6-12 h) and environmental conditions. Instead, sorption onto larger particles and sedimentation are considered the main mechanism to remove MPs from the biological tanks. For examples, Zhang et al. reported that the main removal mechanism of MP in activated sludge is entrapment and adsorption in sludge flocs (Zhang et al., 2020) and Liu et al. showed that the microbead and foam MPs were adsorbed and transferred into sludge and then removed with the sludge in secondary sedimentation (Liu et al., 2019). The adsorption and fixation by biofilm are the main mechanism of MPs removal in biofilm process, in which extracellular polymeric substances played a role (Zhang et al., 2020). As a result, the IFAS technology, a combination of activated sludge and biofilm process, has a better MP removal performance than the OD process. And it can be seen from Table S3, the modified activated sludge processes (such as anaerobic-anoxic-oxic and OD) and the combined processes (such as biofiltration, membrane bioreactor and IFAS) have a higher removal efficiency for MPs than the single conventional activated sludge process.

In the tertiary treatment, WWTP-OD and WWTP-IFAS were treated by coagulation, sedimentation, and disinfection. The integration of these methods can effectively remove MPs. It was speculated coagulation traps MPs in the flocs, and then sedimentation allows them to settle. Previous studies have shown that coagulation and sedimentation had a positive effect on the removal of MPs, while disinfection treatment can hardly remove any MP particles (Long et al., 2019). Furthermore, the density of MPs played a key role in sedimentation process. For example, it was reported that MPs which have high density than wastewater can be removed from wastewater easily via physical sedimentation, resulting in higher MP removal efficiency. It is difficult to remove the low- and moderate-density MPs because they float at the wastewater's

surface or suspend within the wastewater columns (Long et al., 2019). The efficiency of MP removal during tertiary treatment was higher in WWTP-IFAS compared to that in WWTP-OD. The most notable difference between the two WWTPs lies in their disinfection procedures with WWTP-OD utilizing ultraviolet irradiation, whereas WWTP-IFAS relying on the ozone technology and ultraviolet irradiation. It has been reported that ozonation can cause the decomposition of MPs (Zhang et al., 2020), hence, the relatively low MP removal efficiency in WWTP-OD. The results of this study demonstrated that the tertiary treatment could achieve removal on MPs, yet complete MP removal cannot be achieved in the current design of WWTPs.

3.2.2. Removal of MPs with different shapes, colors, and sizes

The number and the relative abundance of MPs by types of shape, color, and size in all sampling locations in the two WWTPs were shown in Table S4 and Table S5. As shown in Fig. 5, the shape, color, and size of MPs in the wastewater treatment processes of both WWTPs varied. At S2, the foam-shaped MPs were completely removed compared with S1, because the foam-shaped MPs can be removed through the screen due to their low density and large particle size.

