Distribution and source of microplastics in China’s second largest reservoir - Danjiangkou Reservoir

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A B S T R A C T

Fresh water microplastic pollution is of pressing concern globally, but its distribution and sources in reservoirs are poorly documented. Danjiangkou Reservoir is the second largest reservoir in China and is divided into the Han Reservoir and Dan Reservoir. In this work, microplastic abundances and morphological characteristics of the reservoir were investigated. The microplastic abundance of 15 main tributaries of the reservoir was also measured. The vertical distribution (in water column and sediment), horizontal distribution (in Han Reservoir and Dan Reservoir) and source of microplastics were analyzed. Microplastics accumulated in the middle layer of the reservoir, and the size and color of the microplastic particles changed from the surface to the bottom, which implies that surveys of surface water are not enough to determine the microplastic contamination for deep water reservoirs. In the surface water, the microplastic abundance in the Han Reservoir was lower than that in the Dan Reservoir ($p < 0.05$), but microplastic abundance did not differ significantly in the intermediate and bottom water. Tributaries were one of the main sources of microplastics for Han Reservoir but not for Dan Reservoir. Agricultural cultivation in the hydro-fluctuation belt might be an important source of microplastics in the Dan Reservoir, which should be given additional attention. The results of this study can provide valuable information for developing microplastic sampling strategies in deep water reservoirs. Further studies are recommended to investigate the process through which microplastics in the hydro-fluctuation belt enter the reservoir and the sinking behavior of microplastics in the reservoir.

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Introduction

The global annual production of plastics has increased rapidly in recent decades, and approximately 359 million tons produced in 2018 (PlasticsEurope, 2019). The widespread use of plastic products increases the level of convenience in people’s daily lives. However, due to the inappropriate disposal of plastic waste, parts of plastic enter aquatic environments via rainfall runoff, sewage discharge and atmospheric precipitation and produce several environmental problems. Microplastics are defined as plastic with diameters less than 5 mm (Thompson et al., 2004). As an emerging pollutant, microplastics have become an extensive concern in recent years. Researchers have found large amounts of microplastics not only in seawater and ocean sediments (Aliabad et al., 2019; Peng et al., 2017; Ronda et al., 2019; Woodall et al., 2020; Zhang et al., 2019a, b; Zhao et al., 2018) but also in inland fresh waters (Xiong et al., 2018; Zhang et al., 2015) and even in the digestive tracts of a variety of organisms (Li et al., 2018a; Lusher et al., 2013).

Microplastics ingested by organisms block the digestive tract, create a false feeling satiety, and cause physiological and reproductive lesions (Ferreira et al., 2016; Lusher et al., 2013; Van Cauwenberghe et al., 2015). In addition, microplastics are carriers of toxic pollutants since they have large specific surface areas and strong adsorption capacities (Fan et al., 2015; Tang et al., 2018). Toxic substances attached to microplastics also pose great hazards to aquatic organisms and eventually pass through the food chain to humans (Wang et al., 2019; Yan et al., 2019).

The problem of microplastics in inland fresh waters has received more attention. Microplastic abundances reported recently showed that microplastic pollution in some freshwater environments of China were serious, especially in areas where anthropogenic activities were frequent. The microplastic concentration range of water was 3400–25,800 n/m³ in Taihu Lake (Su et al., 2016), 900–2800 n/m³ in Dongting Lake and 1250–4650 n/m³ in Hong Lake (Wang et al., 2018). The Three Gorges Reservoir, the largest reservoir in China, also contained a high level of microplastic abundance of 1597–12,611 n/m³ (Di and Wang, 2018). Although there are some studies reported on microplastics in fresh waters, their vertical distribution in reservoirs is poorly documented.

The Danjiangkou Reservoir is the second largest reservoir in China and is the water source for the Middle Route of the South to North Water Diversion Project (SNWDP) (Xin et al., 2015). The SNWDP has been operating since December 2014, and its water supply has become the main water source of more than 20 cities in the Beijing-Tianjin-Hebei Region, serving as a water source for more than 58.59 million people. Thus, the protection of water quality in this reservoir is of great importance. Microplastics were detected in the Danjiangkou Reservoir (Di et al., 2019). However, the vertical distribution of microplastics in the water column and surficial sediment of this reservoir and their sources remain unknown.

