RESEARCH ARTICLE



Occurrence, distribution, and possible sources of microplastics in the surface river water in the Arakawa River watershed

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Abstract

Nowadays, efforts for complementing data concerning microplastics (MPs) in freshwater systems are required as MPs exist in many populated areas. The goal of this study is to investigate the distribution and profiles of riverine MPs along the Arakawa River watershed, which runs through the Tokyo Metropolitan area. The MPs were found in 10 of the 12 sampling sites in the watershed with the mean of 1.8 pieces/m³. Also, the spatial distribution of the MPs displayed the accumulation in the downstream and in the tributary areas with high populations, reflecting the levels of the local anthropogenic activities. In contrast to the heterogeneity of the floating concentrations, polymer type compositions were consistent with the predominance of polyethylene compared with polypropylene and polystyrene. Moreover, the size distributions of the particles were consistent among samples with the predominance of the relatively smaller size fractions. These results suggest that the sources of fragmented plastic debris are likely spread over terrestrial areas and that reducing burden from these land-based MPs is necessary for mitigating MPs pollution in urban aquatic environments.

Keywords Microplastics · Freshwater environments · Non-point sources · Watersheds · Aquatic environments · Polymer · Arakawa River

Introduction

Microplastics (MPs) refer to the plastic debris with a microscopic size of < 5 mm. They include heterogeneous classes of constituting polymers and have different particle sizes, morphological features, and origins. For instance, the polymer particles that are used as ingredients for personal care products are classified as primary MPs (Auta et al. 2017). Another class is the secondary MPs, which are derived from the fragmentation of larger plastic materials and the debris resulting from processes such as UV radiation and mechanical damage (Carr et al. 2016). As the production amounts of plastics have been

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² Graduate School of Science and Engineering, Saitama University, 255 Shimo-Okubo, Sakura-ku, Saitama City 338-8570, Japan increasing, with 359 million tons of plastics produced in 2018 (Plastics Europe 2019), the environmental concerns regarding MPs have significantly increased.

MPs pollution is expected to have a severe negative impact on environments and ecosystems. The uptake possibilities of MPs by organisms have been examined, particularly in aquatic systems (Wang et al. 2020). Watts et al. (2014) showed that the ingestion of MPs occurs via feeding and also via respiration through gills. The potential effects caused by the uptake of MPs include less eating (Cole et al. 2013), abrasion, digestion tracts blockage (Wright et al. 2013), and exposure to pollutants (Mai et al. 2018; Tanaka et al. 2015). As the MPs abundance in the oceans is estimated to increase up to four times by 2060 (Isobe et al. 2019), investigating their occurrence is important to mitigate the environmental impacts.

Plastic emissions via rivers significantly contribute to marine plastic debris (Lebreton et al. 2017). Recently, research studies in freshwater systems were conducted in several regions (Campanale et al. 2020; Cheung et al. 2018; Klein et al. 2015; Lin et al. 2018; Mani et al. 2015). However, the majority of the previous studies have only paid much attention to marine environments (Blettler et al. 2018), so efforts for complementing field data in freshwater systems are required (Horton et al. 2017). MPs

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can be introduced into aquatic environments via both point and non-point sources. However, due to the widespread and complex sources of anthropogenic activities, the pollution characteristics of MPs can be spatiotemporally heterogeneous (Carr et al. 2016; Gies et al. 2018; Wang et al. 2020; Yukioka et al. 2020). The results of the recent studies suggested that freshwater environments surrounded by urbanized areas are hotspots of MPs (Horton et al. 2017; Kataoka et al. 2019; Mani et al. 2015; Sruthy and Ramasamy 2017). The literature also emphasized the need for further studies on populated inland areas to reveal the occurrence of riverine MPs.

The Arakawa River runs through the Greater Tokyo Area. With 2940 km² of catchment areas, this river plays important ecological and hydrological roles in the region. Several tributaries of the river also run in the urban areas of Saitama City (1.3 million people). Moreover, the river is a major drainage leading to Tokyo Bay, in which MPs were detected in various samples (i.e., seawater, sediments, and fish digestion tracts) (Matsuguma et al. 2017; Tanaka and Takada 2016). Tokyo Bay is on the eastern side of the country, and it reaches the Pacific Ocean.

In this study, we aimed at investigating the distribution and profile of riverine MPs along the Arakawa River watershed, including the tributaries. We mainly focused on understanding up to what extent the MPs profiles such as floating concentrations and the compositions of polymer types vary within the watershed. For example, structure, physicochemical properties, and application ways are different among polymers. Comparing size distributions among samples would also be important to understand and identify their sources and entry processes. These processes can lead to the mitigation of plastic pollution in freshwater environments.

