

Distribution, characteristics and daily fluctuations of microplastics throughout wastewater treatment plants with mixed domestic–industrial influents in Wuxi City, China

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HIGHLIGHTS

- MPs were analyzed throughout three WWTPs with mixed domestic – industrial influents.
- White polyethylene granules from plastic manufacturing were the most dominant MPs.
- MPs abundance in random grab-sampling was lower than that in daily dense sampling.
- The production of MPs such as microbeads need to be restricted from the source.

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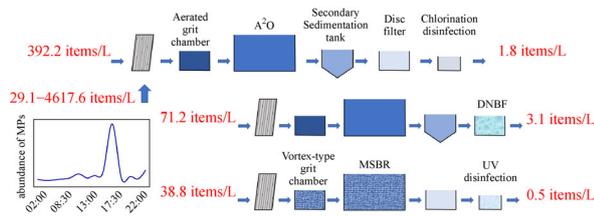
Wastewater treatment plant

Mixed domestic-industrial influent

Characteristic

Daily fluctuation

GRAPHIC ABSTRACT



ABSTRACT

In wastewater treatment plants (WWTPs), microplastics (MPs) are complex, especially with mixed domestic–industrial influents. Conventional random grab sampling can roughly depict the distribution and characteristics of MPs but can not accurately reflect their daily fluctuations. In this study, the concentration, shape, polymer type, size, and color of MPs were analyzed by micro-Raman spectroscopy (detection limit of 0.05 mm) throughout treatment stages of three mixed domestic–industrial WWTPs (W1, W2, and W3) in Wuxi City, China, and the daily fluctuations of MPs were also obtained by dense grab sampling within 24 h. For influent samples, the average MP concentration of 392.2 items/L in W1 with 10% industrial wastewater was much higher than those in W2 (71.2 items/L with 10% industrial wastewater) and W3 (38.3 items/L with 60% industrial wastewater). White polyethylene granules with a diameter less than 0.5 mm from plastic manufacturing were the most dominant MPs in the influent of W1, proving the key role of industrial sources in MPs pollution. In addition, the daily dense sampling results showed that MP concentration in W1 influent fluctuated widely between 29.1 items/L and 4617.6 items/L within a day. Finally, few MPs (less than 4.0 items/L) in these WWTPs effluents were attributed to the effective removal of wastewater treatment processes. Thus, further attention should be paid to regulating the primary sources of MPs.

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1 Introduction

Ubiquitous microplastics (MPs) have been detected in various environments, such as wastewater treatment plants (WWTPs), rivers, lakes, sediments, the very deepest reaches of oceans, and even the Antarctic ice cores (Van Cauwenberghé et al., 2013; Bessa et al., 2019; Bergami et al., 2020). With their size of being smaller than 5 mm in diameter (Mason et al., 2016), MPs can enter the food chain through ingestion by various aquatic animals.

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Potential risks to human and animal health would be posed by mechanical damage, leaching plastic additives, and the reactive oxygen species derived from MPs or adsorbed toxic pollutants associated with MPs (Wright et al., 2013; Hermabessiere et al., 2017; Liu and Wang, 2020; Liu et al., 2021). MPs come from a wide range of sources, including primary microbeads in the personal care and cosmetic products and secondary plastics broken from large pieces of plastics (Mason et al., 2016; Liu et al., 2019). Thus, increasing attention has been paid to MPs pollution (Browne et al., 2011; Avio et al., 2015).

WWTPs are considered latent MPs sources due to the great volume of effluent (Alavian Petroody et al., 2020). Previous studies showed that the distribution and characteristics of MPs in different WWTPs varied greatly (Gatidou et al., 2019; Park et al., 2020), especially in influent. The concentrations of MPs in the influent ranged from 1.57 items/L (Long et al., 2019) to 54000 items/L (Zhou et al., 2020b). The dominant polymer types of MPs in some WWTPs were polyethylene terephthalate (PET) and polystyrene (PS), while those in some other WWTPs were polyamide (PA) or polypropylene (PP) (Long et al., 2019). The big discrepancies in concentration and polymer type, on the one hand, is attributed to a lack of standard sampling and detection methods, as well as important QA/QC (Koelmans et al., 2019; Cowger et al., 2020). On the other hand, WWTPs are complex systems with various treatment processes and different influents, which are related to population served, proportion, and types of industrial wastewater. Which among them has a greater impact on the concentrations of MPs in the influent is still unclear. More investigations should be conducted to gain a wider knowledge of the fate of MPs in WWTPs with the different factors mentioned above.

The concentrations of pollutants in the influents of WWTPs fluctuate constantly in a day, such as pharmaceuticals and personal care products, total phosphorus (TP), suspended solids (SS), and viruses (Li et al., 2018a; Ahmed et al., 2021). However, for MPs, it is not yet known. Most researchers collected samples in a specific period on different dates or by 24 h composite sampling (Lares et al., 2018; Wang et al., 2020). Considering the fluctuation of influent quality within one day, the MPs abundance can be studied through multiple grab samplings within 24 h and the samples of each time interval can be analyzed independently to accurately show the daily fluctuation of MPs in the influents of WWTPs. Whether different sampling strategies will lead to errors in MPs concentration also needs to be studied.

