Pollution characteristics and fate of microfibers in the wastewater from textile dyeing wastewater treatment plant

Xia Xu, Qingtong Hou, Yingang Xue, Yun Jian and LiPing Wang

ABSTRACT

Wastewater discharged from wastewater treatment plants (WWTPs) is suspected to be a significant contributor of microplastics (MPs) to the environment, and fiber is the main shape of MPs in wastewater effluent. A typical textile industry WWTP with 30,000 tons of daily treatment capacity was sampled for microfibers at different stages of the treatment process to ascertain at what stage in the treatment process the microfibers are being removed. The average abundance of microfibers was 334.1 (±24.3) items/litre in influent, and it reduced to 16.3 (±1.2) items/litre in the final effluent with a decrease of 95.1%. Despite this large reduction we calculated that this textile industry WWTP was releasing 4.89 × 10^3 microfibers including microplastic fibers and non-microplastic fibers into the receiving water every day. This study showed that a modest amount of microplastics being released per litre of effluent could result in significant amounts of fibers entering the environment, despite the fact that efficient removal rates of microplastic fibers and non-microplastic fibers were achieved by this modern treatment plant when dealing with such a large volume of effluent. The fate of fibers is described during the textile industry wastewater treatment process.

Key words | microfibers, microplastic fibers, non-microplastic fibers, textile dyeing wastewater, wastewater treatment plant (WWTP)

INTRODUCTION

The production of fibers in the world, including natural fibers and manmade fibers, reached 100 million tons in 2017 (International Cotton Advisory Committee 2017). The manmade fibers include artificial and synthetic fibers. Synthetic fibers are currently responsible for more than 73% of world fiber consumption, with polyester, polyamide, acrylic and polyolefin being the most common. At the same time, only synthetic textiles could be considered plastic. However, the fact is that fibers present in the environment are not always made from plastic (Wesch et al. 2016).

Microplastics (MPs) are defined as plastic particles smaller than 5 mm in length (Thompson et al. 2004) and are derived from a wide range of sources including synthetic fiber from clothing, polymer manufacturing and processing industries, and personal care products (Mahon et al. 2017). MPs can be classified into five different categories due to their shape: fibers, foam, pellets, films and fragments (Miller et al. 2017). The most abundant type of MP found in the environment is microfibers which are defined as a threadlike piece of plastic with a length between 100 μm and 5 mm and a width at least 1.5 orders of magnitude shorter (Zhao et al. 2014; Fischer et al. 2016). The microfiber can come from knitwear (Salvador et al. 2017) or a direct pathway from clothing to water courses via the atmosphere (Dris et al. 2016). In the aquatic environment, microfibers can adsorb a wide variety of pollutants that are widely distributed in marine and freshwater ecosystems, such as polycyclic aromatic hydrocarbons and polychlorinated biphenyls which have been proved to have toxic effects on fish (Keshavarzifard et al. 2013). Because of their small size and length-to-diameter ratio, microfibers are easily ingested by organisms (Dris et al. 2018). These toxic pollutants may eventually enter into the food chain if the contaminated plastic residues are ingested by fish, aquatic invertebrates, and other wildlife (Carr et al. 2016).
The discharge of sewage is one of the main sources of microplastic in freshwater and marine environment. Recently, wastewater treatment plants (WWTPs) have been suggested to act as one of the land-based sources or entrance routes for microplastics to the aquatic environment (Murphy et al. 2016; Ziajahromi et al. 2017; Lares et al. 2018). Compared to other types, microfibers make the most contribution in WWTPs. In terms of available treatments, there is a general consensus that current technology retains the major part of microplastics, where fibers appear to remain mainly in primary sedimentation (Salvador et al. 2017). Even though the removal efficiency of microplastics in WWTPs is found to be around 99%, microplastics like plastic microbeads from toothpaste and textile fibers are continuously being discharged from WWTPs to recipient waters (Carr et al. 2016; Murphy et al. 2016; Talvitie et al. 2017a; Lares et al. 2018). Scientists have already discovered microplastics in coastal areas around the world, such as polyester fiber and acrylic fiber fragments which are smaller than the tip of needles, and these mainly come from the clothes, carpets and curtains.

China is the world’s largest producer and exporter of textiles and garments. The textile industry is a highly labor-intensive industry and a highly polluting industry. Due to the consumption of about 100–200 L of water per kilogram of textile product and the use of an immense range of materials and chemicals in the production chain, the factory will face the production and discharge of a large amount of highly polluted wastewater. The current research on textile wastewater treatment is more concerned with the removal of organic matter and chromaticity (Franca et al. 2015). So far, there have been no reports on the removal of microplastic fiber and non-microplastic fiber from textile wastewater.