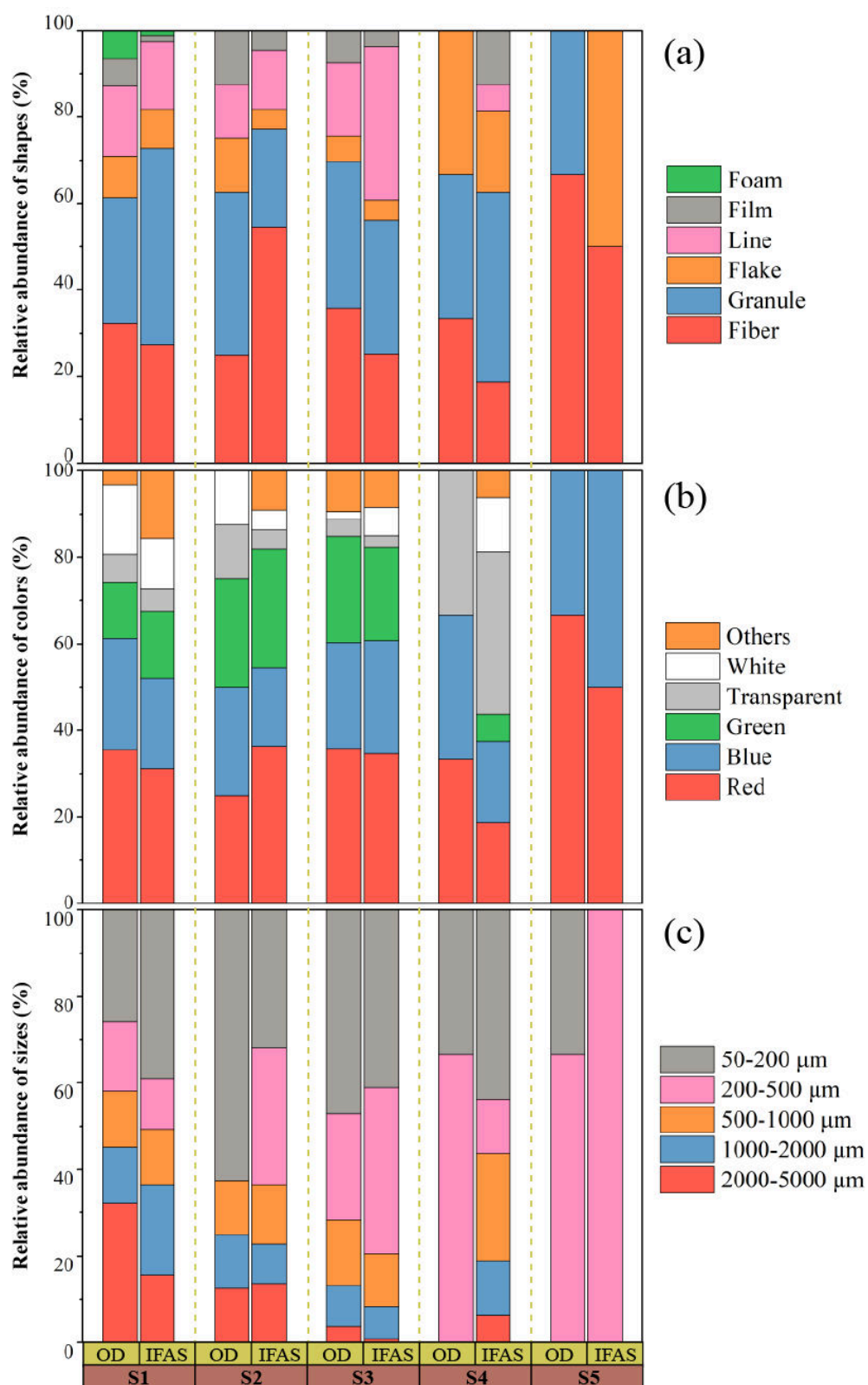


Fig. 5. Relative abundance of MPs by types of shape (a), color (b), and size (c) in all sampling locations in the WWTPs, where OD represents WWTP-OD and IFAS represents WWTP-IFAS. The designation S1-S5 can be found in the title of Fig. 1.

The proportion of line-shaped MPs at S3 increased, and at S4, WWTP-OD plant achieved complete removal of line-shaped MPs, while line-shaped MPs were still detected in WWTP-IFAS. At S5, the line-shaped MPs in both WWTPs were completely removed due to the tertiary treatment. It is evident that the biological treatment exhibits better removal efficiency for line-shaped MPs, and activated sludge is more prone to the accumulation of line-shaped MPs. Most of the diverse shaped MPs were removed by the tertiary treatment, while the percentage of fiber-shaped MPs was significantly increased in the effluent, which may be due to the small cross-section of fibers, allowing them to pass through easily during the treatment processes.

Different MPs with varied colors were quantified to provide further insight into the type of MPs before and after the treatment of both WWTPs. As shown in Fig. 5(b), the results revealed that varied colors were present in the influent before the treatment. MPs in yellow, purple, black, and gray were completely removed from the primary treatment in WWTP-OD, however, they were again detected in the activated sludge application, indicating that activated sludge is responsible for the increment of colors, as discussed in the previous sections. After the secondary treatment, white, green, and other colored MPs were completely removed from WWTP-OD. However, all the colors were still noticed after the secondary treatment in WWTP-IFAS. This might be due to the increase in MPs after the application of activated sludge. Interestingly, most of the colored MPs were removed after the tertiary treatment with the exceptions of those in red and blue. These results of tertiary treatment are efficient compared to the previous studies reported on single activated sludge treatments (Alvim et al., 2020; Long et al., 2019). The variations of MP size were measured to evaluate the efficiency of the treatments in removing MPs. As shown in Fig. 5(c), in WWTP-OD and WWTP-IFAS, the proportion

of MPs with 50~500 μm increased, while the proportion of MPs with 1,000~2,000 μm decreased. It should be noted that MPs with particle size of 200~500 μm were completely removed after the primary treatment from WWTP-OD, yet they appeared again in the activated sludge application. After the secondary treatment with activated sludge, only MPs with the particle sizes of 50~200 and 200~500 μm were present, whereas MPs with larger size were completely removed from WWTP-OD. Interestingly, only 200~500 μm size MPs were noticed in WWTP-IFAS after the tertiary treatment, indicating that these treatments can effectively remove the larger MPs from WWTPs. It can be concluded that the wastewater treatment process units are efficient in removing MP particles with large particle size, primarily due to the high efficiency of precipitation (Lenaker et al., 2019), adsorption, and surface co-precipitation of large-sized particles (Li et al., 2019; Porter et al., 2018). The size of MPs is a factor that affects the removal efficiency through the different treatment units, however, large-sized MPs may break up into smaller ones, thus increasing the concentration in MP number. Cheng et al. also reviewed that MPs larger than 500 μm in size are either removed in different treatment units or fragmented into smaller ones (Cheng et al., 2021).

3.3. Source identification of MPs in WWTPs

As illustrated in Fig. 6(a), the population density within the WWTP-IFAS catchment area considerably exceeds that of the WWTP-OD catchment area. Furthermore, Chen et al. discovered that the population activity intensity near the WWTP-IFAS is considerably higher than that around WWTP-OD (Chen et al., 2022). As shown in Fig. 6(b), the primary land use categories in the WWTP-IFAS area are residential and commercial, whereas the WWTP-OD area comprises agricultural and forest land that

remain relatively less affected by human activities, indicating that the entry of MP pollution is closely related to population density and intensity of human activities.

