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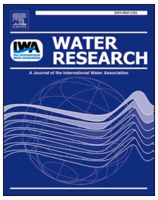

Microplastics discharged from urban drainage system: Prominent contribution of sewer overflow pollution

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ABSTRACT

Urban drainage system is an important channel for terrigenous microplastics (<5 mm in size) to migrate to urban water bodies, especially the input load caused by overflow pollution in wet weather. Investigating how they transport and discharge is essential to better understand the occurrence and variability of microplastics in different water ecosystems. This study evaluated the abundance and distribution characteristics of microplastics in the drainage systems of typical coastal cities in China. The impacts of meteorological conditions and land use were explored. In particular, the prominent contribution of drainage sewer overflow pollution during storm events were investigated. The results showed that the microplastics abundance in daily sewage discharge from different drainage plots ranged between 13.6 and 30.8 items/L, with fibers as the dominant type of microplastics. Sewer overflow discharge can greatly aggravate microplastic abundance to 83.1 ± 40.2 items/L. Road runoff and sewer sediment scouring were the main pollution sources. Systematic estimates based on detailed data showed that the average microplastics emitted per capita per day in household wastewater was 3461.5 items. A quantitative estimation method was proposed to show that the annual emissions load of microplastics via urban drainage system in this research area was 5.83×10^{10} items/km^2, of which the proportion of emissions in wet weather accounted for about 60%. This research provides the first full-process of assessment and source apportionment of the microplastic distribution characteristics in old drainage system. The occurrence of storm events is an important marker of increased microplastic abundance in urban rivers, with a view to urgent need for interception of surface runoff and purification of sewer overflow pollution.

1. Introduction

Microplastics, a type of plastics generally accepted as measuring smaller than 5 mm in size, are widely distributed in the water environment (Rochman, 2018). Microplastics have posed a threat to most parts of ecological system and even transport through the food chain into human bodies (Ross et al., 2021; Wright and Kelly, 2017). Researchers have found microplastic particles in human blood samples, and deep in the lungs of living humans (Jenner et al., 2022; Leslie et al., 2022). Annual global plastic production is estimated to reach 500 million tons in 2025 (Plastics Europe, 2019). 80% of marine microplastics originate from terrestrial systems (Andrady, 2011; Yonkos et al., 2014). Fluvial system are an essential pathway for land-based inputs (Huang et al., 2021; Niu et al., 2021; Xu et al., 2021). Due to the high correlation with human activities, urban areas may be considered of an integral source for microplastics emissions into rivers (Wagner et al., 2019).

The major sources of microplastics in urban agglomerations include sewage discharge, atmospheric deposition, and surface runoff. These sources are all related to the collection and discharge of drainage systems, which are the important channels for pollution transfer between land and water bodies (Browne et al., 2011; Eriksen et al., 2013; Mak et al., 2020). In general, generated microplastics are transported via sewers to the wastewater treatment plants (WWTPs). The removal rates is influenced by a combination of service area characteristics and treatment processes (Blair et al., 2019; Carr et al., 2016; Long et al.,

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2019). Although appreciable removal can be achieved depending on the WWTP types, due to the continuous discharge of treated effluent, high concentrations of microplastics are still detected in both the water column and sediment downstream of the WWTP (Conley et al., 2019; Murphy et al., 2016). Also, rapid and dense urbanization has contributed to increased abundance of microplastic in urban rivers (Esquinas et al., 2020). The sources and pathways of microplastics are closely related to land use and population density. Microplastics are generated from the use of personal care products, washing, illegal waste dumping, etc. (Choi et al., 2021; Ó Briain et al., 2020; Piehl et al., 2018). There is great spatial heterogeneity in microplastic emissions due to differences in production patterns (Jang et al., 2020; Su et al., 2020). However, most previous studies revealed the influence of land use types through the variation of microplastic abundance in water environment in different regions (Dikareva and Simon, 2019; Wagner et al., 2019). While few have reported the abundance of microplastics in directly generated wastewater from drainage plots. Understanding microplastics produced by different drainage plots is critical for identifying sources and managing wastewater treatment facilities.

Another key source of impact to pollution discharge from urban drainage systems is sewer overflow pollution during wet weather. High pollution load caused by sewer overflow has been widely demonstrated, such as COD, nutrients, heavy metals, persistent organic compounds, etc. (Launay et al., 2016; Pilotti et al., 2021; Zhang et al., 2022). During storms or intense rainfall, surface scouring causes microplastics to transport into sewers with runoff and eventually discharge into rivers (Lange et al., 2021; Liu et al., 2019a). The major sources include the form of tire wear particles, road paint plastic waste, etc. Large amounts of runoff cause the water volume to exceed the operational load in the drainage sewers, resulting in overflow pollution. Direct input of untreated wastewater containing high microplastic abundance might occur, which have been detected in overflow effluent in Paris, Italy, and Shanghai (Chen et al., 2020; Di Nunno et al., 2021; Dris et al., 2015; Treilles et al., 2020). In addition, the continuous transport of wastewater in sewers during dry weather results in the settlement or adsorption of large amounts of microplastic particles into the sediments (Sang et al., 2021; Shahsavari et al., 2017). While the increased flow during storm or intense rainfall continuously flushes the sediments at the bottom of the sewers to the receiving water. Sewer sediments may be considered a prominent source of overflow pollution.

