Removal of microplastics in municipal sewage from China’s largest water reclamation plant

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A B S T R A C T:

Municipal sewage treatment plants (STPs) are an important point source of microplastics in domestic waterways. In the present study, effluents from the largest water reclamation plant in China were sampled throughout the treatment process and microplastics were extracted and identified to evaluate their removal. As expected, microplastics were detected in the influent (12.03 ± 1.29 items/L). Following treatment, concentrations of microplastics were reduced by greater than 95% and 0.59 ± 0.22 items/L of microplastics were detected in reclaimed waters. Among detected microplastics, 18 types of polymers of ten colors were identified. Polyethylene terephthalate (PET), polystyrene (PS) and polypropylene (PP) accounted for greater than 70% of detected microplastics. Furthermore, microfibers were the dominant shape detected with an average size of 1110.72 ± 862.95 μm. However, microparticles accounted for only 14.08% of total microplastics with an average size of 681.46 ± 528.73 μm. Results of the present study suggest that current treatment technologies employed at the chosen STP are efficient to remove the majority of microplastics, however consideration of STPs as a point source of microplastics is important due to the large volumes of effluents being released into the aquatic environment on a consistent basis.

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1. Introduction

Microplastics are polymer particles that are less than 5 mm in size. Microplastics can be ingested by aquatic organisms and adversely affect growth, development, and reproduction (Wright et al., 2013). Recently, it has been demonstrated that microplastics accumulate in the liver, kidney and gut of mice and alter energy and lipid metabolism as well as oxidative stress, suggesting potential toxicity to mammals (Deng et al., 2017). Moreover, microplastics might pose a potential threat to human health through exposure to contaminated salts (Karami et al., 2017), aquatic products (Bessa et al., 2018; Van Cauwenbergh and Janssen, 2014), beer (Liebezeit and Liebezeit, 2014), and drinking water (Schymanski et al., 2018).

Recently, primary and secondary sources of microplastics and their contribution to environmental microplastics were evaluated (Sundt et al., 2014, 2016; Verschoor et al., 2016). Among sources, sewage treatment plants (STPs) have been identified as a primary source of microplastics in the environment (Carr et al., 2016; Estahbanati and Fahrenfeld, 2016; Murphy et al., 2016) as significantly greater concentrations of microplastics have been detected downstream of an STP when compared to water upstream of the STP (Estahbanati and Fahrenfeld, 2016; McCormick et al., 2014). Moreover, a strong association between STP and plastic pollution in rivers has been demonstrated (Hoellein et al., 2016). Although STPs have been demonstrated to remove greater than 90% of microplastics from influents (Carr et al., 2016; Talvitie et al., 2017a,b), the large volumes of treated effluents containing low concentrations of microplastics being released daily to receiving waters is cause for concern. It is estimated that between 15,000 and 4.5 million microplastic particles per day are released to surface waters from STPs regardless of removal efficiencies (Hoellein et al., 2016). In addition, it has been demonstrated that microplastic particles have a high affinity for pathogenic bacteria used in wastewater treatment processes which can lead to their release in effluents (Kirstein et al., 2016; Viršek et al., 2017; Raju et al., 2018). Consequently, engineered bacterial communities absorbed to microplastic
particles are more likely to be harmful than natural bacteria found in rivers (Harrison et al., 2017; McCormick et al., 2014).

To mitigate potential risks from the release of microplastics to receiving waters, stringent water quality requirements for effluents are required. However, there currently exists no specific requirements for the removal of microplastics by STPs. Therefore, an understanding of microplastic removal by STPs implementing various wastewater treatment technologies would be helpful for identifying best performing technologies. Recently, removal efficiency of microplastics from effluents by four municipal STPs in Finland implementing different advanced final-stage treatment technologies were investigated. Results of the study demonstrated greater than 99.9% removal of microplastics by membrane bio-reactors (reduction from 6.9 to 0.005 item/L), 97% removal by rapid sand filter (from 0.7 to 0.02 item/L), 95% removal by dissolved air flotation (from 0.1 to 2.0 item/L), and 40–98.5% by disc filter treatment (from 0.5 to 2.0 to 0.03–0.3 item/L) (Talvitie et al., 2017a). In China, widely adopted wastewater treatment systems consist of A2O treatment (anaerobic, anoxic, and aerobic) and subsequent advanced treatment processes (denitrification, ultrafiltration, ozonation and ultraviolet). It has been estimated that 1.56 × 10^14 sludge-based microplastic particles are released per year to surrounding water bodies (Li et al., 2018). However, little attention has been given to the removal of microplastics throughout the treatment process.