In this context, microplastic abundances and morphological characteristics in the surface water, intermediate water, bottom water and surficial sediment of the reservoir were investigated, and microplastic abundances in the surface water of 15 main tributaries of the reservoir were also determined in the present study. Our purpose was to (1) obtain information on the vertical distribution (in the water column and surficial sediment) and horizontal distribution (in the Han Reservoir and Dan Reservoir) and (2) discuss the sources of microplastics in the reservoir. The results of this study can provide foundational data on microplastics in inland freshwater environments and provide a basis for pollution assessment and prevention of microplastics in reservoirs.

1. Materials and methods

1.1. Study area

The Danjiangkou Reservoir (32°36′~33°48′N, 110°49′~110°59′E) is a large reservoir that provides water supply, flood control, irrigation, and power generation. It is in the upper reaches of the Han River, the largest tributary in the middle reaches of the Yangtze River and is the water source of the SNWDP, which provides water to northern China, including Beijing and Tianjin (Li and Zhang, 2010). It has a surface area of 1050 km² and a total storage capacity of 29.05 km³ when the water reaches its normal level of 170 m (Xin et al., 2015). The reservoir catchment area is 95,200 km², and the average annual inflow is approximately 38.78 km³. Average annual precipitation is 833 mm, and 75% of the annual precipitation falls from May to October (Xin et al., 2015). The Danjiangkou Dam is located at the junction of the Han River and Dan River, so the Danjiangkou Reservoir can be divided into the Han Reservoir and Dan Reservoir. The main counties in the reservoir catchment include Xixia County, Xichuan County, Yunxi County, Yunxian County, and Danjiangkou City (Fig. 1). There are 16 main tributaries in the reservoir, accounting for approximately 94.5% of the whole drainage area (Xin et al., 2015).

1.2. Sampling

In the present study, 28 sampling sites were set up, including 13 sites in the reservoir (R1-R13) and 15 sites in the tributaries (T1-T15). The locations of the sampling sites are shown in Fig. 1. For the 16 main tributaries, the sampling work was carried out in 15 tributaries. The Taogou River is the smallest of the 16 tributaries, and the sampling site on this river had to be abandoned because there was a mudslide during the sampling days. The sampling campaign was carried out in August 2019, corresponding to the wet season in the study area. In the reservoir, surface, intermediate and bottom water (a total of 39 samples) and surficial sediments (13 samples) were collected from all sites (R1-R13). The water depths of the sampling sites in the Han River (R1-R6) ranged from 20 to 51 m, and those in the Dan River (R7-R11) ranged from 13 to 54 m (Appendix A Table S1). The surface water was collected at 0.5 m below the water surface, the intermediate water was collected at the middle depth of the water, and the bottom water was collected 0.5 m above the bottom of the reservoir. In the tributaries (T1-T15), surface water was collected. The sampling sites and basin characteristics of the 16 tributaries of the Danjiangkou Reservoir are shown in Appendix A Table S2.
Water samples were collected by a 5 L stainless-steel water sampler and stored in 5 L brown glass bottles. After sampling was completed, samples were refrigerated at 4°C, and filtration was completed within 24 hr. All samples were collected during stable, non-precipitation-driven hydrologic periods. During field collection, the brown glass bottles were cleaned thoroughly by rinsing them with 70% ethanol at least three times before use. Each water sample was filtered by a 75 μm stainless steel sieve with a 12 V DC Teflon™ pump (Wu et al., 2019).

Approximately 500 g of surficial sediment was collected with a Van Veen grab (0.25 m² sampling area) at each sampling site in the reservoir and placed in a stainless-steel cylinder. Then, the stainless-steel cylinders were covered by aluminum foil to avoid air contamination and preserved at 4°C before analysis. For both the water and sediment samples, two replicates were taken for each sample. All sampling tools were cleaned with deionized water between each sample.

1.3. **Sample analysis**

After filtration through a stainless-steel sieve, the retained particles on the sieves were rinsed with deionized (DI) water and combined into a 250 mL beaker and treated with 50 mL 30% H₂O₂ for 24 hr at room temperature. Then, the samples were placed on an electric heating plate at 75°C until no organic matter was found in the beaker (Nuelle et al., 2014). Since the sampling campaign was carried out in the wet season, there was a certain amount of sediment in the water samples. After sample digestion was completed, the residue was floated twice with a NaCl-saturated solution (ρ = 1.19 g/cm³) and ZnCl₂ solution (ρ = 1.55 g/cm³) (Meng et al., 2020). The supernatant of each flotation was collected and filtered with a gridded 0.45 μm glass microfiber filter membrane (GF/F, 47 mm Ø, Whatman) under vacuum. The filter membrane was preserved in a Petri dish.

