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# Occurrence, characteristics and removal of microplastics in wastewater treatment plants with different treatment processes

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ABSTRACT

The abundance of microplastics (MPs) in wastewater from three wastewater treatment plants (WWTPs) were determined in Hangzhou, Zhejiang Province, China. The MPs abundance was 140–350 particles per litre in the influent and 10–30 particles per litre in the effluent. Four shapes of MPs in the influent were observed, while mainly only debris was left in the effluent. The percentage of small ($\leq$100 μm), medium (100–500 μm), and large-sized ($\geq$500 μm) plastics in the raw leachate of the three WWTPs were 54.3%, 8.6%, and 37.1%, 28.6%, 64.3%, and 7.1%, and 41.4%, 24.1%, and 34.5%, respectively. Mainly only the size of $\leq$100 μm was left in the effluent of all. The removal efficiencies of MPs in a range of 78.6% to 96.6% were achieved. Polypropylene, polystyrene, polyethylene, polyethylene terephthalate and polyvinyl chloride were the main types and detected in all wastewater samples, accounting for over 75% of all types. The plastic components contained in different industrial wastewater were more complex. The distribution of MPs was significantly positively correlated with most conventional indicators such as chemical oxygen demead, ammonia nitrogen, and total phosphorus, but not with heavy metals. Similar wastewater, different treatment processes, or similar processes but different wastewater (industrial wastewater proportion varied) could all lead to differences in MPs removal. The MPs abundance measured in this experiment was similar to some previous studies, but relatively high. The three WWTPs can discharge up to $6.0 \times 10^8$–$1.8 \times 10^9$ plastics of MPs per day, which poses potential ecological risks. This study indicates that the source control of MPs and optimizing the process design of existing WWTPs are crucial for preventing and controlling MPs pollution.

## 1. Introduction

Microplastics (MPs) refer to plastic fragments and particles with a diameter less than 5 mm, which can be divided into primary and secondary MPs [1]. MPs are widely distributed with other pollutants in environments such as water bodies, air and soil, and are difficult to degrade [2–6]. Large amounts of MPs accumulation can alter the physical properties of soil and sediment, thereby disrupting the balance of ecosystems [7]. Moreover, MPs have strong adsorption properties and can adsorb harmful substances such as heavy metals and persistent organic pollutants in the environment [8]. These harmful substances accumulate in organisms when MPs enter them and spread through the food chain, ultimately endangering human health [9].

The distribution and pollution of MPs in marine, river, soil, atmosphere and other environmental media have been widely studied [10–12]. The Yangtze River in China carried up to 1.5 million tons of MPs into the East China Sea every year, with the highest MPs load among many rivers [13]. The spatial distribution of MPs in surface soil of different land use types in Lahore, Pakistan showed that the range of MPs content in surface soil was as high as 1750–12200/kg, with the highest concentration in park soil [7]. It can be seen that MPs pollution in the environment is widespread and high-risk.

Numerous studies have shown that the wastewater treatment plants (WWTPs) are important sinks and sources of MPs [14,15]. Researches have shown that MPs can be effectively removed (90% to 99%) after treatment in WWTPs, but the removal situation varied greatly among different treatment processes in different regions. In

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addition, there are various other organic-inorganic pollutants in drinking water, such as fluoride and nitrate pollution found in researches [16–20], which make the removal of MPs more complex due to their combined pollution. The study has shown that the secondary treatment step of a WWTP in Haikou city of China eliminated most of the MPs, with an average concentration of 12.98 ($P \cdot L^{-1}$) in the effluent, indicating a 94.09% of total MPs removal rate. It mainly fixed MPs in activated sludge flocs through microbial adsorption and flocculation, and then discharged them out of the system with the remaining sludge [15]. Another study showed that the MPs abundance decreased from 288.5 particles per litre of the influent to 22.9 particles per litre of the effluent, corresponding to the removal rate of 92.1% in a WWTP in Xi'an city of China [21]. Zhang *et al.* [22] found that most MPs were transported from the wastewater to the sludge, and the concentration of MPs in dehydrated sludge was 27.2 ± 3.1 particles per litre. Different processes have different removal mechanisms for MPs. Biological treatment utilizes microbial flocs to adsorb MPs, combined with membrane filtration or sludge settling to achieve interception; Third level treatment (sand filtration, ozone oxidation, activated carbon adsorption) further removes residual MPs through deep filtration, chemical degradation, or surface adsorption [14,15].

However, the above studies were mostly focused on individual WWTP in a certain area. There is relatively little research on the removal of MPs in different plants in the same region. Therefore, it is worth studying the impact of using similar or different treatment processes on the removal of MPs from wastewater generated in the same region in different WWTPs. In addition, Zhejiang Province is one of the most developed provinces in China. Large amounts of plastics are used in the daily life and industrial production in Zhejiang Province, and the environment in many parts of Zhejiang Province is contaminated with MPs [23]. However, MPs in WWTPs in Zhejiang Province have been studied little. Therefore, the objectives of this study were to determine the abundance and removal of MPs in WWTPs in Hangzhou of Zhejiang Province of China, analyze the removal patterns and effects of different processing techniques on MPs of different types, sizes, and shapes, and to compare the results with the results of previous studies.

## 2. Materials and Methods

### 2.1. Sample collection

Wastewater samples were collected from three WWTPs in Hangzhou City, Zhejiang Province, China. The three WWTPs sites are in Liangzhu (site 1), Jingshan (site 2), and Yuhang (site 3). Among them, sites 1 and 3 mainly collect local residents' domestic sewage and a small amount of industrial wastewater (80% domestic sewage, 20% industrial wastewater), while site 2 mainly collects residents' domestic sewage and industrial wastewater from Jingshan industrial zone (60% domestic sewage, 40% industrial wastewater). The introduction of the WWTPs is listed in Table S1, Supplementary Material. The main sources of MPs in domestic sewage include washing wastewater, personal care products, and daily plastic products, mainly including polyethylene terephthalate (PET), polyethylene (PE), polyamide (PA), polypropylene (PP), polystyrene (PS) and polyvinyl chloride (PVC), *etc.* According to the distribution of industries in Jingshan industrial zone, the plastic types vary greatly, with common types including PET, PA, PVC, PS, polyurethane (PU), acrylonitrile butadiene styrene (ABS), ethylene-vinyl acetate (EVA) and polybutylene terephthalate (PBT), *etc.*