In the present study, we aimed to gain an understanding of the physical and chemical properties and composition of microplastics in STPs, and to evaluate their removal throughout the treatment process. Thus, the removal of microplastics at different treatment stages were quantified continuously for 3 months at the largest water reclamation plant in China.

2. Materials and methods

2.1. Effluent sampling

The Gaobeidian sewage treatment plant (gSTP), located in the Chaoyang District of Beijing, is the largest STP and largest water reclamation plant in China. The plant treats primarily domestic effluents with a mean flow of 1,000,000 m^3/day and services a population of approximately 2,400,000 people. Treatment processes include an aerated grit chamber, primary sedimentation tank, secondary sedimentation tank following A2O treatments (anaerobic, anoxic, and aerobic), and a series of advanced treatment processes (denitrification, ultra-filtration, ozonation and ultraviolet). Overall, hydraulic retention time is 4–12 h, and sludge retention is 5–15 days.

To evaluate the removal of microplastics throughout the treatment process, 30 L samples were collected from April to June in 2018 at the beginning of each month from the influent, effluent from the primary tank, secondary sedimentation tank and draining pool, respectively (Fig. 1). Water samples were collected by use of glass bottles, and then stored at 4 °C for further analysis. During our experiment, all tools including beakers, bottles, sieves, and funnels before and after each use were thoroughly washed and rinsed with tap water filtered through a 1 μm nylon membrane (Jiudinggaoke Filter (Beijing) Corporation, China), and then covered with aluminum foil.

2.2. Isolation and purification of microplastics

Influent and effluent samples were treated by use of a slight modified wet peroxide oxidation as recommended by National Oceanic and Atmospheric Administration of U.S. for the isolation and purification of microplastics in water (Masura et al., 2015). Briefly, water samples were filtered in 10L batches by using of a 5 mm and 50 μm (300 mesh) stainless steel sieve (Φ = 20 cm, n = 3). All solids retained on the 50 μm sieve were transferred to a 1L beaker filled with tap water and then filtered through a 10 μm nylon membrane (Φ = 5 cm). Membranes laden with filtered microplastics were soaked in 200 mL beakers with 50 mL ultra-purified water and sonicated for 20min. Next, membranes were rinsed 3 times with fresh ultra-purified water and the final volume of water in the beaker was less than 100 mL. Beakers were covered with aluminum foil and placed in a 90 °C drying oven for 12 h or until samples were dry.

20 mL of a freshly prepared FeSO4 solution (0.05 M) was added to the dried beakers. Subsequently, 20 mL of hydrogen peroxide solution (30%) was added and mixed vigorously to oxidize organic materials and beakers were left at room temperature for 5min without disturbance. Next, beakers were heated to 60 °C on a hotplate for 30min while stirring continuously by use of a glass rod. An additional 20 mL of hydrogen peroxide was added to beakers with visible solid particles, subsequent aliquots of hydrogen peroxide were added to beakers until no solid particles were visible. The resulting solution was cooled to room temperature and ZnCl2(s) was added on a weight to volume basis (97 g/100 mL = 1.5 g/cm^3) to facilitate density separation. Next, 5–10 mL HCl was added to facilitate dissolution of ZnCl2. Solutions were transferred to a separatory funnel and settled overnight. Settled solids were collected from the bottom of the separatory funnel and discarded following visual inspection for microplastics by use of a microscope. The remaining solution was filtered by use of a PTFE membrane (10 μm).

To prevent contamination of samples throughout the treatment process, all water and solutions were filtered by use of a 1 μm filter membrane prior to use. In addition, all tools and bottles were rinsed three times with filtered water. Moreover, containers were covered with lids to prevent contamination by air. Three negative controls, each consisting of 10 L filtered tap water (1 μm), were treated and analyzed as described above to investigate potential sources of contamination.