Materials and methods

MPs sampling was conducted at 12 sites in the Tokyo Metropolitan area and Saitama Prefecture along the Arakawa River watershed (Fig. 1). Five samples were collected from the upstream (A1) to the downstream sites (A5). A1 is located in the Saitama Prefecture rural area, in which a low anthropogenic level was expected. A5 is located near the central area of Tokyo City and the river mouth. Seven samples were also collected from Saitama City, namely, Ayase River (T1), Toemon River (T2), Shibakawa River (T3 and T4), Kamo River (T5 and T6), and Sasame River (T7). The sites of the tributaries are regularly monitored by the Saitama City Office, and the sampling campaign was conducted from November 2019 to February 2020 during normal weather conditions (Table S1). Several points in the middle stream (A3, T1-T7) and downstream areas (A4-A5) of the Arakawa River basin belong to tidal reach areas in which the sampling was conducted when the downflow was predominant over the tidal reverse flow to minimize the MPs effects in seawater.

To collect floating materials, a nylon planktonet (mesh size: 0.335 mm, mouth opening area: 0.16 m^2) (5502A. RIGO Co., Ltd., Tokyo Japan) was submerged at the sampling sites for 10 min. In the sampling duration, the net was set on the river surface so that half of the net mouth was submerged under the water to collect the materials (Cheung et al. 2018; Matsuguma et al. 2017). The filtered water volume was calculated from the flow velocity, which was measured using calibrated electromagnetic current meters to express the MPs concentrations as particles per cubic meter. After the collection, the net was rinsed on-site to transfer the caught materials to amber glass bottles, and the samples were then transferred to the laboratory. Shortly after, the samples were vacuum filtered using glass fiber filters (GF/A. Whatman). The retained materials on the filters were subjected to a subsequent sorting process in which the materials suspected to be MPs were picked up using a microscopic system with a digital camera (Meiji Techno Co., Ltd., Saitama Japan). The materials were sorted based on their visual features (size, color, and shape) and were then photographed. The maximum axis size was measured to fractionize them according to their sizes (0.5-1 mm, 1-2 mm, 2-3 mm, 3-4 mm, and 4-5 mm). Sizes of less than 0.5 mm were cut off in the present study. The MPs candidates were next subjected to digestion processes of organic materials by adding 5 mL of H_2O_2 solution (30%) (Fujifilm Wako Pure Chemical Industries, Japan) in the test tubes. The samples were set in a thermostatic bath (25 °C) overnight according to Mai et al. (2018). After this treatment, they were again filtered using glass fiber filters, and the retained candidates were individually analyzed using an attenuated total reflectance Fourier transformation infrared spectrometer (AT-FTIR) of FT/IR-6100 (JASCO, Japan). The analysis covered wave numbers ranging 4000-600 cm⁻¹. Background was checked every day. The FTIR analysis was repeated three times for each particle to obtain robust results. The IR spectrum obtained from the samples was checked and compared with the known spectrum from PE, PP, and PS (AS ONE (Osaka, Japan)) to identify polymer types. Although we referred to the hit quality indices (HQI) of the spectrum, the strict thresholds (e.g., > 70%of HIQ) were not defined, as the HQI cannot always be the absolute index for the MPs identification (Renner et al. 2019). We prepared clean blank samples for fields and the laboratory to check contamination from airborne plastics. The results showed no MPs in the blank samples. Statistical analysis using the R software was performed to examine the spatial variations of MPs. The significance level of t test was defined at p < 0.05.

Results

Totally, 221 particles in the samples were identified as MPs. Table 1 shows the results of the MPs in the river (A1–A5) and in its tributaries (T1–T7). MPs were found in most of the river



Fig. 1 Sampling locations in this study. The map was based on "Chiri-in-Chizu" provided by Geospatial Information Authority of Japan (GSI) website (https://www.gsi.go.jp/). The terms A (A1–A5) and T (T1–T7) represent Arakawa River mainstream sites and tributary sites, respectively

water, and the mean MPs concentrations in the river samples were 1.8 pieces/m^3 . We noted that the absence of the MPs in the T6 and A3 was due to the low river velocity, so they were excluded from the calculations. The highest MPs concentrations were found in the tributary sites T5 (4.7 pieces/m³)

followed by T3 (4.5 pieces/m³), and A5 (2.5 particle/m³), respectively. In the tributary sites (T1–T7), MPs were found at relatively higher concentrations compared with the Arakawa main stream site (A1–A5). The mean concentration in the T1–T7 was 2.3 pieces/m³, while that in the A1–A5 was

