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Assessment of the sources and inflow processes of microplastics in the river environments of Japan☆

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ABSTRACT

The numerical and mass concentrations of microplastics collected at 36 sites on the surfaces of 29 Japanese rivers were mapped and compared with four basin characteristics (basin area, population density, and urban and agricultural ratios) and six water quality parameters (pH, biochemical oxygen demand (BOD), suspended solids (SS), dissolved oxygen (DO), total nitrogen (T-N), and total phosphorus (T-P)) in each river basin. Microplastics were found in 31 of the 36 sites, indicating that some plastics fragment into small pieces before reaching the ocean. The microplastic concentrations are significantly correlated with urbanisation and population density, indicating that the microplastic concentrations in the river depend on human activities in the river basin. Furthermore, we found a significant relationship between the numerical and mass concentrations and BOD, which is an environmental indicator of river pollution. This result demonstrates that microplastic pollution in river environments has progressed more in polluted rivers with poor water quality than in rivers with good water quality, leading to the conclusion that the sources and inflow processes of microplastics in river environments are similar to those of other pollutants. Our findings can help identify potential sources (i.e., point and non-point sources) of fragmented microplastics to improve waste management in Japan and model the transport fluxes of fragmented microplastics in Japanese rivers using water quality parameters and basin characteristics.

1. Introduction

Very small plastics, ranging in size from 0.3 mm to 5 mm in length (Kershaw and Rochman, 2016) and commonly referred to as 'microplastics', have become a major concern in aquatic environments (Andrady, 2011; Cole et al., 2011). Aquatic organisms can easily ingest microplastics because their size is similar to that of the larvae of several organisms, including plankton (Besseling et al., 2014; Boerger et al., 2010; Browne et al., 2008; Cole et al., 2013; Kaposi et al., 2014; Tanaka and Takada, 2016; Thompson, 2004). Since microplastics contain toxic chemicals from various processes, such as production and absorption in marine environments (Koelmans et al., 2013; Mato et al., 2001; Nakashima et al., 2016), aquatic organisms and mammals are exposed to these chemicals via the ingestion of microplastics. Recently, numerous studies have reported that toxic chemicals are transferred into natural organisms via microplastics (Besseling et al., 2017; Browne et al., 2013; Koelmans et al., 2016; Koelmans et al., 2013; Tanaka et al., 2013, 2015). Eventually, the transition of toxic chemicals originating from microplastics into natural organisms results in a chemical hazard; ultimately, toxic chemicals derived from microplastics can reach humans through the food web (Rochman et al., 2015; Thompson et al., 2009; Van Cauwenberghe and Janssen, 2014; Wright and Kelly, 2017).

Microplastic sources are categorised as primary and secondary sources (Andrady, 2011; Cole et al., 2011). A primary source refers to particles that were manufactured with small particle sizes (e.g., cosmetics and skin scrubbers). Secondary sources are microplastics produced by the breakdown or fragmentation of larger plastic items due to exposure to solar ultraviolet radiation, weathering, or gradual loss of weight due to physical damage. Beaches are the most likely source of secondary microplastics in marine environments (Andrady, 2011; Kataoka and Hinata, 2015; Kataoka et al.,

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2013a).

Rivers are major pathways by which waste plastics enter oceans (Carr et al., 2016; Jambeck et al., 2015; Lebreton et al., 2017) and transport primary microplastics released from plastic manufacturing and other human activities. In addition, rivers can become secondary sources of microplastics through two processes. One process is the fragmentation of plastics on land. Plastic litter on land can be efficiently fragmented via processes similar to those on beaches, such as photooxidative degradation and physical damage due to human activity (Andrady, 2011). Plastic fragments generated on land can be released into the marine environment via rainwater drainage to rivers. Alternatively, fragmentation of plastics can occur on riverbanks or flood plains. When the plastic debris released from land are repeatedly washed ashore on riverbanks and flood plains, depending on the magnitude of the flooding, the degradation of the plastics proceeds based on the residence time out of water (Andrady, 2011; Kershaw and Rochman, 2016). In particular, macrophytes that flourish on the flood plain would significantly contribute to the degradative process because plastics can get trapped by them. However, the movement of microplastics from land to the ocean is poorly understood (Eerkes-Medrano et al., 2015).

The scientific concern for microplastics in rivers has rapidly increased, resulting in the publication of many microplastic studies on topics such as monitoring microplastic concentrations (Dris et al., 2015a; Mani et al., 2014; Verster et al., 2017; Yonkos et al., 2014), estimating the outflow flux of microplastics to the oceans (Lebreton et al., 2017; Nizzetto et al., 2016; Schmidt et al., 2017; Siegfried et al., 2017), and understanding microplastic sources (Carr et al., 2016; Leslie et al., 2013; McCormick et al., 2016; Murphy et al., 2016; Ziajahromi et al., 2016) and their impact on freshwater species (Besseling et al., 2017; Dris et al., 2015b; Hoellein et al., 2017; Wagner et al., 2014). For example, the microplastic concentrations along the Rhine River, one of Europe's major rivers that enters the North Sea, significantly increase going downstream, except in the tidal zone of the river (Mani et al., 2014). The microplastic concentrations in four estuarine rivers entering the Chesapeake Bay were found to depend on population density and land use (Yonkos et al., 2014). These studies have clearly demonstrated that microplastics are concentrated near densely populated areas (Mani et al., 2014; Yonkos et al., 2014). Recently, microplastic transport in river catchments has been mathematically modelled using an upgraded catchment hydrology, soil erosion and sediment budget model (Nizzetto et al., 2016). In another approach, the global input of microplastics from rivers to oceans was estimated based on waste management, population density, and hydrological information (Lebreton et al., 2017). These recent studies imply a significant relationship between microplastic concentration and waste management, basin characteristics, and the hydrological regime.