2.3. Identification of microplastics

Suspected microplastics were confirmed, counted and imaged by use of a LEICA DM4 M (Germany). First, suspected microplastics were classified as a fiber or particle. Next, these microfibers were classified into ten groups according to their color. Classified microfibers and microparticles were analyzed by use of a Spotlight 200 FT-IR Microscopy Systems (μ-FT-IR) (PerkinElmer, USA). The full spectrum of each particle was analyzed by use of a Bio-Rad KnowItAll® Informatics System 2018 (64-bit)-IR Spectral Library (Bio-Rad Laboratories, California, USA) to confirm microplastic identities. Following noise and baseline correction, each spectrum was searched and automatically matched to potential spectra from a database of known polymers or compounds. Identities of microplastics were then determined by matching peak wavenumber positions. Finally, the number of microplastics of each type were recalculated after removal of non-plastic particles. Daily releases of microplastics from gSTP were estimated by use of equation (1):

\[
\text{Microplastics released / day} = \frac{\text{Concentration}_{\text{microplastics}} \times \text{Volume}_{\text{effluents}}}{\text{Volumeeffluents}/C2/C14} \times \text{C drying oven for 12 h or until samples were dry.}
\]

2.4. Data analysis

Numbers of microplastic among samples were compared by use
of one-way analysis of variance (ANOVA), and when necessary, data were transformed appropriately to meet assumptions of ANOVA. The post-hoc Tukey test was used to identify significant differences among samples (p < 0.05). Pearson correlation analysis was performed to compare stage of treatment and concentrations of microplastics.

3. Results and discussion

3.1. Contamination of samples

Use of negative controls are important to assess potential contamination of samples by the treatment or extraction process. Although a number of precautions were taken to prevent contamination such as; minimal exposure of samples to air, use of nitrile gloves and laboratory coats, covering of filter membranes etc., contamination of negative controls by microfibers was detected and ranged from 1 item/filter to 7 items/filter. However, other sources of microplastics from room fiber and equipment cannot also be discounted. Detected microfibers attributed to contamination were mainly PET, polyvinyl chloride (PVC), and PS as identified by use of μ-FTIR. As a result, microplastic counts in each sample were corrected by of the average background counts for the negative control samples. In addition, cotton fiber contamination was detected in samples and might originate from experimental clothes, these were not classified as microplastics. Although a number of precautions were taken to prevent contamination, it could not be avoided completely. Thus, implementation of more stringent contamination control measures is advised to ensure reliability of results, such as treatment of samples in a clean bench. For example, 96.5% aerial microfiber contamination had been eliminated by the used clean bench (Wesch et al., 2017).

3.2. Microplastic composition in effluents from gSTP

A total of 18 types of polymers were detected in samples collected from gSTP between April and June 2018. However, only 2 polymers including PET and phenol-Formaldehyde resin (PF) were detected in all samplings (Table 1), which has previously been reported in other wastewater samples (Murphy et al., 2016). As expected, the polymer types decreased with the treatment train, that is, 6–10 types of polymers were detected in influents, and only 4–6 types of polymers were detected in draining effluents, indicating that most microplastics were removed effectively, especially the microparticles whatever their materials. Moreover, no obvious difference on polymer composition was found among samplings. Overall, PET (42.25%), polyester (PES) (19.09%), and polypropylene (PP) (13.05%) were the most prevalent microplastics, accounting for greater than 70% of total microplastics. PET and PES were mainly detected as fibers and might originate from textile fibers in domestic washing effluents. Interestingly, the fraction of PE which accounted for only 1.64% was rather lower compared to previous studies from Europe (av. 14%) (Talvitie et al., 2017a) and the US (>90%) (Carr and Tesoro, 2016), implying the potential differences in microplastic use between China and Europe/US. Moreover, these PE particles were detected in influent and effluents Fig. 1. Sampling diagram for microplastics from Gaobeidian sewage treatment plant (gSTP). Samples ①-④ were collected from the influent, primary effluent, secondary effluent, and outlet effluent, respectively.
collected in May only, but not in April and June. In addition, some microplastics (2.09%) also demonstrated mixed characteristics of polyethylene (PE) and PP. Synthetic rubber microparticles (1.30%) were identified in effluents and might originate from tire wear. As a whole, the majority of microplastics were black (36.60%), transparent (33.84%) or blue (11.88%; Fig. 2), suggesting these microplastics came from a number of differing sources.