In this paper, the effectiveness of the textile dyeing wastewater treatment process in the removal of microfibers from the effluent at different stages during the treatment process of a typical textile industry WWTP with daily treatment capacity of 30,000 tons was investigated. The physical and chemical composition of microfibers found at each treatment stage was identified. The first systematic overview of the fate of microfibers (microplastic fiber and non-microplastic fiber) in the textile dyeing WWTP was provided by identifying and quantifying where microfibers are removed at various stage of the treatment. It will provide a reference for the research of the effect of industrial wastewater on microfiber pollution and risk assessment, and look forward to the development of sewage treatment plant processes in the future.

MATERIALS AND METHODS

Information about sampled WWTP

A typical textile industry WWTP was selected as the key research object. This WWTP received the production wastewater of 33 printing and dyeing enterprises in a textile industry park and the domestic sewage of a residential area, with the printing and dyeing wastewater accounting for about 95%. An average of 50,000 tons per day of treated wastewater is discharged from the WWTP. Multiple treatment steps based on physicochemical and biological treatment were used in the WWTP. The wastewater treatment process is shown in Figure 1.

There were two times for sampling in January 2018, and the water quality indicators are shown in Table 1. The suspended solids (SS) were measured by gravimetric method, and the water sample was filtered through a filter membrane and dried at 90 °C to obtain a total unfilterable residue. The pH was measured directly with a portable pH meter (FiveEasy Plus FE28, Mettle Toledo, Switzerland). The chemical oxygen demand (COD) was determined by the titration method. The ammonia nitrogen (NH₃-N) and total phosphorus (TP) were measured by Nash reagent and ammonium molybdate spectrophotometry (Pharo 300, Merck, Germany) respectively. The colorimetry was determined by multiple-dilution method. All the water quality indexes of the effluent conform to the water pollutant emission standard for Taihu River Basin WWTPs and key industries (DB32/1072-2007).

Sample collection

Four stages of the treatment process were sampled (Figure 1): Influent (S1), effluent of primary sedimentation tank (S2), effluent of secondary sedimentation tank (S3), and the final effluent (S4). Steel buckets (10 L) were used for sample collection by an on-site technician. The collected water samples were stored in glass bottles and brought back to the laboratory for processing. Three groups of parallel samples were taken in each process section, of which 6 L were collected from sites 1–3 and 12 L from site 4 respectively.

Isolation of microfibers

Microfibers were extracted from water by a two-step filtration process. Briefly, the volume of water was first recorded and particles in the water were filtered onto a stainless steel mesh filter with different pore sizes of 180,
Any particles on the filter were washed into a glass flask using 100 mL of hydrogen peroxide (30%, V/V) to digest the organic substances. The flasks were covered and placed in an oscillation incubator at 65°C and 120 rpm for 48 h. The liquid in the flask was filtered onto a 5 μm nylon net filter using a vacuum system again, and the filter was dried and stored in dry glass petri dishes for further observation.

**Examination and identification of microfibers**

**Optical microscopy**

All suspected particles on the filters were observed and photographed using a microscope (SMZ18, Nikon, Japan) with ×10–80 magnification. Residues and other processed samples were visually examined using the microscope and analyzed via successive magnifications. All collected fibers, including plastic and non-plastic material, were subjected to a second round of visual identification in which all recovered fibers were photographed and measured for the largest dimension using the built-in NIS-Elements BR 4.50.00 software. Spherical or irregularly shaped fragments, fibers and other ambiguous microfibers were isolated from the samples. Fibers were rod-like and flexible strips. The color and size of the microfibers were also measured and recorded in visual assessments. It should be noted that items of similar color to the background filter paper may have been overlooked because of the complexity of the samples (Murphy et al. 2016).

**FTIR microscopy**

Fourier transform infrared (FTIR) spectroscopy was used to confirm the microfibers' plastic or non-plastic nature. Representative samples of most abundant groups (i.e. supposed plastic and other materials) were analyzed with an FTIR microscope (Nicolet iN10, Thermo Fisher, USA) in the reflectance mode. The particles were exposed one by one to infrared radiation in the wavelength region of 600–4,000 cm⁻¹ using single point transmission mode. To analyze the spectra obtained with FTIR, the Thermo Scientific™ Hummel Polymer and Additives FTIR Spectral Library was used.

