RESEARCH ARTICLE



Presence of microplastics in drinking water from freshwater sources: the investigation in Changsha, China

Maocai Shen¹ · Zhuotong Zeng² · Xiaofeng Wen¹ · Xiaoya Ren¹ · Guangming Zeng¹ · Yaxin Zhang¹ · Rong Xiao²

Received: 10 November 2020 / Accepted: 29 March 2021 / Published online: 3 April 2021 © The Author(s), under exclusive licence to Springer-Verlag GmbH Germany, part of Springer Nature 2021

Abstract

We investigated the abundance of microplastics in freshwater, treated water, and household tap water from the drinking water supply chain in Changsha, China. The abundance was 2173–3998 (mean = 2753), 338–400 (mean = 351.9), and 267–404 (mean = 343.5) particles L^{-1} in freshwater, treated water, and tap water, respectively. Fibrous and fragments made up the majority (> 70%) in all water samples, and most polymers were composed of polyethylene, polypropylene, and polyethylene terephthalate. Microplastics in tap water were related to materials of transportation pipelines in drinking-water supply chain. Although plastics are corrosion-resistant, the slight fragmentation and abrasion may occur during drinking water treatment transportation. This study provided a proof for the occurrence of microplastics in drinking water, which may offer a reference for microplastic removal during drinking water treatment, and the formulation of standards for microplastic content in drinking water.

Keywords Microplastics · Drinking water · Freshwater · Drinking water treatment · Potential risks

Introduction

Currently, microplastics, as an emerging pollutant, have attracted great attention from global environmentalists (Shen et al. 2019; Shen et al. 2019). Microplastics are normally defined as plastic particles < 5 mm (Shen et al. 2019; Thompson et al. 2004); however, there is still some controversy about the definition (Frias and Nash 2019; Hartmann et al. 2019). Microplastics have been widely found in global freshwater system, including water (Eriksen et al. 2013; Hu

| Maocai Shen and Zhuotong Zeng contributed equally to this work. | | | | |
|---|--|--|--|--|
| Responsible Editor: Christian Gagnon | | | | |
| | Guangming Zeng zgming@hnu.edu.cn | | | |
| | Yaxin Zhang zhang_yx@hnu.edu.cn | | | |
| | Rong Xiao xiaorong65@csu.edu.cn | | | |
| 1 | College of Environmental Science and Engineering, Hunan University and Key Laboratory of Environmental Biology and Pollution Control (Hunan University), Ministry of Education, Changsha 410082, People's Republic of China | | | |
| 2 | Department of Dermatology, Second Xiangya Hospital, Central South University, Changsha 410011, People's Republic of China | | | |

et al. 2019; Zhang et al. 2017), sediment (Wen et al. 2018; Zhao et al. 2018), and organisms (Fossi et al. 2014; Fossi et al. 2018; Setälä et al. 2014; Su et al. 2016; Vendel et al. 2017). In addition to the secondary microplastics formed by decomposition of bulk plastics, the small plastic particles, such as microbeads usually used in cosmetics and bath lotions, are also a major source of microplastics to freshwater ecosystems. Due to its small particle size and low density, these primary microplastics can escape the filter device of wastewater treatment plants and be discharged into the surface water (Ziajahromi et al. 2017).

Open and closed freshwater systems such as rivers and lakes can be used as microplastic pipelines and sink tanks respectively (Negrete Velasco et al. 2020; Shen et al. 2021; Shen et al. 2020). The abundances of microplastics in freshwater systems vary greatly from almost zero to millions per cubic meter. A research done by Lechner et al. (2014) reported that the average microplastic concentration of surface water from The Danube was 316.8 item per 1000 m³. Evidence showed that microplastics have been found in freshwater (lakes and rivers) in European countries, with the greatest occurrence in Lake Geneva, Switzerland, reaching 48146 items/km² (Eerkes-Medrano et al. 2015). The average occurrence floating on the surface water was measured to be 43000 items/km² in the Great Lakes Basin (Eriksen et al. 2013). Su et al. (2016) detected the microplastic abundancet in Taihu

Lake, China, and results showed that microplastic concentration was measured to be 3.4–25.8 items/L. In addition, Di and Wang (2018) also found that the microplastic concentrations were in a range of 1597–12611 items/m³ in surface water from Three Gorges Reservoir, China. It is noteworthy that Free et al. (2014) also detected microplastic pollution in Lake Hovsgol in northern Mongolia, and the average abundance of microplastics in surface water was 20246 items/km². The geographical location of the region is remote and the population is sparse, which indicates in the absence of effective management methods, microplastics can be migrated and expanded through factors such as runoff and monsoon. Scattering to all kinds of waters eventually poses an inestimable risk to ecosystems.

The quality of drinking water is related to human health, which may also be one of the ways for human body to be directly exposed to microplastics. Although the toxicological and ecological effects of microplastics are unclear, microplastics still have been considered as emerging contaminants to human health. Once surface freshwater is collected as raw drinking water, microplastics have to be removed before consumption. Drinking water treatment plants (DWTPs) are the most important barriers to the entry of microplastics into human body. Now, interestingly, only few studies on the investigation of the microplastic abundance in drinking water were performed around the world (Kosuth et al. 2018; Mintenig et al. 2019; Pivokonsky et al. 2018). Kosuth et al. (2018) investigated the occurrence of microplastics in 159 global tap water samples. The authors reported that microplastics were found in 81% of water samples with a concentration of 0–61 particles L^{-1} (mean = 5.45 particles L^{-1}), most of which were fibers (0.1–5mm). Mintenig et al. (2019) tested groundwater and drinking-water derived from the groundwater for the presence of microplastics. The concentrations of microplastics were measured to be 0-0.007 particles L^{-1} (mean = 0.0007 particles L^{-1}) in both raw water and drinking water. Pivokonsky et al. (2018) analyzed the microplastic concentration in freshwater and drinking water. Concentrations ranged from 1473 to 3605 particles L^{-1} in raw water and 338–628 particles L^{-1} in treated drinking water, respectively, with a typical removal efficiency of 70-80%. To assess the potential risks to human health, it is necessary to determine the actual exposure of microplastics (Wright and Kelly 2017). The potential impact of daily food and packaging materials on drinking water also should be investigated.