Microplastic abundance in rivers and estuaries can surge several or even tens of times following intense rainfall and storm events (Chen et al., 2020). Given accelerated urbanization process and progressively larger paved areas, the risk of microplastic generation and emission is likely to rise (Dalu et al., 2021; Esquinas et al., 2020). Meanwhile, in China, as well as other countries or regions, there are numerous problems with drainage systems, such as broken, blockages, and separation of rainwater and sewage drains (Schilperoort et al., 2013; Ellis and Butler, 2015; Tan et al., 2019; Xu et al., 2020, 2019). These exacerbate the leakage or overflow of untreated wastewater and sediment transmitted, which also meant that more land-based microplastics would migrate into the nearby water bodies. There is consensus that the most effective way to reduce microplastics is to control the sources and migration pathways (Woodward et al., 2021). However, this microplastic migration pathway has not attracted enough attention.

The aim of this study was to determine the migration and emission of microplastics in urban drainage systems during normal and storm flows. The urbanized catchments of typical coastal cities in southern China are selected. Representative microplastic samples were collected from surface runoff, overflow effluent, sewer sediment, and receiving river during multiple rainfall events. The objectives of this study were to (1) determine the microplastics abundance in the daily sewage discharge from drainage plots of different land use types; (2) evaluate the microplastics abundance of sewer overflows, and investigate the migration characteristics of microplastic in receiving river after storm events; (3) propose a quantitative evaluation method for microplastics emission load in normal and wet weather. Understanding the sources of microplastics in these catchments is crucial for deciding priorities for management interventions and reducing inputs load to waterways.

2. Materials and methods

2.1. Study areas

We selected the drainage system of the built-up area of Nanning to determine the characteristics of microplastic distribution in a typical coastal city (Fig. 1). Nanning located in Southern China, a typical subtropical region with annual average rainfall of about 1300 mm. In normal weather, samples were collected from the influent and effluent of three WWTPs. The collection area of the WWTPs was mainly dominated by the separated drainage system (more than 80% of the area), which mainly collected domestic wastewater (more than 90%) and a small amount of industrial wastewater. Five pumping stations within the service area of the largest WWTP were selected to collect wastewater samples (two wastewater pumping stations and three stormwater pumping stations). Specific information on WWTPs and pumping stations is provided in Table S1. Meanwhile, wastewater samples were collected from 17 different functional area drainage plots (including 4 residential areas, 4 industrial areas, 4 commercial areas, 3 administrative areas and 2 urban villages, Table S2 for details). All samples were collected in the summer of 2021 and on dry days (at least three days after rain).

During wet weather, a separated drainage catchment A was selected for the collection of surface runoff, overflow discharge, and sediments at the bottom of sewers. The catchment A was in the service area of the same WWTP collected in dry weather. Catchment A covered an area of about 14.6 ha and was predominantly residential, with a population density of 335 p/ha. The sewer overflow from catchment A is discharged into an urban river that eventually flows into Yongjiang River. In order to ensure that the untreated wastewater was not directly discharged into the river in dry weather, intercepted sewers were set up along the river to transport the misconnected wastewater to the WWTP. When it rains, a large amount of runoff is mixed in, exceeding the load of the intercepted sewer, thus causing overflow discharge. Water and sediment samples were collected simultaneously from four river cross-sections during and after rainfall. Rainfall information was provided in the Table S3.

2.2. Sample collection

Before sampling, all sampling tools were rinsed with ultrapure water. Water samples were collected using 10 L stainless-steel bucket. WWTPs samples were collected at the collection wells and effluent outlets. Pumping station samples were collected at the collection pond. Drainage plots were collected in the manholes before connected to the municipal sewer network. All samples were collected 15–20 min and volume of 5 L. On rainy days, samples from runoff and overflow were collected every 30 min of the first hour, and every hour thereafter until runoff and overflow ended. River samples were collected three times simultaneously, taking 10 L of mixed water samples at 30 cm below the water surface. After collection, the water samples were passed through a 30 μm stainless-steel meshes repeatedly rinsed 3–5 times, and all residual material on the meshes was transferred to a clean glass bottle using ultrapure water. Sewer sediment samples were collected using a stainless-steel grabber prior to rainfall and approximately 1 kg was collected into a glass container. We collected water samples simultaneously to perform the other chemical analysis. Upon completing the sample collection, all the samples were kept in 500 mL polyethylene bottles, immediately transported to the laboratory, refrigerated at 4℃, and analyzed within 12 h.

