

Microplastic Emission Dynamics in an Urban River under Rainfall Events

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Abstract—Urban rivers are key pathways for land-sourced microplastics (MPs) to the oceans. However, their real-time emission dynamics during rainfall events remain poorly understood. This study presents high-frequency monitoring of MPs in an urban river in Japan over three rainfall events (light, moderate, and heavy). The event mean concentrations (EMCs) of MPs were influenced by the number of antecedent dry days, while total loads linked with the amount of rainfall. During moderate and heavy rainfall events, microplastic (MP) loads increased by up to 100 times and event mean concentrations (EMCs) by up to 350 times, compared to dry weather. MPs smaller than 2000 μm displayed first-flush effect during light and moderate rainfall, with over 50% of their total mass released within the initial 20–40% of runoff flow. The smallest particles (10–40 μm) were mobilized rapidly even at low rainfall intensities, whereas larger MPs (>2000 μm) were discharged around or after the peak rainfall. The polymer composition of riverine MPs was significantly impacted by rain, with polymer characteristics suggesting a likely origin from local road dust. The levels of MPs and total suspended solids (TSS) in river water during rainfall were well-correlated, suggesting the possibility to use TSS measurements as a useful indicator for predicting MP levels. Finally, the findings highlighted the importance of targeting moderate to heavy rainfall events and intercepting stormwater first flush for effective control of MP pollution in urban rivers.

I. INTRODUCTION

The rapid growth in plastic production and poor waste management have intensified global microplastic (MP) pollution, with rivers acting as key conduits transferring microplastics (MPs, 1 μm – 5 mm in size) from land to oceans [1]. Urban rivers receive MPs from both point and non-point sources, among which wastewater treatment plants have been extensively studied as point sources of MP pollution [2]. However, rainfall-induced surface runoff, a major non-point source, has been poorly investigated to date despite the elevated MP concentrations (as high as 8580 items/L) reported in surface runoff [3]. Numerous studies have linked rainfall events, storms, and floods to sharp increases in MP fluxes in freshwater and marine systems, and wet seasons have been found to contribute more to annual MP emissions than dry periods [4,5]. Additionally, strong correlations have been confirmed

between rainfall characteristics and MP abundances in freshwater sources [6,7].

Despite these findings, most studies have been conducted in lentic systems or with limited temporal resolution, providing little insight into flushing effects and real-time mobilization of MPs in rivers during rainfall onset. Moreover, the influence of particle size of MPs on wash-off processes remains poorly understood. Addressing these gaps requires high-frequency sampling of MPs during rainfall events to better capture the characteristics of MP emissions via rivers. Ultimately, this study aims to examine how rainfall events influence the dynamic emissions of different size classes of MPs, with a particular focus on determining whether such events trigger a first flush effect in riverine MP discharges.

II. MATERIALS AND METHODS

A. Study area

The study was conducted in an urban sub-catchment of the Kamo River Basin in Kyoto City, Japan. Sampling took place at the Mikage Bridge, located in the downstream section of the Takano River (Fig. 1). The typical baseflow discharge at the sampling point is 4.75 m³/s [8].

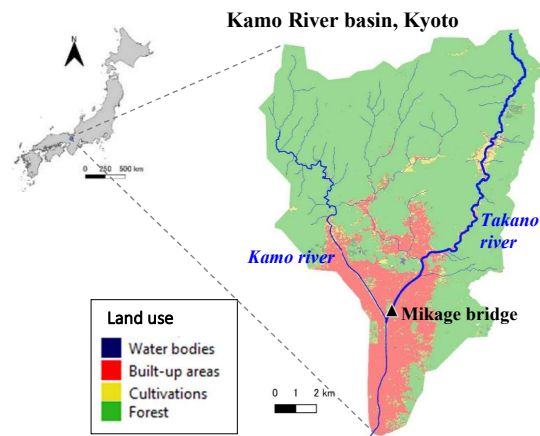


Figure 1. Study area and sampling location

With a population density of 1782 persons/km² [9], the study area is highly urbanized, contributing to

increased human activities around the river. The area is served by a separate sewer system with 99.5% coverage, ensuring wastewater is directed to the municipal treatment plants while stormwater runoff is discharged directly into the river via stormwater drains.