For the sediment, the collected samples were dried in an oven at 60°C to constant weight, and then, 20 g sediment was resuspended in approximately 5 mL of deionized water and treated with 50 mL of 30% H₂O₂ for 24 hr at room temperature. Then, the samples were placed on an electric heating plate at 75°C until no organic matter was found in the beaker (Nuelle et al., 2014). Then, the residue was floated twice with a NaCl-saturated solution (ρ = 1.19 g/cm³) and ZnCl₂ solution (ρ = 1.55 g/cm³) (Meng et al., 2020). The supernatant of each flotation was collected and filtered with a gridded 0.45 μm glass microfiber filter membrane (GF/F, 47 mm Ø, Whatman).
under vacuum. The filter membrane was preserved in a Petri dish.

After water and sediment sample processing, an electric stereoscopic fluorescence microscope (SMZ25 Nikon, Japan) was used to record microplastic numbers and morphological characteristics (shape, size, and color). A laser confocal microscope (DXR2 Thermo Fisher Scientific, USA, 532 nm laser, Raman shift 50–3500 cm$^{-1}$) was used to identify the chemical composition of the microplastics. The chemical compositions of the selected items were identified by comparing their Raman spectra with the Raman polymer spectrum library (Araujo et al., 2018). Of the particles analyzed under Raman Spectroscopy, 30.8%–73.6% were able to be matched to spectra in a known database, which was consistent with literature (Watkins et al., 2019). The microplastics abundance was obtained by multiplying the total particles number by the percentage of successful identification as microplastics.

There is no uniform standard for the classification of microplastics, and the common categories are shape, size, and color (Hidalgo-Ruz et al., 2012). The shape of the microplastics was an important factor reflecting the pollution pattern of the studied areas (Wu et al., 2019). According to their shape, the microplastics were categorized into fragments, fibers, films, microbeads and pellets in our study. Briefly, a fiber is a microplastic with a long and thin appearance, which includes linear fibers; a film is a small piece that has a very thin layer; microbeads are usually solid particles, mainly from personal care products and with small size; a pellet has an ovoid shape or is disc-shaped or cylindrical with a rough surface; when a microplastic cannot be defined as a fiber, film, microbead or pellet, it is defined as a fragment (Rochman et al., 2016; Su et al., 2016). Typical images of these five shapes of microplastics in the present study are shown in Appendix A Fig. S2.

1.4. Data analysis

ArcGIS 10.5 software was used to draw the map of the locations of the sampling sites (Fig. 1) and calculate the land use type data (Appendix A Table S2). Statistical analysis was performed using SPSS Statistics 20 software, and the difference in the microplastic abundance between two samples was analyzed using independent samples t-test (significance level, $p < 0.05$). Other figures were drawn by Origin 8.0 software.

1.5. Quality assurance and quality control

Quality assurance and quality control (QA/QC) procedures were applied at all stages from collection to detection to reduce potential sample contamination, according to a previous study (Courtene-Jones et al., 2017) with some modification. During field collection, a stainless steel sieve was applied to filter the water samples to avoid possible contamination by a nylon screen. All sample tools and laboratory glassware were pretreated with DI water and dried before use. All the water and solutions were filtered with a 0.45 μm glass microfiber filter membrane before use (Mu et al., 2019).

A separate small laboratory was only used by the scientist carrying out the analysis of microplastics. The door remained closed for the duration of the experiment to reduce air-borne contamination sources. The filter membrane was placed in a covered glass dish and dried in the oven at 50°C for analysis. A clean 100% cotton laboratory coat was worn and stored in the laboratory during the analysis to avoid contact with external synthetic fibers. In all the processes, plastic materials were not used. Moreover, the samples were always covered by aluminum foil to avoid air contamination during sample collection, pretreatment and chemical analyses.