The sampling points of site 1 included the raw wastewater in the main influent (Inf) (*i.e.*, the feed to the treatment process), and the effluents from the anaerobic-anoxic-aerobic biological systems (AAO), the membrane bioreactor (MBR) unit, and the disinfection contact tank (DCT) unit. The effluent from DCT was the main effluent (Eff). The sampling points of site 2 was similar with the site 1, which included the Inf, the effluents from AAO and MBR, and Eff. The sampling points of site 3 included the Inf, and the effluents from the oxidation ditch (OD) unit, the secondary sedimentation tank (SST) unit and the biological aerated filter (BAF), and Eff. The sampling diagram is shown in Fig. 1. The wastewater samples were collected three times in one day. Three samples (each 1500 mL) were collected at each sampling point each time. The wastewater was collected using clean glass bottles and then promptly transferred to dark flasks. These samples were immediately sent to the laboratory for storage at 4 °C. Chemical analyses were carried out in

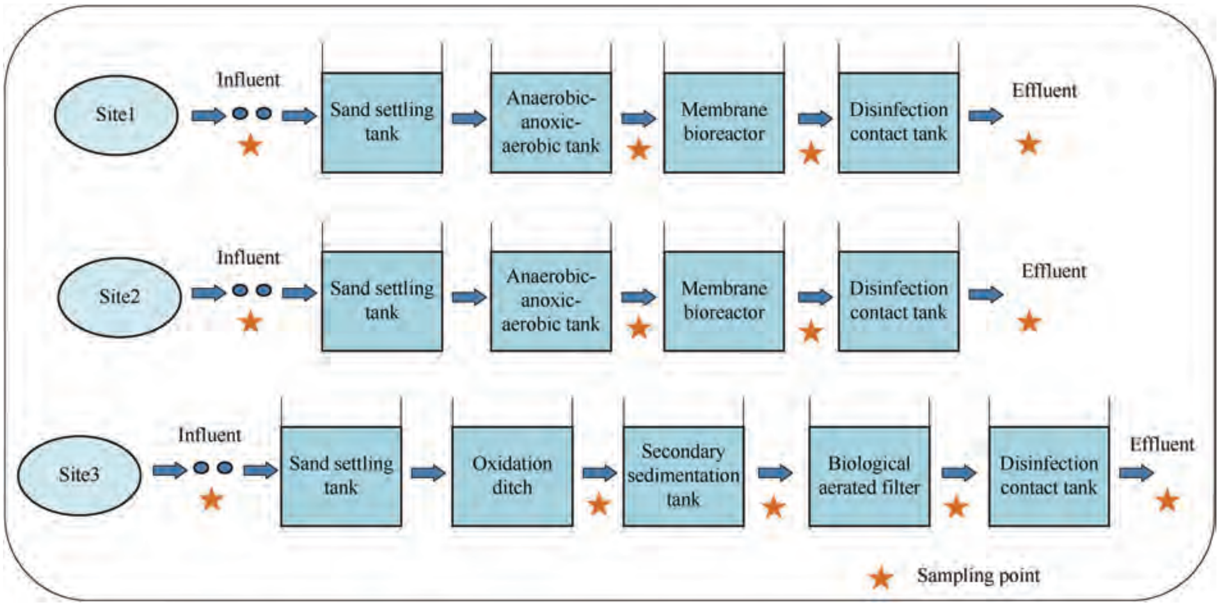

**Fig. 1.** Diagrams of sampling points of three WWPTs.

triplicate during the subsequent two days. The chemicals used in the experiment were of analytical purity.

### 2.2. Sample pretreatment

To avoid potential plastic contamination during the experiment, all containers were rinsed with deionized water. The pretreatment process of MPs determination in wastewater was referred to Zhang *et al.* [1]. Briefly, the 500 ml wastewater samples were poured through the stainless steel filter membrane with pore sizes of 1.0 μm. All particles left on the filter membrane were transferred to a beaker with $H_2O_2$ (30% (vol)). Then these wet particles were placed in covered culture dishes and dried in an oven (60 °C). This digestion process lasted for three days until it was completely digested. Then the samples were filtered again and the filter membrane was placed into the flotation device. The potassium formate solution with a concentration of 1.50 g·m/L was added to the flotation device to submerge the filtration membrane. The flotation device was placed in an ultrasonic device for 20 min and was left to stand overnight. Then the supernatant in the flotation device was collected by filtering through a 1.0 μm stainless steel filter membrane (Fig. S1). Finally the membrane was dried for 5 min before being stored in a glass culture dish for testing.

### 2.3. Detection method

The quantity, shape and particle size of the dried samples were determined on each filter membrane using the visual method by a fluorescence microscope (NIB910-FL). The detail determination method was referred to Long *et al.* [24]. Briefly, 20 circles ($d = 4.4$ mm) per filter membrane as subsamples were randomly defined. The number of MPs within the 20 circles was averaged and the average value was used to estimate the quantity of MPs across the whole filter membrane. The total area covered by the 20 circles exceeded 70% of the effective area of filter membrane. The identification of MPs was made using a Raman spectrometer (Shimadzu, AIRsight) (Fig. S2). The spectra of suspected MPs was compared with the Raman spectral database to determine the polymer type [22].

The removal of MPs can also be affected by the character of wastewater, so the physicochemical indicators and heavy metals of wastewater have been also tested. The pH, chemical oxygen demand (COD), ammonia nitrogen ($NH_3$-N), and total phosphorus (TP) concentrations in the wastewater samples were determined in accordance with the "Standard Procedure for Water and Wastewater Detection" [25], followed by measurements using a visible ultraviolet spectrophotometer (TU1901). The content of heavy metals in wastewater was determined by inductively coupled plasma spectrometry (ICP–MS) after pretreatment with aqua regia [26].

### 2.4. Quality control and statistical analysis

Before sampling, the sampling tools were appropriately cleaned. During the whole experiment, no plastic item was used. The cotton lab coats and nitrile gloves were used during the entire process of sample collection and analysis [15]. To verify the validity of the experimental data, the recovery experiments using the internal