Therefore, with research blank on the data of microplastics in drinking water, the occurrence of microplastics in freshwater, treated water, and household tap water in Changsha, China, were investigated. Purposes of this study are as follows: (1) to quantify microplastics from drinking water and compare their content in different samples, and (2) to identify material composition and provide their particle size distribution.

Methods and materials

Sampling

Samples of freshwater were obtained from a river (XiangJiang River) used as water source, and treated water was collected from a DWTP. In addition, a conventional household tap water was selected in the water distribution system. All sample sites are located at Changsha, Hunan province. The drinking water treatment processes mainly include aeration, coagulation/sedimentation, sand filtration and granular activated carbon filtration (Fig. 1), and daily water supply capacity can reach up 100000 m³. Sand filtration and granular activated carbon filtration were combined into filtration and subsequent treatment methods (disinfection and storage tank) are also added in Fig. 1. After disinfection, the treated water is directly fed into the water supply chain or stored in water tanks. Water pipes are made of polyvinyl chloride (PVC), cast iron, and high-density polyethylene (HDPE). The water quality and transportation of each house are the responsibility of individual consumers. The water quality of raw water, treated water, and tap water is listed in Table S1 (Supporting information).

Sample collection

All water samples were obtained from April to July 2019. The annual average rainfall in Changsha area is 1200–1400 mm, and the rainfall is abundant from April to July. Freshwater was collected at the DWTP inlet, and treated water at the outlet of DWTP. Tap water was obtained from a conventional household. Freshwater, treated water, and tap water were picked into clean glass bottles with a volume of 10 L. Each sampling was repeated three times, and then all water samples were mixed as a sample. Sampling time is set once a month. All samples were kept at 4°C, and attention was paid to avoid sample contamination within the whole process.

Water sample treatment

Firstly, in the laboratory, wet peroxide oxidation was used to removal organic matter from water samples. A vacuum pump connected with other glass filtration equipment was used for microplastic filtration. Treated water and tap water were passed through polytetrafluoroethylene membrane filters ($d = 0.22 \mu m$). Polytetrafluoroethylene membranes were chosen because these filters with a diameter of 5 cm did not seem to interfere with SEM analysis process. Three filters were used (one for every 10 L of water) in order to avoid clogging of the filter caused by other substances in the waters. These filters were immersed in hydrochloric acid with a concentration of 0.02 mol L⁻¹ to dissolve calcium carbonate. Afterward, the filters were rinsed with ultrapure water and 30% ethanol. Finally, each sample was filtered by alumina filter



Fig. 1 Drinking water treatment plant processes and supply chain with sampling locations. (1) The freshwater (raw water); (2) the treated water; and (3) a conventional water tap in a selected household

(Whatman, UK) with a pore size of 0.2 μ m for further qualitative analysis (Mintenig et al. 2019). Thereafter, these filters were dried at 40 °C in an oven and stored at a clean culture glass dish prior analysis.

Freshwater samples contained a large amount of suspended particles and needed to be treated before filtration. Density separation is a common approach to separate microplastics from matrices (Su et al. 2016). Saturated ZnCl₂ solution was chosen in this research. The sample was placed in a glass column with a diameter of 20 cm and a height of 1 m. After 24 h, the supernatant was filtered as mentioned above and the sediment was rinsed with ultrapure water and 30% ethanol. The rinsed solution was also filtered as described above. After that, samples were also filtered by alumina filter with a pore size of 0.2 μ m.

Contamination control

Control and mitigation of contamination are particularly important in experiments. To avoid contamination, experiment was carried out in a closed laboratory, and minimized the access of experimenters as possible. Any direction contact between the sample and the plastic material was avoided during sample collection, water treatment, and further analysis. Only clean cotton coats were worn in the laboratory during the whole experiment. All glassware used in this experiment was clean by sonication and rinsed with Milli-Q three times to avoid contamination before analysis. The surface of the laboratory was wiped with 30% ethanol, and the device was washed with Milli-Q and covered with aluminum foil prior use. Also to ensure that any additional microplastic contamination occurred during sample filtration, the same volume of Milli-Q was used as a blank to measure additional contamination.

Sample analysis

Quantification analysis

Plastic particles on the filters were inspected visually by a Carl Zeiss Discovery V8 Stereo microscope (MicroImaging GmbH, Goottingen, Germany) with a digital camera (M165 FC, Leica, Germany). The suspected microplastic materials were differentiated based on classification criteria developed in previous experiments (Di and Wang 2018). Briefly, a subset of suspected microplastics was randomly selected and examined usingµ-Fourier transform infrared spectroscopy (µ-FTIR, Thermo Scientifific Nicolet). However, simple visual observation was not enough to identify microplastics from other particles (Eriksen et al. 2013). The filters were furtherly observed by a scanning electron microscopy (SEM, SIGMA HD, Nova450) andµ-FTIR. The spectral range was set to $4000-675 \text{ cm}^{-1}$, and the spectral resolution was set to 6 cm⁻¹. The number of scans was 16 times and the data interval

was 0.482 cm⁻¹ (Shen et al. 2021). The particle size, color, and morphology of the particles contained in each sample were recorded. The microplastics were classified into three morphotypes: fibers, fragments and spheres, and four size categories (1–10 μ m; 10–50 μ m; 50–100 μ m; > 100 μ m). Fibers were defined as microplastic materials with an elongated appearance, and the remaining microplastics were defined as others.

