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Microplastics pollution in inland freshwaters of China: A case study in urban surface waters of Wuhan, China

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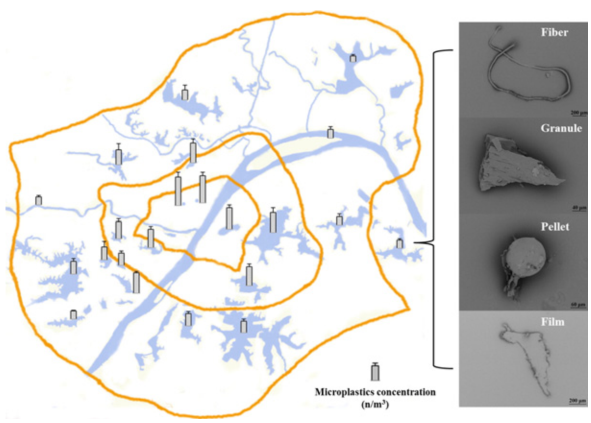
HIGHLIGHTS

- Microplastics were studied in urban waters of the largest city in central China.
- Anthropogenic factors greatly affected the abundance of microplastics in water.
- Fibrous, colored and small-sized were main features of the detected microplastics.

GRAPHICAL ABSTRACT



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ABSTRACT

Microplastics have been considered as an emerging pollutant in the aquatic environment. However, research about microplastic pollution in inland freshwaters of China is insufficient. The present study investigated the levels of microplastics in surface water of 20 urban lakes and urban reaches of the Hanjiang River and Yangtze River of Wuhan, the largest city in central China. Microplastic concentrations ranged from 1660.0 ± 639.1 to 8925 ± 1591 n/m^3 for the studied waters, with the highest concentration found in Bei Lake. Microplastic abundance in lakes varied markedly in space, and negatively correlated with the distance from the city center ($p < 0.001$), which confirmed the important role of anthropogenic factors in microplastic distribution. Urban reaches of the Hanjiang River and Yangtze River were found to have relatively lower levels of microplastics than most of the studied lakes. The major type of microplastics among the studied waters was colored plastic, with fiber being the most frequent shape. More than 80% of microplastics in number had a size of <2 mm. Polyethylene terephthalate and polypropylene were the dominant polymer-types of microplastics analyzed. This study provided important reference for better understanding microplastic levels in inland freshwaters.

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1. Introduction

Due to its lightweight, durable nature and impressive ratio of cost to performance, plastic has become an impartible role in sustaining and delivering the quality, comfort and safety of modern life-styles (Phuong et al., 2016). The global production of plastics increased rapidly since the realization of mass production in 1950s, and the annual yield reached 311 million tons in 2014 (PlasticsEurope, 2015). However, difficulty in degradation also makes the heavily generated plastic waste a serious environmental issue (Barnes et al., 2009). Although a large proportion of the waste has been recycled or landfilled, still a considerable amount of plastic waste entered into the aquatic system (Cole et al., 2011; Eerkes-Medrano et al., 2015). For instance, approximately 4.8–12.7 million tons of plastic waste was estimated to get into the ocean from land per year (Jambeck et al., 2015). Microplastics are defined as small plastic particles with a size of <5 mm (Arthur et al., 2009). They can originate primarily from plastics that are manufactured to be of a microscopic size, or secondarily from the fragmentation of larger plastic debris (Cole et al., 2011). These tiny plastic items, as reported by a large number of researchers, are ubiquitously present in the marine ecosystems worldwide (Derraik, 2002; do Sul and Costa, 2014; Wang et al., 2016). Concerns about potential threats of microplastics to aquatic organisms, birds, mammals and even human beings via mistaken ingestion or food web are rising (Graham and Thompson, 2009; Miranda and de Carvalho-Souza, 2016; Wright et al., 2013). Nevertheless, little information is available about microplastics in freshwater systems (Eerkes-Medrano et al., 2015; Wagner et al., 2014), especially in China, the largest producer of plastics around the world (PlasticsEurope, 2015).