**Data analysis**

All the microfibers on each filter membrane were analyzed, and the mean values of three parallel samples
were collected. Then a part of a typical microfiber was selected for FTIR analysis. Using the equation below an estimated amount of microfibers released at each site was recorded.

$$\text{Microfiber released} = \frac{\text{microfiber present/litres filtered}}{\text{average volume of effluent released (litres/day)}}$$

A one-way analysis of variance was used to determine the differences in the quantities of microfibers among individual sampling sites and the distribution of microfibers’ shape, size and color. A linear regression analysis was used to test whether there was a significant correlation between the abundance of microfiber in wastewater and water-quality index. Pearson correlation coefficient determined the goodness of fit and significance of the correlation. The size distribution of microfibers in different fractions was plotted using a cumulative curve to compare different fractions. The process of degradation in the environment and the digestion with hydrogen peroxide might result in the bleaching of microfibers and a subsequent overestimation of the number of colorless particles. However, our current digestion process was not strong enough to lead to a complete bleaching of microfibers, and only parts of individual items were discolored (Li et al. 2016). To avoid an overestimation of colorless items, only whole white and transparent items were considered to be ‘white’ and ‘transparent’. And some graded items were considered to be ‘others’.

**Quality control of experiments**

In the present work, a serious consideration was given to the risk of contamination (Lares et al. 2018). When analyzing filter papers, a blank filter paper was exposed to the open laboratory conditions to assess the possibility of air-borne contamination. The blank contamination on the processing day for each sample day was subtracted from the microfiber total for each sample. During these steps clean white cotton laboratory coats were worn at all times, and all equipment used was cleaned three times with distilled water. All Petri dishes, filter papers, and forceps were examined underneath a dissection microscope before use to ensure no contamination was present. The tools were sealed in an aluminum foil bag and kept clean before using. All work surfaces were wiped down with 70% ethanol three times prior to work commencing.

**RESULTS AND DISCUSSION**

**Abundance and correlation analysis of microfibers**

**Abundance and removal rates of microfibers with sampling site**

For the wastewater of textile dyeing, fibers were most dominant, accounting for 80–100% of particles across all sampling sites. The average abundance of microfibers in wastewater from four different sampling sites (S1, S2, S3 and S4) was in the range 334.1–16.3 items/litre (Figure 2). There was a large fluctuation at each sampling, which indicated that the abundance of microfiber was influenced by the production volume and types of textile products. The water quality indexes met the water pollutant emission standards of the textile dyeing and finishing industry; however, the microfiber could not be removed completely in the traditional wastewater treatment process.

The abundance of microplastics reported in WWTP effluent varied between 0 and 27 items/litre (Dris et al. 2015; Mintenig et al. 2017; Talvitie et al. 2017a; Ziajahromi et al. 2017; Lares et al. 2018). The large variation in previous results was mainly caused by different types of wastewaters and processes used in studied WWTPs together with different size limitations and sampling, preparation and identification methods.

The average abundance of microfibers at S1 was 334.1 (±24.3) items/litre, and the removal efficiency reached 76.0%, 83.7% and 95.1% respectively from S2 to S4 which was consistent with other previous reports (Dris et al. 2015; Mintenig et al. 2017). The microfibers decreased obviously from S1 to S2 (Table 2). Most of the textile fibers were attached to the gravel and flocs, and would be removed at the process of primary sedimentation. In most related

![Figure 2](https://iwaponline.com/wst/article-pdf/78/10/2046/516474/wst078102046.pdf)

**Figure 2** | The abundance of microfiber at each sampling site.
investigations, the majority of microfibers were removed in the beginning of the wastewater treatment process during mechanical and chemical pretreatments (Carr et al. 2016; Talvitie et al. 2017a; Lares et al. 2018). The result showed that 76.0% of microfibers were removed before the aeration process. Although there was only a small amount of fibers in effluent, a large amount of microfiber would enter into the aquatic environment because of the large discharge of wastewater. The effluent sampled at S4 contained on average 16.3(±1.2) items/litre, and the removal efficiency of microfibers was about 95%; however, the discharge number of microfibers into the receiving water environment could reach 4.89×10^8 per day and 1.78×10^{11} per year.

**Correlation analysis**

The correlation between microfiber concentration and water quality indexes in four sampling sites was analyzed. The result showed that the microfiber concentration had a significant correlation with SS (p < 0.01), and had non-significant correlation with COD, pH, total nitrogen, NH_{3}-N, chroma and other indexes. The correlation between SS and microfibers concentration is shown in Figure 3. Although the abundance of microfibers in wastewater was similar to the SS water quality index, there were still some differences. The number of microfibers increases with the increase of SS. It should be noted that the accumulation of microfibers was a bit different from that of chemical pollutants, and the bioavailability of microfibers was largely determined by shape and size rather than their thermodynamic behavior (Wright et al. 2013).