B. Sampling

Three rainfall events (Events 1, 2 and 3) with varying characteristics (refer Table 1) were monitored between October 2021 and April 2022. Depending on the maximum rainfall intensity, Events 1, 2 and 3 were classified as light (<2.5 mm/h), moderate (2.5–7.6 mm/h) and heavy (>7.6 mm/h) rainfall events, respectively [10]. Sampling began before the onset of rainfall and continued until the river returned to the steady-state, following the event. For baseline comparison, two samples were collected during dry weather in August and October 2021.

TABLE I. CHARACTERISTICS OF RAINFALL EVENTS.

Rainfall characteristics	Event 1 (Light)	Event 2 (Moderate)	Event 3 (Heavy)
Date and time of the event	2021/10/16 04:00–09:00	2021/11/09 01:00–08:00	2022/04/29 10:00–17:00
Total rainfall	6 mm	24 mm	52 mm
Duration of rainfall (hours)	5 h	7 h	7 h
Max. rainfall intensity	2.0 mm/h	7.5 mm/h	17.4 mm/h
Total river flow	193,600 m ³	404,900 m ³	1,052,500 m ³
Number of antecedent dry days (ADDs)	3 days	14 days	2 days

MPs were categorized as small (SMPs, 10–300 µm) and large (LMPs, 300–5000 µm), and were collected using in-situ pump filtration through 10 µm and 300 µm plankton nets, respectively. Samples were carefully transferred to re-sealable bags and transferred to the laboratory for analysis. The hourly precipitation, river water level, and in-situ water quality were also recorded. Additionally, 2 L of river water was collected for the analysis of suspended solids [11].

C. Extraction and analysis

Extraction and analysis of MPs followed the established protocols from Rodrigues et al. (2018) [12]. Samples from plankton nets were transferred to pre-rinsed glass bottles (1 L) using ~200 mL of 30% H₂O₂. Organic matter was removed via wet peroxidation by adding 0.05 M FeSO₄ to the sample in H₂O₂ and oven-drying overnight at 55 °C. Subsequently, density separation with 5.3 M NaI (1.53 g/cm³) was used to isolate MPs from sediments. LMPs were extracted by filtering the supernatant from density separation, through 10 µm nylon nets, and the recovered LMPs were stored in clean Petri dishes. SMPs were isolated by repeated centrifugation and filtration of the supernatant through a 0.45 µm polycarbonate membrane. The filter residues were suspended in 50–100 mL of ethanol (95.1–96.9 vol%) until analysis.

LMPs were characterized for size, shape, and color using a stereoscopic microscope (SMZ-161, Shimadzu), and polymer types were identified with

ATR-FTIR spectroscopy (Cary 630, Agilent Technologies). For SMPs, a 5 mL aliquot of samples stored in ethanol were filtered through 0.2 µm alumina filters (Anodisc, Whatman™) and the particles on filter were analyzed using µ-FTIR (Cary 600 Series). MPs were morphologically classified as fragments, films, or spheres (fibers were excluded).

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

To minimize contamination, all equipment and containers were pre-rinsed with filtered water and covered with aluminium foil when not in use. Recovery tests and procedural blanks were included to detect any potential contamination. All reagents used were of analytical grade. During sample processing, workspaces were regularly cleaned, and cotton lab coats and nitrile gloves were worn to reduce fiber shedding.

E. Methods of calculations

Concentrations and loads: The concentrations of MPs were expressed as the number of items per unit volume or mass per unit volume of water. The mass of the particles was calculated by multiplying the polymer density by the particle volume [4,13].

The load of MPs was calculated using equation (1):

$$\text{Load} = CQ\Delta t \quad (1)$$

where, "C", "Q" and "Δt" represent the concentration, flow rate, and sampling time interval, respectively.