In this study, the concentration, shape, polymer type, size, and color of MPs were investigated in different treatment units throughout three mixed domestic-industrial WWTPs with different influent characteristics in Wuxi City, China. A dense sampling campaign within 24 h was performed to examine the diel variations of MPs in the influent of one WWTP. Furthermore, correlations between

the MP concentration and water quality parameters were analyzed.

2 Materials and methods

2.1 WWTPs description

Three WWTPs (labeled as W1, W2, and W3) are located in the urban areas of Wuxi City, China, with daily wastewater treatment capacities of 100000, 25000, and 90000 m³, respectively (Table 1). Industrial wastewater treated by W1 was mainly from citric acid and cast-iron pipe factories. The proportion of industrial wastewater received by W2 was close to W1 (10%) and was mainly printing and dyeing wastewater. W3 receives abundant industrial wastewater (60%) from the electronics, machinery, and dyeing industries. All three WWTPs adopt a three-stage treatment process (Fig. 1). The pretreatment process includes coarse and fine grids and aerated or vortex-type grit chambers, but without primary sedimentation tanks. W3 adopted modified sequencing batch reactor (MSBR) as the secondary treatment process, whose hydraulic retention time (HRT) was almost half the anaerobic-anoxic-oxic (A²O) processes of W1 and W2 (20 h). Advanced treatment technologies of W1 and W3 were equipped with disc filters with filtration limit of 10 μm. The white filter cloth was made from polyacrylonitrile (PAN). The advanced treatment process in W2 is denitrifying biological filter (DNBF). More information about population and area served, proportion, and type of industrial wastewater of the three WWTPs is showed in Table 1.

2.2 Wastewater sampling

In the summer of 2020, wastewater samples were irregularly collected for six times in W1, three times in W2, and four times in W3 (Table S1). Four sampling points were set in each WWTP: influent (labeled as S1), primary effluent (labeled as S2), secondary effluent (labeled as S3), and final effluent (labeled as S4), as shown in Fig. 1. In one of the three WWTPs, i.e., W1, multiple samplings of the influent within 24 h were conducted in October and a total of 14 samples were collected at 2:00, 4:00, 7:00, 8:30, 10:00, 11:30, 13:00, 14:30, 16:00, 17:30, 19:00, 20:30, 22:00, and 24:00, respectively. Detailed information about sampling dates and volumes of the wastewater samples is shown in Table S1. Wastewater samples were collected in triplicate at a depth of approximately 50 cm for each sampling point and poured into glass bottles.

2.3 Sample processing

Each wastewater sample was sequentially filtered through three sieves with mesh sizes of 5, 0.2, and 0.025 mm (mesh numbers are 4, 64, and 400, respectively). Residues on

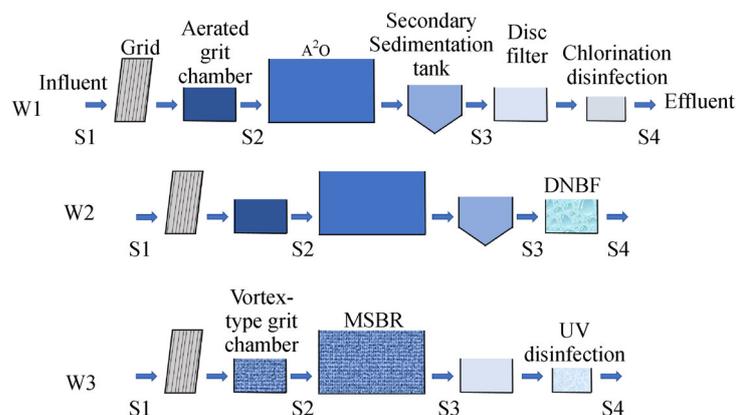


Fig. 1 Sampling points of the three mixed industrial–domestic WWTPs in Wuxi City, China. The same geometrical shape represents the same treatment process.

Table 1 Overview of the three WWTPs in Wuxi City, China

WWTPs	Design capacity (tons/d)	Area served (km ²)	Population served (thousand)	Proportion of industrial wastewater (%)	Main industries	Main treatment process
W1	100000	70	50	10	Citric acid, polyethylene and nodular cast, iron pipe	A ² /O and disc filter
W2	25000	47	>20	10	Pharmaceuticals, cotton printing and dyeing	A ² /O and denitrifying biological filter
W3	90000	80	33	60	Integrated circuit, chip, electronic components, cotton printing and dyeing, auto parts	MSBR and disc filter

0.2 mm and 0.025 mm sieves surface were washed into a beaker by deionized water. The beaker was then stored at 70°C to concentrate the volume of the water sample to 50 mL.

Wet peroxide oxidation (WPO) was adopted to digest organic materials in the concentrated samples according to the guidelines of the National Oceanic and Atmospheric Administration (Hidalgo-Ruz et al., 2012). Briefly, 5 mL of aqueous 0.05 M Fe (II) solution and 5 mL of 30% hydrogen peroxide (H₂O₂) were added into a beaker and stood for 5 min. Then, the beaker was shaken for 12 h (55°C, 80 r/min). After WPO, 200 mL of saturated NaCl solution ($\rho = 1.27$ mg/mL) was added into the beaker and treated by ultrasonication for 30 min, and then the samples were placed into a separating funnel and stood overnight. After that, the supernatant was passed through a 0.45 μ m nitrocellulose membrane (11406-47-acn, 47 mm diameter, Sartorius, Germany). Filtered membranes were stored in clean containers for the following analysis.