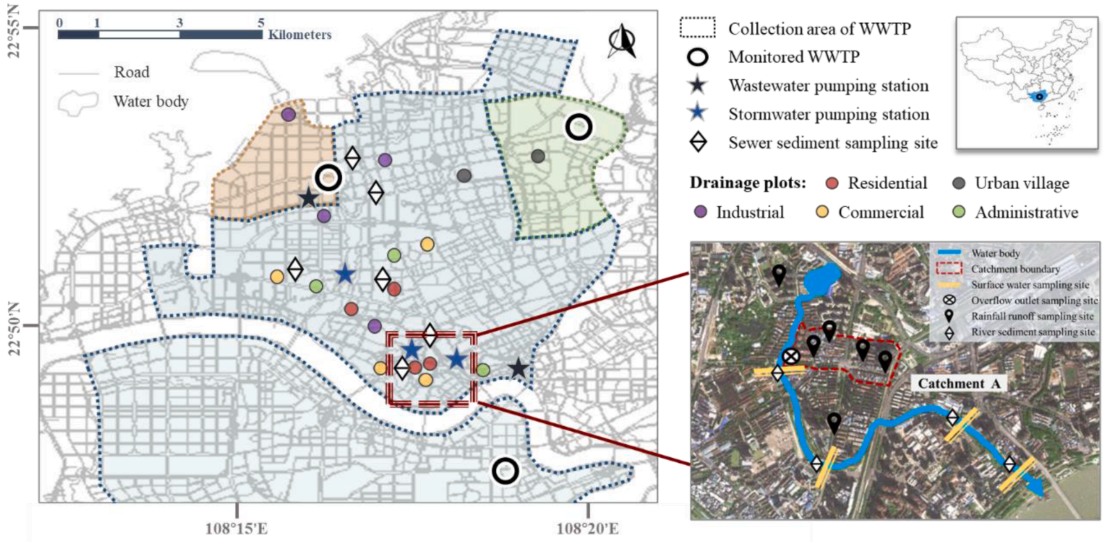


Fig. 1. Water and sediment sampling sites in the studied urban drainage system during dry and wet weather.

2.3. Microplastic identification

Water samples were pretreated to digest using 60 ml of H_2O_2 to remove organic matters and the digestion process was carried out in a thermostat with a stable temperature of 60 °C for 24 h reaction. After digestion, the solution was filtered through a 5-μm polycarbonate filter membrane with a vacuum pump, and the membranes were stored in covered glass dishes and dried in an oven at 40 °C. Sediment samples were sieved (5 mm mesh) and freeze dried for 3 d to a constant weight. 20 g of samples were weighed and then separated using a saturated $ZnCl_2$ solution (1.6 g/cm^3) for 8 h, repeated 3~4 times. The obtained sediment supernatant was then passed through a 30 μm stainless-steel mesh. The subsequent operation was the same as the above water sample treatment process.

The obtained microplastic samples were observed using a stereomicroscope (PXS6555T-J3, Shanghai Cewei Photoelectric Technology Co., Ltd.) and TCapture software, including the quantitative and morphological analysis. The shape can be classified as fiber, particle, fragment, film, etc. The size is divided into five categories, 0.03–0.1 mm, 0.1–0.5 mm, 0.5–1 mm, 1–3 mm, 3–5 mm. After quantitative analysis, 5–10% of typical microplastics samples in each sampling site are selected for micro infrared spectroscopy (Nicolet iN 10, Thermo Scientific). The polymer type of the samples was determined by matching the measured spectra with the standard spectral database at least 80% (OMNIC spectral library) In this research, approximately 43.4% of the suspected plastic items in daily wastewater samples, 38.5% in the surface runoff samples, 55.6% in the overflow samples, 59.1% in the sediment samples, and 58.3% in the receiving river samples were verified as microplastics. Typical microplastic morphology and spectra were described in the Figs. S1 and S2.

To minimize potential plastic use and dust contamination during the experiment, quality control measures were taken with reference to previous studies (Su et al., 2020). All contaminated plastics in the experimental blank samples were fibers with an average concentration of about 6.5 items/L, subtracting the background values from the results.

2.4. Physicochemical analysis

Rainfall characteristics were recorded by a tilting bucket rainfall sensor. The flow rate of wastewater in sewers was monitored by doppler ultrasonic flowmeters (DX-LSX-2). Temperature, DO, conductivity and pH were monitored by portable YSI detector in the field. Turbidity was detected by turbidity meter (HJ041–3, Shanghai Xinrui Instrument Co., Ltd.). Indicators of COD, ammonia nitrogen (NH_3—N), total nitrogen (TN), and SS in water samples were analyzed. The detection methods were all based on China's national standard guidelines. Particle volume concentration was analyzed using the LISST-200X, a field-portable laser particle size analyzer. To ensure precision and accuracy, three identical water samples were collected during each sampling.

2.5. Estimation of microplastics emission via urban drainage systems

Based on the existing research (Bollmann et al., 2019; Wang et al., 2021), a quantitative evaluation method was proposed considering per capita discharge, wastewater treatment rates, sewer system layout, and local precipitation. Randomly selected residential plots and overflow discharge data were analyzed directly to estimate microplastic emission during normal and storm weather from urban drainage catchments. Quantification of microplastics discharge into the aquatic environment from a particular separated drainage system were calculated by the following equations. The detailed description was presented in Table S4.