The sources and inflow processes of microplastics in rivers need to be better understood to efficiently prevent the release of microplastics in aquatic environments. For instance, the inflow processes from non-point sources of microplastics, as well as their point sources (Carr et al., 2016; Leslie et al., 2013; McCormick et al., 2016; Murphy et al., 2016; Ziajahromi et al., 2016), should be discussed considering river pollutants. Since microplastics are generated from land according to human activities, similar to river pollutants, a comparison between the microplastic concentration and water quality could be used to identify microplastic sources and inflow processes. In addition, the microplastic concentrations in Japanese rivers have not been reported often. As demonstrated by Isobe et al. (2015) there are significant concentrations of microplastics in the East Asian seas surrounding Japan, suggesting that this area is a microplastics hotspot with concentrations 27 times greater than those in other oceans (Isobe et al., 2015). Hence, estimating the emissions from Japan is important for clarifying the contributions of Japan to the microplastic concentrations in the East Asian seas surrounding Japan. Understanding the relationships between microplastic concentration and basin characteristics and water quality would allow for more accurate estimates of microplastic emissions from Japan to the surrounding seas via rivers.

Here, we describe the spatial distribution of microplastics at 36 sites on the surfaces of 29 rivers in Japan from August 2015 to May 2018 (Table S1). We discuss the sources and the inflow processes through which microplastics reach the rivers from each source by comparing the microplastic concentrations with four basin characteristics (basin area, population density, and two ratios of the urban and agriculture areas to the basin area) and six water quality parameters (pH, biochemical oxygen demand (BOD), suspended solids (SS), dissolved oxygen (DO), total nitrogen (T-N), and total phosphorus (T-P)). Our research is the first report of the microplastic concentrations on Japanese river surfaces, which may be a source of microplastics for the microplastic hotspot in the East Asian seas. In addition, our research uses the relationship between the microplastic concentrations and the basin characteristics in the river basin by surveying more sites than any previous study. Our results can be used as a basis in the future to estimate microplastic transport from inland areas and to clarify the contributions of Japan to the microplastic abundance in the East Asian seas.

2. Materials and methods

2.1. Field survey for collecting microplastics

Microplastics were collected at 36 sites on 29 rivers in Japan (Table S1). The target rivers were selected considering river basins that include various land uses (i.e., urban and agriculture). The collection sites were selected to avoid the tidal reach. In the tidal reach of a river, microplastics can be transported from both the sea and the upstream area due to tidal movements. Sampling within the tidal reach should thus be avoided because of the difficulty in discussing the inflow processes by which microplastics reach each river.

Microplastics were collected on bridges over the rivers according to a survey of water quality (Fig. S1). Although the collection of microplastics has previously been conducted using ships in rivers and oceans (Isobe et al., 2015; Mani et al., 2014; Yonkos et al., 2014), bridge surveys are advantageous because the microplastics can be safely collected regardless of the flow conditions (e.g., flooding). In rivers, the concentrations of microplastics depend strongly on the flow conditions. In fact, Kataoka et al. (2013b) reported that the fluxes of macro-debris with sizes >20 mm in rivers rapidly increase under flood conditions and greatly contribute to the annual fluxes of microplastics in rivers. Nonetheless, we investigated the amount of microplastics under normal flow conditions. It is difficult to collect microplastics at multiple sites simultaneously when collecting samples under flood conditions, and the amount of microplastics depends strongly on the magnitude of the flood. Consequently, it is difficult to compare the amounts of microplastics among multiple sites or to assess the potential for generating microplastics from a river basin (e.g., the major sources of microplastics in the basin). In addition, to compare the amounts of microplastics with the water quality parameters that are regularly measured under normal flow conditions, we also sampled microplastics under normal flow condition.