Qualitative analysis

Because particle size $< 1 \mu m$ cannot be reliably identified in material composition, these particles were not included from the results of microplastic identification (Pivokonsky et al. 2018). A μ -FTIR analysis was carried out to verify microplastics, as described as Yang et al. (2015). The spectral range was set to $4000-675 \text{ cm}^{-1}$, and the collection time was 3 s. All samples had a spectral resolution of 8 cm^{-1} with 6 coadded scans, and the pore size range was of $50 \times 50 \ \mu m$ to 150 × 150 µm. Raman spectroscopy (Renishaw 2000, 532 nm laser, Raman shift 100–3500 cm⁻¹) was used to analyzed the small microplastics having a particle size < 20 µm (Käppler et al. 2016). The laser wavelength was set as 785 nm to identify microplastics (Zhang et al. 2017). The polymer types were determined by comparing the spectra of each sample with that of Hummel and Raman sample libraries (Hummel polymer Sample Library, Organics by Raman Sample Library, Raman Sample Library, Sigma Biological Sample Library, User Example Library).

The collected spectra were processed by software (Nicolet Omnic 8.0). Spectra were compared to an instrument database to determine their chemical composition of the obtained particles (Di and Wang 2018). In addition, considering that microplastics in aquatic environment have been eroded, the threshold of matching factor was calculated to be 0.70 in this study (Klein et al. 2015).

Results and discussions

Background value

The blank samples showed that contamination of microplastics happened in the process of sample treatment. Microfibers were found in the blank samples (n = 4). Table 1 shows the occurrence of microplastics determined in blank samples. According to the subsequent identification, some particles produced spectra in the infrared band with PE, PEst (polyester), and PP characteristics. Evidence showed that fiber contamination is one of the most frequently discussed and treated inconsistencies (Woodall et al. 2015). Enhancement of laboratory conditions such as clean air condition seems to prevent such fiber contamination. To reduce

Table 1 Occurrence of microplastics determined in blank samples (particles L^{-1})

| Blank samples | | PE | PEst | РР | Summation |
|-----------------|------|----|------|----|-----------|
| Blank 1 (April) | 1 | 0 | 2 | 0 | 2 |
| | 2 | 2 | 3 | 1 | 6 |
| | 3 | 1 | 0 | 0 | 1 |
| | Mean | 1 | 2 | 0 | 3 |
| Blank 2 (May) | 1 | 2 | 1 | 2 | 5 |
| | 2 | 1 | 2 | 1 | 4 |
| | 3 | 1 | 1 | 0 | 2 |
| | Mean | 1 | 1 | 1 | 4 |
| Blank 3 (June) | 1 | 2 | 1 | 1 | 4 |
| | 2 | 0 | 1 | 0 | 1 |
| | 3 | 2 | 3 | 2 | 7 |
| | Mean | 1 | 2 | 1 | 4 |
| Blank 4 (July) | 1 | 2 | 2 | 1 | 5 |
| | 2 | 1 | 1 | 2 | 4 |
| | 3 | 1 | 0 | 1 | 2 |
| | Mean | 1 | 1 | 1 | 4 |

the impact of contamination, the average number microplastic particles of blank sample was deducted from the data obtained in all water samples in this study.

Occurrence of microplastics in samples

The occurrence of microplastics in all water samples is listed in Table 2. The results showed that microplastics were found in all the samples. The SEM images of microplastics detected in raw water, treated distributed water, and tap water are given in Fig. 2. The amount of microplastics in freshwater, treated water, and household tap water varied with sampling time. The microplastic abundance in freshwater samples was measured to be 2173 \pm 112, 2258 \pm 172, 2584 \pm 113, and 3998 \pm 246 particles per liter at sequential sampling (April, May, June, and July), respectively. This difference could be influenced by various factors, such as human activities, surrounding environment, and current weather conditions. June and July are the rainy season in Changsha, Hunan Province. Floods triggered by rainstorms can bring large quantities of pollutants, including microplastics, into the water environment (Gündogdu et al. 2018). The results of this research coincided with the theory. The microplastic abundance of freshwater significantly increased by 1.84 times compared with that before the flood. The Xiangjiang River, as a source of raw water, flows through industrial and residential areas. Although there is no evidence that these factors may have an impact on microplastic abundance in raw water, the fate of microplastics in monitoring sources and aquatic environment requires to be studied in detail.

 Table 2
 Quantification of microplastics in raw water, treated water, and tap water within April–July 2019

| Sampling time | Type of water | Microplastic a (particles L^{-1}) | Removal efficiency | | |
|---------------|----------------------------|--|----------------------|-----|--|
| | | Mean Range | | - | |
| April | Raw water Treated water | $\begin{array}{c} 2173\pm112\\ 282\pm13 \end{array}$ | 2013–2315 263–295 | 87% | |
| | Tap water | 267 ± 11 | 254-284 | | |
| May | Raw water Treated water | $\begin{array}{c} 2258\pm172\\ 338\pm21 \end{array}$ | 1967–2407 305–360 | 85% | |
| | Tap water | 321 ± 11 | 309-335 | | |
| June | Raw water Treated water | $\begin{array}{c} 2584\pm113\\ 388\pm14 \end{array}$ | 2403–2712 368–406 | 85% | |
| | Tap water | 381 ± 18 | 356-400 | | |
| July | Raw water Treated water | $\begin{array}{c} 3998\pm246\\ 400\pm13 \end{array}$ | 3635–4316 384–420 | 90% | |
| | Tap water | 405 ± 30 | 367–446 | | |

The content of microplastics in treated distributed water was much lower compared with that in freshwater, and was measured to be 282 ± 13 , 338 ± 21 , 388 ± 14 , and 400 ± 13 particles L^{-1} , respectively. The results showed that current drinking water treatment processes have good removal efficiency for microplastics (Table 2). The microplastic content in treated distributed water was determined by its abundance in raw water. The difference of microplastic removal rate at different sampling times may be related to the water quality. The DWTP operates aeration, coagulation/sedimentation, sand filtration, and granular activated carbon filtration. Coagulation/ sedimentation and subsequent filtration are the main treatment processes for microplastic removal from freshwater (Ma et al. 2018; Ma et al. 2019). Additionally, due to the physical of many plastic polymers such as light and buoyant, aeration and air floatation seem particularly suitable for microplastic removal (Di and Wang 2018). Of course, further research is needed to demonstrate the relationship between the process layout of DWTP and microplastic removal.