Three blanks were performed to determine level of ambient environmental contamination. The field blank samples of water (DI-water) and sediment (blank quartz sand) were treated the same as the sampled water and sediment in the sampling and laboratory pretreatment processes. In the six blank field experiments (three samples for surface water and three samples for sediment), the mean particle numbers of microplastics on the filter membrane were 7 particles per filter membrane for the blank water samples and 11 particles per filter membrane for the blank sediment samples, indicating that contamination of microplastics during the experimental process could be ignored. These mean particle numbers were subtracted from all sample counts for all analysis and reporting. In the laboratory, three deionized water samples were put into glass bottles, and the same steps of the measurement process as those for laboratory blank samples were carried out; the mean particle number of microplastics on the filter membrane was 4 particles per filter membrane. For 54 water samples (surface, intermediate and bottom water samples in R1-R13 in the reservoir and 15 surface water samples in T1-T15) and 13 surficial sediment samples (R1-R13 in the reservoir), 6 water samples and 2 sediment samples were tested as duplicate samples. The difference in the microplastic particles between the duplicate samples and the environmental samples was less than 50%.

2. Results

2.1. Microplastics in the reservoir

2.1.1. Water

Whole reservoir. Microplastics were detected in all 39 water samples (including the surface, intermediate and bottom water samples) (Appendix A Table S3), and the microplastic abundance varied from 530 to 24,798 n/m$^3$ in the reservoir (mean value 7205 n/m$^3$, Fig. 2). The microplastic abundances in the surface, intermediate and bottom waters ranged from 637 to 12,466 n/m$^3$ (mean value 6081 n/m$^3$), 5058 to 24,798 n/m$^3$ (mean value 9777 n/m$^3$) and 530 to 15,720 n/m$^3$ (mean value 5757 n/m$^3$), respectively (Fig. 2). The mean microplastic abundance in the intermediate water was significantly higher than that in the surface water and bottom water ($p < 0.05$) in the Danjiangkou Reservoir.

In comparison to the other microplastic types, fragments were observed in all 39 water samples in the reservoir and were higher in numbers in most of the water samples, accounting for 16.9%–100% of the microplastics; fibers were distributed in 36 water samples and accounted for 0.7%–64.7%; films were present in 25 of the water samples, ranging from 0.2% to 57.5%; and microbeads and pellets were only present...
in 15 and 13 of the water samples, accounting for 0.5%–15.4% and 0.3%–8.3%, respectively (Fig. 3).

Microplastics were divided into four size groups: 75–200 μm, 200–500 μm, 500–1000 μm and 1000–5000 μm. Of all the particles (including those in the surface, intermediate and bottom water), 61.4% were in the 200–500 μm size range, 20.7% were in the 500–1000 μm size range, 7.9% were larger than 1000 mm, and particles with sizes of 75–200 μm accounted for 10.0% (Fig. 3). When the size was larger than 200 μm, the microplastic abundance decreased with size. From the surface to the bottom of the reservoir, the percentage of particles with sizes of 75–200 μm decreased from 11.0% to 9.0%, and the percentage of particles with sizes of 1000–5000 μm increased from 6.6% to 12.4%.

The most abundant particle color in the 39 water samples (including the surface, intermediate and bottom water samples) was transparent (42.8%), followed by brown (40.9%), black (5.9%), green (3.7%), gray (3.3%), etc. (Fig. 3). In the water column, transparent particles were dominant in the surface and intermediate water, accounting for 43.4% and 47.4% of the particles, respectively. However, brown (51.6%) was most abundant color in the bottom water, and transparent particles accounted for only 34.5% of the microplastics. The size and color of the microplastic particles also changed from the surface to the bottom of the water column.

In the Han Reservoir, 18 water samples (R1-R6, including surface, intermediate and bottom water samples) were collected (Appendix A Table S4), and the mean microplastic abundances in the surface, intermediate and bottom waters were 4260 n/m³, 7819 n/m³, and 4737 n/m³, respectively. In the Dan Reservoir, 21 water samples (R7-R13, including surface, intermediate and bottom water samples) were collected (Appendix A Table S4), and the mean microplastic abundances in the surface, intermediate and bottom waters were 7643 n/m³, 11,456 n/m³, and 6632 n/m³, respectively. Microplastic abundance in the surface water from the Han Reservoir was lower than that from the Dan Reservoir (p < 0.05) but did not differ significantly in the intermediate and bottom water.

2.1.2. Sediment
The microplastic abundance in the sediment varied from 708 to 3237 n/kg (mean 1818 n/kg, Fig. 4). The microplastic particles observed in the sediment samples were dominated by fibers (52.8%). Other particles observed included fragments (42.3%), films (3.87%), and pellets (1.2%), and no microbeads were detected (Fig. 3, Appendix A Table S3). The majority (35.2%) of the particles in the sediment samples were 200–500 μm in size; 23.4% were 500–1000 μm in size; 21.7% were smaller than 200 μm in size; and 19.6% were larger than 1000 μm in size (Fig. 3).