2.4 MPs analysis

MPs were first examined using a stereomicroscope (PXS8-T, magnification $\times 63$), and all suspected MPs on the

surface of each membrane were recorded. Raman technique is the widely used method for MP composition identification (Zhao et al., 2017; Long et al., 2019). An MP sample would produce a unique molecular fingerprint during the interaction of radiation with molecular vibration. We can determine the composition of MP by comparing the fingerprint of the MP sample with those of standards. Micro-Raman spectroscopy (Dxr2xi, 785 nm laser, voltage 3–5 MW, residence time 0.1 s, Raman shift 100–3500, Thermo Scientific, USA) was then used to identify the polymer type of the suspected MPs. Particles were randomly selected on the surface of each membrane, and no less than 100 suspected MPs were tested for each influent sample. Particles that showed clear characteristic peaks similar to plastic polymers and matched >60% of reference spectra, were confirmed as MPs. Nicolet Standard Collection of Raman Spectra was used as the reference spectra in the OMNIC 9 software obtained from Thermo Scientific (USA). Their characteristics, such as shape, color, and polymer type, were recorded. MPs were classified into fibers, granules, and fragments (including films) in shape, and 1–5, 0.5–1, 0.2–0.5, and 0.05–0.2 mm (detection limit of 0.05 mm) in size. The total number of MPs on each membrane was calculated as follows (Eq. (1)):

$$N = a/b \times n, \quad (1)$$

where, N – the total number of MPs, items.

a – the number of MPs confirmed by micro-Raman spectroscopy, items.

b – the number of suspected particles analyzed by micro-Raman spectroscopy, items.

n – the number of suspected particles examined by stereomicroscope, items.

2.5 Quality assurance and quality control

Before sampling, all containers and tools were cleaned with deionized water. Wastewater was collected with a stainless steel bucket. To reduce the interference of ambient airflow, all bottles were immediately covered with aluminum foil and transported to the laboratory for further treatment. Blank samples were prepared by using the same bottles and deionized water. PS MPs with a diameter of 1000 μm were used as positive control, and the recovery rate was 99.02%. Quality control was strictly conducted by wearing cotton clothes, using nitrile gloves, and wrapping openings with aluminum foil according to previous methods (Wei et al., 2020; Zhou et al., 2020b). As a result, no MPs but only several cotton fibers were detected in blank samples.

2.6 Wastewater quality analysis

Concentrations of total nitrogen (TN), TP, and SS were analyzed with a HACH DR 6000 spectrophotometer according to the standard methods provided by HACH Company, USA. Chemical oxygen demand (COD) was determined by APHA closed reflux titrimetric method (Greenberg et al., 2005). Statistical analysis was completed using SPSS Statistics 20.0 and a value of $P < 0.05$ was regarded as statistically significant.

3 Results and discussion

3.1 Abundance and shape of MPs

The abundance and shape distributions of the MPs throughout the three WWTPs are shown in Table 2. For influent samples, the average MP concentration in W1 was 392.2 items/L, which was much higher than those in W2 and W3 with average concentrations of 71.2 and 38.3 items/L, respectively. The MP concentrations in W2 and W3 influents were similar to those in previous studies (Liu et al., 2019; Ziajahromi et al., 2021), and fibers and granules accounted for the majority of identified MPs. Among the three WWTPs, W1 and W2 have the same proportion of domestic wastewater in their influents (about 90%). However, the MP concentrations were significantly different. If MPs mainly come from personal care and

cosmetic products, then such a large difference in the MP concentration would not exist. Remarkably, more than 90% of MPs were granules in the influent of W1. Previous studies showed that granular MPs were often derived from toiletries related to industrial production (Fendall and Sewell, 2009; Carr et al., 2016). W1 and W3 have similar service populations and areas but with different proportions of industrial influent (10% in W1 and 60% in W3). Substantial amounts of MPs were only found in W1 not in W3, thereby suggesting that industrial wastewater source and not volume may be an important contributing factor for MP pollution.

From influent to primary effluent, the average MP concentrations in the three WWTPs decreased to 52.4, 47.3, and 16.1 items/L, respectively, with the removal rates of 86.6% (W1), 33.6% (W2), and 58.5% (W3) (Table S2). Despite the high MP concentration in the influent of W1, most granules were removed and only 32.8 items/L remained in the primary effluent. Screening and grit chamber as primary treatment units can remove grits, cinders, and suspended solids. Small granules can be more easily absorbed by these solids than fibers or fragments due to their small size (Tang et al., 2020). Bilgin et al. showed that the settling tanks of the grit chamber are principal units that capture MPs rather than aeration tanks or vortex-type tanks (Bilgin et al., 2020). The lowest removal of MPs in W2 was possibly due to the short hydraulic retention time of 10 min of settling tank.