The microplastic abundance in tap water was no significant different from the treated distributed water, and was 267 ± 11 , 321 ± 11 , 381 ± 18 , and 405 ± 30 particles L⁻¹, respectively (Table 2). The results showed that drinking water transfer processes from the storage tank to the tap have no significant contribution to microplastics. Based on the data from the National Academy of Medicine, humans should consume more than 2.2 L of drinks per day to maintain normal metabolism of the body (Kosuth et al. 2018). If these drinks are made of tap water or tap water itself, more than 587.4 particles will be consumed by humans per day, 214401 per year. Although there is no evidence in drinking water can cause health damage, research should be carried out to assess the exposure level and health effects of microplastics in drinking waters.



Fig. 2 SEM images of microplastics detected in raw water (a), treated distributed water (b), and tap water (c)

Particle size distribution and morphology

In this study, the particle sizes of microplastics were divided into four categories: $1-10 \ \mu\text{m}$, $10-50 \ \mu\text{m}$, $50-100 \ \mu\text{m}$, and > $100 \ \mu\text{m}$. Figure 3 illustrates the particle size distribution of microplastics found in raw water (A), treated distributed water (B), and tap water (C). The results showed that microplastics with particle sizes of $1-10 \ \mu\text{m}$ accounted for the majority (>



Fig. 3 Particle size distribution of microplastics detected in raw water (a), treated distributed water (b), and tap water (c)

85%) of all water samples, which were similar to the results of Pivokonsky et al. (2018). The abundance of microplastics decreased with the increase of particle sizes, and the content of microplastics with large particle sizes (> 100 μ m) was not very high. In July, the total content of microplastics and the large microplastics significantly increased compared with other months. The most probable causes are a series of impacts of heavy rains and floods, it need to be further verified. Interestingly, particle size analysis $< 5-10 \mu m$ was seldom used in most current studies on microplastics in aquatic environment (Di et al. 2019; Mason et al. 2018). These may be influenced by various sampling and analysis methods. However, artificially ignoring the presence of small particle size microplastics in water may underestimated the level of total microplastics pollution. This will adversely affect the removal of microplastics in drinking water and development of new drinking water treatment processes, as well as related human health risk assessment. Evidence has demonstrated that the particle size of microplastics in water tends to be smaller, possibly due to the decomposition of larger microplastics (Zhao et al. 2014), which will exacerbate the above risks. In addition to microplastics in water samples, other unknown small particles were also detected in all water samples. Because these particles were difficult to be certified as microplastics, no records were allowed, which may also reduce the abundance of microplastics in drinking water. Recently, a new research showed that the fragmentation of microplastics into nanoplastics occurred during the water treatment processes and increased the amounts of micro(nano)plastics

in water (Enfrin et al. 2019). Therefore, water treatment processes may have an impact on micro(nano)plastic contamination in drinking water, especially nanoplastics.

Regarding the shape of microplastic, three categories were set: fragments, fibers, and spheres. Figure 4 shows the shape of microplastics detected in all water samples. Fragments were the main shapes in raw water, followed by fibers and spheres. The Xiangjinag River flows through living and industrial areas, and microplastics in water come from many sources. The fragments mainly came from the decomposition of various discarded plastics, and the spheres came from personal care products and other cleaning media (Di and Wang 2018). Sources of fibers were usually from domestic laundry wastewater (Browne et al. 2011). According to Fig. 4, we could find that the proportion of microplastic shape has changed. The spheres were almost all removed by treatment process, while the proportion of fibers increased. The removal efficiency of fibers was low, which implied that the water treatment processes have a certain effect on the shape and removability of polymers. In addition, the color of microplastics detected in water is described in Fig. S1 (Supplementary materials).

Chemical composition analysis

Visually inspected microplastics were identified by μ -FTIR and Raman spectroscopy. The chemical composition of microplastics is shown in Fig. 5. Polyethylene (PE, 26.8%),



Fig. 4 Microplastic shapes detected in raw water (a), treated distributed water (b), and tap water (c)



Fig. 5 Chemical composition of microplastics detected in raw water (a), treated distributed water (b), and tap water (c). PA, polyamide; PE, polyethylene; PET, polyethylene terephthalate; PMMA, polymethyl methacrylate; PP, polypropylene; PS, polystyrene; PVC, polyvinyl chloride

polypropylene (PP, 13.2%), polystyrene (PS, 16.5%), and polyethylene terephthalate (PET, 16.1%) accounted for the main types of microplastics detected in raw water. These plastics are widely used in many daily consumption plastics such as disposable plastic cups and bags and other plastic packings (PlasticsEurope. 2018). The large-scale production and wide application increases the opportunities for plastics to enter the environment. In addition, PVC, polyamide (PA), and polymethyl methacrylate (PMMA) were also detected in raw water. The chemical composition of microplastics detected in raw water, treated distributed water, and tap water by μ -FTIR is illustrated in Fig. 6.

In treated water, PE, PP, and PET were also detected, accounting for 24%, 14%, and 25%, respectively. Research has pointed out that the content of PP microplastics in treated water was not only related to the concentration of microplastics in freshwater but also might be related to the application of coagulants containing polyacrylamide (Pivokonsky et al. 2018). Certainly, this needs further verification. Several PVC and PA were found in treated water. And within the tap water, the content of PE, PP, PET, and PA was no significant change compared with the treated water, while the abundance of PVC obvious increased (Fig. 5c). The results showed drinking water supply chains might contribute to the increase of PVC. Water storage tank in DWTPs is often coated with epoxy resin to avoid corrosion. Pipe from DWTP to individual household is often made of cast iron, HDPE (high-density polyethylene), and PVC, and the corresponding accessors are made of PA materials (Mintenig et al. 2019). Although plastics are corrosion-resistant materials, the wear may occur during the treatment processes and the transportation. In this study, the water pipes in household are made of PVC materials, which may be a reasonable explanation for the increase of PVC in tap water compared with in treated water.