A small plankton net (No. 5512-C; RIGO Co. Ltd., Japan), originally designed for sampling plankton, fish larvae, and fish eggs near the sea surface, was used for microplastic sampling (Fig. S1). The diameter and length of the net were 30 cm and 75 cm, respectively. The net mesh size was originally selected as 100 μm but was

changed to 335 μm, which has often been used in microplastic surveys in the oceans (Isobe et al., 2015), after the survey on 27 July 2016 because the 100-μm net was easily clogged with suspended materials. The net change did not significantly affect the numerical concentrations evaluated at Noda Bridge on 5 September 2016 (Supplementary Notes). The net was hung from the bridge at the centre of the stream for 5–30 min, according to how clogged the net became. When the water volume that passed through the net (hereinafter, 'filtered water volume') was low, the microplastic sampling was repeated three times at the same location to collect a filtered water volume. Since the net is not fixed in the river water, the upper edge of the net was kept on the water surface by carefully adjusting the length of the rope (Supplementary Video). The filtered water volume was estimated by multiplying the projected area of the mouth of the net ($0.25 \times 0.30 \times 0.30 \times \pi = 0.0707\ m^2$) by the flow velocity inside the net, measured using a flow metre (No. 5571-A, RIGO Co. Ltd., Japan) installed at the mouth of the net or an electromagnetic current metre (AEM1-D; JFE Advantech Co., Ltd., Japan) (Supplementary Notes). Notably, the filtered water volume was potentially overestimated because the net frame was not fully submerged during sampling. When the height of the upper edge of the net frame that emerged above the water surface during the sampling was checked using the video (Supplementary Video), it was found to range from 3 cm to 5 cm. Hence, the filtered water volume in the present study may have varied by 5%–10%.

Supplementary video related to this article can be found at https://doi.org/10.1016/j.envpol.2018.10.111.

2.2. Evaluation of the microplastic concentrations in the laboratory

All of the samples were placed into stainless-steel bottles by rinsing the net during the field surveys. Next, the samples were brought back to the laboratory to separate the microplastics from other suspended materials. River water was collected *in situ* to rinse the net and was filtered using the 100-μm net to prevent contamination from unexpected plastics in the rinse water. In the laboratory, all the samples in the stainless-steel bottles were filtered using the 100-μm net. If large suspended materials were collected, the samples underwent salinity-based density separation using a saturated sodium chloride solution made with tap water (specific gravity (SG) ≈ 1.20) to enhance the efficiency of the microplastic identification analysis (Supplementary Notes). Then, the settled particles were checked visually. This process can be used to extract only microplastics with a lower SG than the solution (i.e., polyethylene (PE; SG = 0.91–0.94), polypropylene (PP; SG = 0.83–0.85), and polystyrene (PS; SG = 1.05); Andrady (2011)). Microplastics with a higher SG (e.g., polyethylene terephthalate (PET; SG = 1.37) and polyvinyl chloride (PVC; SG = 1.38); Andrady (2011)) are not typically recovered using this process.

The filtered samples were transferred to a glass Petri dish with a lid and then were dried at 60 °C for 24 h. Thereafter, particles that appeared to be plastics were picked up with stainless-steel tweezers from the filtered samples by visual observation based on their colour and shape (Hidalgo-Ruz et al., 2012). Each particle was then processed following three steps: First, the size of each particle was measured by analysing a picture of the particle taken by a stereoscopic microscope (SZX7; Olympus Co. Ltd., Japan) installed with a USB camera (HDCE-20C; AS ONE Co. Ltd., Japan) using image-processing software (ImageJ, downloaded from http://imagej.nih.gov). Second, the mass of each particle was measured using an XPR Ultra-Microbalance (XPR2UV, Mettler Toledo Co. Ltd., Japan) with maximum and minimum weights of 2.1 g and 0.1 μg, respectively. Third, the compositions of the particles were identified using Fourier transform infrared spectrophotometry (FTIR; FT-IR alpha, Bruker Optics K.K., Japan or IRAffinity-1S, Shimadzu Co. Ltd., Japan). For each collection site, the number of particles analysed using FTIR (i.e., candidates) is listed in Table S1. The ratio of microplastics to total candidates was 35%. In the FTIR identification, we did not define a threshold for matching the IR spectra of candidates with the reference spectra of pure polymers to avoid incorrectly identifying polymers. This method was used because the spectra of the sampled plastics were usually contaminated by unknown materials, such as vegetation and other particles, that adhered to their surface. Instead, we identified the plastics by visually checking the wavenumber (or wavelength) of the IR spectrum peaks of the sampled plastics. The numerical and mass concentrations were then calculated by dividing the number and mass of the microplastics identified with FTIR by the filtered water volume.

Contamination with unexpected plastics was prevented by the following measures: Plastic utensils, such as beakers, Petri dishes, and sieves, were avoided when possible, and a white robe made of 100% cotton was worn throughout the sorting and processing procedure of identifying plastic particles from the suspended materials. In addition, it is important to recognise blanks during the sorting and processing. To estimate the contamination from the air and tap water, two experiments were conducted. In one experiment, airborne particles were collected by placing glass Petri dishes (ϕ 91 mm) on the tables for sorting and processing for 1 day. In the other experiment, waterborne particles were collected by sampling tap water in a 10-L glass beaker. In both experiments, no plastics were found. Nevertheless, two measures were taken during the sorting and processing to prevent contamination as follows: lids were generally placed on all utensils to prevent air blanks in the laboratory, and the tap water was used only after it was passed through a 75 μm filter.