**Table 1**
Treatment efficiency of conventional indicators of sewage.

| WWPTs | Parameter | Influent (except pH)/mg·L$^{-1}$ | Effluent (except pH)/mg·L$^{-1}$ | Discharge standard value (except pH)/mg·L$^{-1}$ | Whether it met the standard |
|---|---|---|---|---|---|
| Liangzhu (site 1) | pH | 7.08 ± 0.02 | 7.01 ± 0.01 | 6–9 | Y |
| | COD | 139.95 ± 2.78 | 28.06 ± 0.05 | 40 | Y |
| | $NH_3$-N | 43.46 ± 1.02 | 0.27 ± 0.02 | 4 | Y |
| | TP | 5.66 ± 0.01 | 0.17 ± 0.01 | 0.3 | Y |
| | Cd | 0.0004 ± 0.0001 | 0.0003 ± 0.0001 | 0.01 | Y |
| | Cr | 0.008 ± 0.001 | 0.007 ± 0.001 | 0.1 | Y |
| | As | 0.003 ± 0.001 | 0.002 ± 0.001 | 0.1 | Y |
| | Pb | 0.012 ± 0.002 | 0.010 ± 0.001 | 0.1 | Y |
| | Ni | 0.006 ± 0.001 | 0.006 ± 0.001 | 0.05 | Y |
| | Cu | 0.045 ± 0.001 | 0.029 ± 0.001 | 0.5 | Y |
| | Zn | 0.217 ± 0.003 | 0.119 ± 0.004 | 1.0 | Y |
| | Mn | 0.203 ± 0.002 | 0.161 ± 0.006 | 2.0 | Y |
| Jingshan (site 2) | pH | 7.38 ± 0.01 | 6.85 ± 0.03 | 6–9 | Y |
| | COD | 126.90 ± 3.00 | 10.13 ± 1.46 | 40 | Y |
| | $NH_3$-N | 21.28 ± 0.10 | 0.06 ± 0.01 | 4 | Y |
| | TP | 4.40 ± 0.02 | 0.22 ± 0.01 | 0.3 | Y |
| | Cd | 0.0003 ± 0.0001 | 0.0003 ± 0.0001 | 0.01 | Y |
| | Cr | 0.010 ± 0.001 | 0.009 ± 0.001 | 0.1 | Y |
| | As | 0.005 ± 0.001 | 0.003 ± 0.001 | 0.1 | Y |
| | Pb | 0.011 ± 0.002 | 0.010 ± 0.001 | 0.1 | Y |
| | Ni | 0.020 ± 0.003 | 0.017 ± 0.002 | 0.05 | Y |
| | Cu | 0.100 ± 0.002 | 0.073 ± 0.001 | 0.5 | Y |
| | Zn | 0.292 ± 0.002 | 0.294 ± 0.001 | 1.0 | Y |
| | Mn | 0.522 ± 0.001 | 0.091 ± 0.003 | 2.0 | Y |
| Yuhang (site 3) | pH | 7.18 ± 0.01 | 7.00 ± 0.01 | 6–9 | Y |
| | COD | 271.38 ± 2.00 | 0.84 ± 0.03 | 40 | Y |
| | $NH_3$-N | 32.06 ± 1.16 | 0.18 ± 0.03 | 4 | Y |
| | TP | 7.37 ± 0.05 | 0.12 ± 0.01 | 0.3 | Y |
| | Cd | 0.0003 ± 0.0001 | 0.0002 ± 0.0001 | 0.01 | Y |
| | Cr | 0.009 ± 0.002 | 0.008 ± 0.001 | 0.1 | Y |
| | As | 0.003 ± 0.001 | 0.002 ± 0.001 | 0.1 | Y |
| | Pb | 0.011 ± 0.002 | 0.011 ± 0.001 | 0.1 | Y |
| | Ni | 0.009 ± 0.001 | 0.007 ± 0.001 | 0.05 | Y |
| | Cu | 0.066 ± 0.001 | 0.060 ± 0.002 | 0.5 | Y |
| | Zn | 0.321 ± 0.006 | 0.278 ± 0.003 | 1.0 | Y |
| | Mn | 0.173 ± 0.003 | 0.072 ± 0.001 | 2.0 | Y |

standard method were conducted. Three procedural blank control groups were set up in the experiment. Each experiment was performed three times in parallel, and the statistical average of all indicators was used to present the results. Statistical analysis was conducted using Excel 2016 (USA) and SPSS 26.0 (USA) software including the calculation of average and standard deviation. The graphing was performed using Origin 2022 (USA).

## 3. Results and Discussion

### 3.1. Conventional indicator removal rate

The physicochemical indicators of wastewater, such as ionic strength and organic content, indirectly regulate the removal efficiency of MPs by affecting their surface properties and adsorption