As indicated in Table 2, secondary treatment units further removed MPs to the average concentrations of 3.9, 4.8, and 5.4 items/L in the secondary effluents of the three WWTPs with the removal rates of 92.6%, 89.9%, and 15.8%, respectively (Table S2). The activated sludge system managed to further decrease the MPs in the wastewater to 0.2%–14% (Sun et al., 2019), and it was 1.0%–13.9% in our study. Most MPs could be trapped by sludge flocs and extracellular polymeric substances in the activated sludge system and further removed by water–sludge separation process during the settling stage, which was related to HRT. In W3, the highest concentration and lowest removal rate of MPs might be due to the absence of an independent secondary sedimentation tank and short HRT (10.5 h), which was half of those of W1 and W2. The proportion of fiber MPs increased significantly at this stage, suggesting that fiber MPs could not be easily adsorbed by activated sludge. Fiber MPs were usually from laundry wastewater with a large diameter (Tang et al., 2020). In addition, fiber MPs are easier to twist with the current and escape from activated sludge due to its slender shape in comparison with granule and fragment MPs.

After tertiary treatment by disc filters, the average MP concentration decreased to 1.8 items/L in W1 and 0.5 items/L in W3 with the removal rates of 53.8% and 90.7%, respectively. However, DNBF did not remove MPs efficiently in W2, especially for granules with a small diameter. Long, thin fibers could also pass through packing

Table 2 The average abundance of MPs throughout the three WWTPs by irregular grab-sampling during the summer of 2020

Sampling points		The concentration of MPs (items/L)			
		Fibers	Granules	Fragments	Total
W1 ^{a)}	S1	25.1±13.5	356.0±230.3	11.1±2.6	392.2±223.1
	S2	14.0±6.7	32.8±17.0	5.6±5.9	52.4±29.4
	S3	1.0±1.14	0.9±1.0	2.0±1.9	3.9±2.6
	S4	0.7±0.5	0.2±0.2	0.9±0.8	1.8±1.2
W2	S1	23.5±7.2	37.5±3.4	10.4±5.1	71.2±8.2
	S2	12.3±9.1	28.3±9.6	6.7±4.1	47.3±25.5
	S3	3.7±0.4	1.1±0.1	ND ^{b)}	4.8±0.5
	S4	2.1±0.6	1.0±0.2	ND	3.1±0.5
W3	S1	8.1±2.8	22.0±4.3	8.7±2.7	38.8±1.8
	S2	2.6±0.2	10.3±8.4	3.2±0.4	16.1±11.1
	S3	3.0±0.3	2.1±0.9	0.3±0.1	5.4±1.7
	S4	0.3±0.1	0.1±0.1	0.1±0.1	0.5±0.2

Notes: a) W1, W2, and W3 represent the three WWTPs, respectively. b) ND means no detection.

media in filter directly (Claessens et al., 2013). DNBF was filled with 3–4 mm quartz sand coated with biofilm. Talvitie et al. (2017) found that DNBF could not significantly decrease MP concentration. However, the underlying reasons need to be further explored. In general, the MP concentration in the effluents of the three WWTPs occurred at moderate levels in other studies (Liu et al., 2019; Sun et al., 2019; Yang et al., 2019).

3.2 Characteristics of MPs

Various polymer types were identified in this study, including polyethylene oxides (PE), PP, polyester, PA, PS, and PET (Figs. S1 and 2(a)). As one of the most productive plastics in China (Ning, 2020), PE was detected as the most abundant polymer in the influents of all three WWTPs. The main shapes of PE MPs were granule and fragment, especially in W1, where PE MPs occupied 99.0% of the detected particles. PE MPs had been extensively reported in the influents of WWTPs due to their widespread application in film, pipe, wire, and cable production (Carr et al., 2016; Li et al., 2019a). To further explore the potential source of MPs in W1, we checked the list of enterprises that discharge wastewater to W1 and found a plastic-related manufacturer nearby. The company is a PE pipe manufacturer, where abundant MPs could be produced in the copolymerization of original ethylene, mold cleanup, and pipe cutting. The MP concentration in W1 varied significantly from 154.1 items/L to 690.5 items/L across six sampling activities, which was consistent with the intermittent discharge of industrial wastewater from the PE-pipe manufacturer. Moreover, no treatment facilities were available to prevent MPs from flowing into wastewater in the factory, thereby resulting in abundant MPs in the influent of W1.

Other common polymer types were PP, polybutene (PB), and PA. They were detected in the three WWTPs and reported WWTPs (Rezania et al., 2018). All identified PET showed similar morphology to the extracted fibers from synthetic cloths (Hernandez et al., 2017). However, PET was not detected in the influent of W3, which was possibly due to industrial wastewater taking up a large proportion (60%) of the influent. Another fibrous polymer type, polyester, was mainly found in W3, probably because of the fiber manufacturing wastewater flowing into W3. Some unusual polymer types, such as polyformaldehyde (POM) and polyethylene oxide (PEO), were also detected in W3 influent, and the overall diversity of polymer types in W3 was higher than those in W1 and W2. In general, the detected polymer types in the W1 influent were similar to those in W2 influent as the two WWTPs mainly receive and treat domestic wastewater. In the effluents, PE was still common in all WWTPs. The proportion of PET in W1 and W2 and polyester in W3 greatly increased, corresponding to abundant fibers in the effluents (Table 2). Monomers of PE, PET, and polyester obtain a low hazard score according to hazard ranking of polymers on the basis of monomer classifications (Lithner et al., 2011). However, PE, PET, and PA, including PVC, can also cause potential ecological risks by releasing endocrine disruptors (bisphenol A, styrene, and phthalates), solvents (benzene and methanol), and catalysts (potassium persulphate and zinc oxide) (Lithner et al., 2011).