2.3. Basin characteristics data

We obtained river basin characteristics from the National Land Numerical Information (NLNI) download service (http://nlftp.mlit.go.jp/ksj-e/index.html). The NLNI has various data classified by the designated regional area, coastal zone, nature, land use, national average, facility, census, and hydrology. The vector data for the river basin and land use for every square of a 100-m mesh was downloaded from the NLNI. The basin, urban, and agricultural areas in the upstream region of each microplastic collection site were calculated using these vector data, and then the proportions of the urban and agricultural areas to the basin area (i.e., urban and agricultural ratios, respectively) were computed. In addition, the populations in the upstream region of each microplastic collection site were calculated using 250-m mesh population data downloaded from a portal site for Japanese Government Statistics (e-Stat: http://www.e-stat.go.jp/SG1/estat/eStatTopPortalE.do). The population density was calculated by the ratio of the population in the upstream region to the basin area. QGIS (version 2.14) was used to establish the basin characteristics data.

2.4. Water quality data

The water quality is regularly monitored at a frequency of one to three months in public waters, such as rivers, lakes, groundwater, and coastal seas, by national and local governments based on the Basic Environment Law. In the present study, the four indices of water quality, pH, BOD, SS, and DO, were compared with the numerical and mass concentrations of the microplastics collected at each site. In addition to the four indices, T-N and T-P were compared with the microplastic concentrations because they reflect human activities in a river basin. Here, the annual mean of the water quality data was used because the water quality parameters were not simultaneously measured *in situ* during the

microplastics sampling. It is difficult to relate the microplastic concentration to the water quality because the microplastics and pollutants originate from different human activities, and both the microplastic concentration and water quality parameters have large temporal variabilities. Hence, to remove the temporal variability and characterise the anthropogenic effects in the river basin for comparison with the microplastic concentration, we used the annual means of the water quality parameter in the same year that the microplastics were sampled. Notably, the microplastic concentrations at three sites (Site Nos. 12, 13, and 18; Table S1) were compared with the averages of the water quality parameters during the several years when microplastics were sampled. At Kaihei Bridge (Site No. 17), the annual mean in 2017 was used for the comparison because the annual mean in 2018 has not yet been obtained at present. The microplastic concentrations were also classified based on the class of the nearest water quality station, as shown in the Supplementary Notes. The numbers of Class AA, A, B, C, and D sites were 1, 19, 8, 3, and 3, respectively (Table S1). There was no class E site.

2.5. Statistical analysis

Two types of statistical analyses were applied to investigate the relation of the microplastic concentrations with the basin characteristics (basin area, population density, urban ratio, and agricultural ratio) and water quality parameters (pH, BOD, SS, DO, T-N, and T-P). First, the microplastic concentrations were regressed with the basin characteristics and water quality parameters, and the significance of the regression was tested by a *t*-test at a 95% confidence level. Second, the microplastic concentrations and basin characteristics at all collection sites were classified under the water quality classes (Table S1) and then averaged for the same class. To test whether the averages of the microplastic concentrations and basin characteristics are the same in all water quality classes, the Kruskal-Wallis test, which is a non-parametric method, was applied because the variance of the data under the water quality class was heterogenous at a 95% confidence level, as shown by the Bartlett test (Table S2). The null hypothesis in the Kruskal-Wallis test is that there is no difference in the means. A comparison by the Kruskal-Wallis test can investigate the significant differences between the water quality classes. R (version 3.3.2) was used to carry out the statistical analysis.

3. Results

3.1. Concentrations of microplastics in Japanese rivers

Three polymer materials with lower SG (i.e., PE, PP, and PS) were found in the suspended materials from Japanese rivers collected with a net. As with the East Asian seas (Isobe et al., 2015), these materials were predominant among the polymer materials found at many sites, with the exception of five sites (i.e., Site Nos. 2, 16, 26, 27, and 34; Table S1) (80% by number and 72% by mass; Fig. 1). Various other polymer materials (e.g., acryl, nylon, ethylene-vinyl acetate, and polyacetylene) were listed as “others”. Thus, we focused on the three major polymers, and microplastics refer to the three major polymers throughout this paper, unless otherwise noted. In addition, we focused on secondary microplastics (i.e., fragments) because primary microplastics (e.g., resin pellets) were only collected at the Yoshikoshi Bridge (i.e., Site No. 15; Table S1) on 4 December 2015. The concentrations of plastic pieces smaller than 1.0 mm were higher than those of other sizes of plastic pieces in Japanese rivers, corresponding to those collected from the East Asian seas (Supplementary Notes).

The numbers of identified microplastics and mean microplastic concentrations at each site on all collection dates are listed in Table S1. To investigate the seasonal variation of the microplastic concentration under normal flow conditions, microplastics were sampled repeatedly at five collection sites (i.e., Site Nos. 11, 12, 13, 15, and 18; Table S1). For these sites, the standard deviations are presented in Table S1. As seen from Fig. 2, both the numerical and mass concentrations of the three major polymer materials vary considerably among the sites. The averages of these concentrations are 1.6 pieces m^{-3} and 0.44 mg m^{-3}, respectively; these values are smaller than their standard deviations (2.3 pieces m^{-3} and 0.77 mgm^{-3}, respectively; Table 1), which indicates that a large spatiotemporal variability exists in the microplastic concentrations. The average and median numerical concentrations in the Japanese rivers are on the same order of magnitude as those in the East Asian seas, while the variance of the former was smaller than that of the latter by one order of magnitude (Table 1). The maximum numerical (mass) concentration of 12 pieces m^{-3} (3.2 mgm^{-3}) was recorded at Kachi Bridge (Site No. 11) (Kisaki Bridge (Site No. 12)). Note that the collection site that recorded the maximum numerical concentration is different from the one with the maximum mass concentration because the mass concentration is dependent on the size and polymer types of the collected plastic particles. Overall, the numerical concentration tends to be high in the Kanto region, which is close to metropolitan areas (Fig. 2c), and high concentrations of microplastics can also be found in the Kyushu region (Fig. 2d). This spatial distribution of microplastics can be seen on the map of mass concentration (Fig. S2).