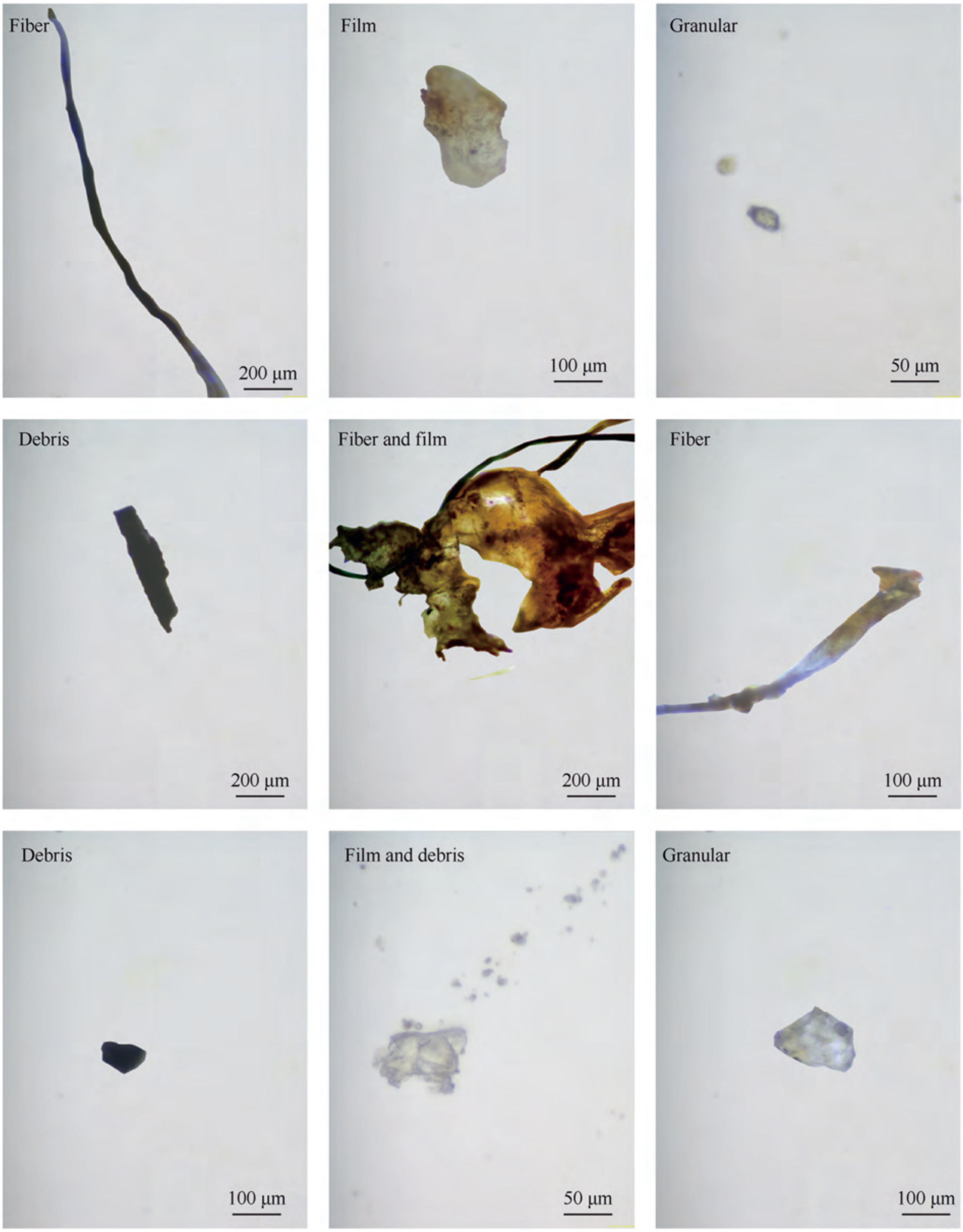

**Fig. 2.** Microscopic photos of various shapes of MPs.

behavior [27]. The presence of heavy metals may enhance the aggregation stability of MPs or hinder their effective removal pathways through competitive adsorption, charge neutralization, and complexation shielding effects, ultimately affecting the removal efficiency of MPs by traditional physicochemical treatment processes [28,29]. The treatment efficiency of conventional indicators of sewage from three WWTPs is shown in Table 1. The COD, $NH_3$-N and TP of the influent of the three WWTPs were 126.90–271.38, 21.28–43.46 and 4.40–7.37 mg/L, respectively. After processing technology, all indicator values could meet the discharge standards. For example, The COD concentrations were 28.06, 10.13 and 0.84 mg/L in the effluent of site 1, 2 and 3 respectively, which were all less than the discharge standard value (40 mg/L). In addition, the concentrations of $NH_3$-N and TP of the effluent were both much less than their respective discharge standard value (4 and 0.3 mg/L). Similarly, all heavy metals could ultimately meet the discharge standards (Table 1). These three sites complied with the strictest discharge standards for WWTPs in China, indicating that their treatment processes were effective enough in removing conventional pollutants. However, many emerging contaminants, including MPs, have not been included in the discharge standard indicators, so their existence and removal should be given more attention.

### 3.2. MPs morphological characters

MPs were detected in all samples from the three WWTPs sites. The abundance of MPs was 140–350 particles per litre in the influent of the different WWTPs. MPs presented various shapes, mainly divided into four categories, namely debris, granular, film and fiber, respectively (Fig. 2). From the figure, it can also be seen that the surface color and transparency of MPs were different, which may be due to the varying degrees of aging of different types of MPs in sewage environments. In order to further analyze the changes in the surface of MPs, Fig. 3 shows the MPs morphology of different treatment units in the three WWTPs. It can be seen that four different shapes of MPs were detected in the influent of site 1 and site 3, which may be related to the similar wastewater composition (80% domestic sewage, 20% industrial wastewater) of the two WWTPs (Table S1). Comparing the effluent and influent, it can be clearly seen that MPs shape types have decreased, but there were certain fluctuations in each unit during the treatment process. The reason for this situation may be that MPs encountered different physical and chemical reactions in different sewage environments. For example, the shape of fiber which did not appear elsewhere, appeared in the effluent of the MBR of site 2. This may be due to the detachment of substances from the biofilm or the release of MPs from the membrane themselves. Previous studies have also shown that while membrane treatment processes remove pollutants including MPs, they may also release MPs due to aging and other reasons [30,31]. After treatment, the shape of MPs in the effluent became simple, with only debris and granular remaining in sites 1 and 2, accounting for both 66.7% and 33.3%, while in the effluent in site 3, only debris remained.

The different size distribution of MPs from the three WWTPs is shown in Fig. 4. Various sizes of MPs were distributed in the influent from the different sites. The percentage of small (≤100 μm), medium (100–500 μm), and large-sized (≥500 μm) plastics in the influent of the three sites were 54.3%, 8.6%, and 37.1%, 28.6%, 64.3%, and 7.1%, and 41.4%, 24.1%, and 34.5%, respectively. During the processing of site 2, a large size of MPs (≥500 μm) appeared, which may be related to the release of MPs from the sludge in the AAO and MBR. The similar phenomenon of MPs rising instead of decreasing after biological treatment of wastewater was also found in previous studies [15,16]. Ultimately, the large-sized MPs was effectively removed, while the MPs size in the effluent was smaller. Except for the size of ≤100 μm and 100–500 μm