Event mean concentration (EMC): EMC was employed to express the flow-weighted concentrations of MPs for each rainfall event [14], and calculated with equation (2):

$$\text{EMC} = \frac{\text{Total load per event}}{\text{Total volume per event}} = \frac{\sum_{i=1}^N C_i Q_i \Delta t_i}{\sum_{i=1}^N Q_i \Delta t_i} \quad (2)$$

where, "N" represents the total number of samples, and the definitions of other variables remain the same.

The statistical analysis was conducted using R 4.2.1. All weather information was obtained from the website of the Japan Meteorological Agency [15].

III. RESULTS AND DISCUSSION

A. Effects of rainfall characteristics on MP emissions

As illustrated in Fig.2 the MP emissions in the river during rainfall events closely followed variations in river flow, which peaked with hourly rainfall intensity. Events 1, 2, and 3 yielded nearly 30%, 100%, and 170% flow increments compared to the baseflow. The EMCs of events 1, 2, and 3 were 35,000 items/m³ (3 mg/m³), 929,000 items/m³ (49 mg/m³), and 331,000 items/m³ (33 mg/m³), respectively. The corresponding total loads were 0.5 kg (7 billion items), 19.8 kg (380 billion items), and 35.0 kg (350 billion items) for events 1, 2, and 3, respectively (Fig. 3). Event 1, with the lowest and shortest rainfall, exhibited significantly lower MP concentrations and loads (Kruskal-Wallis test, $p < 0.05$). Events 2 and 3, however, indicated comparable concentrations and loads regardless of the differences in rainfall. Despite the highest total load occurring during

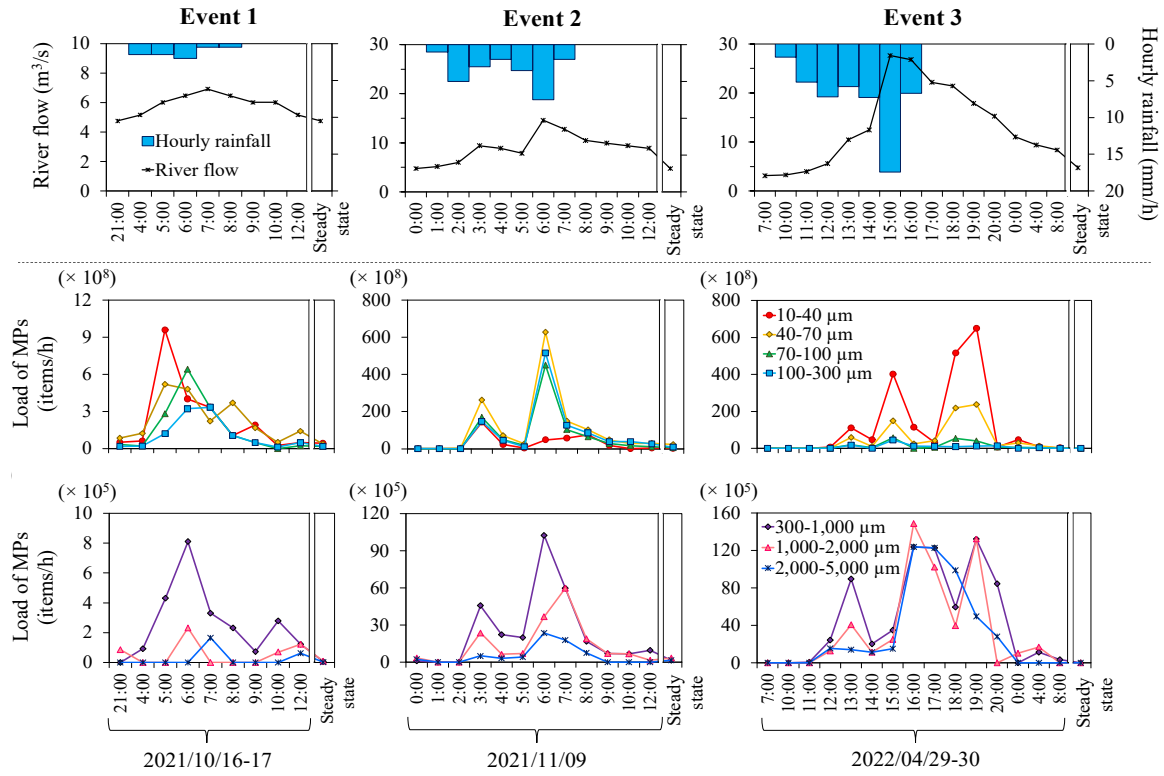


Figure 2. Temporal variations in rainfall, river flow, and the loads of MPs of different size classes