(1) Microplastics discharged during normal weather (W_{ND}) can be termed as direct discharge of untreated wastewater (W_{UW}) and effluent discharge of wastewater treatment plant (W_{WE}).

$$W_{ND} = W_{UW} + W_{WE} \tag{1}$$

$$W_{UW} = (F * P - V_{WT} * C_{WI}) * (1 - \%_{INT-N}) \tag{2}$$

$$F = (V_{WC} * C_{WW} * \varphi)/P \tag{3}$$

$$W_{WE} = V_{WT} * C_{WI} * (1 - R_{WWTP}) \tag{4}$$

Where F is per capita microplastic discharge (items/p·d), P is the population of the survey area, V_{WT} is the treatment capacity of WWTP (m^3/d), C_{WI} is the microplastic abundance of influent in WWTP (items/L), $\%_{INT-N}$ is the proportion of terminal interception volume in normal weather, V_{WC} is per capita daily domestic water usage (m^3/d), C_{WW} is the microplastic abundance of daily wastewater (items/L), φ is the pollution production coefficient (Usually 0.8–0.9, 0.85 for this study area), R_{WWTP} is the microplastic removal rates in WWTP (%).

(2) Microplastics discharged during wet weather (W_{WD}) can be termed as direct discharge of stormwater runoff (W_{SR}) and sewer overflow (W_{SO}).

$$W_{WD} = W_{SR} + W_{SO} \tag{5}$$

$$V_{SC} = \sum V_R * A * \beta * \%_{SC} \tag{6}$$

$$W_{SR} = V_{SC} * \%_{SS} * EMC_{SR} \tag{7}$$

$$W_{SO} = (V_{SC} * (1 - \%_{SS}) + V_{MW}) * (1 - \%_{INT-W}) * EMC_{SO} \tag{8}$$

$$EMC = \frac{\sum_{t=1}^{n} C_t Q_t \Delta t}{\sum_{t=1}^{n} Q_t \Delta t} \tag{9}$$

Where V_{SC} is the volume of stormwater runoff collected into the sewers (m^3), V_R is the rainfall (mm), A is the area of research catchment (ha), β is the runoff coefficient of each surface (0.7 for this study), $\%_{SC}$ is the proportion of produced stormwater runoff collected into the sewers, $\%_{SS}$ is the proportion of sewer system separation, EMC is the event mean concentration of stormwater runoff and sewer overflow (items/L), V_{MW} is the volume of misconnected or confluent wastewater into the stormwater sewer (m^3), $\%_{INT-W}$ is the proportion of terminal interception volume in wet weather, C_t is the microplastic abundance at time t (items/L); Q_t is the discharge flow rate at time t (m^3); n is the sampling number of a single overflow event; Δt is the interval time between two samples (min).

The treatment capacity, influent abundance, and removal rates of the WWTPs used in this research were all average values. Studied WWTP is the largest in Nanning City, and its treatment capacity for about 50% of the total wastewater volume (diurnal treatment capacity is 0.7 million m^3, official data in 2021). Its removal rates (AAO process) can typically represent the general level here. Per capita emissions were measured based on the actual flow and abundance of household drainage sewers. The event mean concentration of stormwater runoff and sewer overflow was the average of multiple sampling. Annual rainfall used official data for 2021.

2.6. Data analysis

Quantitative expression of the similarities and differences of microplastic discharge using multiple statistical analysis methods. Principal component analysis (PCA) was used to determine the correlation structure between microplastics and other water quality indicators. Regression analysis and Pearson correlation analysis were used to elucidate the correlation between microplastic and physicochemical indicators related to particulate matter. All statistical and data analyses were carried out with the help of the Origin 2018 and IBM SPSS 23.0.

3. Results

3.1. Microplastics abundance in wastewater generation – transport – treatment processes

Fig. 2a revealed the microplastics abundance collected from different drainage plots and pumping stations (1, 2 and 3 are stormwater pumping stations, 4 and 5 are wastewater pumping stations). Microplastics in wastewater discharged from drainage plots generally showed urban villages (30.8 ± 1.3 items/L) > residential areas (28.4 ± 4.0 items/L) > administrative areas (25.5 ± 4.9 items/L) > industrial areas (19.1 ± 6.5 items/L) > commercial areas (13.6 ± 4.5 items/L). Fibers were the dominant microplastic shape, accounting for more than 70%. The microplastics abundance in wastewater pumping stations was larger than the stormwater pumping stations, with concentrations of 32.3 ± 5.1 items/L and 20.7 ± 6.2 items/L, respectively. Particles and films accounted for a larger proportion in the stormwater pumping stations than in the wastewater pumping stations. Fig. 2b showed the mean value of microplastic abundance in WWTP influent was 42.3 ± 9.2 items/L and the effluent was 15.8 ± 5.1 items/L. The removal rates of WWTPs were about 53.6~70.8%. Fibers were the main type of microplastics in