In the Han Reservoir, plastic particles were found in all 6 surficial sediment samples (R1-R6) and the abundance was 708–3237 n/kg (mean value 2062 n/kg) (Fig. 4). In the Dan Reservoir, 7 surficial sediment samples (R7-R13) were collected and the microplastic abundance was 1173–2217 n/kg (mean value 1609 n/kg) (Fig. 4). The microplastic abundance, shape, size and color in the sediment did not differ significantly between the Han Reservoir and Dan Reservoir.

2.2. Microplastics in the tributaries
Microplastic abundance in the surface water from the tributaries ranged from 457 to 27,734 n/m³ (mean value 7056 n/m³, Figs. 5 and Appendix A Fig. S1). The water samples from the tributaries had larger microplastic abundance fluctuations. Fragments were dominant (93.0%) in the surface water samples from the tributaries (T1-T15, Appendix A Table S3), followed by fibers (4.1%), films (2.4%) and microbeads (0.4%), and these results were similar to those in the water of the reservoir, which implied that tributary was one of the sources of microplastics in the reservoir. Brown microplastics (49.4%) were most abundant in the surface water from the tributaries (T1-T15), followed by transparent (35.8%), black (7.68%) and green (2.77%) microplastics.

In the tributaries of Han Reservoir, microplastic abundance ranged from 457 to 27,734 n/m³ (mean value 7702 n/m³) (Fig. 5, Appendix A Table S4), and the most abundant colors were brown (56.0%) and transparent (29.6%). In the tributaries of Dan Reservoir, microplastic abundance ranged from 1141 to 9001 n/m³ (mean value 5278 n/m³) (Fig. 5, Appendix A Table S4), and transparent (51.4%) and brown (33.0%) particles were the dominant colors.

2.3. Chemical identification of microplastics
In terms of chemical identification, 638 particles of microplastic (128 from the surface water in the reservoir, 131 from the
Fig. 3 – Morphological characteristics (shape, size and color) of microplastics in (a) surface water, (b) intermediate water, (c) bottom water and (d) surficial sediment of Danjiangkou Reservoir. R1-R6 in Han Reservoir, R7-R13 in Dan Reservoir.

Fig. 4 – Microplastic abundances in sediment in Danjiangkou Reservoir. R1-R6 in Han Reservoir, R7-R13 in Dan Reservoir.

intermediate water in the reservoir, 120 from the bottom water in the reservoir, 150 from the sediment in the reservoir, and 109 from the surface water in the tributaries) were randomly selected and identified by Raman spectroscopy. The results are shown in Table 1. Polyamide (PA, 24.8%), polyethylene (PE, 24.0%) and polypropylene (PP, 17.1%) were the most abundant polymers in the reservoir, and the Raman spectra of these are shown in Appendix A Fig. S3.

3. Discussion

3.1. Occurrence of microplastics

3.1.1. In the reservoir
Comparisons of microplastic abundance in the surface water and sediment of Danjiangkou Reservoir with other freshwater areas are shown in Appendix A Tables S5 and S6, respectively. The mean microplastic abundance in the surface water from the Danjiangkou Reservoir (6081 n/m³) was higher than that from the Three Gorges Reservoir (4703 n/m³) (Di and Wang, 2018) and Hong Lake (2867 n/m³) (Wang et al., 2018) but lower than that from Beihu Lake (8925 n/m³) and Huanzi Lake (8550 n/m³) in Wuhan city (Wang et al., 2017) and the Gallatin River (12,000 n/m³) in the USA (Barrows et al., 2018), which ex-
Table 1 – Composition of the collected microplastics identified by Raman spectroscopy.