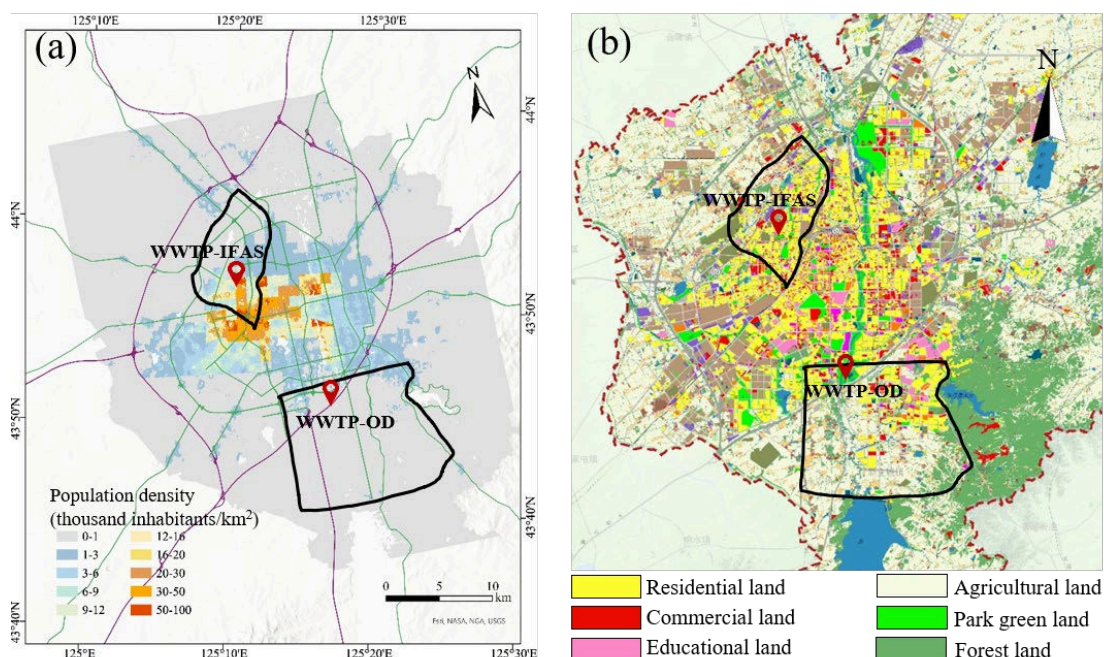


Fig. 6. Population density distribution map created using ArcGIS Pro (3.0.2 Version, USA, resolution: 100 m × 100 m) (a) and current land use classification map in Changchun (2020) obtained from Changchun Municipal People's Government (<http://www.changchun.gov.cn/>) (b).

As shown in Fig. 7, MCA enables to clarify the correlation among two or more qualitative variables, offering key insights on multiple relationships among various MPs to reveal MP sources. The MCA revealed three major clusters as shown in Fig. 7, in which the correlations between polymer type, shape, color, and size were identified. Fiber, small size (50~500 μm) and polychromatic colors have a high correlation with PET. PET fiber is the most widely used in clothing home textiles, which are color rich and small-sized, indicating that PET is likely to come from residential laundry wastewater in wastewater treatment (Hartline et al., 2016; Tian et al., 2021). These results are in line with those from Lim et al. (2022), who reported that the fibers are

manufactured from waste PET, such as textile products and garments.

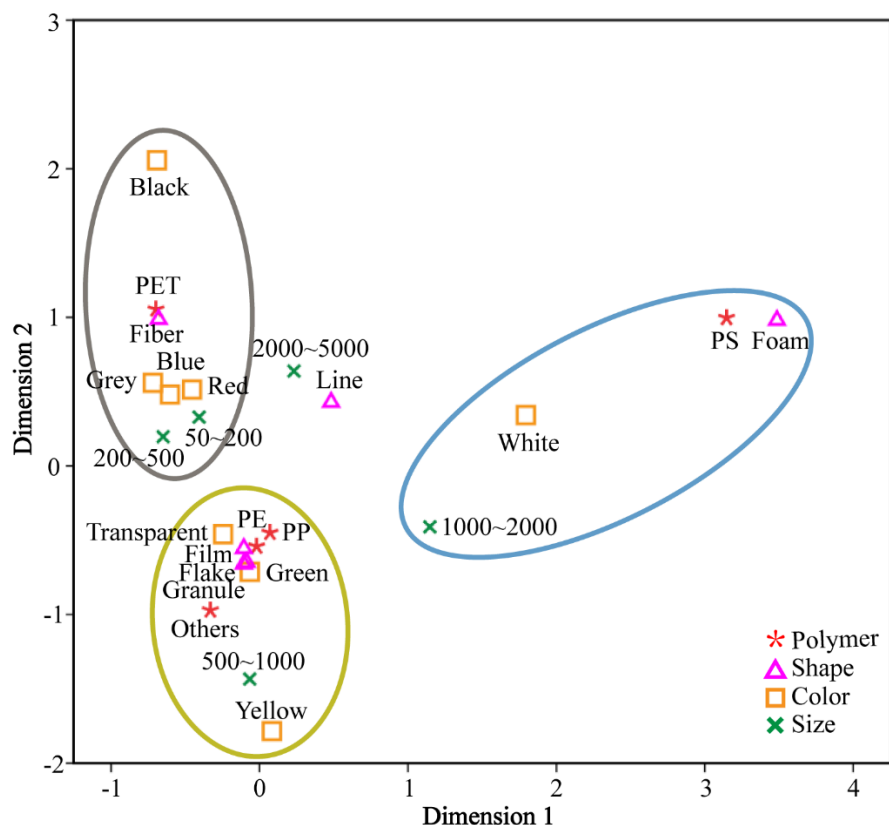


Fig. 7. MCA factor map with the contribution of polymer type, shape, color, and size (unit: μm).

Even though all fibers can be changed into microfibers, the complete elimination of microfibers was difficult. This has been noted in Fig. 5 that the percentage of fiber in small size (50-500 μm) was high even after the tertiary treatment, which warrants further study on the effective removal technologies in the future. In addition, considering the correlation between fibers and lines, it is likely that broken lines are the source of fiber MPs. In 2021, China's chemical fiber production was 65.24 million tons, with PET fiber accounting for 82.2 %, surpassing other textile fibers such as nylon (or PA) fiber (CNTAC, 2022).