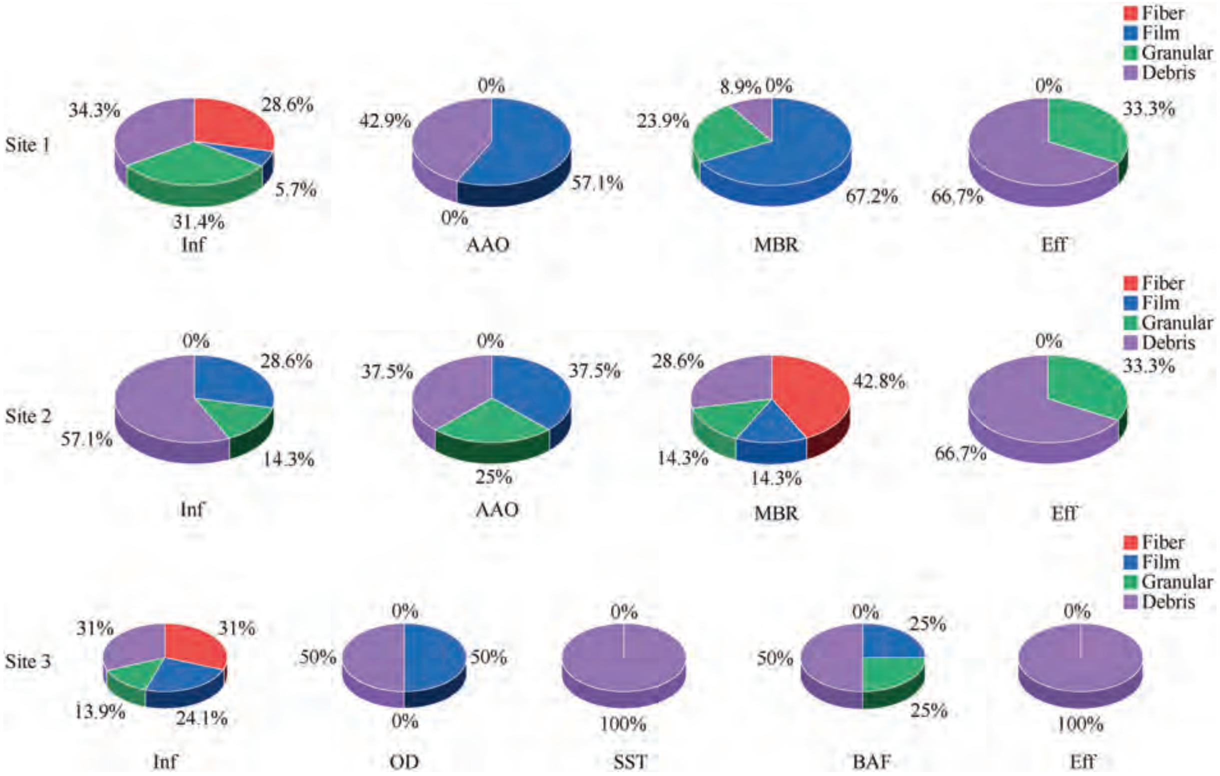

**Fig. 3.** Distributions of shape of MPs. The data is presented as the average value. Inf: influent; AAO: anaerobic-anoxic-aerobic biological systems; MBR: membrane bioreactor; OD: oxidation ditch; SST: secondary sedimentation tank; BAF: biological aerated filter; Eff: effluent.

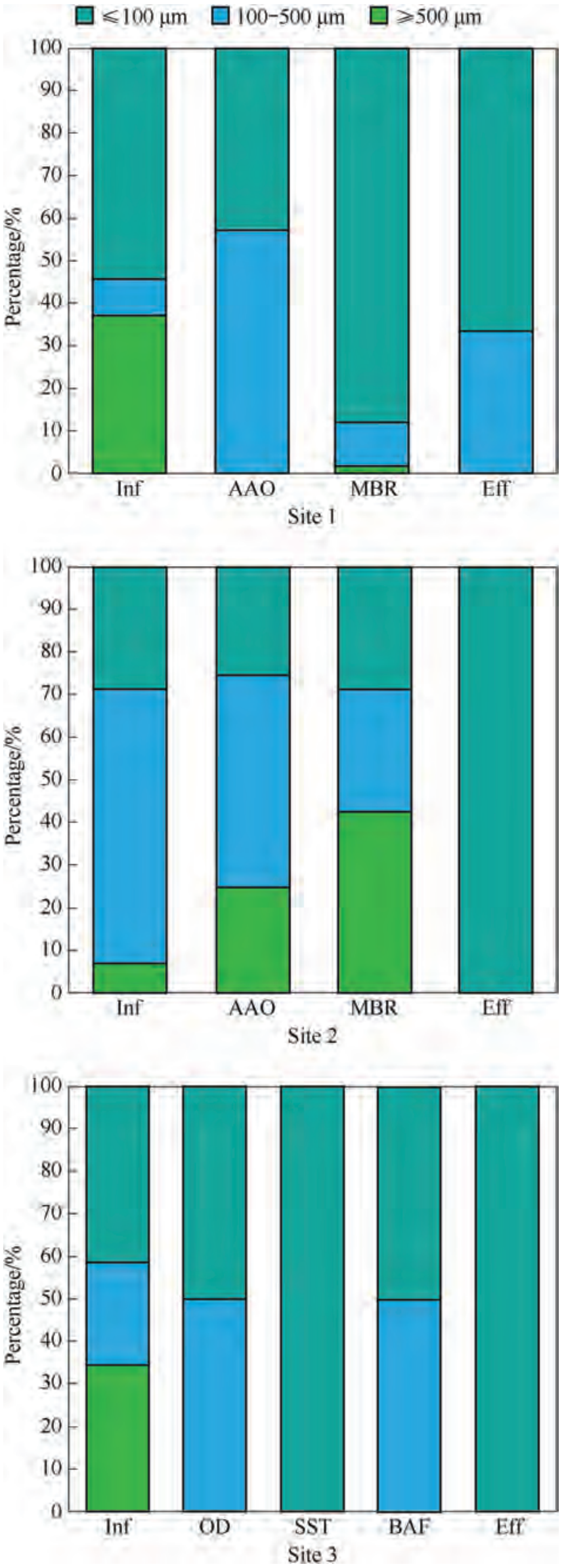

**Fig. 4.** Distributions of size of MPs. The data is presented as the average value. Inf: influent; AAO: anaerobic-anoxic-aerobic biological systems; MBR: membrane bioreactor; OD: oxidation ditch; SST: secondary sedimentation tank; BAF: biological aerated filter; Eff: effluent.

remaining in sites 1, which account for 33.3% and 66.7% respectively, the other two only have the smallest size (≤100 μm) left.

### 3.3. MPs polymers types

In order to further clarify the composition of MPs, The MPs polymers types in the influent and effluent of the three WWTPs have been analyzed, and the results are shown in Fig. 5. The MPs extracted from the wastewater of three sites comprised PP, PS, PE, PET, PVC, PA, PU, EVA, ABS, PBT and acrylic. Among them, PP, PS, PE, PET and PVC were the main types and detected in all wastewater samples, accounting for over 75% of all types. As shown in Fig. 5(a), PP with 36.9% was the major constituent of total MPs found in the influent of site 1 followed by PS (28.9%), PE (18.6%), PET (10.6%), PVC (2.6%), and others (2.6%, PBT), respectively. Similarly, the same main types were found in two other sites. This result is similar to previous research findings. For example, PP accounted for 40% and PET for 56% among the identified MPs detected in the influent of a WWTP from Korea [32]. For another, PET, PS, PE and PP accounted for greater than 70% of detected MPs in a WWTP from Beijing of China [33]. This may be due to the worldwide use of PP, PE, PS and PET in daily products such as films, bags, and containers. Mainly, those plastic products are easy to break down in various ways under environmental exposure and would be transferred or discharged into the wastewater pipes [34].

From Fig. 5(b), the proportion of 'other' types (including PA, PU, EVA, ABS and acrylic) of MPs in the influent and effluent from site 2 was 21.4% and 16.7%, respectively, which was significantly higher than the other two sites. This may be related to the composition of the wastewater from this site containing a large amount of industrial wastewater (40%, Table S1). Although the proportion of 'other' types in the effluent from site 3 was high (20%, mainly PU) (Fig. 5(c)), their concentration was also low due to their small total abundance (10 particles per litre). This result indicated the plastic components contained in different industrial wastewater were more complex. In addition, the variation in MPs types in sewage from WWTPs could be related to factors such as different populations, commercial patterns and consumer behavior in different regions [34].