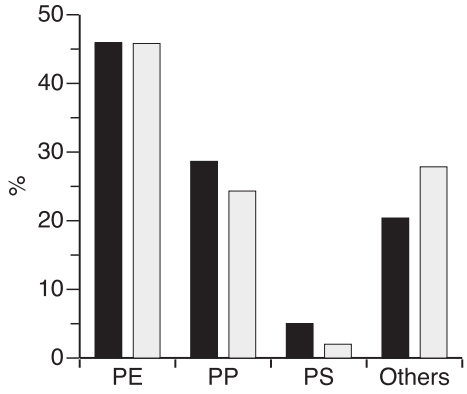

Fig. 1. Number (black) and mass (grey) proportions of all polymers collected in all survey rivers.

3.2. Relationship with basin characteristic data

To discuss the sources and inflow processes through which microplastics reach rivers, both the numerical and mass concentrations were compared with the basin area, population density, urban ratio, and agricultural ratio in the upstream region from the collection sites (Fig. 3a–d). Here, the urban (agricultural) ratio is defined as the proportion of urban (agriculture) area in the basin area. Both the numerical and mass concentrations were significantly related to the population density and urban ratio at the 95% confidence level (Fig. 3b–c; Table S3). The positive slopes of the regression lines for the population density and urban ratio suggest that microplastics are abundant both in count and mass in river basins where the population density and urban ratio are high. A significant relationship between the microplastic concentrations and the population density within the range of 10^1 to 10^2 persons km^{-2} has also been determined for four estuarine rivers that flow into the Chesapeake Bay (Yonkos et al., 2014). On the other hand,