PP and PE showed good connection with a medium size (500~1,000 μm), the colors of

transparent, green and yellow, and the shapes of film, flake, and granule. PP and PE are used as raw materials for food or cosmetic containers, various household items, bags, toys, or food packaging films, etc., which are daily necessities. This is why PP and PE are connected with polychromatic colors and shapes of film and flake (Fig. 7). The demand for PE film in 2020 accounted for 54% of the total demand for PE, due to its wide use for the production of plastic packaging, agricultural film, and shopping bags (CPPIA, 2021).

The correlation among PS, foam, white, and 1,000~2,000 μm indicated that the source of PS foam MPs might be waste packaging. Expanded polystyrene (EPS) is widely used in shock-proof packaging and fast-food packaging because of its high impact resistance, heat insulation, and moisture resistance (Jang et al., 2018; Sulong et al., 2019). In 2020, the output of EPS raw materials reached 5.61 million tons (CPPIA, 2021). These waste EPS plastics are light in weight, large in volume, slow in aging, and corrosion resistant, hence, the EPS has become a major challenge in the garbage disposal.

3.4. Implications for MP reduction and WWTP upgrade

Given the refractory, persistent, and migratory nature of MPs in the environment, one effective approach to tackle MPs in WWTP is to reduce MP concentrations in the influent. As the result showed, anthropogenic activities and consumer behaviors played a significant role in the concentration of MPs in the influent, therefore, it is recommended that the use of biodegradable plastic products instead of traditional plastic products in daily life should be implemented more strictly to achieve source reduction. In addition, it is recommended to implement policies that restrict the use of disposable plastic products and prohibit the addition of plastic microbeads in personal care products. Furthermore, it was found that one of the main sources of MPs in

WWTPs was laundry washing. Previous studies have demonstrated that fiber-trapping devices fitted to washing machines perform well in catching microfibers at the laundry outlet before they enter the sewage network (Erdle et al., 2021; Napper et al., 2020). Nevertheless, manufacturers have not widely implemented or upgraded these devices in residential washing machines (Napper et al., 2023). Though extensive studies have been conducted to explore the impacts of MPs to human health and the ecosystem, there is a lack of regulations in the industry worldwide. Therefore, the installation of fiber-trapping devices in washing machine upgrading is recommended.

Although WWTPs can remove most MPs, the large discharge volumes of effluent still contribute a considerable number of MPs to the receiving water, posing a severe risk to the environment. As evidenced in this study, WWTP-OD and WWTP-IFAS have a treatment capacity of $1 \times 10^5 \text{ m}^3$ and $2 \times 10^4 \text{ m}^3$ per day, respectively, which can result in up to 54.8 billion and 7.30 billion MP particles being released into the receiving water annually. In addition, a large number of MPs can enter the soil environment via sludge applications on the farmland, as the MPs are retained in the sludge. Therefore, it is important to upgrade the current WWTPs, to incorporate the MP removal capabilities into the treatment processes, with consideration of MP removal from both discharged water and sludge. Proactive strategies to prevent the MPs from entering the sludge and effluent were proposed. For example, it is recommended that an ultrafine screen with a smaller space between its wires (e.g., 0.05 mm) be installed at the end of primary treatment, through which more MPs with smaller sizes can be removed from the system. The primary sedimentation tank also played an important role in removing MPs, but the effect of operational conditions (e.g., flow and hydraulic retention time) on the removal efficiency is unknown and worth further exploration in the future. As

shown in the current study, the IFAS process was more effective than the OD process in removing MPs because IFAS applies the combination of activated sludge and biofilm process. However, it is worth noting that the choice of fillers should be taken into consideration (e.g., fillers made of non-plastic materials such as activated carbon, metals, ceramics), due to their potential contributions to the MPs.

Previous studies showed that tertiary treatment, such as membrane filtration and rapid sand filtration, can remove MPs effectively through interception or retention (Hidayaturrehman and Lee, 2019). It is recommended to adopt membrane filtration during tertiary treatment because of its smaller pore size, which is beneficial for removing MPs. Moreover, compared to filtration, it has been found that ozone oxidation provides even higher MPs removal efficiency, and it is predicted that MPs as polymers could be decomposed by ozone (Chen et al., 2018). Therefore, the use of ozone instead of UV in the disinfection process warrants further investigation.

4. Conclusions

This study investigated the occurrence, removal, and source identification of MPs within two distinct WWTPs employing different processes (OD and IFAS). The removal efficiency of WWTP with IFAS process in removing MPs is 7% higher than the one with OD process. Notably, primary treatment processes played a key role in MP removal, with removal efficiency greater than 70%, exhibiting higher efficacy in eliminating foam, film, and line-shaped MPs compared to the more challenging removal of fiber-shaped MPs. Moreover, larger-sized MP particles demonstrated higher removal efficiency within the treatment units. The study further highlighted that activated sludge acts as a primary sink for accumulated MPs within WWTPs. The

findings elucidated correlations, revealing that the abundance of MPs in influent correlated more closely with the population density of the served area rather than the sewage volume or the served equivalent population. The identification of MP sources pointed predominantly towards the laundry washing and daily necessities. Recommendations include implementing strategic approaches such as source reduction, the use of ultrafine screens or membrane filtration, and the optimization of tertiary treatment processes to limit MP discharge into the environment. Further research is needed to understand the impact of operational parameters such as flow rates and hydraulic retention times on MP removal in WWTPs. Investigating how MP density and polymer characteristics influence their fate in treatment plants is an important aspect too. Addressing these research gaps is critical to advancing our understanding and developing effective strategies to mitigate the environmental impact of MPs. The findings in this study provide technical support for the management of MP sources in WWTPs, as well as the strategies for future WWTP design and upgrade.