<table>
<thead>
<tr>
<th>Composition of particles</th>
<th>Number of particles</th>
<th>In reservoir</th>
<th>Intermediate water</th>
<th>Bottom water</th>
<th>Sediment</th>
<th>In tributaries</th>
<th>Total</th>
<th>(%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Particles measured</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Polyamide (PA)</td>
<td>128</td>
<td>120</td>
<td></td>
<td></td>
<td>109</td>
<td></td>
<td>638</td>
<td></td>
</tr>
<tr>
<td>Polyethylene (PE)</td>
<td>39</td>
<td>25</td>
<td></td>
<td></td>
<td>29</td>
<td></td>
<td>158</td>
<td>24.8</td>
</tr>
<tr>
<td>Polypropylene (PP)</td>
<td>21</td>
<td>53</td>
<td></td>
<td></td>
<td>26</td>
<td></td>
<td>153</td>
<td>24.0</td>
</tr>
<tr>
<td>Polyvinyl fluoride (PVF)</td>
<td>15</td>
<td>13</td>
<td></td>
<td></td>
<td>15</td>
<td></td>
<td>109</td>
<td>17.1</td>
</tr>
<tr>
<td>Polyvinylchloride (PVC)</td>
<td>29</td>
<td>9</td>
<td></td>
<td></td>
<td>7</td>
<td></td>
<td>75</td>
<td>11.7</td>
</tr>
<tr>
<td>Polystyrene (PS)</td>
<td>6</td>
<td>10</td>
<td></td>
<td></td>
<td>12</td>
<td></td>
<td>53</td>
<td>8.3</td>
</tr>
<tr>
<td>Others**</td>
<td>16</td>
<td>6</td>
<td></td>
<td></td>
<td>5</td>
<td></td>
<td>20</td>
<td>3.1</td>
</tr>
</tbody>
</table>

* The percentage of each plastic type in all the measured particles.
** Others refer to the other compositions of particles except PA, PE, PP, PVF, PVC and PS.

The mean microplastic abundance in the surface water in the Danjiangkou Reservoir (6081 n/m²) was higher than the reported concentrations of 2594 n/m² in 2017 (Di et al., 2019). The sampling campaigns in the 2017 study and our study were carried out in December (dry season) and August (wet season), respectively. In the wet season, plastic wastes from land can be transported to rivers and reservoirs via rainfall-induced surface runoff (Xiong et al., 2018), and microplastic abundance should be at a higher level compared with that in the dry season. The increased microplastic abundance in the reservoir indicated that a large amount of microplastics entered the reservoir in the last 2 years, and the degradation of microplastics into smaller pieces might be another reason for the increased microplastic abundance (Browne et al., 2010).

Fibers were numerically dominant in the sediment in the Danjiangkou Reservoir, which was in accordance with the result in 2017 (Di et al., 2019), but their abundance was higher than that in 2017. This may be due to two reasons. One reason is that fibers were the most common microplastics in the water in 2017 (Di et al., 2019), and they have a relatively larger surface area-to-volume ratio and are more prone to biofouling and sinking (Meng et al., 2020); thus, the fibers in the water settled gradually to the sediment. Thus, this result indicates that the settling of microplastics from the surface waters occurs within the slower water of the reservoir (Watkins et al., 2019).

Another freshwater study also found that particles from fibers can distribute through the water column and settle in the surficial sediment (Lenaker et al., 2019). The other reason for the dominance of fibers is that microplastics can be present and accumulate in sediment for a long time (Wu et al., 2019). Researchers have reported that relatively larger particles (i.e., 300–500 μm) can be retained in sediment during base flow periods (Meng et al., 2020; Nizzetto et al., 2016). Microbeads and pellets were detected at most sampling sites in the water column but were not present in the sediment. This may have been due to the lower surface-to-volume ratio of spherical debris; thus, these types of microplastics are unlikely to biofoul and sink (Meng et al., 2020).

Microplastics that were < 500 μm were most dominant (71.4%) in the water column in the present study. The reason for this result might be that the particle size of microplastics in the wastewater discharged from most sewage treatment plants is less than 500 μm (Mason et al., 2016; Li et al.,
Similarly, microplastics will continue to degrade into smaller plastics, and the smaller the particle size is, the easier it is to enter the tributary with rainfall and ultimately enter into the reservoir (Zhang et al., 2015). The particle sizes of the microplastics in the 15 surface water samples from the tributaries were measured, and particles with sizes < 500 μm were found to be dominant (accounting for 72.7%), which was assistant with that in the reservoir. Since small particles have a greater impact on the ecosystem (Xiong et al., 2018), more attention should be focused on the microplastics in the reservoir.