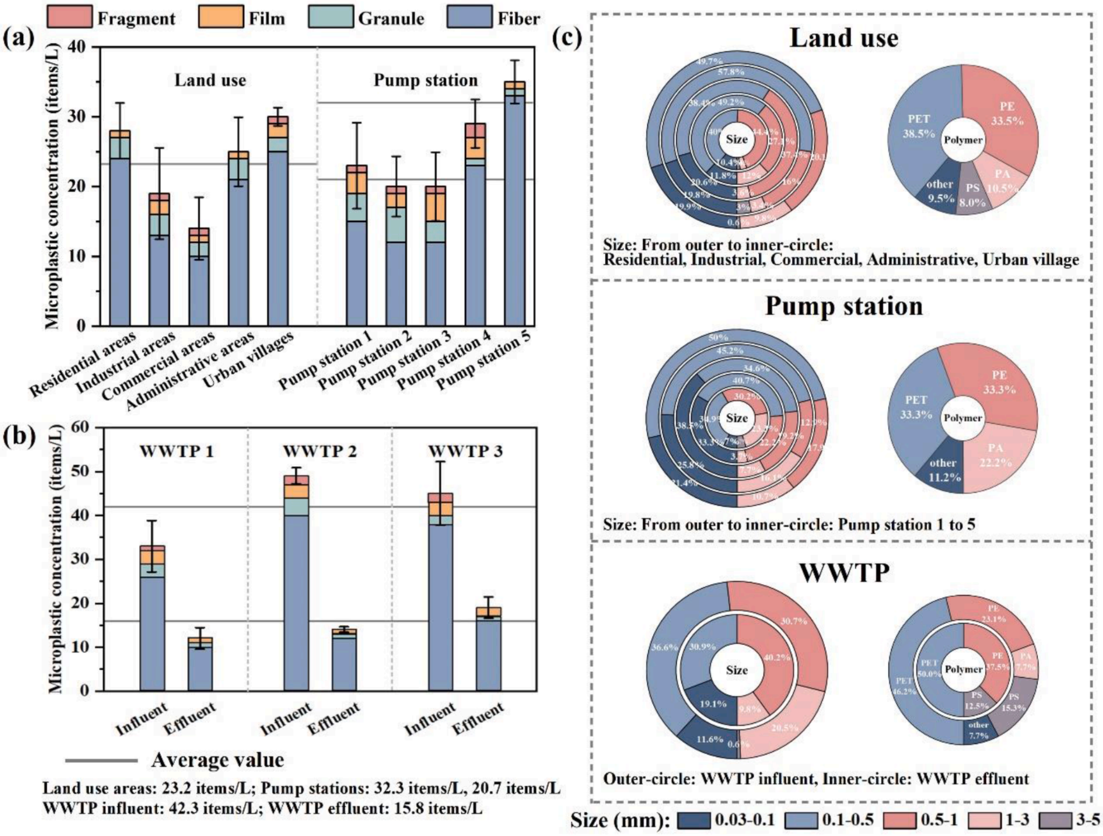


Fig. 2. Microplastics concentration and shape in daily wastewater from (a) drainage plots of different land use types and pumping stations of different drainage systems, (b) influent and effluent of the WWTPs. (c) Size and polymer types of microplastics in drainage plots, pumping stations and WWTPs, respectively.

the influent (26.1–40.2 items/L) and effluent (10.2–16.8 items/L).

Fig. 2c revealed that microplastics in daily wastewater were mainly 0.03–1 mm in size, accounting for more than 85% of the total. There was no significant difference in the size distribution of microplastics from different drainage plots. The average proportion of less than 0.5 mm was about 62.0%. Polyethylene terephthalate (PET) and Polyethylene (PE) were the main types, accounting for more than 65%, followed by Polyamide (PA) and Polystyrene (PS). The types of microplastics in wastewater transport process have slight differences, mainly because microplastics in wastewater were mainly affected by human activities, and there were time-varying characteristics.

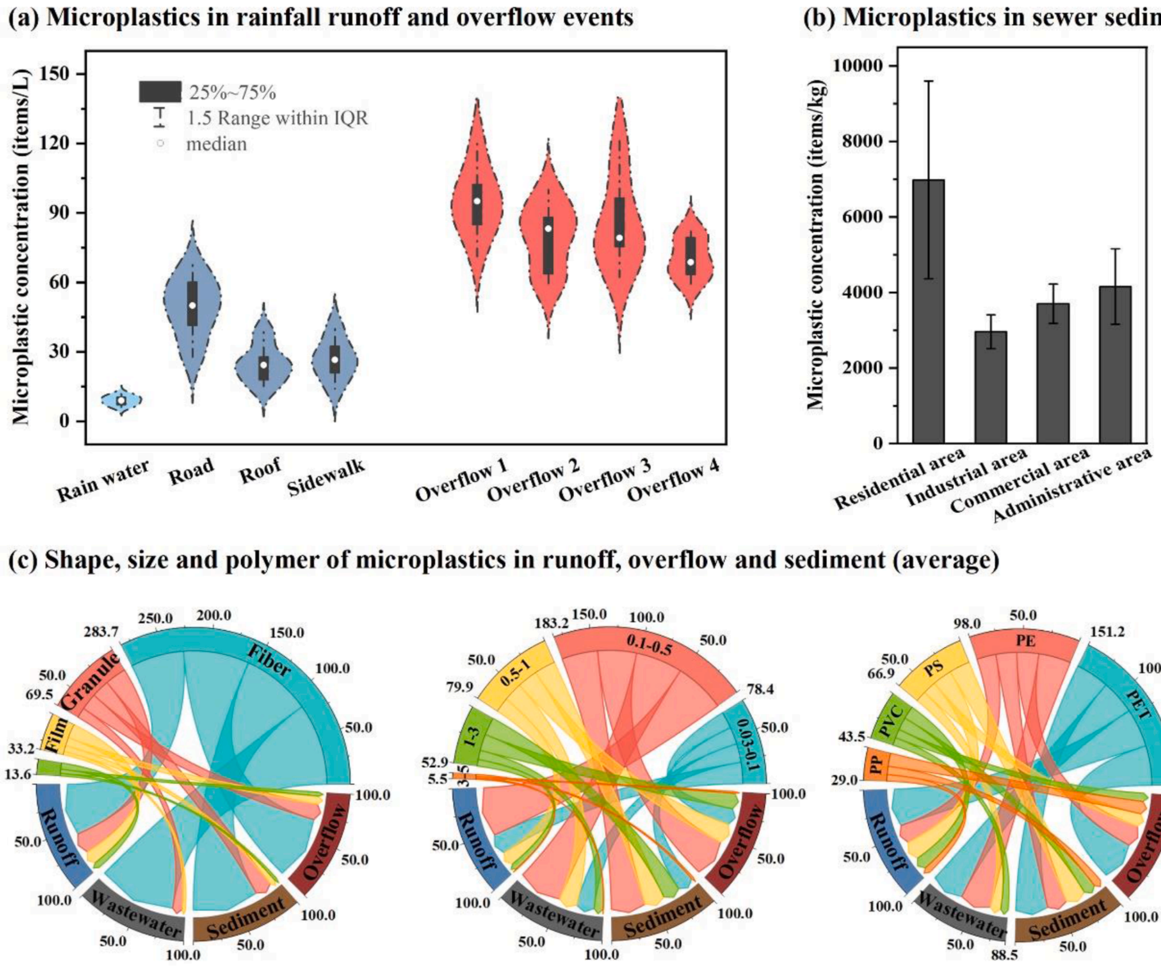
3.2. Microplastics discharged from drainage sewer overflow