Event 3, Event 2 showed the highest EMC, likely due to its longer ADDs, which allowed for greater surface accumulation of MPs. Overall, the data indicate that the total loads of MPs being transported are linked with the total rainfall (or average rainfall intensity of the event), while the concentrations are related to the number of ADDs (Fig. 3).

The relatively lower concentration observed for Event 3, despite heavy rainfall, can be attributed to the short ADD (2 days) coupled with the high intensity of the proceeding rainfall (34 mm rainfall continued for 17 h; maximum intensity - 24 mm/h) (Fig. 3). It had likely flushed accumulated MPs from the catchment. The lag in peak emissions of MPs after rainfall in Event 3 (Fig. 2) suggests sediment resuspension as a secondary emission pathway, which is consistent with findings from [5] and [16]. This highlights the complex hydrodynamics of river systems [17], where both overland and in-stream processes contribute to MP transport.

Rainfall intensity and duration also impacted the mobilization of MPs significantly. While [18] reported mobilization thresholds at >2.5 mm/h for over 2 hours, this study found that even a lower intensity of 1.5 mm/h continued for 2 hours could transfer nearly 2 billion MPs into the river during a low-rainfall event.

The observations highlight the need to consider the entire hydrological response period of the river, but not just the rainfall duration, when assessing MP emissions. Therefore, high-frequency sampling

during, as well as after rainfall is essential to accurately capture the dynamics of MP emissions.

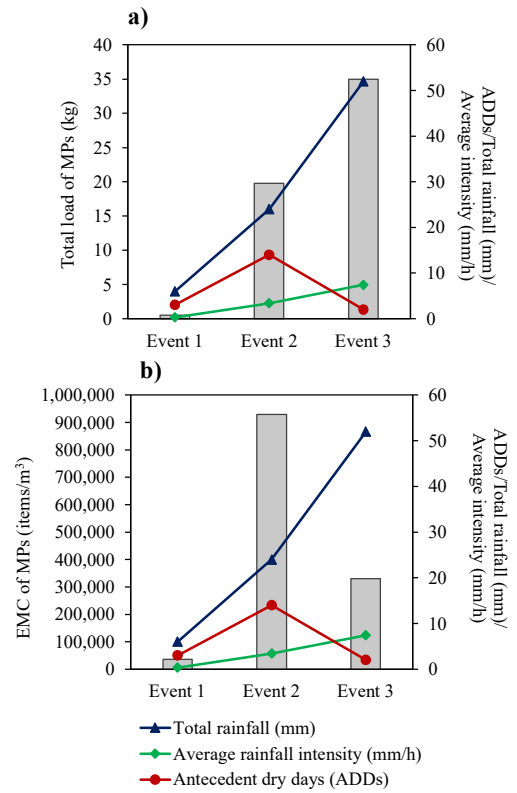


Figure 3. Relationship between a) total loads of MPs and rainfall characteristics; b) event mean concentrations (EMCs) of MPs and rainfall characteristics

B. Effects of particle size on MP emissions

MP emissions during rainfall events showed strong size-dependent patterns, with SMPs consistently dominating the total loads by three to four orders of magnitude, over LMPs (Fig. 2). In light rain (Event 1), the smallest MPs (10–40 μm) were rapidly mobilized, peaking even before the peak intensity of rainfall, indicating potential first flush effects. Medium-sized particles followed, while the largest MPs (>2000 μm) were prominently released after peak rainfall. In moderate rainfall (Event 2), early peaks in small MPs were again evident, but most MPs peaked near the maximum rainfall intensity. Heavy rainfall (Event 3) exhibited more complex behavior: MPs <70 μm showed a tertiary peak after rainfall, and there was a notable increase in larger MP emissions, likely due to sediment scouring and post-rainfall turbulence [5].