More than 90% of all recovered microplastics from the influent samples were smaller than 0.5 mm, and so were MPs in the effluents (Fig. 2(b)). In the effluents of W2 and W3, the proportions of MPs with a size between 0.2 and 0.5 mm were highest, which was attributed to the abundant fibers in the effluents. Similarly, a recent study showed that microfibers of 0.1–0.3 mm accounted for the highest

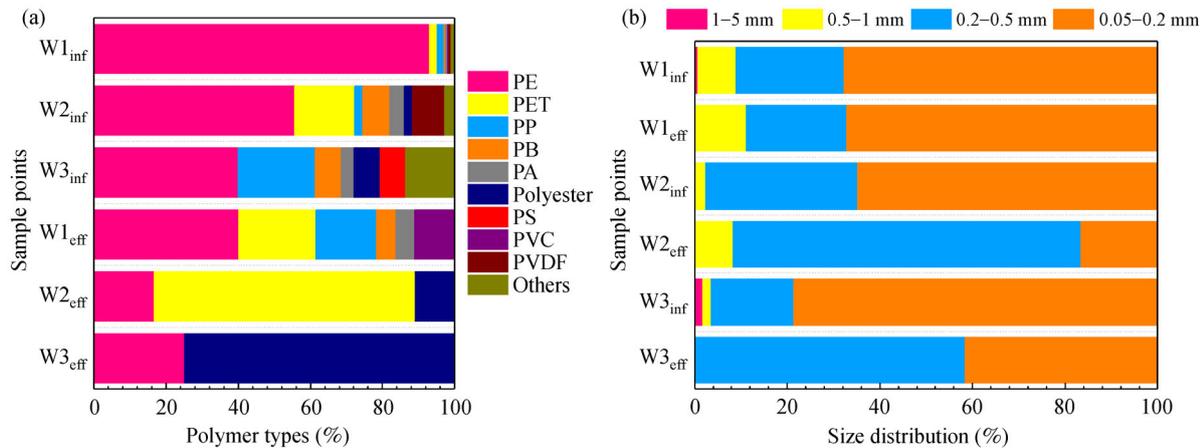


Fig. 2 Relative abundances of typical MP types (a) and size percentage distribution (b) in the influents and effluents of the three WWTPs. Here, “inf” and “eff” represent influent and effluent, respectively.

percentage in textile wastewater and surface water samples (Zhou et al., 2020b).

The color of MPs was also examined for the collected wastewater samples. As shown in Fig. 3(a), diverse colors were observed in the influents, and the most common color of the identified MPs was white (90.9%, 70.4%, and 57.8% in W1, W2, and W3, respectively). The transparent color was counted as white because of their similarity. The results further confirmed our speculation that MPs in W1 came from industrial wastewater because primary PE was mostly white or transparent in color. Other identified colors of influent MPs were red, black, green, and brown (Fig. S2). Compared with the influent, the diversity of the color of MPs was lower in the effluent and the main colors were white, red, and black (Fig. 3(b)). Previous studies showed that MPs with different colors could be selectively ingested by a series of aquatic fauna. For example, most MPs in the digestive tracts of Japanese anchovy were white or transparent (71%) in Tokyo Bay (Tanaka and Takada, 2016). The juveniles of planktivorous palm ruff prefer to capture black MPs rather than blue or yellow MPs because black MPs are similar to their food pellets (Ory et al., 2018). Therefore, the color, size, and type of MPs in the effluents of WWTPs are important to assess the ecological risk of MPs on the food web in water systems.

3.3 Fluctuation of influent MPs in W1 within 24 h

The above results showed that the MP concentration in W1 influent was typically higher and varied greatly, and approximately 90% of MPs were white PE granules (size between 0.05 and 0.2 mm) (Table 2, Figs. 2 and 3). To more accurately assess the MPs load and further track the potential sources of the MPs, a dense grab sampling within 24 h was conducted for W1 influent. Similar to the high concentrations and fluctuations of MPs in the random grab sampling campaigns, white PE granules with a small size

(0.05–0.2 mm) were still the main contributors to MPs and great variations were observed within 24 h. The MP concentration gradually increased from 2:00 to 17:30 and then sharply decreased at 19:00. From 19:00 to 22:00, MP concentration gradually decreased and finally increased to higher level at 24:00. The lowest concentration was detected between 2:00 and 8:30, which did not exceed the average value (392.2 items/L) from the above random grab sampling. The peak and subpeak concentrations of MPs occurred at 17:30 and 16:00 with 4617.6 and 1054.1 items/L, respectively, which were conspicuously higher than in previous studies (Liu et al., 2021). The results further verify the possibility of industrial sources of MPs in W1 influent. Notably, a peak also occurred at 24:00, which may be related to residents’ activities. It generally takes about 2–3 h for domestic wastewater to flow into WWTPs, and 21:00–23:00 is usually the time for urban residents to wash before going to bed. Therefore, the emergent MPs at 24:00 might come from PE granules in body and facial scrubs. We speculate that the primary production of MPs may be an important pathway to environments on the basis of the daily fluctuation pattern and industrial wastewater source of W1 (Fig. 4 and Table 1). However, policy restrictions on the primary sources of MPs, such as plastic pipe processing and plastic microbeads, have not been imposed in China. Some states in the USA had proposed to prohibit the use and sale of consumer products that contain MPs (Carr et al., 2016). Continuous attention should be paid to industrial sources of MPs in WWTPs (Wu et al., 2017).