Comparison with other studies

Freshwater is the main raw water source for agricultural, industry, energy production, and human consumption. The ubiquitous occurrence of microplastics in global freshwater has gained more and more attentions. Table 3 shows the abundance of microplastics in global freshwater obtained from published reports. Due to the difference of sampling and subsequent analysis, the lower size limit of detected microplastics is also different. The presence of microplastics in River Seine, France, was in a range of 3-106 (mean = 30) particles L^{-1} and the particle size of all microplastics was more than 100 µm (Dris et al. 2015). The finding also showed that sampling with the plankton net had a predominance of fibers, while great diversity of both microplastic shapes and types was found during manta trawl samplings. The microplastic occurrence in Dongting Lake, Taihu Lake, and Poyang Lake, China, was measured to be < 1-2.8, 3.4-25.8, and 5-34 particles L^{-1} , respectively. The morphology of microplastics was mainly fragment type, and the chemical composition was mostly PE and PP. However, most of current studies did not involve small microplastic particles in the corresponding water samples. At present, trawl is the main sampling method to determine the concentration of microplastics in freshwater, which also leads to the reason that small particle size microplastics cannot be detected. The microplastic abundance in bottled water was also investigated (Mason et al. 2018; Oßmann et al. 2018; Schymanski et al. 2018). Different bottles were investigated, including glass bottles, single use PET bottles, and reusable PET bottles. The microplastic content in different bottles was obviously different. So they suggested that plastic packing material had an un-ignorable contribution to the microplastic abundance in bottled waters. Oßmann et al. measured the particle size > 1 μ m in waters, while the lower size limit of detected microplastics was > 5 μ m (Schymanski et al. 2018) and > 6.5 μ m (Mason et al. 2018), respectively. Compared with freshwater, the concentration of microplastics in drinking water (tap water and bottled water) increased significantly because of the lower detection limit of microplastics. In this study, the concentration of microplastics in three different water, freshwater (raw water), treated distributed water, and household tap water, was 2753, 351.9, and 343.5 particles L^{-1} , respectively, which was similar to the

42319



Fig. 6 Analysis of chemical composition of microplastics detected in raw water, treated distributed water, and tap water by μ -FTIR

study of Pivokonsky et al. (2018). The identification of the concentration of small and medium-sized microplastics in samples plays an important role in evaluating the pollution status of microplastics in the whole environment. Due to different detection limits and lack of

microplastic data in drinking waters, the contrast among different research is particularly difficult. Therefore, more efforts are desired to investigate the microplastic content in global drinking water and to gradually standardize the lower detection limits.

🖄 Springer

| Table 3 | Comparison | on microplastic | abundance | with o | other published | l studies |
|---------|------------|-----------------|-----------|--------|-----------------|-----------|

| Sampling source | | astic ace (particles | Particle size range (µm) | Reference |
|---|------------------------|-------------------------|---|--------------------------|
| | Mean | Range | | |
| Raw water | | | | |
| Raw water from a surface water reservoir, Czech Republic | 1812 | 1648–2040 | 1–100 (92% of particles between 1 and 10 $\mu m)$ | (Pivokonsky et al. 2018) |
| Raw water from a surface water reservoir, Czech Republic | 1473 | 1384–1575 | 1–100 (86% of particles between 1 and 10 $\mu m)$ | |
| Raw water from a river, Czech Republic | 3605 | 3123-4464 | 1–100 (81% of particles between 1 and 10 $\mu m)$ | |
| Raw water from a surface water reservoir, China | 2.6 | 0.47–15 | 48–5mm (5.7–44.4%, 48–500 μm) | (Di et al. 2019) |
| Raw water from groundwater, USA | 6.4 | 0-15.2 | $> 0.45~(100\% \text{ of particles} > 0.45~\mu\text{m})$ | (Panno et al. 2019) |
| Raw water from groundwater, Germany | $0.7 \\ m^{-3}$ | $0-7 \text{ m}^{-3}$ | 50–150 (100% of particles between 50 and 150 μm) | (Mintenig et al. 2019) |
| River Seine, France | 30 | 3-106 | > 100 µm (100%) | (Dris et al. 2015) |
| Amsterdam canal water, Netherlands | _ | 48–187 | (61% 10–300 μm, 39% > 300 μm) | (Leslie et al. 2017) |
| Elbe River, Germany | - | 100–900 | < 20 µm (96%) | (Triebskorn et al. 2019) |
| Dongting Lake, China | _ | < 1–2.8 | < 330 µm (27%) | (Wang et al. 2018, b) |
| Taihu Lake, China | _ | 3.4-25.8 | < 100 µm (18%) | (Su et al. 2016) |
| Poyang Lake, China | _ | 5–34 | < 500 µm (73%) | (Yuan et al. 2019) |
| Raw water from a river, China | 2753 | 2173-3998 | > 1 µm | This study |
| Public supply water | | | | |
| Treated water, Czech Republic | | 243–466 | 1–100 (79% of particles between 1 and 10 μm, microplastic removal efficiency at 81%) | (Pivokonsky et al. 2018) |
| Treated water, Czech Republic | 443 | 369–485 | 1–50 (90% of particles between 1 and 10 μm, microplastic removal efficiency at 70%) | |
| Treated water, Czech Republic | 628 | 562–648 | 1–100 (90% of particles between 1 and 10 $\mu m,$ microplastic removal efficiency at 82%) | |
| Treated water, Germany | < 1 m ⁻³ | - | - | (Mintenig et al. 2019) |
| Treated water, China | 351.9 | 338-400 | > 1 µm | This study |
| Tap water, Ecuador Tap water, UK | 4.02 7.73 | 0–9.04 3.66–13.0 | > 2.5 μm > 2.5 μm | (Kosuth et al. 2018) |
| Tap water, France | 1.82 | _ | > 2.5 µm | |
| Tap water, Germany | 0.91 | 0-1.82 | > 2.5 µm | |
| Tap water, India | 6.24 | 0–20 | > 2.5 µm | |
| Tap water, Indonesia | 3.32 | 0-10.8 | > 2.5 µm | |
| Tap water, Ireland | 1.83 | - | > 2.5 µm | |
| Tap water, Italy | 0 | - | > 2.5 µm | |
| Tap water, Lebanon | 6.64 | 0–23.3 | > 2.5 µm | |
| Tap water, Slovakia | 3.83 | 0-10.9 | > 2.5 µm | |
| Tap water, Switzerland | 2.74 | 0-5.47 | > 2.5 µm | |
| Tap water, Uganda | 3.92 | 0-12.7 | > 2.5 µm | |
| Tap water, USA | 9.24 | 0–60.9 | > 2.5 µm | |
| Tap water, China | 343.5 | 267-404 | > 1 µm | This study |