By use of physical shape, microplastics were further classified into two groups: microfibers or microparticles. Microfibers (85.92% of microplastics) (Fig. 3(a)–2(d)) averaged 1110.72 ± 862.95 μm in size (Fig. 4(a)) and implies they are likely from domestic washing. Microplastics (Fig. 3(e)–2(h)), which included spherical, granule, fragment, film, and irregular shapes, accounting for 14.08% of microplastics by color in effluent and quantification by FTIR, and the highest ratio of 85% of suspected microplastics from effluents were natural particle or fiber mainly. This was similar to a previous study which had reported 22 and 90% of the suspected microplastics to be non-plastic particles (Ziajahromi et al., 2017).

### 3.3. Removal of microplastics throughout the treatment process

Microplastics were detected in all effluents collected from gSTP, and their concentration decreased substantially throughout the treatment processes (Fig. 5), suggesting their efficient removal by current treatment technologies implemented at gSTP. Briefly, 12.03 ± 1.29 items/L of microplastics were detected in incoming influents and was similar to influents from Scotland (15.70 ± 5.20 items/L) (Murphy et al., 2016), but was lesser slightly than wastewater collected from Canada (31.1 ± 6.7 items/L) (Gies et al., 2018). Following primary aerated grit treatment, 58.84 ± 8.05% of microplastics were removed and was significantly less than the influent (p < 0.05); subsequently, 54.47 ± 14.73% microplastics and 71.67 ± 11.58% microplastics were removed consecutively after A2/O and a series of advanced treatments (p < 0.05), respectively. Overall, 95.16 ± 1.57% of microplastics in the incoming influents were removed following treatment at gSTP. Although the removal efficiency of microplastics in gSTP was lesser than that of a membrane bioreactor (99.9%) (Talvitie et al., 2017a), it was similar to previous reports (Carr et al., 2016; Talvitie et al., 2017b) for rapid sand filters and dissolved air flotation treatment technologies (Talvitie et al., 2017a). Throughout the treatment process, no single treatment step had a significantly greater removal efficiency, thus indicating that microplastic removal was reliant on the treatment process as a whole. Finally, only 0.59 ± 0.22 items/L were detected in the final effluent (reclaimed water) which was similar to final effluents (0.25 ± 0.04 items/L) in Scotland (Murphy et al., 2016) and in Australia (0.28 items/L) (Ziajahromi et al., 2017), but was higher slightly than the effluents in Europe (Talvitie et al., 2017a) and the US (Mason et al., 2016). Moreover, microfibers comprised the majority of microplastics in reclaimed waters to be used for industrial cooling, street flushing, urban greening, or in effluents discharged into surrounding water bodies.

It is important to note that treatment technologies currently used throughout gSTP were not designed for the removal of microplastics. Regardless, a large fraction of microplastics were removed from the influent at gSTP. Further improvements would be difficult as advanced technologies are currently employed at gSTP. For example, it has been reported that installed post-filtration units can be used to reduce synthetic fiber loads (Mintenig et al., 2017), a technology already employed at gSTP (ultra-filtration) following the secondary sedimentation treatment. As a greater understanding of the composition of microplastics in incoming influents is gained, microplastic sources can be better controlled to limit their

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**Table 1**

Relative abundance of polymers and microplastics in effluents collected from gSTP.