The average MP concentration during dense sampling within 24 h was 661.8 ± 1176.8 items/L, much higher than the results of random grab sampling (392.2 ± 223.1 items/L). In comparison with daily dense sampling, the random grab sampling method is more widely applied in previous studies (Lares et al., 2018; Hu et al., 2019; Ziajahromi et al., 2021). For WWTPs with little variations in MPs

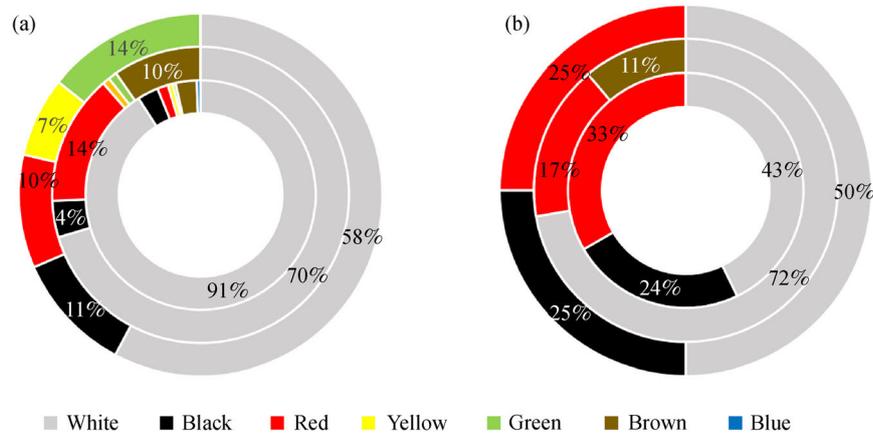


Fig. 3 Colors of MPs in the influents (a) and effluents (b) of the three WWTPs. Each color represents the color of MPs. Inner ring, middle ring, and outer ring indicate W1, W2, and W3, respectively.

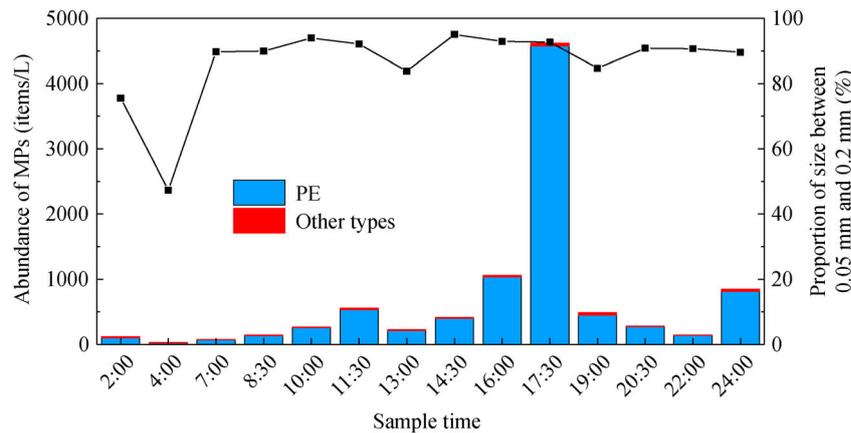


Fig. 4 Concentration, type, and size distributions of MPs in the influent of W1 during 24 h.

abundance, such as W2 and W3, the random grab sampling method could simply and efficiently reflect the MPs load. But for WWTPs with great load fluctuations such as W1, the random grab sampling needs to be carefully used. To accurately evaluate the MPs load in the influent, one or more daily dense sampling campaigns need to be conducted.

Previous studies found that the MP concentration was correlated with urban characteristics, such as population density and GDP of primary or secondary industries (Li et al., 2018b; Zhou et al., 2020a). Long et al. (2019) showed that MP abundance was positively correlated with SS in the influents of different WWTPs. However, the relationship between MP abundance and influent quality parameters within one day is not known. Here, possible relationships between MP concentration and influent parameters were further evaluated based on the 24 h samples. Results show no significant relationships between the MP concentration and contents of COD ($r = -0.003$, $P > 0.05$), TN ($r = -0.006$, $P > 0.05$), and TP ($r = 0.487$, $P > 0.05$). A significant positive correlation existed

between SS concentration and MPs abundance for influent samples ($r = 0.722$, $P < 0.01$), consistent with Long's study (Long et al., 2019). However, the correlation was too dependent on the maximum MP concentration. After the maximum value was deduced, the correlation between SS concentration and MPs abundance was not statistically significant ($r = 0.119$, $P > 0.05$). Different types of MPs reportedly showed inconsistent heteroaggregation with SS (Li et al., 2019b). The heteroaggregation of PE MPs and small SS had a minor effect on the settling of PE MPs (Li et al., 2019b), thereby suggesting the weak correlation between PE MPs and SS, as in the influents of W1. This finding indicates that measuring influent SS may not be a potential way to reflect MP abundance. More research should be conducted to analyze the relationship between MPs and SS in the future.