The sources of urban drainage sewer overflow pollution in wet weather mainly included rainfall runoff, combined or misconnected wastewater and sewer sediments (Gasperi et al., 2010; Li et al., 2022). The microplastic abundance in runoff and sewer overflow from different storm events were presented in Fig. 3a. Microplastic abundance in overflow (83.1 ± 40.2 items/L) was significantly higher than that in runoff (34.2 ± 33.5 items/L). The highest concentration was found in road runoff (49.1 ± 21.4 items/L), followed by sidewalk (26.9 ± 14.7 items/L) and roof (24.9 ± 12.1 items/L). The runoff samples were much higher than stormwater (8.8 ± 2.9 items/L). Microplastic abundance in overflow pollution differed under different rainfall conditions. The highest average overflow effluent concentration of 71.1 to 122.2 items/L on July 21 and the lowest average overflow effluent concentration of 59.0 to 82.3 items/L on September 14. The concentrations of single overflow event showed a trend of high in the early stage and low in the late stage (Fig. S3). The overall microplastic abundance in the sewer sediment of the stormwater sewers showed that the residential area (6982.5 ± 3049.5 items/kg) > administrative area (4161.0 ± 711.0 items/kg) > commercial area (3705.0 ± 285.0 items/kg) > industrial area (2964.0 ± 1824.0 items/kg) (Fig. 3b).

The shapes, sizes, and polymer of microplastics in sewer overflow during wet weather shown in Fig. 3c. Fibers were the most dominant type, accounting for 56.1% in runoff, 85.7% in misconnected wastewater, and 72.6% in sewer sediment, respectively, and a total of 69.2% in the overflow effluent. Particles accounted for a higher proportion in runoff and sewer sediment, 22.6% and 18.7%, respectively, and a total of 17.6% of the overflow effluent. In comparison, the size of microplastics in runoff and daily wastewater was small, with the particle size (0.03–0.5 mm) accounted for 76.8% and 69.5%, respectively. It is noteworthy that the distribution of large size microplastics (>1 mm) in sewer sediments was significantly higher than that of runoff and daily wastewater, which was about 21.2%. PET (38.5%), followed by PE (21.5%) and PS (18.4%) were the most abundant, and other polymers were Polyvinyl chloride (PVC), Polypropylene (PP) and Cellophane.

3.3. Microplastics pollution in the receiving urban river after sewer overflow

The variation process of microplastics abundance in an urban river during and after the sewer overflow pollution was observed. The



(a) Microplastics in rainfall runoff and overflow events

(b) Microplastics in sewer sediment

(c) Shape, size and polymer of microplastics in runoff, overflow and sediment (average)

Fig. 3. Abundance of microplastic emissions from urban drainage system overflow pollution during wet weather and microplastic characteristics in rainfall runoff, daily wastewater, and sewer sediment.

samples were collected when the overflow occurred, and 10 h, 24 h, 72 h after. From Fig. 4, the highest microplastics abundance in receiving river occurred during the overflow, which was much higher than the average abundance in runoff and daily wastewater. Then gradually decreased, with the peak abundance lasting about 10–24 h. And it returned to normal level after 2–3 days. The microplastics abundance in the upstream river was larger than downstream. Section 1 and Section 2 were 46.4~68.44 items/L and 33.6–77.7 items/L at the time of overflow, which may relate to the reason that the overflow outlets were mostly located upstream.