Wuhan is the largest city and center of economy, culture and education in central China, with a population of >10 million in the urban area. Because of richness of water resources, including 166 lakes and the Yangtze River (and its largest tributary – the Hanjiang River) flowing through the city, Wuhan is also known as the "city of hundreds of lakes" and "river city". However, along with the fast growing economy, population and urbanization since 1970s, the ecological health of aquatic environments in this city is threatened by various kinds of pollutants. Lakes and rivers were successively reported with toxic organic or/and inorganic pollutants, some of which were proved to pose risks to the aquatic organisms and even human (Cui et al., 2015; Yang et al., 1997; Yang et al., 2009). Nevertheless, pollution status of the new pollutant - microplastics in aquatic ecosystems within Wuhan and even China, is still unclear at present. Hence, in this study, we investigated the abundance, distribution, and morphological characteristics of microplastics in surface water of 20 urban lakes and also the urban sections of the Yangtze River and Hanjiang River in Wuhan. This study could provide useful reference for monitoring microplastic pollution in inland freshwater systems.

2. Method and materials

2.1. Study areas and sample collection

There are 166 lakes in Wuhan City, among which 40 lakes are dotted in the urban areas. In the present study, 20 major lakes from different administrative districts of Wuhan and urban sections of the Yangtze River and Hanjiang River intersected by the City Ring Expressway were chosen as sampling sites (Fig. 1). Sampling work was carried out in April 2016. A total of 123 water samples were collected, including 107 lake water samples and 16 running water samples in the Yangtze River ($n = 11$) and the Hanjiang River ($n = 5$) (Table 1). The sampling points for each site were equally distributed and located by the global position system (GPS) (Table S1). Twenty liters of surface water sample (0–20 cm in depth) was collected using a pre-cleaned 12 V DC Teflon pump and passed through a 50 μm stainless steel sieve. Two replicates were collected at each sampling point. The residue on the sieve was rinsed with distilled water into a 50 mL glass jar and preserved with 5% formalin solution at 4 °C before analysis.