4 Conclusions

In this study, the distribution, characteristics, and diel

variations of MPs throughout three WWTPs with mixed domestic–industrial influents in Wuxi City, China, were evaluated by using two sampling strategies. Irregular grab sampling results showed that the MP concentration of 392.2 items/L in W1 influent was much higher than those of 71.2 and 38.3 items/L in W2 and W3 influents, respectively. Approximately 90% of MPs in W1 influent were small white PE granules. MP concentrations in these WWTPs effluents were less than 4.0 items/L. For the dense grab sampling of W1 influent within 24 h, we observed surprising large daily fluctuations (from 29.1 to 4617.6 items/L) of MP concentrations, indicating the limitation of random grab sampling. Moreover, long-term daily dense sampling campaigns should be conducted to obtain more precise information about MPs.

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References

- Ahmed W, Bivins A, Bertsch P M, Bibby K, Gyawali P, Sherchan S P, Simpson S L, Thomas K V, Verhagen R, Kitajima M, Mueller J F, Korajkic A (2021). Intraday variability of indicator and pathogenic viruses in 1-h and 24-h composite wastewater samples: Implications for wastewater-based epidemiology. *Environmental Research*, 193: 110531
- Alavian Petroody S S, Hashemi S H, van Gestel C A M (2020). Factors affecting microplastic retention and emission by a wastewater treatment plant on the southern coast of Caspian Sea. *Chemosphere*, 261: 128179
- Avio C G, Gorbi S, Milan M, Benedetti M, Fattorini D, d'Errico G, Paoletto M, Bargelloni L, Regoli F (2015). Pollutants bioavailability and toxicological risk from microplastics to marine mussels. *Environmental Pollution*, 198: 211–222
- Bergami E, Rota E, Caruso T, Birarda G, Vaccari L, Corsi I (2020). Plastics everywhere: first evidence of polystyrene fragments inside the common Antarctic collembolan *Cryptopygus antarcticus*. *Biology Letters*, 16(6): 20200093
- Bessa F, Ratcliffe N, Otero V, Sobral P, Marques J C, Waluda C M, Trathan P N, Xavier J C (2019). Microplastics in gentoo penguins from the Antarctic region. *Scientific Reports*, 9(1): 14191
- Bilgin M, Yurtsever M, Karadagli F (2020). Microplastic removal by aerated grit chambers versus settling tanks of a municipal wastewater treatment plant. *Journal of Water Process Engineering*, 38: 101604
- 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*, 91: 174–182
- Claessens M, Van Cauwenberghe L, Vandegehuchte M B, Janssen C R (2013). New techniques for the detection of microplastics in sediments and field collected organisms. *Marine Pollution Bulletin*, 70(1–2): 227–233
- Cowger W, Booth A M, Hamilton B M, Thaysen C, Primpke S, Munno K, Lusher A L, Dehaut A, Vaz V P, Liboiron M, Devriese L I, Hermabessiere L, Rochman C, Athey S N, Lynch J M, De Frond H, Gray A, Jones O A H, Brander S, Steele C, Moore S, Sanchez A, Nel H (2020). Reporting guidelines to increase the reproducibility and comparability of research on microplastics. *Applied Spectroscopy*, 74(9): 1066–1077
- Fendall L S, Sewell M A (2009). Contributing to marine pollution by washing your face: Microplastics in facial cleansers. *Marine Pollution Bulletin*, 58(8): 1225–1228
- Gatidou G, Arvaniti O S, Stasinakis A S (2019). Review on the occurrence and fate of microplastics in Sewage Treatment Plants. *Journal of Hazardous Materials*, 367: 504–512
- Greenberg A E, Trussell R R, Clesceri L S, Association A W W (2005). Standard methods for the examination of water and wastewater: Supplement to the sixteenth edition. *American Journal of Public Health & the Nations Health*, 56(3): 387
- Hermabessiere L, Dehaut A, Paul-Pont I, Lacroix C, Jezequel R, Soudant P, Duffos G (2017). Occurrence and effects of plastic additives on marine environments and organisms: A review. *Chemosphere*, 182: 781–793
- Hernandez E, Nowack B, Mitrano D M (2017). Polyester textiles as a source of microplastics from households: A mechanistic study to understand microfiber release during washing. *Environmental Science & Technology*, 51(12): 7036–7046
- Hidalgo-Ruz V, Gutow L, Thompson R C, Thiel M (2012). Microplastics in the marine environment: a review of the methods used for identification and quantification. *Environmental Science & Technology*, 46(6): 3060–3075
- Hu Y, Gong M, Wang J, Bassi A (2019). Current research trends on microplastic pollution from wastewater systems: A critical review. *Reviews in Environmental Science and Biotechnology*, 18(2): 207–230
- Koelmans A A, Mohamed Nor N H, Hermsen E, Kooi M, Mintenig S M, De France J (2019). Microplastics in freshwaters and drinking water: Critical review and assessment of data quality. *Water Research*, 155: 410–422
- 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
- Li L, Geng S, Wu C, Song K, Sun F, Visvanathan C, Xie F, Wang Q (2019a). Microplastics contamination in different trophic state lakes along the middle and lower reaches of Yangtze River Basin. *Environmental Pollution*, 254(Pt A): 112951
- Li W L, Zhang Z F, Ma W L, Liu L Y, Song W W, Li Y F (2018a). An evaluation on the intra-day dynamics, seasonal variations and removal of selected pharmaceuticals and personal care products