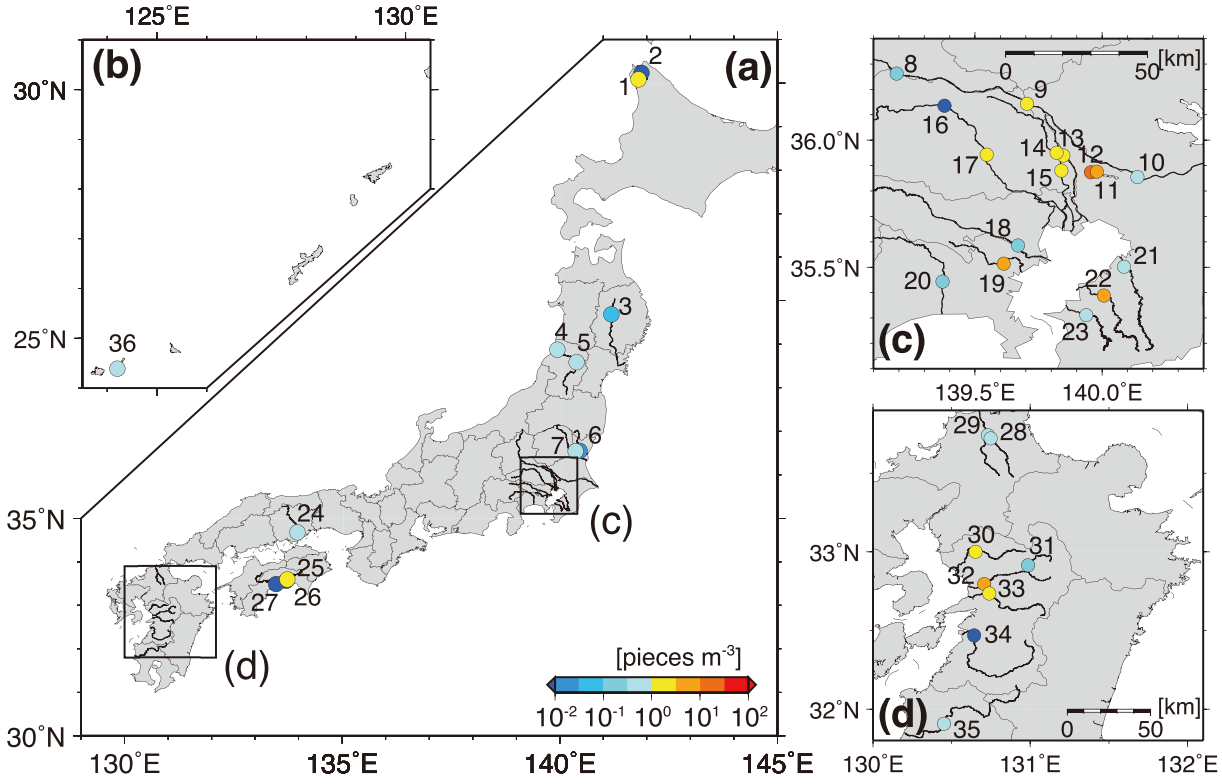

Fig. 2. Maps of the mean numerical concentrations of the three major polymers on all sampling days at each site. Panels (c)–(d) are enlarged maps of the areas shown in squares in panel (a). The number around each circle indicates the survey site number shown in Table S1. The bold and thin lines denote the river streams and prefectural boundaries, respectively. The colours of the circles show the concentration according to the scale in the bottom-right of panel (a). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Table 1
Statistics of microplastic concentrations in all sampled rivers and the East Asian seas.

Statistics	Numerical conc. [pcs m^{-3}]	Mass conc. [mg m^{-3}]	Numerical conc. in the East Asian seas [pcs m^{-3}][a]
Maximum	1.2×10^{1}	3.2×10^{0}	4.91×10^{2}
Minimum	0.0	0.0	3.00×10^{-2}
Median	7.9×10^{-1}	4.9×10^{-2}	7.40×10^{-1}
Average	1.6×10^{0}	4.4×10^{-1}	3.74×10^{0}
Standard deviation	2.3×10^{0}	7.7×10^{-1}	1.04×10^{1}

[a] Isobe et al., 2015.

we sampled microplastics in rivers where the population in the basin is more dense than that studied by Yonkos et al. (2014) (i.e., the range of the population density was between 10^{1} and 10^{3} persons km^{-2}; Table S1). Hence, our results offer more evidence that microplastic concentrations strongly depend on urbanisation and the population density.

3.3. Relationship with water quality data

Both microplastic concentrations were compared with the pH, BOD, SS, DO, T-N, and T-P (as water quality parameters) at 34 collection sites (Fig. 3e–j). Note that the water quality parameters were not obtained at two sites (i.e., Site Nos. 1 and 2; Table S1), and T-N and T-P were not monitored at two sites (i.e., Site Nos. 25 and 36; Table S1). Both the numerical and mass concentrations were significantly related to the BOD, as well as the DO, T-N, and T-P (Fig. 3f, h-j; Table S3). The microplastic concentrations have a positive relationship with BOD, which is interesting because BOD is an environmental indicator of river pollution, while they have a negative relationship with DO.

However, the linear relationship with the water quality parameters may be questionable because of the temporal variability of the microplastic concentrations. Thus, to justify the significant relationship of the microplastic concentrations with the water quality parameters, we analyse the averaged microplastic concentration for each water class by applying the Kruskal-Wallis test. Microplastic concentrations tended to be relatively high in more polluted water (Fig. 4). The Kruskal-Wallis test indicates a statistically significant difference in the mean numerical and mass concentrations among the water quality classes (Table S2). On the other hand, the higher the mean population density and mean urban ratio are, the more polluted the riverine environment tends to be (Fig. S3). These significant differences demonstrate that microplastics are abundant on the surface of highly polluted rivers that flow through river basins with high population densities.

4. Discussion and conclusions

Our findings clearly demonstrate that microplastics already exist in river environments in Japan, indicating that a certain