Sample ID	Conc. (particles/m ³)	Particle number								
		Total particle	PE	PP	PS	0.5– 1 mm	1– 2 mm	2– 3 mm	3– 4 mm	4– 5 mm
T1	1.4	20	18	1	1	9	9	2	0	0
T2	0.8	9	3	4	2	1	6	1	1	0
Т3	4.5	48	19	15	14	13	16	12	3	4
T4	1.8	8	5	1	2	0	3	5	0	0
T5	4.7	26	13	9	4	8	11	5	0	2
T6	ND	ND	0	0	0	0	0	0	0	0
T7	0.6	1	1	0	0	0	1	0	0	0
A1	0.1	1	1	0	0	0	1	0	0	0
A2	0.2	5	0	4	1	1	3	1	0	0
A3	ND	ND	0	0	0	0	0	0	0	0
A4	1.2	26	17	9	0	9	13	2	0	2
A5	2.5	77	50	26	1	20	33	20	3	1

Table 1 Concentration and particle numbers of MPs in each sampling site

PE polyethylene, PP polypropylene, PS polystyrene

Fig. 2 Floating concentrations of MPs along the Arakawa River mainstream (A1–A5) as a function of distances from the mouth river. Data in A3 are plotted as 0 in this figure



1.0 particles/m³. However, there were no significant differences in the MPs concentrations between the main stream and the tributaries (*t* test, p > 0.05). Figure 2 illustrates the MPs concentrations in the main stream area as a function of the distance from the river mouth. The MPs concentrations clearly increased toward the downstream areas. At the upstream site of the Arakawa River watershed (A1), only one particle was found in the sample, and the concentration was low (0.1 particles/m³), but it increased to 2.5 particles/m³ at the downstream area of A5, which is located at the downstream of the central area of Tokyo.

Heterogeneous shapes of MPs, such as fragments, fibers, films, sheets, granules, and flakes, were reported in previous studies (Koelmans et al. 2019). However, in this study, the shape distributions of the MPs were not quantitatively summarized due to the difficulty and ambiguity in distinguishing

the three-dimensional shapes of some MPs (Lin et al. 2018). Nevertheless, most of the MPs were clearly characterized as a fragment type, which is among the secondary microplastics, regardless of the sampling sites. The MPs were categorized based on their size and polymer types. As for the size distribution, many of the MPs were found in smaller size fractions. Figure 3 shows the size distributions of the MPs at each sampling site and all the MPs particles. The 1-2 mm of the MPs particles were predominant, accounting for 43% of the total MPs particles on the particle number basis, followed by 0.5-1 mm (27.6%) and 2-3 mm (21.7%) MPs. In addition, the distribution sizes of the MPs with the predominance of the smaller fractions were similar regardless of the upstream sites, tributary sites, and downstream sites. Of the 10 samples, the 1-2 mm MPs showed the highest contribution at nine sampling sites. The percentages of the smallest particle fraction



Fig. 3 Size distributions of MPs in samples collected from the Arakawa River watershed

(0.5-1 mm) in A5 (26%) were at the same level as those observed in the tributary sites (0–45%). In contrast, the contributions of the large-size MPs were at low levels within the watershed. The 4–5 mm MPs were only found in four sites, and their average percentage was 3.6%.

The polymer type of each MP particle was identified by ATR FTIR, and PE was found as the major polymer in the samples. The contribution of each polymer type (PE, PP, and PS) is shown in Fig. 4. On the particle number basis, 57.5% of the total MPs were identified as PE, followed by PP (31.5%). However, the PS types only accounted for 11.5% of the total MPs particles. Figure 5 describes the polymer-specific size distribution of the MPs in the samples. The PE and PP showed similar distributions with the accumulation in 1-2-mm size fractions, accounting for 48% and 35%, respectively. The contributions of the 0.5-1 mm size of the MPs were also similar in the PE and PP, with 24% and 28%, respectively. As for the PS, although its specific size distribution was insufficiently described due to the lower sample abundances (n = 25), remarkable differences compared to the PE and PP were not observed. Thus, the results show that the detected MPs accumulate in surface water with similar size distributions regardless of the polymer types.

Discussion

This study showed the wide distribution of riverine MPs in the Arakawa River basin. Although MPs concentrations in watersheds have been recently reported (Campanale et al. 2020; Cheung et al. 2018; Lin et al. 2018; Mani et al. 2015), a quantitative comparison of the MPs concentration data with different studies can be inappropriate, especially when