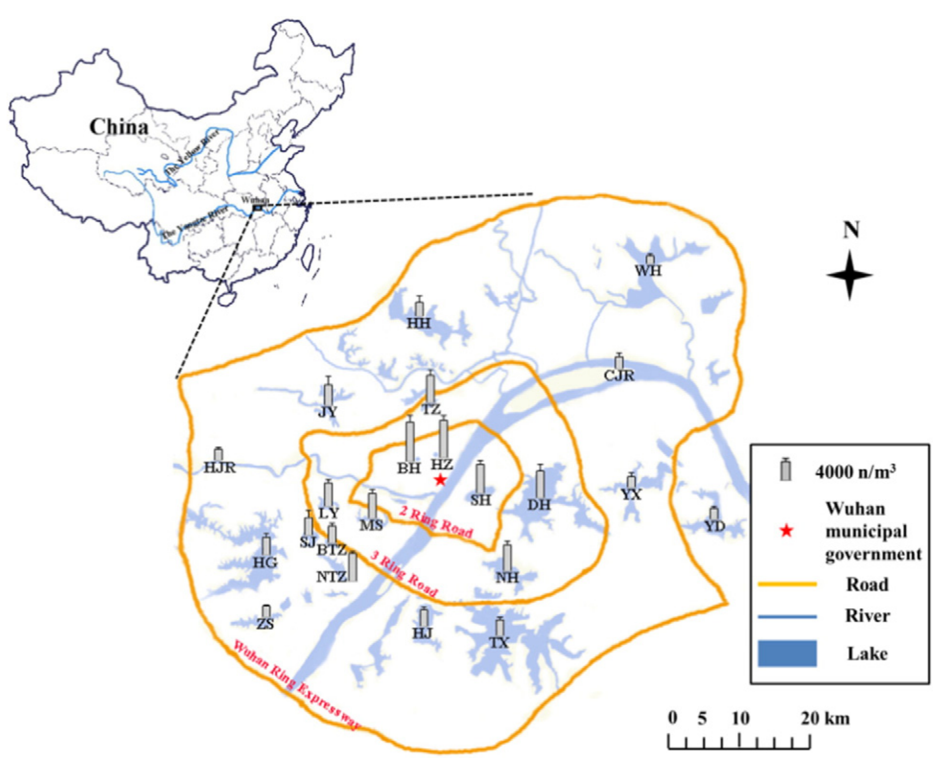

Fig. 1. Geographic location, sampling sites, and spatial distribution of microplastics in surface waters of Wuhan, China.

Table 1
Information about sampling sites.

Location	Code	n[a]	Size (km^2)[b]
Bei Lake	BH	2	0.093
Beitaizi Lake	BTZ	2	0.52
Dong Lake	DH	10	33
Houguan Lake	HG	10	34
Hou Lake	HH	7	16.1
Huangjia Lake	HJ	5	8.12
Huanzi Lake	HZ	2	0.094
Jinyin Lake	JY	5	8.16
Longyang Lake	LY	3	1.68
Moshui Lake	MS	4	3.64
Nan Lake	NH	5	7.67
Nantaizi Lake	NTZ	4	3.57
Sha Lake	SH	4	3.08
Sanjiao Lake	SJ	3	2.39
Tangxun Lake	TX	13	47.62
Tazi Lake	TZ	2	0.31
Wu Lake	WH	9	27.8
Yandong Lake	YD	6	9.11
Yanxi Lake	YX	7	14.23
Zhushan Lake	ZS	4	4.93
Yangtze River	CJR	11	The section intersected by Wuhan city ring expressway
Hanjiang River	HJR	5	As above

[a] Number of water samples collected from each site, the two duplicates collected in one sampling point were considered as one sample.
[b] According to Records of Lakes in Wuhan (Wuhan Water Authority, 2015).

2.2. Laboratory analysis

In the laboratory, water samples were treated with 30% H_2O_2 at room temperature under dark for 24 h to digest organic matter, including biological and abiological materials (Nuelle et al., 2014). Then each sample was diluted with distilled water and filtered through a glass microfiber filter paper with a pore size of 0.45 μm (GF/F, 47 mm Ø, Whatman) under vacuum. After filtration, the filter paper was placed in a clean petri dish and inspected visually using a stereoscopic microscope (M165 FC, Leica, Germany) at up to 160-fold. Subsamples of plastic particles were examined using scanning electron microscopy (SEM) (TM3030, Hitachi, Japan) to investigate their surface characteristics. Microplastics on the filter papers were identified mainly according to their morphological characteristics (such as color, surface structure, and shape) and detailed criteria described by previous studies (Hidalgo-Ruz et al., 2012; Nor and Obbard, 2014). The number, shape, color and size of microplastics for each sample were recorded. Based on their sizes, microplastics were classified into six size classes: class 1 (50–500 μm), class 2 (<500–1000 μm), class 3 (<1000–2000 μm), class 4 (<2000–3000 μm), class 5 (<3000–4000 μm), class 6 (<4000–5000 μm). They were divided into four categories: fiber, granule, film, and pellet, according to the shapes. The abundance of microplastics was calculated as number of particles per cubic meter of water.

In addition, a set of plastic-like items (two particles for each water) were selected based on the proportions of different shapes in the total microplastics, and confirmed by identifying their polymer composition using the Fourier transform infrared (FT-IR) spectrometer (Nicolet 6700; Thermo Fisher Scientific; USA) with a MCT detector running in a wave number range of 8000–50 cm^{-1}. The synthetic polymer type for each sample was identified by comparing its IR spectrum with that of the FT-IR polymer spectrum library.