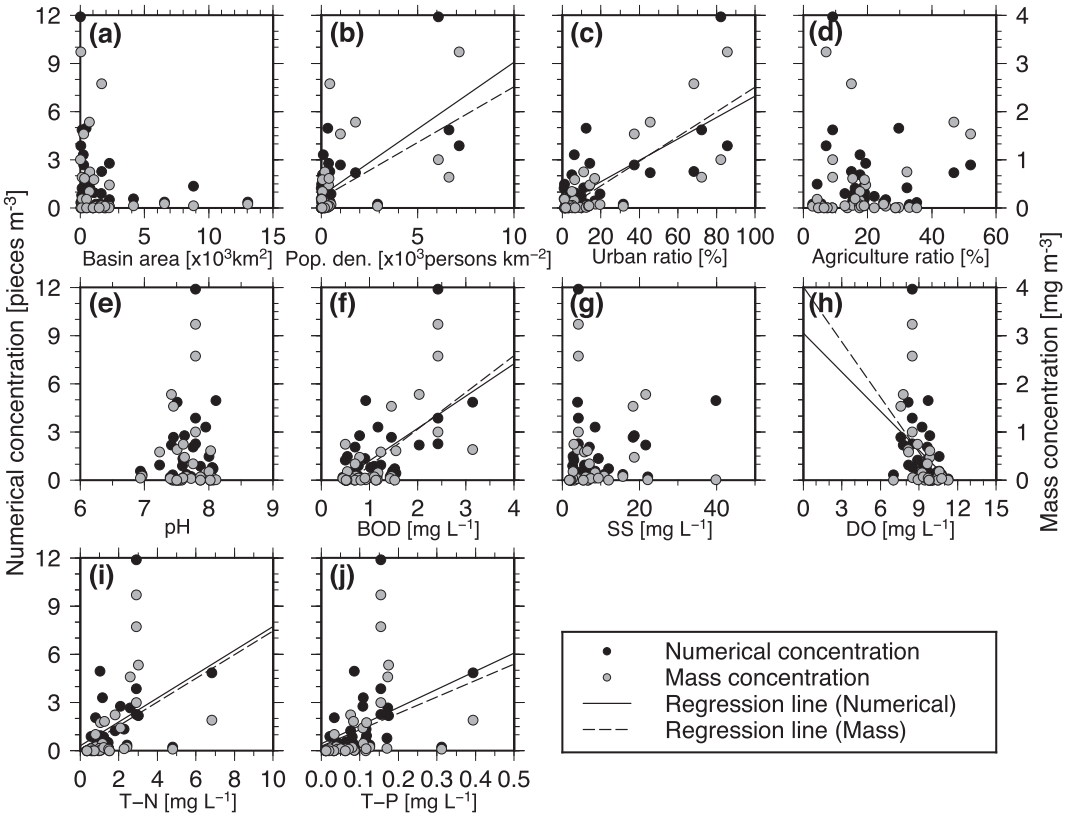

Fig. 3. Correlation of the numerical and mass concentrations with four basin characteristics and six water quality parameters: (a) basin area, (b) population density, (c) urban ratio, (d) agricultural ratio, (e) pH, (f) biochemical oxygen demand (BOD), (g) suspended solids (SS), (h) dissolved oxygen (DO), (i) total nitrogen (T-N) and (j) total phosphorus (T-P). Only significant regression lines are included in this figure, and the statistical significance of the regression lines is shown in Table S3. The legend of the symbols and lines is shown on the right side of panel (j).

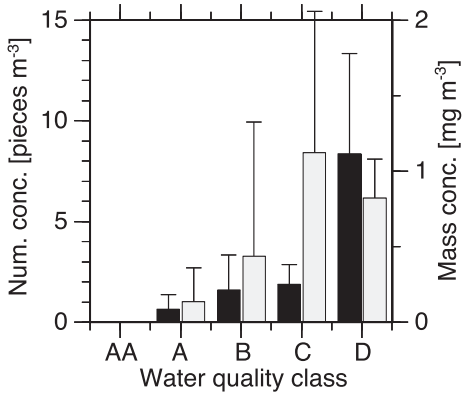

Fig. 4. Mean numerical and mass concentrations of each water quality class. The error bars are the standard deviation of each class. The statistical significance of the differences among water quality classes is shown in Table S2.

amount of plastics fragment into small pieces before entering the ocean. In particular, the pollution of river environments by microplastics has progressed the most in polluted rivers with poor water quality (Fig. 4) due to the high pollution density and urbanisation (Fig. S3). It is interesting that the microplastic concentrations have increased significantly and have a significant relationship with the BOD, DO, T-N, and T-P (Fig. 3) as the water quality decreases. The large spatiotemporal variation in the microplastic concentrations is due to the seasonal variation that is dependent on the hysteresis of flooding and land use in a river basin (Borah and Bera, 2004). The mean numerical and mass concentrations at Noda Bridge over the Edo River from April to September (3.8 pieces m^{-3} and 0.62 mg m^{-3}, respectively) were 3.5 and 2.7 times higher, respectively, than those from October to March (1.1 pieces m^{-3} and 0.23 mg m^{-3}, respectively). This result indicates that there may be seasonal variations in the microplastic concentrations.

Based on these results, the question that arises naturally is why the microplastic concentrations are relatively high in polluted river basins (Fig. 4). One possible reason is that the sources and inflow processes of microplastics are similar to those of pollutants. In general, pollutants are classified as having 'point' or 'non-point' sources. Point sources include domestic outputs, livestock and manufacturing industries, commerce, and wastewater treatment plants (WWTPs), and non-point sources include forest, urban, and agricultural areas. For instance, pollutant loads from domestic wastewater (or urban areas) corresponding to a point source (or a non-point source) are evaluated by their generation rates per unit person (or per unit urban area) (Borah and Bera, 2004). Hence, the pollutant loads from these point and non-point sources increase according to the population and urbanisation. The significant relationships between the microplastic concentrations and the population density and urban ratio (Fig. 3b–c) suggest that the microplastics floating on river surfaces could be generated on land and reach public water through pathways similar to those of pollutants.

Another question based on the results of this research considers where the small plastic fragments that float on river surfaces come from; in other words, what are the sources of secondary microplastics? Domestic wastewater (a point source) drains directly into

public water without passing through WWTPs when there is no sewer system. When a sewer system is developed, domestic wastewater drains into public water after most of the debris is removed at a WWTP (Dris et al., 2015a). Livestock and industrial wastewater also drain into rivers after most of the debris is removed at private WWTPs. In this case, the generation of microplastics in public water would depend on the debris filter size in the WWTPs. On the other hand, some microplastics floating on the river surface may originate from non-point sources. For example, in urban areas, this may include physical damage from human activities such as plastic that is broken into fragments from being crushed by vehicles. Microplastics were found to be abundant in the soil under roadside trees in Kawasaki, one of the largest cities in Japan that contributes to the high population density in the basin of the Tama River (Fig. S4). Microplastics from urban areas are mostly transported via the rainwater in roadside gutters. Open landfill sites near rivers could also be a candidate source of plastic fragments; for example, the plastics littered on the landfill site may be fragmented and then released into the river due to rainfall and/or strong wind. Riverbanks and floodplains could be other sources; for example, large quantities of plastic debris including microplastics have accumulated on river walls surrounding a tidal flat, such as the river wall near the mouth of the Ara River (Fig. S4). Plastic debris deposited onto riverbanks would not be flushed downstream unless the water level reaches the debris. Similar to the residence time of debris on beaches (Kataoka et al., 2013a, 2015), the residence time of debris at non-point sources is an important parameter that influences the generation of plastic fragments in a river basin.