### 3.4. MPs abundance and removal efficiency

The abundance of MPs in the influent of the three WWTPs was 350, 140, and 290 particles per litre, respectively. This is similar to the total amount of MPs contained in other WWTPs in previous studies [16,35,36]. The abundance of MPs generally decreased with the treatment process (Fig. 6). For example, the removal rate of MPs after the treatment of AAO and MBR and in the final effluent were 42.9%, 50.0% and 78.6%, respectively in site 2. This demonstrated that this treatment process had a good removal efficiency for MPs. It is worth noting that, although the treatment processes for site 1 and site 2 were similar, their MPs abundance and removal efficiency differed obviously. The removal contribution rates of AAO and MBR in site 1 and site 2 were 80.0% and 2.9%, 42.9% and 7.1%, respectively. The reason for this should be that there was a significant difference in the influent between the two WWTPs. Site 1's influent contained 20% industrial wastewater, while site 2's influent contained 40% industrial wastewater (Table S1). The difference in influent quality directly leads to differences in the initial MPs content and its removal efficiency [15]. The wastewater quality of site 1 and site 3 was similar, but the treatment process was different, also resulting in different MPs final removal rates. The overall removal rate of MPs of site 3 was higher than that of site 1. Moreover, the removal efficiency of the waste water conventional indicators of site 3 was also better than that of site 1. This indicated

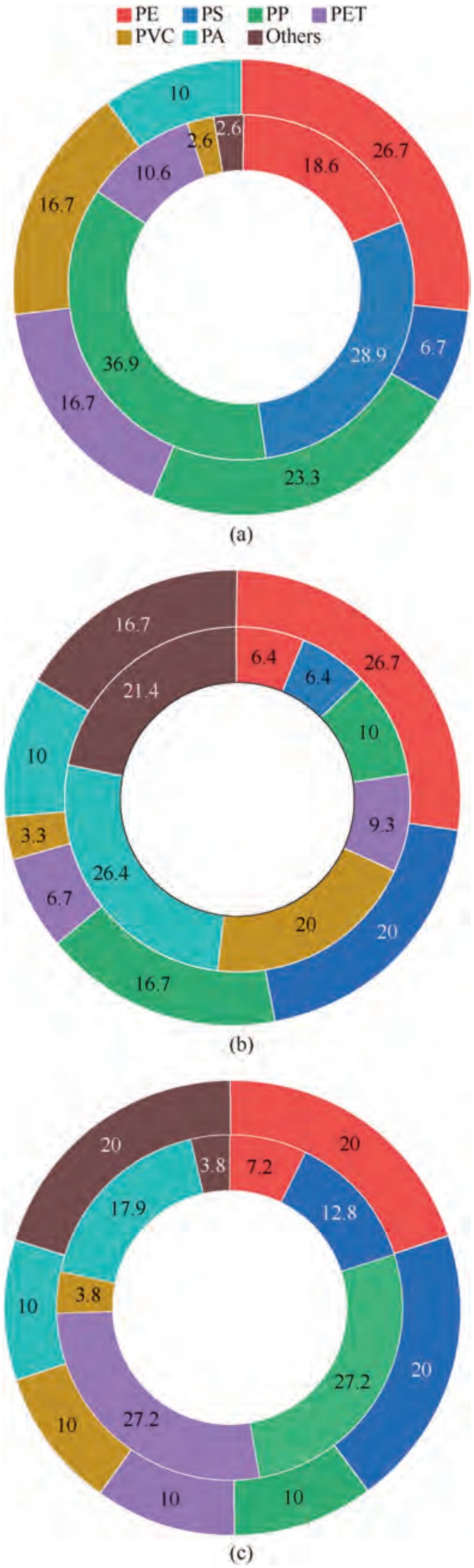

**Fig. 5.** Type of polymer characteristics of MPs in WWTPs (the data is presented as the average value): (a) site 1, (b) site 2, (c) site 3. Inner ring means influent; Outer ring means effluent. Results are shown as percentage. PP: polypropylene; PS: polystyrene; PE: polyethylene; PET: polyethylene terephthalate; PVC: polyvinyl chloride; PA: polyamide.

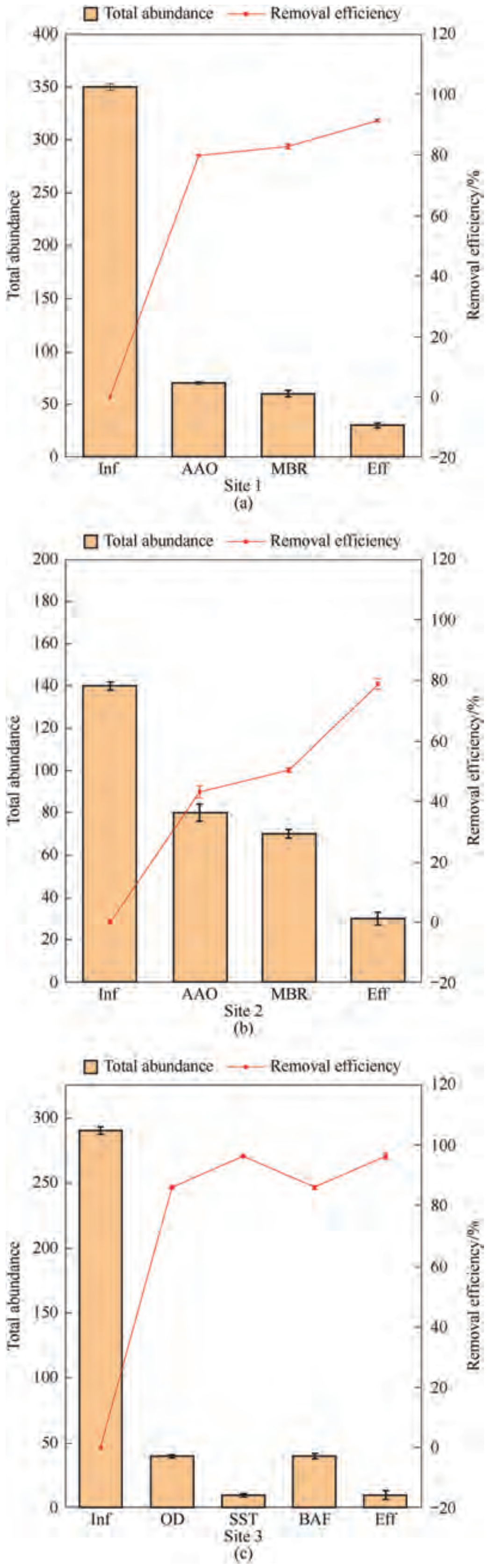

**Fig. 6.** The abundance changes (particles per litre) and removal characteristics of MPs: (a) site 1, (b) site 2, (c) site3.

that the process of OD had better pollutant removal efficiency than MBR for the urban sewage in this area. The OD process can effectively remove MPs from wastewater through the synergistic effect of aeration and sedimentation. The aeration process may break down some large-sized MPs into smaller sizes, and the subsequent settling process can further remove them [9]. In addition, the hydraulic retention time of the OD process is longer, which can promote full contact between MPs and microorganisms, enhance the adsorption and settling effects of MPs [37]. In contrast, although MBR technology exhibits higher efficiency in removing MPs through membrane retention and adsorption, its backwashing process may lead to the release of some MPs, thereby affecting its stability [9]. However, this result is exactly opposite to the previous conclusion that OD had a better MPs removal rate than MBR [37]. The reason for this result may be related not only to the differences in the removal mechanisms of MPs between the two processes mentioned above, but also to the different properties of the wastewater. This further illustrates the complexity of removing MPs from wastewater.