### The distribution of size of the microfibers

The particle size distribution of microfibers was calculated and is shown in Figure 4. The abundance of microfiber shows a linear decrease from S1 to S4, especially from S1 to S2. The abundance percent of microfibers (>30 μm) was above 85%, and the abundance of microfibers between 30 μm and 74 μm was the largest. There was no significant difference for the percent of microfibers with different size in influent and effluent. The microfibers which were larger than 30 μm accounted for 88.8% in influent, and the microfibers in effluent accounted for 76.7%.

The size distribution of microfiber with different size in different processes was calculated and is shown in Figure 4. Except for the microfibers with 5–10 μm, there was obvious decline in the percent of microfibers from S1 to S2. The microfibers with size distribution between 5 μm and 10 μm accounted for 4.6% in S1; however, it increased to 14.2% in S2. The result showed that the processes of grille and primary sedimentation tank were not very appropriate for the removal of microfiber (5–10 μm), and the proportion of this size of microfibers decreased to 10.5% after second sedimentation tank. At the same time, there was an obvious decline for the microfibers with size distribution of more than 10 μm from S1 to S2. This result showed that grille and sedimentation were more appropriate for larger fiber.

From the viewpoint of the length, it was similar to the characteristic of particle size, and the superfine fiber occupied the main position. Among the four sampling points, microfibers with a length of 0.1–1 mm accounted for the main part, and the cumulative rate increased from S1 to S4. The proportion of microfibers in S2–S4 was similar. The gap between S1 and S2 was larger, which meant that the removal of the larger-sized microfibers is mainly in the pretreatment. In addition, fibers with length >5 mm were also found in S1 and S2, which might be related to the fiber's unique aspect ratio.

So far, the main size of microplastics in final effluent reported has been between 0.5 and 1 mm (Talvitie et al. 2017a). It has been reported that 64% of all recovered microplastics from samples were smaller than 1 mm, half of which were also smaller than 0.5 mm (Carr et al. 2016). The
difference was produced by the different sampling methods, mesh with different size was used. Because the size and shape of the fibers are not symmetrical, even if one of the sizes is longer than the mesh size, part of the microfibers could also pass through the filter membrane. Therefore, the number of microfibers counted from samples only reflects the retention fraction of microplastics (Mintenig et al. 2014). On the other hand, the sample may also have microfibers in size smaller than the size of the filter, if they are entangled together or attached to larger particles.

The distribution of color of the microfiber

More than 10 microfibers with different color were observed, and these colors were black, red, blue, dark blue, light blue, red, pink, grey, brown, purple, yellow, black, transparent and others (Figure 5). Blue series (blue, dark and light blue) was the dominant color which accounted for 35% in influent. There were some changes in the color and proportion of color between influent and effluent. Compared with the influent, there was obviously less purple, brown and yellow in effluent. The blue and pink were the main colors in influent, and the blue and transparent were the main colors in effluent. Thus the blue series dominated in all color classes in influent and effluent. In particular, the blue microplastic items frequently found in clams have been reported in field studies of marine fishes (Güven et al. 2017). Therefore, knowing the color of microfibers can predict its source and can be used as a part of monitoring the contamination from microfibers. From S1 to S4, the percent of microfibers with pink and red decreased. The black and transparent microfiber increased to 16% and 24%. The color of microfibers would be changed in wastewater treatment with the change of season and product.

Characterization of microfiber

Microfiber morphology and nature

Microfibers of various colors are shown in Figure 5. The fibers found in the WWTP were a dark and irregular...
color. However, the integrity of fibers in the WWTP was better than those in the aquatic environment. These fibers may be mostly primary sources and have not been weathered for a long time. It was necessary to divide microfibers into primary and secondary items which could help to find the sources and identify solutions to reduce their input to the environment (Talvitie et al. 2017a). It could be considered that most of the microfibers originated from the textile industry and households. In the textile dyeing wastewater, the results of this study indicated the high impact of textile and clothing materials on the microfiber emissions (Talvitie et al. 2017b). At the same time, the storm water runoffs may also be the source of microfiber in WWTP.

From the exterior shape, there were both flat and cylindrical microfibers, and some of them were flattened. Compared with the non-microplastic fiber, the toughness of the microplastic fiber was stronger, and it had the ability to resist mechanical damage in the aquatic environment. At the same time, fibers were easily intertwined and agglomerated to form a fiber ball. It is apparent that microplastic fibers seem to be a main contaminant of the WWTP.