3.1.2. In the tributaries
In the tributaries of the Han Reservoir (Appendix A Fig. S1), lower microplastic abundance occurred in the water samples from the lower reaches of the Han River (T4-T11, except T7), which was probably due to relatively less agricultural and human activity in this area. The microplastic abundances in the water of samples T6 (in the Quyuan River) and T8 (in the Si River) were lower than 1000 n/m³. However, the microplastic abundance of the sampling sites in the upper reaches of the Han River (samples in T1-T3) exceeded 8000 n/m³. The microplastic abundance in sample T1 (in the Han River) was 10,462 n/m³. Sample T3 (in the Tian River) was downstream of Yunxi County, whose agriculture is well developed. Thus, the microplastic abundance of T3 reached as high as 27,734 n/m³. Sample T7 (in the Shending River) was downstream of Shiyian City, a developed area due to the automobile industry. The water quality of the Shending River does not meet the standard throughout the year due to the overflow of sewage and the discharge of a large amount of water treated by sewage treatment plant (Hu et al., 2019). The microplastic abundance of T7 (in the Shending River) was 9608 n/m³, and the relatively high microplastic pollution was due to industrial activities and domestic effluents, as well as plastic waste upstream of Shiyian City.

3.2. Distribution of microplastics
The mean microplastic abundance in the intermediate water was significantly higher than that of the surface water and bottom water (p < 0.05) in the Danjiangkou Reservoir (Fig. 2). Researchers found that monitoring multiple water layers rather than surface water was more reliable for obtaining the spatial distribution of microplastics in seawater (Dai et al., 2018; Lusher et al., 2015) and fresh rivers and lakes (Lenaker et al., 2019). Lusher et al. (2015) observed more microplastics in the water column (6 m depth) than in the surface water in the Arctic Ocean. Lenaker et al. (2019) demonstrated the uneven distribution of polymer types through water column compartments and sediment in freshwater; in addition, the presence of low-density particles decreased from the water surface to the subsurface to the sediment, while the presence of high-density particles had the opposite result. In present study of an inland freshwater reservoir, we found that microplastics accumulated in the middle-layer water. Surface currents in the Danjiangkou Reservoir have a higher velocity (up to 0.48 m/s) than the subsurface currents (Duan, 2014; Duan et al., 2018), which may easily lead to the transport of microplastics from the surface to the subsurface with long residence times at the subsurface (Dai et al., 2018; Song et al., 2020). Thus, for large and deep-water reservoirs, surveys of surface water are not enough to determine microplastics contamination.

The mean microplastic abundances in the water column (including surface, intermediate and bottom water) of the Han Reservoir and Dan Reservoir were 5605 n/m³ and 8577 n/m³, respectively (Fig. 2). As most of the water in the Han Reservoir is discharged through the Danjiangkou Dam, water exchange in the Han Reservoir is more frequent, and the flow rate is faster (Xin et al., 2015). The water-flow speed in the Dan Reservoir is slower than that in the Han Reservoir (Duan et al., 2018), which is more conducive to the aggregation of microplastics (Zhang et al., 2017). In the Dan Reservoir, the microplastic abundance at the sampling sites on the left bank (R7, R8, R10 and R11) was higher than that on the right bank (R9 and R12) (p < 0.05), which may be related to the slower water velocity on the left bank than on the right bank (Duan et al., 2018). Furthermore, R10 and R11 were near a big town named Xianghua Town, their relatively higher microplastic abundance probably due to frequent agricultural and human activities in the town.

The mean microplastic abundances in the surficial sediment of the Han Reservoir and Dan Reservoir were 2062 ± 827 n/kg and 1609 ± 366 n/kg, respectively (Fig. 4), and no significant difference was found. Sediments have been proposed as the destination of microplastics and some other pollutants in aquatic environments (Woodall et al., 2020). Natural processes, buoyancy and density changes eventually lead to the deposition of microplastics in sediments (Bergmann et al., 2017). It has been reported that a high microplastic abundance in sediment may be related to a decrease in flow velocity, which facilitates the sedimentation of dense plastic particles (Zhang et al., 2017). The relatively lower flow velocity in the Dan Reservoir was beneficial to the settlement of microplastics in the water column (Di et al., 2019; Zhang et al., 2017). However, the transport and settling of microplastics in water bodies are mainly driven by hydrodynamic conditions such as flow velocity, river geometry, and flood events (Sagawa et al., 2018; Song et al., 2020), which will change microplastic residence times in the water column and sediment. It will be necessary to estimate the residence times to elucidate the behaviors of microplastics in reservoir environments in the future.

3.3. Source of microplastics
It has been reported that human activity, agriculture, and industrial production are the main sources of plastic waste in natural waterbodies, and microplastic abundance is closely related to local economic development (Wang et al., 2017). The way microplastics enter aquatic environments includes atmospheric deposition, tributary input, sewage discharge, and rainfall runoff.