The abundance of MPs in the effluent of the three sites was 30, 30, and 10 particles per litre, with overall removal efficiencies of 91.4%, 78.6%, and 96.6%, respectively. The initial abundance of MPs in the wastewater from site 2 was not high, but its removal efficiency was not as good as the other two, which may be related to its 40% industrial wastewater content. The removal efficiency of MPs was not only related to the treatment process, but also to the characteristics of wastewater quality. The removal of MPs, organic pollutants, and some heavy metals showed a strong correlation [14,38]. Therefore, the use of advanced combination technologies is necessary to improve the removal efficiency of pollutants, including MPs, in water [18,19]. Furthermore, the various treatment facilities had high removal efficiency for MPs and their content in the effluent was already very low in this study, but combined with the amount of wastewater (Table S1), the three WWTPs sites can

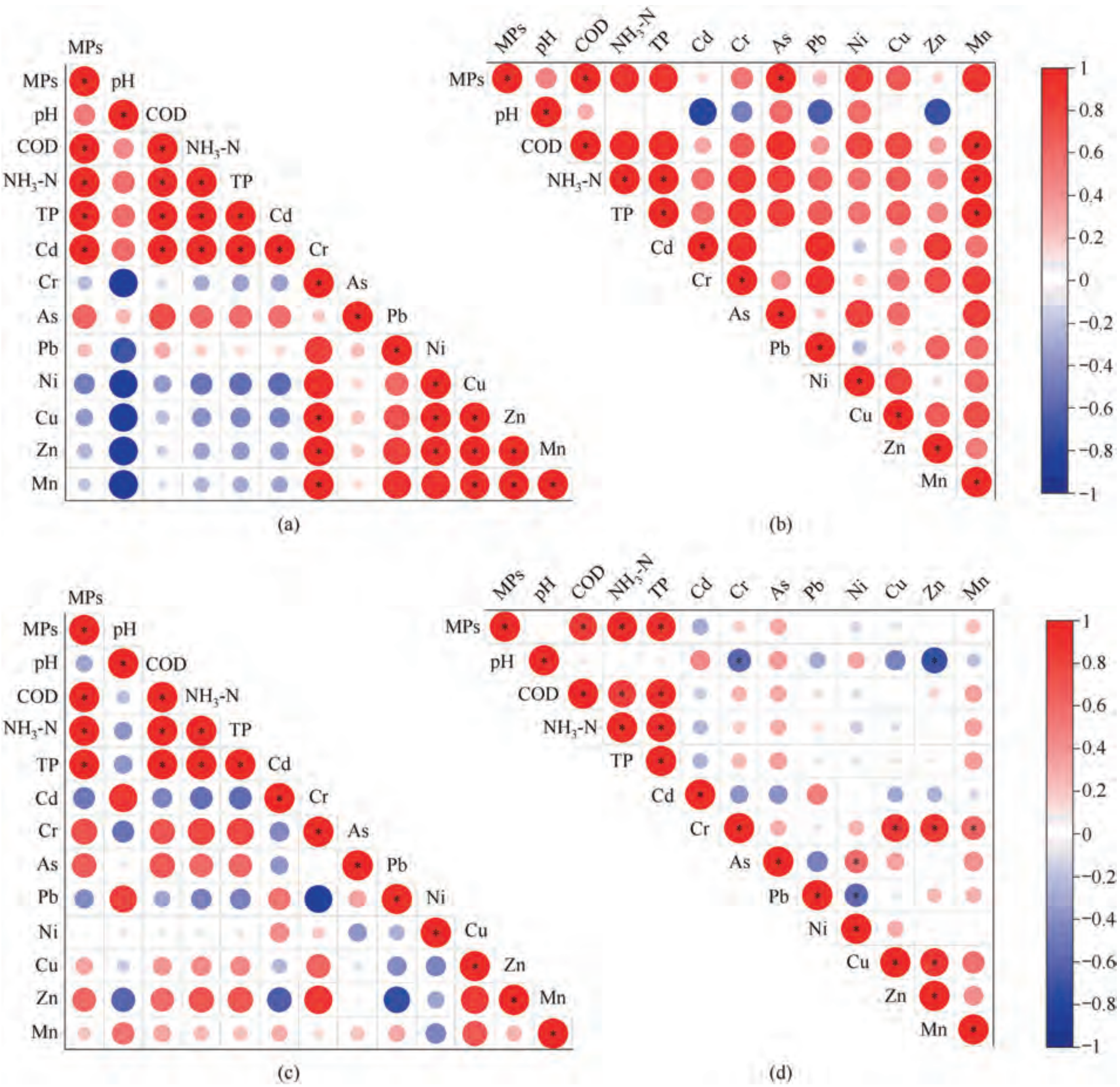

**Fig. 7.** The Person correlation analysis among MPs abundance, conventional indicators, and heavy metals was conducted in each unit of site 1 (a), site 2 (b), site 3 (c), and three sites (d). $p \le 0.05$.

discharge up to $1.8 \times 10^9$, $9.0 \times 10^8$, and $6.0 \times 10^8$ particles of MPs per day, which poses potential ecological risks that cannot be ignored.

### 3.5. *Correlation analysis*

The removal of MPs in wastewater is related to the properties of the wastewater itself. In order to clarify which factors were related to the removal of MPs, further correlation analysis was conducted on the distribution of MPs, conventional water quality indicators, and heavy metal concentration. The results are shown in Fig. 7. The distribution of MPs was significantly positively correlated with most of the conventional indicators including COD, $NH_3$-N and TP. Specially, there was a significant correlation between MPs and COD in all three sites, indicating that MPs, like COD, have been effectively removed with the step-by-step treatment of the processes.