Authorship contribution statement

Min Ma: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Writing - Original Draft, Writing – review & editing. **Mingxin Huo:** Conceptualization, Data curation, Methodology, Supervision, Funding acquisition. **Frederic Coulon:** Conceptualization, Writing – review & editing. **Mukhtiar Ali:** Visualization, Methodology, Writing - Original Draft. **Zhiwen Tang:** Visualization, Methodology. **Xin Liu:** Visualization, Methodology. **Zhian Ying:** Methodology, Investigation. **Bin Wang:** Resources, Investigation. **Xin Song:** Conceptualization, Methodology, Supervision, Writing – review & editing, Funding acquisition.

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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References

- Alvim CB, Bes-Pia MA, Mendoza-Roca JA. Separation and identification of microplastics from primary and secondary effluents and activated sludge from wastewater treatment plants. *Chemical Engineering Journal* 2020; 402: 126293. <https://doi.org/10.1016/j.cej.2020.126293>.
- An LH, Liu Q, Deng YX, Wu WN, Gao YY, Ling W. Sources of microplastic in the environment. In: He D, Luo Y. (eds) *Microplastics in terrestrial environments. The Handbook of Environmental Chemistry* 2020; 95. https://doi.org/10.1007/698_2020_449.
- Arthur C, Baker J, Bamford H. Proceedings of the international research workshop on the occurrence, effects, and fate of microplastic marine debris. University of Washington Tacoma, Tacoma 2009; September 9-11. <http://www.MarineDebris.noaa.gov>.
- Barrett J, Chase Z, Zhang J, Holl MMB, Willis K, Williams A, et al. Microplastic pollution in deep-sea sediments from the great Australian bight. *Frontiers in Marine Science* 2020; 7: 576170. <https://doi.org/10.3389/fmars.2020.576170>.
- Bilgin M, Yurtsever M, Karadagli F. Microplastic removal by aerated grit chambers versus settling tanks of a municipal wastewater treatment plant. *Journal of Water Process Engineering* 2020; 38: 101604. <https://doi.org/10.1016/j.jwpe.2020.101604>.
- Boerger CM, Lattin GL, Moore SL, Moore CJ. Plastic ingestion by planktivorous fishes in the North Pacific Central Gyre. *Marine Pollution Bulletin* 2010; 60: 2275-2278. <https://doi.org/10.1016/j.marpolbul.2010.08.007>.
- Cai YP, Yang T, Mitrano DM, Heuberger M, Hufenus R, Nowack B. Systematic study of microplastic fiber release from 12 different polyester textiles during washing. *Environmental Science & Technology* 2020; 54: 4847-4855. <https://doi.org/10.1021/acs.est.9b07395>.
- Cao YR, Wang Q, Ruan YF, Wu RB, Chen LL, Zhang K, et al. Intra-day microplastic variations in wastewater: A case study of a sewage treatment plant in Hong Kong. *Marine Pollution Bulletin* 2020; 160: 111535. <https://doi.org/10.1016/j.marpolbul.2020.111535>.
- Chand R, Kohansal K, Toor S, Pedersen TH, Vollertsen J. Microplastics degradation through hydrothermal liquefaction of wastewater treatment sludge. *Journal of Cleaner Production* 2022; 335: 130383.

- <https://doi.org/10.1016/j.jclepro.2022.130383>.
- Chen R, Qi M, Zhang GH, Yi CG. Comparative experiments on polymer degradation technique of produced water of polymer flooding oilfield. *IOP Conf. Series: Earth and Environmental Science* 2018; 113: 012208. <https://doi.org/10.1088/1755-1315/113/1/012208>.
- Chen Y, Hu Y, Lai L. Demography-oriented urban spatial matching of service facilities: Case study of Changchun, China. *LAND* 2022; 11: 1660. <https://doi.org/10.3390/land11101660>.
- Cheng YL, Kim JG, Kim HB, Choi JH, Tsang YF, Baek K. Occurrence and removal of microplastics in wastewater treatment plants and drinking water purification facilities: A review. *Chemical Engineering Journal* 2021; 410: 128381. <https://doi.org/10.1016/j.cej.2020.128381>.
- China National Textile and Apparel Council (CNTAC). 2021/2022 China textile industry development report, Beijing, 2022.
- China Plastics Processing Industry Association (CPPIA). China plastic industry yearbook. 2021.
- Conley K, Clum A, Deepe J, Lane H, Beckingham B. Wastewater treatment plants as a source of microplastics to an urban estuary: Removal efficiencies and loading per capita over one year. *Water Research* 2019; 3: 100030. <https://doi.org/10.1016/j.wroa.2019.100030>.
- Daziroglu MEC, Bilici S. The hidden threat to food safety and human health: microplastics. *Environment Development and Sustainability* 2023. <https://doi.org/10.1007/s10668-023-03565-7>.
- de Carvalho AR, Garcia F, Riem-Galliano L, Tudesque L, Albignac M, Ter Halle A, et al. Urbanization and hydrological conditions drive the spatial and temporal variability of microplastic pollution in the Garonne River. *Science of the Total Environment* 2021; 769: 144479. <https://doi.org/10.1016/j.scitotenv.2020.144479>.
- Ding J, Li J, Sun C, Jiang F, Ju P, Qu L, et al. Detection of microplastics in local marine organisms using a multi-technology system. *Analytical Methods* 2019; 11: 78-87. <https://doi.org/10.1039/c8ay01974f>.
- Ding N, An D, Yin XF, Sun YX. Detection and evaluation of microbeads and other microplastics in wastewater treatment plant samples. *Environmental Science and Pollution Research* 2020; 27: 15878-15887. <https://doi.org/10.1007/s11356-020-08127-2>.
- Dyachenko A, Mitchell J, Arsem N. Extraction and identification of microplastic particles from secondary wastewater treatment plant (WWTP) effluent. *Analytical*