Studies on airborne microplastics have recently garnered increased attention. Airborne microplastics are considered to be an important contributor to microplastics in aquatic environments, and they enter aquatic environments via atmospheric bulk deposition (Cai et al., 2017; Dris et al., 2016; Huang et al., 2020). There are few studies on atmospheric deposition fluxes of microplastics in China, and only atmo-
spheric fallout in Dongguan City and Yantai City of China has been reported. It was found that the dry and wet atmospheric deposition fluxes in Dongguan City and Yantai City of China were 175–313 n/(m² day) and 130–624 n/(m² day), respectively, and the mean value was 228 n/(m² day) (Cai et al., 2017; Huang et al., 2020), which showed a relatively high atmospheric deposition fluxes. Rainfall is reported as an important factor influencing the fallout flux (Dris et al., 2016). In the wet season, large amounts of microplastics enter the reservoir via atmospheric rainfall. Due to such a large water area of Danjiangkou Reservoir (1050 km²), atmospheric deposition will be a source of microplastic that cannot be ignored in this reservoir. The atmospheric deposition fluxes of microplastics in the Danjiangkou Reservoir needs to be measured and the amount of microplastics originated annually from the atmosphere deposition should be calculated in the future work.

The main water source of the Danjiangkou Reservoir is the 16 main tributaries (Xin et al., 2015). The total runoff of 11 tributaries in the Han Reservoir accounted for 90.7% of the total water inflow to the Danjiangkou Reservoir, and that in the Dan Reservoir accounted for only 9.3% (Appendix A Table S2). In addition, according to the microplastic abundances measured in each tributary in the present study, the calculated annual input microplastic particle number from the tributaries in the Han Reservoir was 14 times that in the Dan Reservoir. Thus, tributaries were one of the main sources of microplastics for the Han Reservoir but not for the Dan Reservoir.

A special zone on the bank of the Danjiangkou Reservoir, which is called the hydro-fluctuation belt (HFB), is periodically exposed due to the seasonal impoundment of the Danjiangkou Dam. This belt has a vertical height of 13 m (from 157 m to 175 m water level) with a total area of 485.6 km², of which 66.1% is farmland, 6.2% is residential area, 4.7% is forest, and the remaining 11.7% is not used (Appendix A Fig. S4). The HFB area of the Danjiangkou Reservoir (485.6 km²) is even larger than that of the largest Chinese reservoir, the Three Gorges Reservoir (349 km²) (Zhang et al., 2019a,b). Farmland was dominant in the HFB and mostly distributed in Xichuan County, which is in the Dan Reservoir area (Figs. 1 and Appendix A Fig. S4) (Ye et al., 2016). The agricultural production in this area is still active in recent years.

The HFB has been reported to be an important microplastic sink when the water level is low, and the belt can turn into a potential source when the water level is high (Zhang et al., 2019a,b). The application of plastic film and organic fertilizers during agricultural activities is a nonnegligible source of microplastics in the environment (Di et al., 2019; Nizzetto et al., 2016). In the Dan Reservoir, the detected particle numbers of the film and fragments were much larger than those of the Han Reservoir (Appendix A Table S3). This might be related to the high proportion of vegetable greenhouses and plastic mulch as part of agricultural production in the HFB around the Dan Reservoir. The sampling campaign was carried out in August in present study, corresponding to the wet season in the study area. The large amount of microplastics in the farmland can be transported to reservoirs with rainfall runoff (Ding et al., 2019; Xiong et al., 2018). Therefore, microplastics in the HFB might be an important source of microplastics in the Dan Reservoir and we need to conduct more research to sort this out.

4. Conclusions

This study investigated the distribution of microplastics in the water column and surficial sediment of the Danjiangkou Reservoir and the source of microplastics. The microplastic abundance in the water column of the reservoir showed that microplastics accumulated in the middle layer of the reservoir. The size and color of the microplastic particles also changed from the surface to the bottom of the vertical water column. Tributaries were one of the main sources of microplastics in the Han Reservoir but not in the Dan Reservoir; microplastics in the HFB of the Dan Reservoir need to be given increased attention. Further studies are recommended to investigate how the microplastics in the hydro-fluctuation belt enter the reservoir and the sinking behavior of microplastics in the reservoir, which could be used for policies to reduce microplastic transport.

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Appendix A Supplementary data

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.jes.2020.09.018.

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