Further research

In this study, our results showed that there are microplastics in the drinking water supply chain, and the current drinking water treatment process can remove most of the microplastics in raw water. Drinking water is closely related to human health and the water quality should be guaranteed. Plastic products are indispensable materials in our daily life. Controlling plastics from the sources into freshwater is the key to solve the problem of microplastics in drinking water. Currently, some countries and organizations have already begun to promulgate laws and regulations to reduce the use of plastics and (micro)plastic emissions (Hu et al. 2019). Wastewater treatment plant effluent is an important source of microplastics (Ziajahromi et al. 2017). The removal rate of microplastics in wastewater treatment process can be improved by improving existing processes (Perren et al. 2018; Xu et al. 2012). Additionally, microplastic fate during drinking water treatment processes is not yet fully understood. Evidence showed that the fragmentation and decomposition of microplastics happens during wastewater treatment processes (Enfrin et al. 2019; Wang et al. 2018, b), which would significantly increase the content of microplastics with small particle size or nanoplastics in waters. This fragmentation may also occur in drinking water treatment processes. At present, the conventional water treatment processes are not designed for microplastic removal, and the occurrence of microplastics may affect the whole drinking water treatment process such as coagulation, filtration, and disinfection. Therefore, the fate of microplastics during the drinking water supply chain should be valued and given attentions, and standards of the content of microplastics in drinking water should be formulated. Furthermore, the existence of micro-plastics in drinking water has been confirmed, and it is necessary to implement terminal treatment of drinking water. Membrane technology (ultrafiltration, nanofiltration, reverse osmosis) has been successfully applied in water treatment (Wang et al. 2019). It may be a good choice to add membrane water filter device after tap before human consumption to remove microplastics with small particle size in tap water. Moreover, drinking water has become one of the main sources of human exposure to microplastics. However, unfortunately, there are few studies on the effect of microplastics from drinking water on human health and the impacts are not clear. More efforts are required to determine the toxicity of microplastics and the route of exposure to assess the associated potential risks (Shen et al. 2019; Wright and Kelly 2017). Plastics widely used in our daily life should be paid special attention, of course, because plastic packaging materials may also lead to contamination of drinks and foods.

Conclusions

In this study, the content of microplastics in three different water, freshwater (raw water), treated distributed water, and household tap water, was investigated from April to July 2019. The overall average content of microplastics in freshwater was measured to be 2753 (2173–3998) particles L^{-1} , and 351.9 (338–400) and 343.5 (267–404) particles L^{-1} in treated distributed water and household tap water,

respectively. Microplastics can be significantly removed through current drinking water treatment processes, while the concentration of microplastics in tap water was not ignorable. Microplastics with small particle size (< 10 μ m) accounted for the majority, which were difficult to be quantified and artificially neglected in most research. Fibers and fragments made up the majority in all water samples, which also demonstrates that current drinking water treatment processes are not effective in removing small fibers and fragments. In addition, we found that the plastic materials applied to the drinking water supply chain could contribute to the content of microplastics in drinking water. At present, research on microplastics in drinking water has just begun; the determination and quantification of small microplastics in drinking water, the removal of microplastics during water treatment, and potential risks to human health should be further studied in order to better understand the microplastics in drinking waters.

Author contribution All co-authors have seen and approved the manuscript and have agreed to its submissions for publication:

Maocai Shen: Writing, methodology, investigation, analysis, original draft

Zhuotong Zeng: Investigation, analysis, review, and editing

Xiaofeng Wen: Methodology, review, and editing

Xiaoya Ren: Investigation, review, and editing

Guangming Zeng: Writing, methodology, investigation, review, and editing

Yaxin Zhang: Investigation, review, and editing Rong Xiao: Methodology, review, and editing

Supplementary Information The online version contains supplementary material available at https://doi.org/10.1007/s11356-021-13769-x.

Funding The study is financially supported by the Program for the National Natural Science Foundation of China (U20A20323, 51521006, 51909090), the Program for Changjiang Scholars and Innovative Research Team in University (IRT-13R17), the Three Gorges Follow-up Research Project (2017HXXY-05), and the Natural Science Foundation of Hunan Province, China (2019JJ50409).

Declarations

This manuscript was only submitted on *Environmental Science and Pollution Research*.

The authors make sure they have permissions for the use of software, questionnaires/(web) surveys, and scales in their studies (if appropriate). This research may not be misapplied to pose a threat to public health or national security.

There was no animal experiment in this manuscript.

Consent to publish Results in this manuscript were presented clearly, honestly, and without fabrication, falsification, or inappropriate data manipulation (including image-based manipulation).