- Methods 2017; 9: 1412-1418. <https://doi.org/10.1039/c6ay02397e>.
- Eerkes-Medrano D, Thompson RC, Aldridge DC. Microplastics in freshwater systems: A review of the emerging threats, identification of knowledge gaps and prioritisation of research needs. *Water Research* 2015; 75: 63-82. <https://doi.org/10.1016/j.watres.2015.02.012>.
- Erdle LM, Parto DN, Sweetnam D, Rochman CM. Washing machine filters reduce microfiber emissions: evidence from a community-scale pilot in Parry Sound, Ontario. *Frontiers in Marine Science* 2021; 8: 777865. <https://doi.org/10.3389/fmars.2021.777865>.
- Estahbanati S, Fahrenfeld NL. Influence of wastewater treatment plant discharges on microplastic concentrations in surface water. *Chemosphere* 2016; 162: 277-284. <https://doi.org/10.1016/j.chemosphere.2016.07.083>.
- Fahrenfeld NL, Arbuckle-Keil G, Beni NN, Bartelt-Hunt SL. Source tracking microplastics in the freshwater environment. *Trac-Trends in Analytical Chemistry* 2019; 112: 248-254. <https://doi.org/10.1016/j.trac.2018.11.030>.
- Galgani F, Hanke G, Werner S, De Vrees L. Marine litter within the European Marine Strategy Framework Directive. *Ices Journal of Marine Science* 2013; 70: 1055-1064. <https://doi.org/10.1093/icesjms/fst122>.
- Group of experts on the scientific aspects of marine environmental protection (GESAMP). Guidelines for the monitoring and assessment of plastic litter in the ocean. United Nations Environment Programme (UNEP), 2019; No. 99, 130p. <http://gesamp.org>.
- Hamidian AH, Ozumchelouei EJ, Feizi F, Wu CX, Zhang Y, Yang M. A review on the characteristics of microplastics in wastewater treatment plants: A source for toxic chemicals. *Journal of Cleaner Production* 2021; 295: 126480. <https://doi.org/10.1016/j.jclepro.2021.126480>.
- Hartline NL, Bruce NJ, Karba SN, Ruff EO, Sonar SU, Holden PA. Microfiber masses recovered from conventional machine washing of new or aged garments. *Environmental Science & Technology* 2016; 50: 11532-11538. <https://doi.org/10.1021/acs.est.6b03045>.
- Hidayaturrehman H, Lee TG. A study on characteristics of microplastic in wastewater of South Korea: Identification, quantification, and fate of microplastics during treatment process. *Marine Pollution Bulletin* 2019; 146: 696-702. <https://doi.org/10.1016/j.marpolbul.2019.06.071>.
- International Union for Conservation of Nature (IUCN). Primary microplastics in the oceans: A global evaluation of sources. Gland, Switzerland 2017.

- <https://doi.org/10.2305/IUCN.CH.2017.01.en>.
- Jang M, Shim WJ, Han GM, Song YK, Hong SH. Formation of microplastics by polychaetes (*Marphysa sanguinea*) inhabiting expanded polystyrene marine debris. *Marine Pollution Bulletin* 2018; 131: 365-369. <https://doi.org/10.1016/j.marpolbul.2018.04.017>.
- Jiang JH, Wang XW, Ren HY, Cao GL, Xie GJ, Xing DF, et al. Investigation and fate of microplastics in wastewater and sludge filter cake from a wastewater treatment plant in China. *Science of the Total Environment* 2020; 746: 141378. <https://doi.org/10.1016/j.scitotenv.2020.141378>.
- Klein S, Worch E, Knepper TP. Occurrence and spatial distribution of microplastics in river shore sediments of the Rhine-Main area in Germany. *Environmental Science & Technology* 2015; 49: 6070-6076. <https://doi.org/10.1021/acs.est.5b00492>.
- Lenaker PL, Baldwin AK, Corsi SR, Mason SA, Reneau PC, Scott JW. Vertical distribution of microplastics in the water column and surficial sediment from the Milwaukee river basin to lake Michigan. *Environmental Science & Technology* 2019; 53: 12227-12237. <https://doi.org/10.1021/acs.est.9b03850>.
- Leslie HA, Brandsma SH, van Velzen MJ, Vethaak AD. Microplastics en route: Field measurements in the Dutch river delta and Amsterdam canals, wastewater treatment plants, North Sea sediments and biota. *Environment International* 2017; 101: 133-142. <https://doi.org/10.1016/j.envint.2017.01.018>.
- Li J, Zhang H, Zhang KN, Yang RJ, Li RZ, Li YF. Characterization, source, and retention of microplastic in sandy beaches and mangrove wetlands of the Qinzhou Bay, China. *Marine Pollution Bulletin* 2018; 136: 401-406. <https://doi.org/10.1016/j.marpolbul.2018.09.025>.
- Li Y, Wang XJ, Fu WY, Xia XH, Liu CQ, Min JC, et al. Interactions between nano/micro plastics and suspended sediment in water: Implications on aggregation and settling. *Water Research* 2019; 161: 486-495. <https://doi.org/10.1016/j.watres.2019.06.018>.
- Lim SJ, Park YK, Kim H, Kwon J, Moon HM, Lee Y, et al. Selective solvent extraction and quantification of synthetic microfibers in textile laundry wastewater using pyrolysis-gas chromatography/mass spectrometry. *Chemical Engineering Journal* 2022; 434: 134653. <https://doi.org/10.1016/j.cej.2022.134653>.
- Lin YD, Huang PH, Chen YW, Hsieh CW, Tain YL, Lee BH, et al. Sources, degradation, ingestion and effects of microplastics on humans: A review. *Toxics* 2023; 11: 747. <https://doi.org/10.3390/toxics11090747>.
- Liu WY, Zhang JL, Liu H, Guo XN, Zhang XY, Yao XL, et al. A review of the removal