<table>
<thead>
<tr>
<th>Polymer</th>
<th>Percentage %</th>
<th>Sampling time</th>
</tr>
</thead>
<tbody>
<tr>
<td>melamine co-polycondensation resin, MCP</td>
<td>0.11</td>
<td>✓ ✓</td>
</tr>
<tr>
<td>ethylene-vinyl acetate resin, EVA</td>
<td>0.17</td>
<td>✓ ✓</td>
</tr>
<tr>
<td>Polyurethane, PU</td>
<td>0.23</td>
<td>✓ ✓</td>
</tr>
<tr>
<td>Polyphenylene Oxide, PPO</td>
<td>0.28</td>
<td>✓ ✓</td>
</tr>
<tr>
<td>Nylon, PA</td>
<td>0.45</td>
<td>✓ ✓</td>
</tr>
<tr>
<td>Phenolic epoxy resin, PER</td>
<td>0.45</td>
<td>✓ ✓</td>
</tr>
<tr>
<td>Polyacrylonitrile, PAN</td>
<td>0.51</td>
<td>✓ ✓</td>
</tr>
<tr>
<td>polyvinyl alcohol, PVA</td>
<td>0.56</td>
<td>✓ ✓</td>
</tr>
<tr>
<td>Synthetic rubber, SR</td>
<td>1.30</td>
<td>✓ ✓</td>
</tr>
<tr>
<td>Phenol-Formaldehyde Resin, PF</td>
<td>1.64</td>
<td>✓ ✓ ✓</td>
</tr>
<tr>
<td>Polyethylene, PE</td>
<td>1.64</td>
<td>✓ ✓ ✓</td>
</tr>
<tr>
<td>Polychlorinated chloride, PVDC</td>
<td>2.43</td>
<td>✓ ✓</td>
</tr>
<tr>
<td>Polycrystalline, PAC</td>
<td>2.77</td>
<td>✓ ✓</td>
</tr>
<tr>
<td>Polymide, PI</td>
<td>3.90</td>
<td>✓ ✓ ✓</td>
</tr>
<tr>
<td>Polyvinyl chloride, PVC</td>
<td>7.06</td>
<td>✓ ✓ ✓</td>
</tr>
<tr>
<td>Polystyrene, PP</td>
<td>13.05</td>
<td>✓ ✓ ✓</td>
</tr>
<tr>
<td>Polyester, PES</td>
<td>19.10</td>
<td>✓ ✓ ✓</td>
</tr>
<tr>
<td>Polytetraethylene terephthalate, PET</td>
<td>42.26</td>
<td>✓ ✓ ✓</td>
</tr>
</tbody>
</table>

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**Fig. 2.** Classification of microplastics by color in effluent collected from gSTP. Results are presented as relative abundance (%). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)
presence in the influent. In the present study, microfibers were the dominant components of influents and effluents, and greater than 60% of microfibers were significantly removed ($p < 0.05$) following aerated grit chamber treatment. Thus, aerated grit chamber treatment might be an effective alternative to be used in reducing microfiber loads in STPs. For example, the use of a microfiber catching bag could be implemented throughout the washing process to limit the initial discharge of microfibers and subsequent downstream release from STPs.

Based on the present investigation data, it is estimated that approximately $0.59 \pm 0.22 \times 10^3$ items of microplastics are released into the aquatic environment daily from gSTP, while the majority of microplastics in influent ($95.16 \pm 1.57\%$) remained in sewage sludge. And it is estimated that the microplastics in dry sludge from 28 STPs across 11 provinces in China ranged between $1.60-56.4 \times 10^3$ microplastics/kg dry sludge with an average of $22.7 \pm 12.1 \times 10^3$ microplastics/kg dry sludge (Li et al., 2018). Additionally, microfibers were the predominant form of microplastics in STPs.
microplastics (Bayo et al., 2016; Talvitie et al., 2017a) which was similar to the present results. It is estimated that 3956 t/d sludge is produced (Wang, 2011), therefore management of sewage sludge laden with microplastics should be the focus of future research. In addition, potential ecological risks of microplastics in sludge should be evaluated before used for commercial or agricultural applications.

4. Conclusion

In the present study, we provide a systematic characterization of microplastics in waters processed at the largest water reclamation plant in China throughout the treatment process. In addition, microplastics were detected, quantified and grouped by type and color. Microfibers in influents and effluents were primarily comprised of PET, PES, and PP. Overall, 95.16 ± 1.57% of microplastics in incoming influents were removed and deposited into sewage sludge, and 0.59 ± 0.22 items/L remained in effluents released to surrounding water bodies. Future studies should focus on the microfibers before entering into STPs and the sewage sludge when land application.

Declaration of interests

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.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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