FTIR identification

There was a high load of fibers in the textile industry WWTP. It should be pointed out that this is based on the statistics of microfibers, which included microplastic fibers and non-microplastic fibers. The type distribution of microfibers in influent and effluent is shown in Figure 6. The comparison using the analyzing software of the obtained spectra with the database showed that 25% of all textile fibers were natural fibers of cotton, linen or wool, and the rest of the textile fibers were polyester (PES), polypropylene (PP), polyacrylonitrile (PAN) and rayon. The maximum proportion of microfibers was PES (34%), followed by rayon (29%), natural fibers (25%), PP (12%) and PAN (2%) in the influent. And the maximum proportion of microfibers was rayon (38%), followed by PES (25%), natural fibers (18%) and PP (19%) in the effluent. It should be noted that there were also some fibers with weak quality infrared spectra, and it may be that the fibers were polluted by the organic matter or biofilm in the wastewater, which blocked the possible peak of the actual material from the fiber and hindering the analysis. In addition, the chemical treatment may harm the materials of natural fibers and make the analyses complicated.

The matching was sometimes not high because of the complexity of environmental samples, and it even showed great changes in the same or similar samples. A match of 70% was proposed as an acceptable value (Yang et al. 2015). The highest spectrum matches reached 98% in Figure 7. PP fiber with a length of 0.1–1 mm is the largest proportion of all PP fibers, and it has the advantages of high strength, good toughness, good chemical resistance and good microbial resistance. Thus PP fibers are widely used in knitting fields, which are an ideal way to make sportswear, T-shirts and other materials. Compared with PP fibers, PES fibers found in the WWTP were more resilient. PAN fiber was a common fiber type in the textile. It could be blended with wool into wool yarn, or woven into blankets and carpets, and it could mixed with cotton, artificial fiber or other synthetic fiber, and woven into a variety of clothing materials and indoor supplies. Most microfibers in effluent were identified as rayon. Rayon is a silk fiber made up of cellulose, and the many properties of rayon are the same as other fibers, such as cotton and linen fibers. The strength of rayon would decrease in the water environment, and it would be easier for rayon to escape the wastewater treatment process into the environment.

Figure 6 | Type distribution of microfibers in influent (a) and effluent (b).
The main color of rayon found in the effluent was yellow, blue and pink.

In most of the studies, non-microplastic fibers were generally excluded from analysis and the focus has been more on synthetic fibers. Because it is generally believed that non-microplastic fibers can be degraded quickly, they will not cause harm to the environment. In fact, some harmful chemicals such as flame retardants are added to non-microplastic fibers during processing and dyeing, which can prolong the fiber’s degradation time in the environment. Both synthetic and non-synthetic fibers presented high occurrences in freshwater (Dris et al. 2018). Rayon has also already been found in previous works in marine environments and even in the gastrointestinal tract of fish in the English Channel (Lusher et al. 2014). These results indicated that non-microplastic fibers could also reach the environment through the WWTP and remain long enough until they are ingested. So both the microplastic and non-microplastic fibers can have negative impacts in the environment; this reinforces our opinion that all fibers need to be assessed and studied.

CONCLUSIONS

The study showed that the textile dyeing WWTP had an effective removal of pollutants including microfibers in wastewater. The abundance of microfibers found from four sampling sites decreased gradually from S1 to S4, and the rate of decline from S1 to S2 was the fastest and similar to the decline in the SS water quality index. It proved that the removal process of microfibers mainly occurs in the initial stage of sedimentation, which could provide a reference for the improvement of sewage treatment facilities in the future. More than 80% of microfibers had a diameter above 30 μm, and the length was between 0.1 and 1 mm. At the same time, the microfibers in the effluent presented a variety of color. More than 60% of the fibers were identified by FTIR as microplastic fibers, which indicated that natural fibers are also part of the contamination of microfibers.

Although the removal rate could be up to 95%, which was at a medium level compared with other research results, the discharge number of microfibers into the receiving water environment could reach $4.9 \times 10^8$ per day. Thus the textile dyeing WWTP was one of the potential sources of microfibers. Even the advanced WWTPs of other countries cannot remove all microfibers, so sewage and sludge treatment also need to improve according to the migration characteristics of new microfiber pollution in sewage and sludge. The most important is to control the use of microplastic fibers at their source in the textile printing and dyeing industries.

ACKNOWLEDGEMENTS

This work was supported by grants from the National Natural Science Foundation of China (21607017), State Key Laboratory Open Project Fund of Pollution Control and Resource Resuse (PCRRF17036) and Environmental Monitoring Fund Project of Jiangsu Province (1702). We are grateful to Professor Hua-Hong Shi and the State Key Laboratory of Estuarine and Coastal Research.

REFERENCES


First received 10 August 2018; accepted in revised form 6 November 2018. Available online 20 November 2018