- of microplastics in global wastewater treatment plants: Characteristics and mechanisms. *Environment International* 2021; 146: 106277. <https://doi.org/10.1016/j.envint.2020.106277>.
- Liu XN, Yuan WK, Di MX, Li Z, Wang J. Transfer and fate of microplastics during the conventional activated sludge process in one wastewater treatment plant of China. *Chemical Engineering Journal* 2019; 362: 176-182. <https://doi.org/10.1016/j.cej.2019.01.033>.
- Long ZX, Pan Z, Wang WL, Ren JY, Yu XG, Lin LY, et al. Microplastic abundance, characteristics, and removal in wastewater treatment plants in a coastal city of China. *Water Research* 2019; 155: 255-265. <https://doi.org/10.1016/j.watres.2019.02.028>.
- Lv XM, Dong Q, Zuo ZQ, Liu YC, Huang X, Wu WM. Microplastics in a municipal wastewater treatment plant: Fate, dynamic distribution, removal efficiencies, and control strategies. *Journal of Cleaner Production* 2019; 225: 579-586. <https://doi.org/10.1016/j.jclepro.2019.03.321>.
- Ma M, Liu SB, Su M, Wang C, Ying ZA, Huo MX, et al. Spatial distribution and potential sources of microplastics in the Songhua River flowing through urban centers in Northeast China. *Environmental Pollution* 2022; 292: 118384. <https://doi.org/10.1016/j.envpol.2021.118384>.
- Mason SA, Garneau D, Sutton R, Chu Y, Ehmann K, Barnes J, et al. Microplastic pollution is widely detected in US municipal wastewater treatment plant effluent. *Environmental Pollution* 2016; 218: 1045-1054. <https://doi.org/10.1016/j.envpol.2016.08.056>.
- Mintenig SM, Int-Veen I, Loder MGJ, Primpke S, Gerdt G. Identification of microplastic in effluents of wastewater treatment plants using focal plane array-based micro-Fourier-transform infrared imaging. *Water Research* 2017; 108: 365-372. <https://doi.org/10.1016/j.watres.2016.11.015>.
- Murphy F, Ewins C, Carbonnier F, Quinn B. Wastewater Treatment Works (WwTW) as a source of microplastics in the aquatic environment. *Environmental Science & Technology* 2016; 50: 5800-5808. <https://doi.org/10.1021/acs.est.5b05416>.
- Napper IE, Barrett AC, Thompson RC. The efficiency of devices intended to reduce microfibre release during clothes washing. *Science of the Total Environment* 2020; 738: 140412. <https://doi.org/10.1016/j.scitotenv.2020.140412>.
- Napper IE, Parker-Jurd FNF, Wright SL, Thompson RC. Examining the release of synthetic microfibres to the environment via two major pathways: Atmospheric deposition and treated wastewater effluent. *Science of the Total Environment* 2023;

- 857: 159317. <https://doi.org/10.1016/j.scitotenv.2022.159317>.
- Nguyen MK, Hadi M, Lin CT, Nguyen HL, Thai VB, Hoang HG, et al. Microplastics in sewage sludge: Distribution, toxicity, identification methods, and engineered technologies. *Chemosphere* 2022; 308: 136455. <https://doi.org/10.1016/j.chemosphere.2022.136455>.
- Okamoto K, Nomura M, Horie Y, Okamura H. Color preferences and gastrointestinal-tract retention times of microplastics by freshwater and marine fishes. *Environmental Pollution* 2022; 304: 119253. <https://doi.org/10.1016/j.envpol.2022.119253>.
- Picó Y, Alvarez-Ruiz R, Alfarhan AH, El-Sheikh MA, Alshahrani HO, Barceló D. Pharmaceuticals, pesticides, personal care products and microplastics contamination assessment of Al-Hassa irrigation network (Saudi Arabia) and its shallow lakes. *Science of the Total Environment* 2020; 701: 135021. <https://doi.org/10.1016/j.scitotenv.2019.135021>.
- Porter A, Lyons BP, Galloway TS, Lewis C. Role of marine snows in microplastic fate and bioavailability. *Environmental Science & Technology* 2018; 52: 7111-7119. <https://doi.org/10.1021/acs.est.8b01000>.
- Prata JC, Godoy V, da Costa JP, Calero M, Martin-Lara MA, Duarte AC, et al. Microplastics and fibers from three areas under different anthropogenic pressures in Douro river. *Science of the Total Environment* 2021; 776: 145999. <https://doi.org/10.1016/j.scitotenv.2021.145999>.
- Prata JC. Microplastics in wastewater: State of the knowledge on sources, fate and solutions. *Marine Pollution Bulletin* 2018; 129: 262-265. <https://doi.org/10.1016/j.marpolbul.2018.02.046>.
- Sadia M, Mahmood A, Ibrahim M, Irshad MK, Quddusi AA, Bokhari A, et al. Microplastics pollution from wastewater treatment plants: A critical review on challenges, detection, sustainable removal techniques and circular economy. *Environmental Technology & Innovation* 2022; 28: 102946. <https://doi.org/10.1016/j.eti.2022.102946>.
- Salam M, Zheng HL, Liu YY, Zaib A, Rehman SAU, Riaz N, et al. Effects of micro(nano)plastics on soil nutrient cycling: State of the knowledge. *Journal of Environmental Management* 2023; 344: 118437. <https://doi.org/10.1016/j.jenvman.2023.118437>.
- Shaw DG, Day RH. Color-dependent and form-dependent loss of plastic micro-debris from the North Pacific-Ocean. *Marine Pollution Bulletin* 1994; 28: 39-43. [https://doi.org/10.1016/0025-326x\(94\)90184-8](https://doi.org/10.1016/0025-326x(94)90184-8).