2.3. Quality assurance and quality control (QA/QC)

Field blank tests were conducted in three randomly selected sampling sites. On each site, 20 L of distilled water was pumped and passed through a 50 μm stainless steel sieve. The duplicates were set in triple. Subsequent treatment for blank samples was identical with that for water samples as above mentioned. Finally, 3.11 ± 1.35 plastic particles were detected under the stereoscopic microscope.

To avoid potential background contamination during the analysis process in the laboratory, the following preventive measures described in literatures (Lusher et al., 2014; Rocha-Santos and Duarte, 2015) were taken in this study. All the materials and vessels were rinsed for three times with distilled water before use and covered with tin foil paper after each step. Cotton lab coats and nitrile gloves were worn during the whole process. Prior to analyzing the samples in the petri dishes, the workplace for stereo-microscopic inspection was carefully cleaned.

Method blank tests were also carried out to check potential contamination (such as contaminants from the distilled water and the air in the laboratory) during the process of analysis (Chae et al., 2015; Dubaish and Liebezeit, 2013; Nuelle et al., 2014). Each test was set in triple. Potential airborne particles were examined by sucking the air in the workplace through the 0.45 μm glass microfiber filter paper for 1 h under vacuum filtration and on average 0.67 ± 0.58 particles were found under the stereoscopic microscope. No plastic particles were detected after passing 20 L of distilled water through the filter paper under vacuum filtration. Therefore, contamination during the analysis process was negligible.

2.4. Statistical analysis

Statistical analysis was conducted using the SAS/PC 9.3 (SAS Institute Inc., USA). Pearson correlation analysis was performed to test the relationship between microplastic concentrations and distance of corresponding lakes from the urban center. Statistical tests were considered significant at p-value < 0.05.

3. Results and discussion

3.1. Abundance and spatial distribution of microplastics

Microplastics were widely detected in surface waters of Wuhan, with concentrations ranging from 1660.0 ± 639.1 n/m^3 to 8925 ± 1591 n/m^3 in the studied areas (Table S2). Microplastic concentrations showed a high heterogeneity, with variation coefficients of higher than 15% in most of the studied waters. Many factors can contribute to the heterogeneous distribution patterns of microplastics in the aquatic environments, such as plastic properties, hydrological situations, surroundings and meteorological conditions (Ballent et al., 2012; Kukulka et al., 2012; Thiel et al., 2003). Pollution status of floating microplastics in water may be better reflected by investigating larger surface areas. In this study, numerous sampling points were equally distributed in each sampling site, largely enhancing the representativeness of sampling. The 50-μm steel sieve used here made it possible for investigation of smaller plastic particles, which are an important part of microplastics. These tiny particles can be easily missed by samplers with a larger mesh size, such as the manta trawl. However, a limited sample value (20 L) could also contributed to the disparity of microplastic densities among sampling sites (Zhao et al., 2015).

The distribution profiles of microplastics in lakes generally presented a decreasing concentration trend away from the urban center (Fig. 1). Almost all the lakes with microplastic concentrations higher than 5000 n/m^3 are located inside or near the area surrounded by the City Third Ring Road. Especially, Bei Lake was found with the highest microplastic concentration at 8925 ± 1591 n/m^3, followed by Huanzi Lake at 8550 ± 989.9 n/m^3. These two lakes are located in the very center of Wuhan City and surrounded by populous residential areas (Wuhan Bureau of Statistics, 2015). Breakdown of larger plastic items in household waste might be the main source of microplastics in the two lakes. In addition, the smaller lake size can concentrate suspended particles, which is another contributor to the high levels of microplastics in surface water (Free et al., 2014). Same interpretation can apply to the heavy microplastic pollution in Tazi Lake (6175 ± 1308.2 n/m^3). Sha