Consistent with the sewer overflow characteristics, microplastics abundance in the river was generally higher on July 21 (61.5 items/L), and the lowest was on August 15 (40.8 items/L). The morphological characteristics of microplastics in the river were mainly fibers (31.6% ~91.7%) and particles (4.6%~35.3%), with lower content of films and fragments. In addition, sediment samples from each river cross-section were synchronously collected. Overall, microplastics abundance in the sediment was higher in Section 1 (6270.0 ± 1779.5 items/kg) and Section 4 (6384.0 ± 1595.9 items/kg) than in Section 2 (4845.0 ± 403.1 items/kg) and Section 3 (4190.0 ± 809.2 items/kg). The reason might be mainly due to the installation of rubber dams at the sampling Sections 1 and 4, which intercepted the particles. The rain-sourced urban rivers have slow velocity (about 0.13–0.26 m/s) in dry weather. Microplastics would be deposited in the riverbed by the movement of aggregation and sedimentation, which makes microplastics accumulate in the sediment (Alimi et al., 2018). Consistent with the surface water,

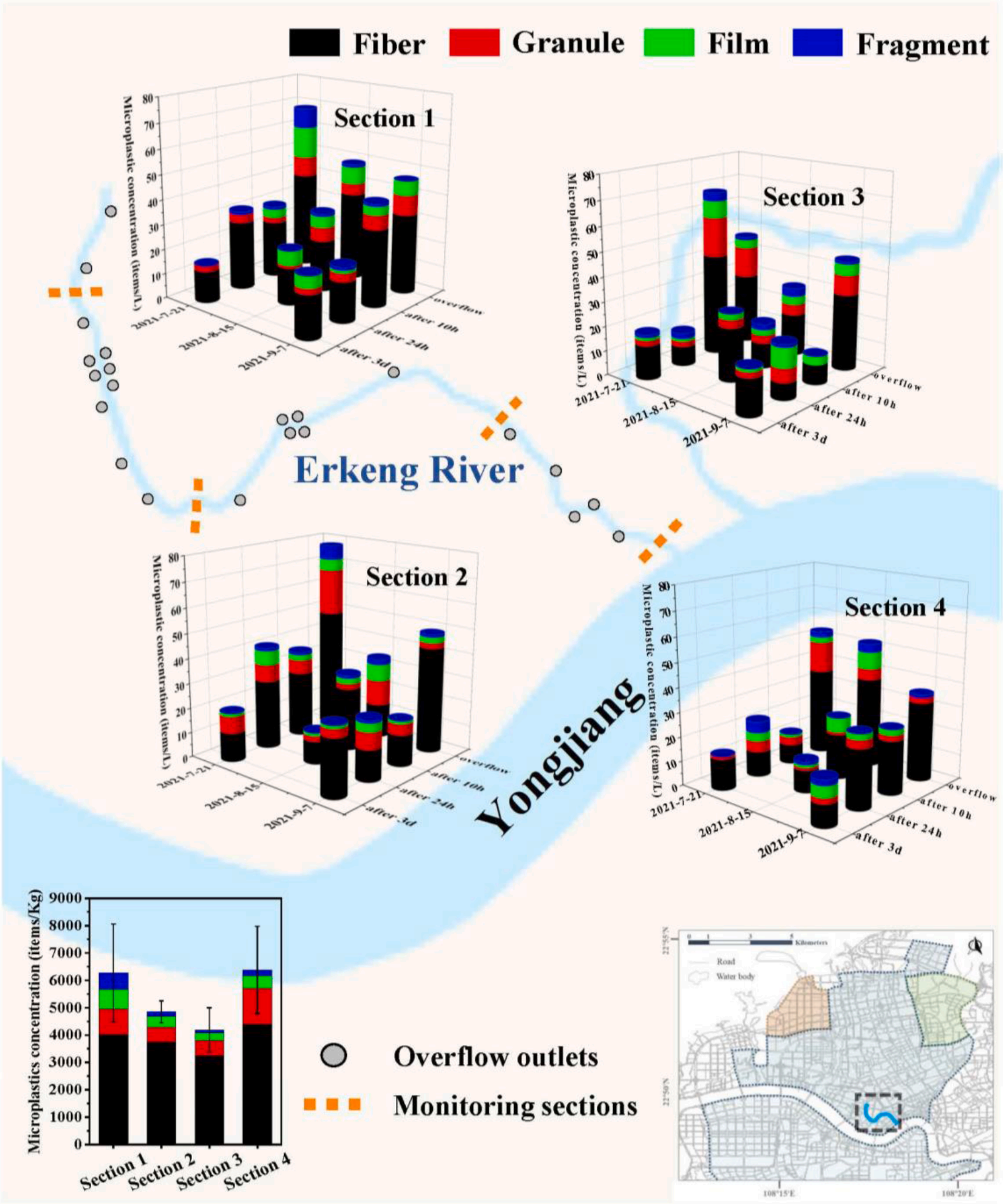


Fig. 4. The variation process of microplastics concentration in an urban river during and after the overflow discharge (when overflow occurred, after 10 h, 24 h and 3 days) via three rainfall events. Microplastics characteristics in the river sediments of the monitoring sections were shown in the lower left corner, all sampled three days after the rainfall.

the morphological distribution generally showed fiber (71.9%) > particle (15.0%) > film (8.4%) > fragments (4.7%).