- from urban wastewater treatment plants. *Science of the Total Environment*, 640–641: 1139–1147
- Li X, Chen L, Mei Q, Dong B, Dai X, Ding G, Zeng E Y (2018b). Microplastics in sewage sludge from the wastewater treatment plants in China. *Water Research*, 142: 75–85
- Li Y, Wang X, Fu W, Xia X, Liu C, Min J, Zhang W, Crittenden J C (2019b). Interactions between nano/micro plastics and suspended sediment in water: Implications on aggregation and settling. *Water Research*, 161: 486–495
- Lithner D, Larsson A, Dave G (2011). Environmental and health hazard ranking and assessment of plastic polymers based on chemical composition. *Science of the Total Environment*, 409(18): 3309–3324
- 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
- Liu X, Wang J (2020). Algae (*Raphidocelis subcapitata*) mitigate combined toxicity of microplastic and lead on *Ceriodaphnia dubia*. *Frontiers of Environmental Science & Engineering*, 14(6): 97
- Liu X, Yuan W, Di M, Li Z, Wang J (2019). Transfer and fate of microplastics during the conventional activated sludge process in one wastewater treatment plant of China. *Chemical Engineering Journal*, 362: 176–182
- Long Z, Pan Z, Wang W, Ren J, Yu X, Lin L, Lin H, Chen H, Jin X (2019). Microplastic abundance, characteristics, and removal in wastewater treatment plants in a coastal city of China. *Water Research*, 155: 255–265
- Mason S A, Garneau D, Sutton R, Chu Y, Ehmann K, Barnes J, Fink P, Papazissimos D, Rogers D L (2016). Microplastic pollution is widely detected in US municipal wastewater treatment plant effluent. *Environmental Pollution*, 218: 1045–1054
- Ning J (2020). Progress of the world's plastics industry from 2018 to 2019(1). *China Plastics Industry*, 48(03): 1–14 (in Chinese)
- Ory N C, Gallardo C, Lenz M, Thiel M (2018). Capture, swallowing, and egestion of microplastics by a planktivorous juvenile fish. *Environmental Pollution*, 240: 566–573
- Park H J, Oh M J, Kim P G, Kim G, Jeong D H, Ju B K, Lee W S, Chung H M, Kang H J, Kwon J H (2020). National reconnaissance survey of microplastics in municipal wastewater treatment plants in Korea. *Environmental Science & Technology*, 54(3): 1503–1512
- Rezania S, Park J, Md Din M F, Mat Taib S, Talaiekhazani A, Kumar Yadav K, Kamyab H (2018). Microplastics pollution in different aquatic environments and biota: A review of recent studies. *Marine Pollution Bulletin*, 133: 191–208
- Sun J, Dai X, Wang Q, van Loosdrecht M C M, Ni B J (2019). Microplastics in wastewater treatment plants: Detection, occurrence and removal. *Water Research*, 152: 21–37
- 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
- Tanaka K, Takada H (2016). Microplastic fragments and microbeads in digestive tracts of planktivorous fish from urban coastal waters. *Scientific Reports*, 6(1): 34351
- Tang N, Liu X, Xing W (2020). Microplastics in wastewater treatment plants of Wuhan, Central China: Abundance, removal, and potential source in household wastewater. *Science of the Total Environment*, 745: 141026
- Van Cauwenberghe L, Vanreusel A, Mees J, Janssen C R (2013). Microplastic pollution in deep-sea sediments. *Environmental Pollution*, 182: 495–499
- Wang R, Ji M, Zhai H, Liu Y (2020). Occurrence of phthalate esters and microplastics in urban secondary effluents, receiving water bodies and reclaimed water treatment processes. *Science of the Total Environment*, 737: 140219
- Wei S, Luo H, Zou J, Chen J, Pan X, Rousseau D P L, Li J (2020). Characteristics and removal of microplastics in rural domestic wastewater treatment facilities of China. *Science of the Total Environment*, 739: 139935
- 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
- Wu W, Yang J, Criddle C S (2017). Microplastics pollution and reduction strategies. *Frontiers of Environmental Science & Engineering*, 11(1): 6
- Yang L, Li K, Cui S, Kang Y, An L, Lei K (2019). Removal of microplastics in municipal sewage from China's largest water reclamation plant. *Water Research*, 155: 175–181
- Zhao S, Danley M, Ward J E, Li D, Mincer T J (2017). An approach for extraction, characterization and quantitation of microplastic in natural marine snow using Raman microscopy. *Analytical Methods*, 9(9): 1470–1478
- Zhou G, Wang Q, Zhang J, Li Q, Wang Y, Wang M, Huang X (2020a). Distribution and characteristics of microplastics in urban waters of seven cities in the Tuojiang River basin, China. *Environmental Research*, 189: 109893
- Zhou H, Zhou L, Ma K (2020b). Microfiber from textile dyeing and printing wastewater of a typical industrial park in China: Occurrence, removal and release. *Science of the Total Environment*, 739: 140329
- Ziajahromi S, Neale P A, Telles Silveira I, Chua A, Leusch F D L (2021). An audit of microplastic abundance throughout three Australian wastewater treatment plants. *Chemosphere*, 263: 128294