Heavy metals may interfere with biological treatment processes by inhibiting microbial activity, leading to a decrease in the efficiency of organic matter degradation. Their binding to the surface of MPs may alter the electrochemical properties of particulate matter, weakening the coagulation and sedimentation effect [27]; MPs, due to their hydrophobicity and large specific surface area, are prone to adsorb heavy metals and form composite pollutants. These composite materials may clog filter media, reduce membrane separation efficiency, and affect the stability of conventional physical separation due to complex changes in size and density, ultimately affecting the processing load and energy consumption [28]. However, there is basically no correlation between MPs and heavy metals in this study, except for a significant positive correlation between MPs and Cd in site 1 and MPs and As in site 2. This result is different from previous studies. Previous study revealed that MPs were acting as heavy metals vector and the interaction between MPs and heavy metals may alter their environmental behaviors, bioavail-ability and potential toxicity, leading to ecological risks [39,40]. The reason why MPs and heavy metals often show a correlation is that the concentration of heavy metals on the biofilm on the surface of MPs has been measured [41,42]. In this study, the distribution of MPs and heavy metals in the wastewater were separately analyzed. The deeper possible reasons will be studied later. Heavy metals cannot be completely degraded and removed in the process, mainly through sludge adsorption or membrane filtration transfer. Although the concentration of heavy metals in the effluent was much lower than that in the influent, there was no gradual decrease in the treatment process, but rather a fluctuation phenomenon, which may be due to the uncertainty of adsorption and desorption of sludge and other pollutants, including MPs themselves.

### 3.6. *Comparison of different processing techniques*

As mentioned above, this experiment tested the distribution and removal efficiencies of MPs in three WWTPs in Hangzhou. Similar water quality, different treatment processes, or similar treatment processes but different water quality could all lead to differences in MPs removal. In order to further understand the distribution and removal of MPs in WWTPs in other regions, Table 2 presents a comparative analysis of their respective processes and removal efficiencies of MPs. It shows that removal efficiencies of MPs in a range of 78.6% to 99.5% were achieved by different processes in WWTPs. For example, the use of AAO with MBR at a WWTP in Wuxi of China showed 99.5% removal of MPs [37], while the sand filter process of a WWTP in Italy, had the potential to remove 84.0% of total MPs [42]. In this study, the abundance of MPs in the effluent was 10–30 particles per litre (Table 2). This was similar to the research results of certain cities, such as 35 particles per litre in Paris [35] and 22.9 particles/L in Xi'an [21]. In contrast, contrasting MPs abundance in the effluent of WWTPs was reported at other areas such as 0.59 particles per litre in Beijing [33] and 0.14 particles per litre in Korea [43].

It can be seen that the abundance of MPs in the influent of WWTPs varies greatly in different regions (Table 2). In most adjacent areas of China, the composition of urban sewage is relatively stable, and in principle, the distribution of MPs should not differ too much. However, the data in Table 2 shows significant differences. This may be because in addition to water quality and treatment processes, the sampling and testing methods for MPs studies may also influence the results. The limitations of the method for determining MPs in wastewater are mainly reflected in multiple aspects: during the sampling stage, uneven distribution of MPs and limitations in the aperture of sampling equipment may lead to insufficient representativeness of the sample, especially for small particle sizes (<1 μm) that are easily overlooked; During the preprocessing process, the filtering and digestion steps may introduce external pollution or damage some MPs structures, affecting detection accuracy, and the steps are cumbersome and time-consuming; When analyzing and identifying, mainstream technologies such as FTIR and Raman spectroscopy have low sensitivity to small-sized microplastics (<20 μm), and organic or particulate matter in complex wastewater matrices can easily

**Table 2**
Comparison of MPs removal by different processing techniques in different regions.

| Area | Main processing techniques | MPs abundance in influent | MPs abundance in effluent | Removal efficiency/% | References |
|---|---|---|---|---|---|
| Mikkeli, Finland | MBR | 57.6 | 1.0 | 98.3 | [31] |
| Paris, France | BAF | 290 | 35 | 88.0 | [35] |
| Northern, Italy | Sand filter | 2.5 | 0.4 | 84.0 | [42] |
| Oldenburg, Germany | Post-filtration | 167 | 5 | 97.0 | [36] |
| Y-City, Korea | SBR | 16.45 | 0.14 | 99.1 | [43] |
| Beijing, China | AAO + UF | 12.03 | 0.59 | 95.1 | [33] |
| Xi'an, China | OD | 288.5 | 22.9 | 92.1 | [21] |
| Xiamen, China | BAF | 1.57–13.69 | 0.20–1.73 | 79.3–97.8 | [24] |
| Guiyang, China | SBR/AAO | 32.5 | 5.0 | 84.6 | [22] |
| Haikou, China | AAO | 219.50 | 12.98 | 94.0 | [15] |
| Wuxi, China | AAO + MBR | 0.28–4 | 0.05–0.13 | 99.5 | [37] |
| Nanjing, China | SBR + BAF | 44 | 2 | 95.5 | [1] |
| Hangzhou, China | AAO + MBR<br>AAO + MBR<br>OD + BAF | 140–290 | 10–30 | 78.6–96.6 | This study |

Note: MBR- membrane bioreactor; BAF- biological aerated filter; SBR- sequencing batch reactor; AAO- anaerobic-anoxic-aerobic biological systems; UF- Ultrafiltration; OD- oxidation ditch; MPs abunclance, particles per litre.

interfere. In addition, the lack of unified standard methods leads to poor data comparability, while expensive equipment and long detection cycles further limit the ability for large-scale monitoring and real-time evaluation [44,45]. These factors collectively constrain the precise quantification and effective control of MPs pollution in wastewater. This indicated an urgent need to standardize the methods of MPs sampling and analysis in future research.

## 4. Conclusions

The abundance of MPs was 140–350 particles per litre in the influent of three WWTPs in Hangzhou, Zhejiang Province, China. Four shapes of MPs in the influent were observed, while mainly only debris left in the effluent, and the size was basically $\leq$100 μm. PP, PS, PE, PET and PVC were the main types and detected in all wastewater samples, accounting for over 75% of all types. The distribution of MPs was significantly positively correlated with COD, $NH_3$-N and TP, but not with heavy metals. After the treatment units of AAO with MBR, or OD with BAF, the removal efficiencies of MPs were 78.6% to 96.6%. However, the three WWTPs can discharge up to $6.0 \times 10^8$–$1.8 \times 10^9$ plastics of MPs per day, which poses potential ecological risks. Future research will further analyze the comparison of different types of MPs removal, in order to better understand the impact and mechanism of different processes on their removal, and to optimize the removal process and operating conditions to reduce MPs emissions. Strengthening microbial adsorption and adding appropriate coagulants may improve the removal rate of MPs in wastewater by OD process. Optimize membrane material and pore size design, improve membrane cleaning and maintenance methods, optimize aeration and backwash frequency of MBR system may improve the removal of MPs by MBR process. These are future research goals to reduce MPs emissions and ecological risks.

## CRediT Authorship Contribution Statement

Yiting Lian: Writing – original draft, Resources. Xianwei Wang: Writing – review & editing, Validation, Data curation. Peng Sun: Software, Methodology, Investigation. Hua Wang: Writing – review & editing, Data curation. Chengran Fang: Supervision, 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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## Supplementary Material

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