Fig. 4 Polymer profiles of MPs in the samples collected from the Arakawa River watershed. PE Polyethylene, PP polypropylene, PS polystyrene

different strategies are used, such as the mesh size of the net. analytical procedure, target MPs size ranges, and reporting units (i.e., particles/m³ vs particles/m²) (Campanale et al. 2020). Nevertheless, our results are at a comparable level with the reported riverine MPs concentrations by Kataoka (average: 1.6 pieces/ m^3), in which MPs were investigated at 36 river sites in Japan with a similar methodology, so our results reflect the occurrence of riverine MPs in freshwater environments under normal river flow conditions. The predominance of the fragment types of MPs in small size fractions is generally in line with previous reports (Campanale et al. 2020; Kataoka et al. 2019; Lin et al. 2018; Matsuguma et al. 2017; Tang et al. 2018). Moreover, the high MPs concentrations in the urbanized downstream areas are in line with previous studies (Kataoka et al. 2019; Mani et al. 2015). Moreover, our results in tributary sites suggested that riverine MPs concentrations can be high in inland areas. Conversely, the MPs level in A1 was low. The land area of the surrounding Chichibu region in A1 is 892 km², but 84% of the area is covered with forests (Chichibu Agriculture and Forestry Promotion Center 2019). Therefore, anthropogenic activities related to the emission of MPs are considered to be low in such areas.

The PE predominance in the surface water is explainable by its wide usage in various consumer products and due to its low density, which differentiates the distributions in water columns from other MPs with high densities (Matsuguma et al. 2017, Plastics Europe 2019, Tang et al. 2018). It is interesting that riverine MPs mainly accumulated in small size fractions regardless of polymer types (PE, PP, and PS) and sampling sites. This suggests the importance of understanding fragmentation processes and the sources of small size MPs. Moreover, these size distributions of the obtained mm and



■PE ■PP ■PS

Fig. 5 Size distributions of MPs in each polymer types and all particles. Black lines indicate cumulative percentages. PE polyethylene, PP polypropylene, PS polystyrene



Cumulative percentages

100%

90%

80%

70%

60%

50%

40%

30%

20%

10%

0%

100%

90%

80%

70%

60%

50%

40%

30%

20%

10%

0%

3-4 mm

4-5 mm

sub-mm sized MPs appeared to be similar to those reported in East Asian Seas (Isobe et al. 2015), which signifies the impact of MPs in freshwater systems on marine environments (Lebreton et al. 2017). Possible reasons to rationalize the ubiquitous distributions of small size MPs including upstream and tributary sites are that the important sources of MPs exist in terrestrial areas, and fragmented plastics entered into aquatic environments. Although the information on MPs from terrestrial environments are still limited, these MPs can enter rivers through both point and non-point sources (e.g., urban, agriculture, and forest runoff). For instance, roadside dust is considered among the important non-point sources of MPs (Su et al. 2020). According to previous studies, the fragment types of MPs, which originate from the litter and debris of plastic materials and worn rubbers, have widely accumulated in residential roadsides in many regions (Kataoka et al. 2019; Su et al. 2020; Yukioka et al. 2020). Consequently, accumulated MPs in roadside areas and impervious surfaces can easily be flushed into aquatic environments by runoff and wind (Su et al. 2020). Trends in MPs polymer type compositions found in surface road dust samples from Japanese residential areas were similar with our findings (Yukioka et al. 2020). MPs loads to rivers via such mechanisms should be clearly increased and spread in populated and urbanized basin areas. This scenario seems to be important in the Arakawa River basin because MPs were abundantly detected in the populated tributary and downstream sites whereas concentrations in upstream areas were low. Potential point sources might include municipal wastewater drainages to rivers (Grbic et al. 2020). MPs have also been identified in the influents of wastewater treatment plants (WWTPs) at high concentrations (Carr et al. 2016; Koelmans et al. 2019), which would be due to the abrasion and unintentional discharge of various plasticcontaining products. However, contributions from WWTPs are not known in the present time because the efficient removal ratios of MPs during primary and secondary treatment processes have

been reported in developed wastewater systems (Carr et al. 2016; Gies et al. 2018; Talvitie et al. 2017). The percentage of the sewered population reached 93.2% and 100% for Saitama City and central Tokyo, respectively, which are higher than the Japanese average value (79.2%) (Saitama Prefecture 2019; Tokvo Metropolitan Goverment 2020). Nevertheless, large effluent amounts from WWTPs and combined sewage overflows during heavy rain events can also be considerable MPs sources (Dris et al. 2018).

Conclusion

In summary, this study suggests that reducing the inputs of land-based MPs from spread sources over inland areas is necessary for mitigating the MPs pollution in urban aquatic environments. Therefore, further studies should be conducted in inland areas so as to further identify a specific source of MPs. Moreover, the flushing behaviors of those MPs during rain events should also be addressed in future research studies.

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Authors' contributions KS performed the conceptualization, investigation, data curation, and writing of the draft. YY was involved in investigation and data curation. Both the authors have read and approved the final manuscript.

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Availability of data and materials Datasets used during the current study are available from the corresponding author on reasonable request.

Compliance with ethical standards

Ethics approval and consent to participate Not applicable.

Consent for publication No applicable.

Competing interests The authors declare that they have no competing interests.

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