Lake, Nantaizi Lake, and Nan Lake were also detected with high microplastic concentrations at 6390 ± 862.7, 6162.5 ± 537.5, and 5745 ± 901.6 n/m^3, respectively. Effluent from the nearby wastewater treatment plants (WWTP) might be an important point source of microplastics in these lakes (Browne et al., 2011; Wagner et al., 2014). Although adjacent to WWTP, Tangxun Lake, Huangjia Lake, and Jinyin Lake was not found to have such high microplastic levels as the three above mentioned lakes, which was probably due to their larger water areas and less populous surroundings. As a famous tourist resort in Wuhan, Dong Lake was found with microplastic concentration at 5914 ± 1580.7 n/m^3, while degradation and fragmentation of plastic litter discarded by tourists and nearby residents could contribute to the high presence of microplastics in the water body (Eerkes-Medrano et al., 2015). Lakes with lower microplastic abundance, such as Hou Lake, Wu Lake, Yandong Lake, Yanxi Lake, and Zhushan Lake, are all located far from the urban center, with Wu Lake having the least microplastic concentration at 1660.0 ± 639.1 n/m^3. This indicated spatial associations between pollution levels of microplastics in waters and proximity to urban centers (Moore et al., 2011). Pearson correlation analysis was employed to investigate the relationship between microplastic concentrations and distance of corresponding lakes from Wuhan municipal government, the hypothesized urban center. The result showed microplastic concentrations of lakes were negatively correlated with the distance of lakes from the urban center ($p < 0.001$) (Fig. 2). Usually marked by higher population densities and anthropogenic activities, urban centers are also producers of considerable amount of municipal waste and effluent which may contain large quantities of plastics (Browne et al., 2011; Fendall and Sewell, 2009; Hidalgo-Ruz et al., 2012). These plastic items can reach the aquatic system directly through poorly managed discharge or indirectly via surface runoff (Wagner et al., 2014), thus contributing to the higher microplastic levels in urban center lakes than those in outer ones. Similar results were also reported in other studies. In surface water of the Laurentian Great Lakes, Northern America, Lake Erie, the most populated lake, was detected with the highest microplastic concentration (Eriksen et al., 2013). In Lake Hovsgol, Mongolia, microplastic densities decreased with distance away from the most populated southwestern shore (Free et al., 2014). Hence, population densities and anthropogenic activities are suggested to be important factors influencing the abundance of microplastics in aquatic environments.

The Yangtze River and its largest tributary - the Hanjiang River also suffered microplastic pollution (Table. S2). Microplastic concentrations in the urban sections of the Yangtze River and the Hanjiang River were 2516.7 ± 911.7 n/m^3 and 2933 ± 305.5 n/m^3, respectively. Potential origins of these plastics can be diverse, such as delivery from upper reaches, input from banks, shipping, tourism, and fishing activities (Eerkes-Medrano et al., 2015; Zhang et al., 2015). Compared with the lakes, the two rivers presented a relatively lower plastics level. This may be due to the stronger hydrodynamics in rivers that facilitates the evacuation of contaminants. Whereas in lakes, because of the relatively closed environment, microplastic concentrations would increase constantly by long-term degradation and accumulation (Eerkes-Medrano et al., 2015). Abundance of microplastics in the urban reaches of the Yangtze River and the Hanjiang River in Wuhan was lower than that reported in the Yangtze Estuary (4137.3 ± 2461.5 n/m^3) (Zhao et al., 2014). Three factors can account for this difference. Relative to the 32 μm pore size of sieve used by Zhao et al. (2014), the 50 μm sieve used in this study may underestimate the microplastic concentrations. Besides, emission of waste from residents and industries along the river shores can result in an increase in densities of plastic particles in the lower reaches of the Yangtze River. Moreover, the complex water circulation patterns induced by tidal currents and riverine inflow may cause re-suspending of plastic debris from deeper water column and sediments to upper water, or re-entering from the ebb tides, which thereby contribute to the accumulation of microplastic items in surface water of estuaries (Sadri and Thompson, 2014; Zhao et al., 2015). However, the two rivers in this study presented a higher level of microplastic pollution than those in three Californian rivers (30 to 109 n/m^3), North America (Moore et al., 2011), Danube river (0.3168 n/m^3), Austria (Lechner et al., 2014) and Tamar estuary (0.028 n/m^3), England (Sadri and Thompson, 2014). Generation of comprehensive monitoring data on the abundance of microplastics in inland freshwaters is still needed.