The first flush phenomenon often describes if a high proportion of non-point source pollutants is washed off during the initial part of a rainfall event. [19] introduced the dimensionless $M(V)$ curve that is plotted between the normalized cumulative mass of pollutants and the normalized cumulative volume of flow to analyze the first flush of typical non-point source pollutants (Fig. 4). The first flush is said to occur simply when the $M(V)$ curve lies above the identity line. The $M(V)$ curves for the present study revealed first flush behavior in Events 1 and 2 for most size classes, especially for MPs <40 μm , where 40–45% of the mass was discharged within the first 20% of flow. However, during Event 3, only MPs in the 100–300 μm size range exhibited a first flush, while other sizes, especially >2000 μm , showed final flush behavior, with over 50% of emissions occurring during the latter part of the flow [20].

These results suggest that smaller MPs are easily mobilized, particularly in light to moderate events, while larger MPs require greater flow energy, typical of intense or prolonged rainfall. There is scarce evidence on the temporal emission patterns of MPs in runoff based on particle size. However, numerous studies on suspended solids and other inorganic pollutants have shown that the finest particles are eroded first during the initial stage of rainfall events, implying more potential to exhibit first flush effects than larger particles [21].

C. Effect of rainfall on polymer composition of MPs

The polymer composition of MPs in river water was distinctly influenced by rainfall-induced runoff (Fig. 5). Consistent with previous studies, polyethylene (PE, 78%) was the dominant polymer type in river water, under dry weather conditions [22]. Nevertheless, the proportion of PE relatively declined during rain (<47%) due to the influx of other polymers. The fraction of polypropylene (PP, dry: 8%, wet: 9%–16%), which is generally lower than PE, only slightly increased during rainfall events. Additionally, runoff has likely introduced exceptionally high levels of polymethyl methacrylate (PMMA, dry: 6% wet: 17–56%), probably derived from road marking paints [23]. Moderate to heavy rainfall increased the mobility of denser polymers, including polyvinyl chloride (PVC) and polyvinyl alcohol (PVA), which are commonly associated with road markings, plumbing works, and drainage pipe sediments [24].

D. Implications for pollution control

In comparison to dry weather, Events 1, 2, and 3 produced 4-, 110-, and 40-fold higher EMCs and 10-, 350-, and 320-fold higher MP loads. Although wet days represent only ~30% of a typical year in Japan, the present estimations suggest that they account for about 98% of the annual MP mass emissions (96% by number) along the selected river. Light rainfall events, while more frequent, contributed only ~1% of annual loads, indicating that emission control efforts should focus on moderate to heavy rainfall episodes, particularly following longer ADDs. These results suggest that wet periods may have a far greater influence on riverine MP emissions than previously estimated [4].

In addition, the MP concentrations (both by number and mass) were positively correlated with TSS during all three rainfall events ($R^2 = 0.66\text{--}0.90$), suggesting that MPs are closely associated with particulate matters transported by surface runoff, such as road dust. While [25] observed no correlation between TSS and MPs (>300 μm) under dry conditions, this study shows that such relationships become evident during rainfall, when external inputs of MPs from catchment surfaces dominate.