- Su L, Xue YG, Li LY, Yang DQ, Kolandhasamy P, Li DJ, et al. Microplastics in Taihu Lake, China. *Environmental Pollution* 2016; 216: 711-719. <https://doi.org/10.1016/j.envpol.2016.06.036>.
- Sulong NHR, Mustapa SAS, Rashid MKA. Application of expanded polystyrene (EPS) in buildings and constructions: A review. *Journal of Applied Polymer Science* 2019; 136: 47529. <https://doi.org/10.1002/app.47529>.
- Sun J, Dai XH, Wang QL, van Loosdrecht MCM, Ni BJ. Microplastics in wastewater treatment plants: Detection, occurrence and removal. *Water Research* 2019; 152: 21-37. <https://doi.org/10.1016/j.watres.2018.12.050>.
- Tadsuwan K, Babel S. Unraveling microplastics removal in wastewater treatment plant: A comparative study of two wastewater treatment plants in Thailand. *Chemosphere* 2022; 307: 135733. <https://doi.org/10.1016/j.chemosphere.2022.135733>.
- Tagg AS, Sapp M, Harrison JP, Sinclair CJ, Bradley E, Ju-Nam Y, et al. Microplastic monitoring at different stages in a wastewater treatment plant using reflectance micro-ftir imaging. *Frontiers in Environmental Science* 2020; 8: 145. <https://doi.org/10.3389/fenvs.2020.00145>.
- Thompson RC, Olsen Y, Mitchell RP, Davis A, Rowland SJ, John AWG, et al. Lost at sea: Where is all the plastic? *Science* 2004; 304: 838-838. <https://doi.org/10.1126/science.1094559>.
- Tian YJ, Chen Z, Zhang JY, Wang ZZ, Zhu YJ, Wang P, et al. An innovative evaluation method based on polymer mass detection to evaluate the contribution of microfibers from laundry process to municipal wastewater. *Journal of Hazardous Materials* 2021; 407: 124861. <https://doi.org/10.1016/j.jhazmat.2020.124861>.
- Vitali C, Peters RJB, Janssen HG, Nielen MWF, Ruggeri FS. Microplastics and nanoplastics in food, water, and beverages, part II. Methods. *TrAC-Trends in Analytical Chemistry* 2022; 157: 116819. <https://doi.org/10.1016/j.trac.2022.116819>.
- Waqas S, Bilal MR, Man Z, Wibisono Y, Jaafar J, Mahlia TMI, et al. Recent progress in integrated fixed-film activated sludge process for wastewater treatment: A review. *Journal of Environmental Management* 2020; 268: 110718. <https://doi.org/10.1016/j.jenvman.2020.110718>.
- Watt R, Roux C, Robertson J. The population of coloured textile fibres in domestic washing machines. *Science & Justice* 2005; 45: 75-83. [https://doi.org/10.1016/S1355-0306\(05\)71632-4](https://doi.org/10.1016/S1355-0306(05)71632-4).
- Wright SL, Thompson RC, Galloway TS. The physical impacts of microplastics on

- marine organisms: A review. *Environmental Pollution* 2013; 178: 483-492.
<https://doi.org/10.1016/j.envpol.2013.02.031>.
- Wu XW, Zhao XL, Chen RZ, Liu P, Liang WG, Wang JY, et al. Wastewater treatment plants act as essential sources of microplastic formation in aquatic environments: A critical review. *Water Research* 2022; 221: 118825.
<https://doi.org/10.1016/J.Watres.2022.118825>.
- Xu X, Zhang L, Jian Y, Xue YG, Gao Y, Peng MG, et al. Influence of wastewater treatment process on pollution characteristics and fate of microplastics. *Marine Pollution Bulletin* 2021; 169: 112448.
<https://doi.org/10.1016/j.marpolbul.2021.112448>.
- Yin K, Wang DX, Zhao HJ, Wang Y, Guo MH, Liu YC, et al. Microplastics pollution and risk assessment in water bodies of two nature reserves in Jilin Province: Correlation analysis with the degree of human activity. *Science of the Total Environment* 2021; 799: 149390. <https://doi.org/10.1016/j.scitotenv.2021.149390>.
- Zhang CH, Quan BX, Tang JW, Cheng KP, Tang YH, Shen WL, et al. China's wastewater treatment: Status quo and sustainability perspectives. *Journal of Water Process Engineering* 2023; 53: 103708.
<https://doi.org/10.1016/j.jwpe.2023.103708>.
- Zhang K, Gong W, Lv JZ, Xiong X, Wu CX. Accumulation of floating microplastics behind the three gorges dam. *Environmental Pollution* 2015; 204: 117-123.
<https://doi.org/10.1016/j.envpol.2015.04.023>.
- Zhang XL, Chen JX, Li J. The removal of microplastics in the wastewater treatment process and their potential impact on anaerobic digestion due to pollutants association. *Chemosphere* 2020; 251: 126360.
<https://doi.org/10.1016/j.chemosphere.2020.126360>.
- Ziajahromi S, Neale PA, Rintoul L, Leusch FDL. Wastewater treatment plants as a pathway for microplastics: Development of a new approach to sample wastewater-based microplastics. *Water Research* 2017; 112: 93-99.
<https://doi.org/10.1016/j.watres.2017.01.042>.



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Understanding microplastic presence in different wastewater treatment processes: removal efficiency and source identification

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