Conflict of interest The authors declare no competing interests.

References

- Browne MA, Crump P, Niven SJ, Teuten E, Tonkin A, Galloway T, Thompson R (2011) Accumulation of microplastic on shorelines woldwide: sources and sinks. Environ Sci Technol 45:9175–9179
- Di M, Liu X, Wang W, Wang J (2019) Pollution in drinking water source areas: microplastics in the Danjiangkou Reservoir, China. Environ Toxicol Pharmacol 66:133–133
- Di MX, Wang J (2018) Microplastics in surface waters and sediments of the Three Gorges Reservoir, China. Sci Total Environ 616:1620– 1627
- Dris R, Gasperi J, Rocher V, Saad M, Renault N, Tassin B (2015) Microplastic contamination in an urban area: a case study in Greater Paris. Environ Chem 12:592–599
- Eerkes-Medrano D, Thompson RC, Aldridge DC (2015) Microplastics in freshwater systems: a review of the emerging threats, identification of knowledge gaps and prioritisation of research needs. Water Res 75:63–83
- Enfrin M, Dumee LF, Lee J (2019) Nano/microplastics in water and wastewater treatment processes - origin, impact and potential solutions. Water Res 161:621–638
- Eriksen M, Mason S, Wilson S, Box C, Zellers A, Edwards W, Farley H, Amato S (2013) Microplastic pollution in the surface waters of the Laurentian Great Lakes. Mar Pollut Bull 77:177–182
- Fossi MC, Coppola D, Baini M, Giannetti M, Guerranti C, Marsili L, Panti C, de Sabata E, Clò S (2014) Large filter feeding marine organisms as indicators of microplastic in the pelagic environment: the case studies of the Mediterranean basking shark (Cetorhinus maximus) and fin whale (Balaenoptera physalus). Mar Environ Res 100:17–24
- Fossi MC, Pedà C, Compa M, Tsangaris C, Alomar C, Claro F, Ioakeimidis C, Galgani F, Hema T, Deudero S, Romeo T, Battaglia P, Andaloro F, Caliani I, Casini S, Panti C, Baini M (2018) Bioindicators for monitoring marine litter ingestion and its impacts on Mediterranean biodiversity. Environ Pollut 237:1023– 1040
- Free CM, Jensen OP, Mason SA, Eriksen M, Williamson NJ, Boldgiv B (2014) High-levels of microplastic pollution in a large, remote, mountain lake. Mar Pollut Bull 85:156–163
- Frias JPGL, Nash R (2019) Microplastics: finding a consensus on the definition. Mar Pollut Bull 138:145–147
- Gündogdu S, Çevik C, Ayat B, Aydoğan B, Karaca S (2018) How microplastics quantities increase with flood events? An example from Mersin Bay NE Levantine coast of Turkey. Environ Pollut 239:342–350
- Hartmann NB, Hüffer T, Thompson RC, Hassellöv M, Verschoor A, Daugaard AE, Rist S, Karlsson T, Brennholt N, Cole M, Herrling MP, Hess MC, Ivleva NP, Lusher AL, Wagner M (2019) Are we speaking the same language? Recommendations for a definition and categorization framework for plastic debris. Environ Sci Technol 53:1039–1047
- Hu D, Shen M, Zhang Y, Li H, Zeng G (2019) Microplastics and nanoplastics: would they affect global biodiversity change? Environ Sci Pollut Res 26:19997–20002
- Hu D, Shen M, Zhang Y, Zeng G (2019) Micro(nano)plastics: an unignorable carbon source? Sci Total Environ 657:108–110

- Käppler A, Fischer D, Oberbeckmann S, Schernewski G, Labrenz M, Eichhorn KJ, Voit B (2016) Analysis of environmental microplastics by vibrational microspectroscopy: FTIR, Raman or both? Anal Bioanal Chem 408:1–15
- Klein S, Worch E, Thomas P, Knepper (2015) Occurrence and spatial distribution of microplastics in river shore sediments of the Rhine-Main area in Germany. Environ Sci Technol 49:6070–6076
- Kosuth M, Mason SA, Wattenberg EV (2018) Anthropogenic contamination of tap water, beer, and sea salt. PLoS One 13:e0194970
- Lechner A, Keckeis H, Lumesberger-Loisl F, Zens B, Krusch R, Tritthart M, Glas M, Schludermann E (2014) The Danube so colourful: a potpourri of plastic litter outnumbers fish larvae in Europe's second largest river. Environ Pollut 188:177–181
- Leslie HA, Brandsma SH, Velzen MJMV, Vethaak AD (2017) Microplastics en route: field measurements in the Dutch river delta and Amsterdam canals, wastewater treatment plants, North Sea sediments and biota. Environ Int 101:133–142
- Ma B, Xue W, Ding Y, Hu C, Liu H, Qu J (2018) Removal characteristics of microplastics by Fe-based coagulants during drinking water treatment. J Environ Sci 78:267–285
- Ma B, Xue W, Hu C, Liu H, Qu J, Li L (2019) Characteristics of microplastic removal via coagulation and ultrafiltration during drinking water treatment. Chem Eng J 359:159–167
- Mason SA, Welch VG, Neratko J (2018) Synthetic polymer contamination in bottled water. Front Chem 6:407
- Mintenig SM, Loder MGJ, Primpke S, Gerdts G (2019) Low numbers of microplastics detected in drinking water from ground water sources. Sci Total Environ 648:631–635
- Negrete Velasco AJ, Rard L, Blois W, Lebrun D, Lebrun F, Pothe F, Stoll S (2020) Microplastic and fibre contamination in a remote mountain lake in Switzerland. Water 12:2410
- Oßmann BE, Sarau G, Holtmannspötter H, Pischetsrieder M, Christiansen SH, Dicke W (2018) Small-sized microplastics and pigmented particles in bottled mineral water. Water Res 141:307– 316
- Panno SV et al (2019) Microplastic contamination in karst groundwater systems. Groundwater 57:189–196. https://doi.org/10.1111/gwat. 12862
- Perren W, Wojtasik A, Cai Q (2018) Removal of microbeads from wastewater using electrocoagulation. ACS Omega 3:3357–3364
- Pivokonsky M, Cermakova L, Novotna K, Peer P, Cajthaml T, Janda V (2018) Occurrence of microplastics in raw and treated drinking water. Sci Total Environ 643:1644–1651
- PlasticsEurope (2018) Plastics-The facts 2018
- Schymanski D, Goldbeck C, Humpf HU, Furst P (2018) Analysis of microplastics in water by micro-Raman spectroscopy: release of plastic particles from different packaging into mineral water. Water Res 129:154–162
- Setälä O, Fleming-Lehtinen V, Lehtiniemi M (2014) Ingestion and transfer of microplastics in the planktonic food web. Environ Pollut 185: 77–83
- Shen M, Song B, Zeng G, Zhang Y, Teng F, Zhou C (2021) Surfactant changes lead adsorption behaviors and mechanisms on microplastics. Chem Eng J 405:126989
- Shen MC, Song B, Zhu Y, Zeng G, Zhang Y, Yang Y, Wen X, Chen M, Yi H (2020) Removal of microplastics via drinking water treatment: current knowledge and future directions. Chemosphere 251:126612
- Shen M, Zeng G, Zhang Y, Wen X, Song B, Tang W (2019) Can biotechnology strategies effectively manage environmental (micro)plastics? Sci Total Environ 697:134200
- Shen M, Zhang Y, Zhu Y, Song B, Zeng G, Hu D, Wen X, Ren X (2019) Recent advances in toxicological research of nanoplastics in the environment: a review. Environ Pollut 252:511–521
- Shen M, Zhu Y, Zhang Y, Zeng G, Wen X, Yi H, Ye S, Ren X, Song B (2019) Micro(nano)plastics: unignorable vectors for organisms. Mar Pollut Bull 139:328–331