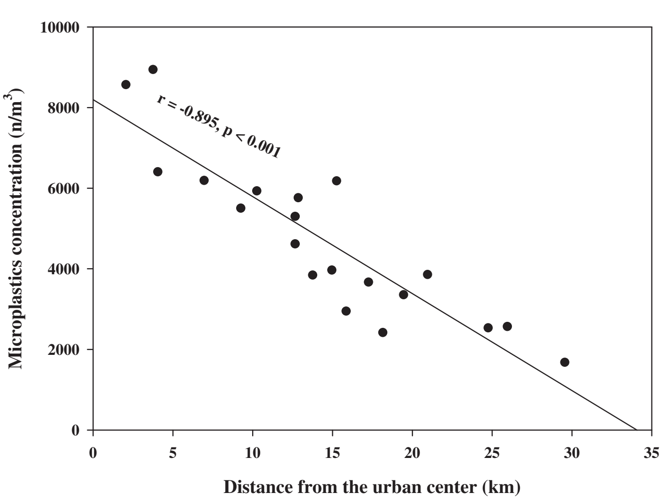

Fig. 2. Pearson correlation between mean concentrations of microplastics and distance of corresponding lakes from the urban center. (The Wuhan Municipal Government was assumed to be the urban center (Fig. 1). Distance from Wuhan Municipal to the center of each lake was measured by Google Earth (http://map.earthol.com/help/)).

3.2. Morphological characteristics of microplastics

Fiber, granule, film, and pellet were commonly detected in urban surface waters of Wuhan. Typical examples of the four types of microplastics under the stereoscopic microscope and SEM were presented in Fig. S1 and Fig. S2, respectively. Fibers were the most frequently detected form of microplastics, accounting for 52.9 to 95.6% of total plastic particles in number in all studied waters (Fig. 3A). These fibers can originate from domestic effluent (Browne et al., 2011), breakdown of fishing nets and lines (Cole et al., 2011), as well as the deposition of airborne matters (Dris et al., 2016). As most of the studied waters are proximal to residential areas (Table S1), household sewage may be an important carrier that conveys fibers to the aquatic system via effluent discharge or surface runoff (Browne et al., 2011; Wagner et al., 2014). Numerous fisheries dotted in the lakes, such as Dong Lake, Wu Lake, Tangxun Lake, Yanxi Lake, Yandong Lake, along with the prevalent fishing activities in lakes and rivers, can also contribute to the high presence of fibers in the these waters (Lin, 2016; Stolte et al., 2015). Furthermore, the fact that colored fibers are the major type of microplastics implies municipal wastewater may be the major source for these fibrous items, for coloration is more likely to be applied in plastic consumer products rather than fishing nets or lines. Although low in proportion, granules, films and pellets were almost ubiquitous in urban surface waters of Wuhan. The small plastic granules are commonly found in many cleaning and cosmetic products or produced by the breakdown of larger degradable plastics (Cole et al., 2011; Fendall and Sewell, 2009). Pellets are typically used in air-blasting or feedstock for plastic production (Eerkes-Medrano et al., 2015). Films may come from fragmentation of larger plastic wrappings and bags (Nor and Obbard, 2014). However, knowledge about accurate determination of microplastic sources and pollution control strategies is still incomplete.