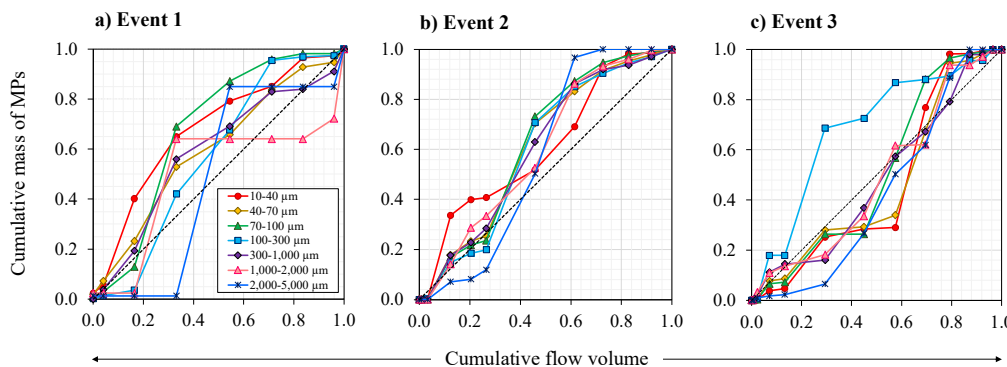


Figure 4. $M(V)$ Curves between cumulative mass emissions of MPs of different size classes with respect to river flow volume

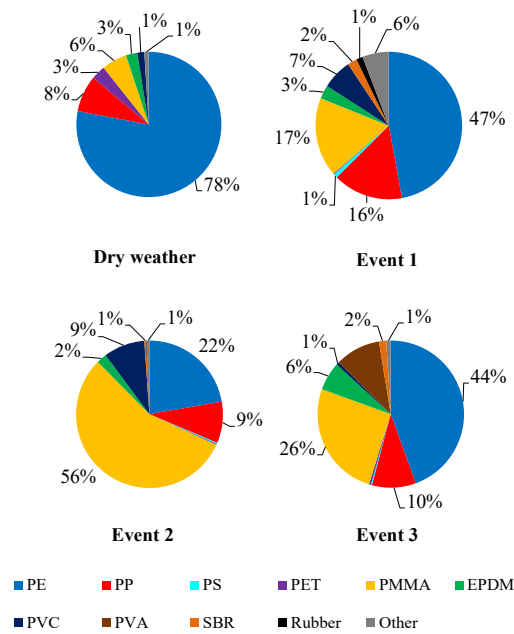


Figure 5. The polymer composition of MPs during the three rainfall events compared to dry weather (PE – polyethylene; PP – polypropylene; PS – polystyrene; PET – polyethylene terephthalate; PMMA – polymethyl methacrylate; EPDM – ethylene propylene diene monomer; PVC – polyvinyl chloride; PVA – polyvinyl alcohol; and SBR – styrene-butadiene rubber).

These findings indicate that MPs behave similarly to non-point source pollutants, with their mobilization and discharge patterns reflecting those of TSS. This resemblance offers practical implications: since real-time monitoring of MPs during storm events is challenging, TSS can potentially serve as an effective indicator for predicting MP dynamics. Additionally, conventional or existing stormwater management strategies can be optimized for size-specific mitigation of MPs in stormwater runoff, coupled with continuous and frequent monitoring of MP build-up and wash-off processes. Eventually, this study set the stage for mitigating rainfall-induced pollution in urban rivers through frequent and continuous monitoring of microplastic discharges at the catchment scale, taking regional rainfall patterns into account.

IV. CONCLUSIONS

This study demonstrated valuable insights on dynamic emissions of MPs in an urban river by sampling at a high frequency over the course of three different rainfall events, classified as light, moderate and heavy. Overall, the findings emphasize that riverine MP emissions are strongly influenced by the synergistic effects of rainfall characteristics (such as rainfall intensity, and antecedent dry days), and particle size. In general, MPs as small as 10–40 μm are shown to be rapidly mobilized at lower rainfall intensities, indicating first flush effects, whereas those larger than 2000 μm are discharged at higher

rainfall intensities. Our findings suggested that intercepting and treating the initial 40% of the runoff that is directly discharged into the rivers may avert MP pollution to a significant extent. We further revealed the opportunity to predict the dynamic emissions of MPs using typical non-point source pollutants like TSS, when planning interventions for MP pollution. Additionally, the findings highlighted the importance of targeting moderate to heavy rainfall events for effective control of MP pollution. These insights are critical for catchment-scale management and the development of effective mitigation strategies for MP pollution in urban river systems.

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