- Su L, Xue Y, Li L, Yang D, Kolandhasamy P, Li D, Shi H (2016) Microplastics in Taihu Lake, China. Environ Pollut 216:711–719
- Thompson RC, Olsen Y, Mitchell RP, Davis A, Rowland SJ, John AW, McGonigle D, Russell AE (2004) Lost at sea: where is all the plastic? Science 304:838–838
- Triebskorn R et al (2019) Relevance of nano- and microplastics for freshwater ecosystems: a critical review. Trac-Trends Anal Chem 110: 375–392. https://doi.org/10.1016/j.trac.2018.11.023
- Vendel AL, Bessa F, Alves VE, Amorim AL, Patrício J, Palma AR (2017) Widespread microplastic ingestion by fish assemblages in tropical estuaries subjected to anthropogenic pressures. Mar Pollut Bull 117:448–455
- Wang W, Yuan W, Chen Y, Wang J (2018) Microplastics in surface waters of Dongting Lake and Hong Lake. China Sci Total Environ 633:539–545
- Wang H, Zeng Z, Xu P, Li L, Zeng G, Xiao R, Tang Z, Huang D, Tang L, Lai C, Jiang D, Liu Y, Yi H, Qin L, Ye S, Ren X, Tang W (2019) Recent progress in covalent organic framework thin films: fabrications, applications and perspectives. Chem Soc Rev 48:488–516
- Wang Y, Zhu Y, Hu Y, Zeng G, Zhang Y, Zhang C, Feng C (2018) How to construct DNA hydrogels for environmental applications: advanced water treatment and environmental analysis. Small 14: 1703305
- Wen X, du C, Xu P, Zeng G, Huang D, Yin L, Yin Q, Hu L, Wan J, Zhang J, Tan S, Deng R (2018) Microplastic pollution in surface sediments of urban water areas in Changsha, China: abundance, composition, surface textures. Mar Pollut Bull 136:414–423
- Woodall LC, Gwinnett C, Packer M, Thompson RC, Robinson LF, Paterson GLJ (2015) Using a forensic science approach to minimize

environmental contamination and to identify microfibres in marine sediments. Mar Pollut Bull 95:40-46

- Wright SL, Kelly FJ (2017) Plastic and human health: a micro issue? Environ Sci Technol 51:6634–6647
- Xu P, Zeng GM, Huang DL, Feng CL, Hu S, Zhao MH, Lai C, Wei Z, Huang C, Xie GX, Liu ZF (2012) Use of iron oxide nanomaterials in wastewater treatment: a review. Sci Total Environ 424:1–10
- Yang D, Shi H, Li L, Li J, Jabeen K, Kolandhasamy P (2015) Microplastic pollution in table salts from China. Environ Sci Technol 49:13622–13627
- Yuan W, Liu X, Wang W, Di M, Wang J (2019) Microplastic abundance, distribution and composition in water, sediments, and wild fish from Poyang Lake. China Ecotoxicol Environ Saf 170:180–187. https:// doi.org/10.1016/j.ecoenv.2018.11.126
- Zhang K, Xiong X, Hu H, Wu C, Bi Y, Wu Y, Zhou B, Lam PKS, Liu J (2017) Occurrence and characteristics of microplastic pollution in Xiangxi Bay of Three Gorges Reservoir, China. Environ Sci Technol 51:3794–3801
- Zhao S, Zhu L, Wang T, Li D (2014) Suspended microplastics in the surface water of the Yangtze Estuary System, China: first observations on occurrence, distribution. Mar Pollut Bull 86:562–568
- Zhao J et al (2018) Microplastic pollution in sediments from the Bohai Sea and the Yellow Sea, China. Sci Total Environ 640–641:637– 645
- Ziajahromi S, Neale PA, Rintoul L, Leusch FD (2017) Wastewater treatment plants as a pathway for microplastics: development of a new approach to sample wastewater-based microplastics. Water Res 112: 93–99

Publisher's note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.