Finally, the inflow processes of microplastics into rivers—or how they reach rivers from their sources—also need to be considered. These inflow processes depend on the development of sewer systems in a given river basin. At the end of March 2017, 78.3% of the population of Japan had access to sewers (http://www.jswa.jp/rate/); therefore, 21.7% of the population did not use sewer systems. In areas with undeveloped sewer systems, domestic wastewater is released into public water via a septic tank, which is a point source. Other areas use either combined or separate sewer systems. In a combined sewer system, a single sewer transports both rainwater and sewage (i.e., combined wastewater) to a WWTP (Dris et al., 2015a). Relatively large pieces of debris would be filtered out, but a portion of the small debris, such as microplastics, would not be filtered out. Even if 83–95% of the microplastics are removed by WWTPs (Dris et al., 2015a), 5–17% would still be released. Furthermore, when the rainwater discharge passing through a combined sewer exceeds a level three times that of the wastewater discharge on a day with good weather conditions, the combined wastewater with microplastics is released directly into rivers as 'combined sewer overflow'. On the other hand, a separate sewer system transports rainwater and sewage to WWTPs in individual sewers. Only sewage is treated at the WWTPs, regardless of the weather conditions, and rainwater is released directly into the rivers or ocean after the large debris is filtered out. Small plastic fragments generated in urban areas (e.g., microplastics from sediment on the roadside in Kawasaki; Fig. S4) could therefore be released into the public water with rainwater.

In summary, microplastics are mainly released into rivers from the following sources: homes that are not connected to sewer systems (i.e., point sources), combined sewer overflow (i.e., point and non-point sources) during heavy rainfall events, and rainwater from separate sewer systems (i.e., non-point sources). The significant relationships of the microplastic concentration with BOD, DO, T-N, and T-P (Fig. 3f, and h-j) indicate that areas in which there are no sewer systems are important sources of microplastics collected under normal river conditions. Furthermore, microplastic concentrations tend to increase when there is a large difference in water levels, implying that massive quantities of microplastics are released from non-point sources during flood events due to heavy rainfall in the river basin (Fig. S5). In the future, we will investigate the relationship between the microplastic concentration and river discharge by sampling microplastics under flood conditions.

Our final goal is to evaluate the outflow of microplastics from inland Japan into the surrounding seas. Evaluating the outflow of microplastics is essential to reducing the generation of microplastics and mitigating the adverse impacts of microplastics on river and marine environments. Considering the above assumptions for the sources and inflow processes of microplastics in river environments, several issues arise. First, we need to regularly collect data on the microplastic concentrations, as well as water quality, in various Japanese rivers. The additional collection of microplastic concentration data would enable us to more accurately evaluate the outflow than that estimated on a global scale (Lebreton et al., 2017). To efficiently collect microplastic concentration data, we need to select effective monitoring sites in river basins. Our results suggest that the microplastic concentrations should be preferentially monitored at stations with poor water quality. The strategic monitoring of the microplastics would enable us to more efficiently address this serious environmental problem in river basins, as well as coastal regions and to detect areas with high microplastic concentrations in river basins by sampling microplastics from the upper reaches to lower reaches along a river.

Second, we need to understand the spatiotemporal behaviours of microplastics in a river. The significant difference in microplastic concentrations according to the water quality classes (Fig. 4) provides a guideline for surveys to understand the microplastic behaviour. In particular, the responses of microplastics at non-point sources are similar to those of pollutants under flood conditions. Flood conditions significantly increase the pollutant flux in rivers because pollutants instantaneously flow out from non-point sources (Borah and Bera, 2004; Kataoka et al., 2013b; Nizzetto et al., 2016; Schmidt et al., 2017). Hence, understanding the fluxes of microplastics in rivers under flood conditions is essential. Additionally, to evaluate the outflow of microplastics under flood conditions, hysteresis must be considered, i.e., when and at what concentration have microplastics been flushed out due to floods in the past? In particular, massive quantities of microplastics will be transported in the first flush in flood events. In addition, we need to understand the spatial distribution of microplastics in the cross-section of rivers. In the present study, we measured the microplastic concentration on the surfaces of rivers at one or a few points in a transverse direction. In the transport process by which microplastics flow down rivers, they may be subjected to turbulence due to horizontal and vertical mixing. In the future, the horizontal and vertical distributions of microplastics in rivers should be investigated.

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Appendix A. Supplementary data

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