Colored particles were the major microplastic type, accounting for 50.4% to 86.9% of total microplastics in number (Fig. 3B). Plastic plays an important role in sustaining the comfort of people's modern life, while coloration is a common means of improving the market appeal of plastic products (Thetford et al., 2003). Large amount of plastic

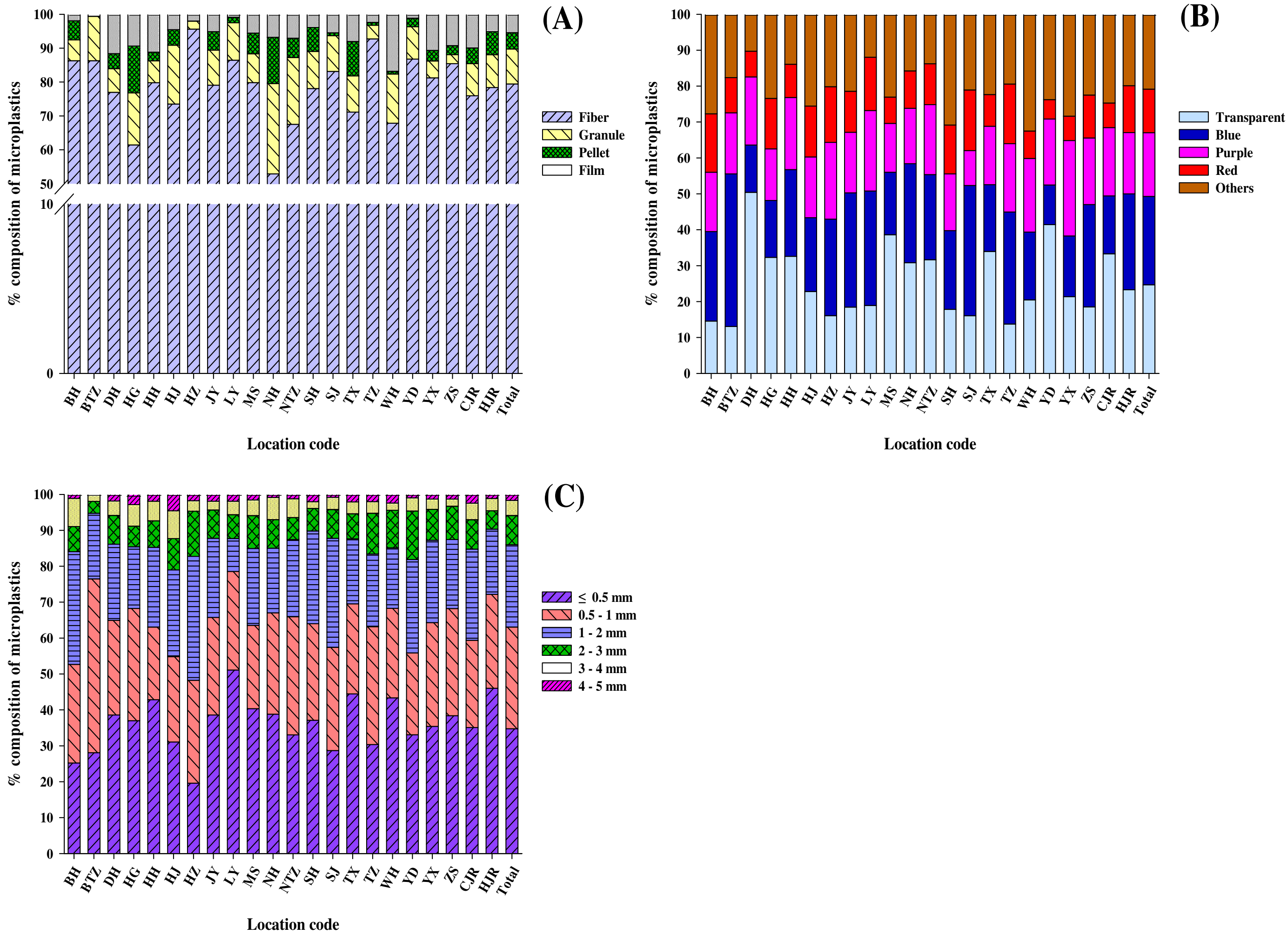

Fig. 3. % composition of microplastics in shape (Fig. 3A), color (Fig. 3B) and size (Fig. 3C) categories.

waste is generated by consumption of these products, which may evolve into microplastics through fragmentation. Breakdown of large plastics can happen on land or after being transported to the waters (Zhang et al., 2015). Clothing and packaging from surrounding residential areas might be potential sources for these colored items in the studied areas. Transparent particles occupied 24.7% of the total microplastics in number, which could be explained by the wide use of transparent plastic materials in fishing nets and lines (Stolte et al., 2015). For example, Dong Lake was detected with the highest proportion (49.6%) of transparent plastics, perhaps greatly due to the numerous fisheries and fishing activities in it. The dominance of colored plastics corresponded to the wide application of coloration in production of plastic consumer goods.

Microplastics with a size of <2 mm accounted for >80% of the plastic particles in number in all sampling sites (Fig. 3C). Similar results were reported in the Yangtze Estuary, China (Zhao et al., 2014), the Laurentian Great Lakes, USA (Eriksen et al., 2013), and Lake Hovsgol, Mongolia (Free et al., 2014). The high proportion of small sized plastic particles was probably due to the fact that large plastic debris could break down into several smaller pieces (Zhang et al., 2015). Given that fibers were of the major type, it could be inferred that most of the detected microplastics were fragmented from larger plastic items (Eerkes-Medrano et al., 2015). Small plastic particles may pose potential threats to the aquatic biota. Owing to their fine sizes as sediments and some plankton, microplastics are considered bio-available to many aquatic organisms through mistaken ingestion and the food-web delivery (Cole et al., 2015; Cole et al., 2011; Wright et al., 2013). However, which characteristics (color, shape, or size) promote the ingestion and what the fate of microplastics is in the biota still need to be further studied.

3.3. Polymer identification of microplastics

The Fourier transform infrared spectroscopy has been widely employed to identify microplastic polymers, for its high reliability in determining the chemical composition of unknown plastic fragments (Hidalgo-Ruz et al., 2012). In this study, a total of forty-four microplastic samples were analyzed by the FTIR (Fig. S3). Eighteen particles were identified as polyethylene terephthalate (PET), thirteen particles were polypropylene (PP), six particles were polyethylene (PE), five particles were polyamide (Nylon), and the other two particles were polystyrene (PS) (Table S3). PET and PP were the main polymer types for the selected microplastics. PET is typically used in the production of textiles, such as clothes, blankets and fleeces. PP, as the most widely produced polymer around the world (PlasticsEurope, 2015), is commonly applied in fishing nets and ropes. PET fibers contained in household effluent and breakdown of PP nets might be important origins for fibrous microplastics in the studied waters. However, it was far-fetched to determinate the origin of plastics by identifying the polymer-types of such a small number of subsamples. Interestingly, PET (1.37 g/cm^3), Nylon (1.15 g/cm^3) and PS (1.05 g/cm^3), whose densities are higher than the freshwater, were detected in the surface waters of Wuhan. More factors other than density can work together to influence the vertical distribution of microplastics in water. Firstly, the higher surface to volume ratio enables these tiny plastic particles to suspend in the water column (Zhao et al., 2015). In addition, physical forces, such as temperature, pressure, wind driven turbulence, from tides or waves, can result in resuspension of the benthic particles (Ballent et al., 2012; Sadri and Thompson, 2014). Moreover, biofouling may change the density of plastic debris, which leads to a more natural drifting or sinking of plastic particles in water (Wang et al., 2016). Therefore, the vertical dispersal of microplastics is driven by a complicated interaction of external factors and properties of the plastics themselves.

4. Conclusions

The urban surface waters in Wuhan were universally contaminated by microplastics. Higher microplastic concentrations in lakes closer to the city center implied an important role of anthropogenic factors in microplastic pollution and distribution. Urban reaches of the Hanjiang River and Yangtze River were found with relatively lower microplastic concentrations than most of the studied lakes. Fibrous, colored and small-sized were the primary characteristics of detected microplastics. Polyethylene terephthalate and polypropylene were the major polymer-types for the selected microplastics. There is still a long way to deeply understand the sources and fate of freshwater microplastics.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2016.09.213.

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