3.4. Correlation analysis of microplastics and water quality

PAC was used to determine the correlation between the microplastics and conventional water quality indexes (COD, NH_3-N, TN, TSS, turbidity and particle volume concentration) in wastewater, runoff/overflow, and surface water. The results showed that two principal components (PC1: 49.5%; PC2: 26.2%) comprised the majority variance. Fig. 5a, Tables S5 and S6 demonstrated that the variables of wastewater, runoff/overflow and surface water formed more distinct independent clusters. Wastewater had a more distinct positive correlation with PC1, while runoff/overflow had a more distinct positive correlation with PC2. In terms of

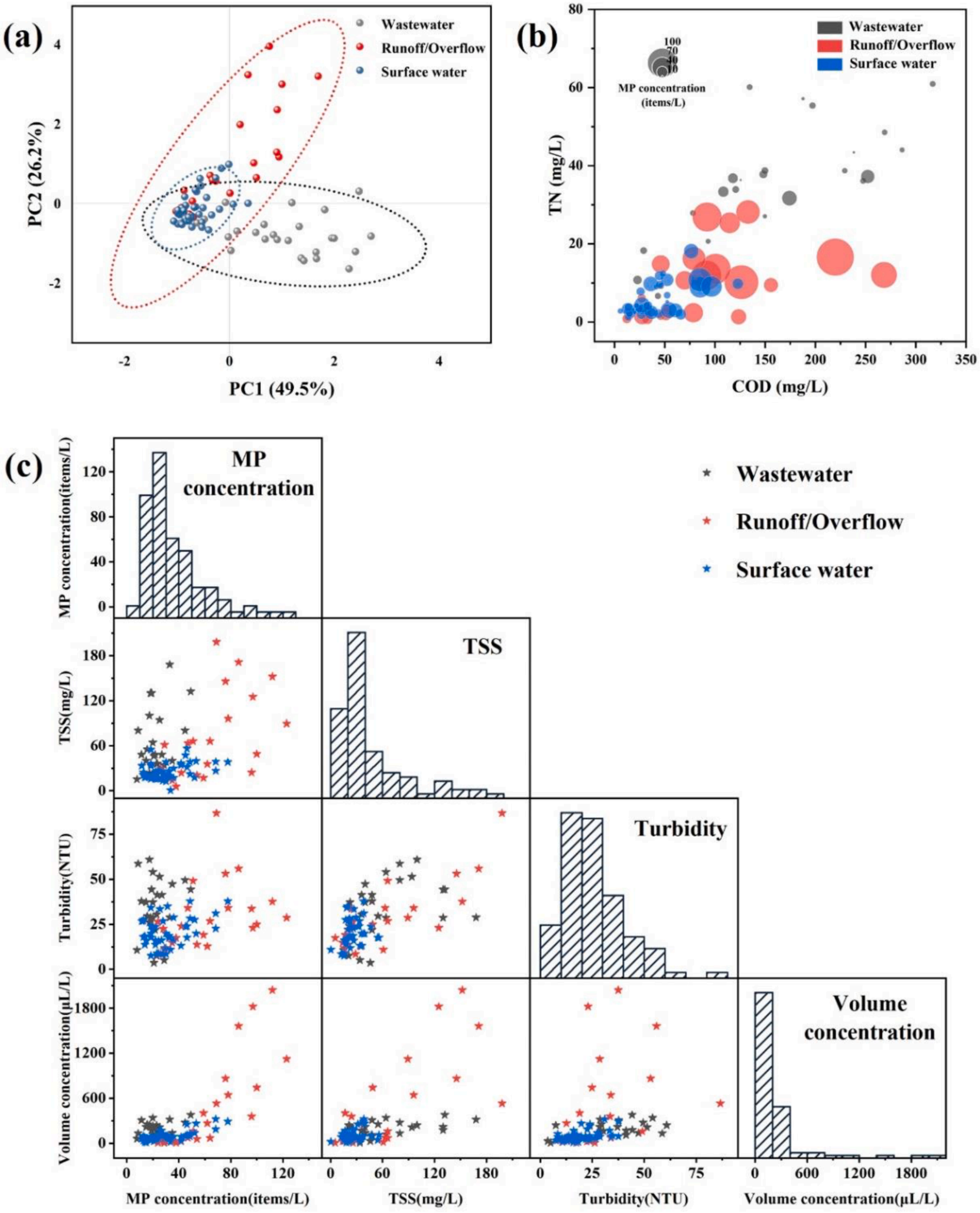


Fig. 5. Statistical analyses of microplastics and conventional water quality indexes discharged via urban drainage system. (a) Principal Component Analysis of the pollution characteristics in the wastewater, runoff/overflow, and surface water. (b) Correlation between microplastic abundance and COD and TN. (c) Correlation between microplastic abundance and the TSS, turbidity, and particle volume concentration.

different indicators, positive PC1 correlated with COD, TN, and NH_3-N, whereas positive PC2 correlated with microplastics concentration and particle volume concentration (due to the instrument range, the particle volume concentration was below 0.495 mm).

Fig. 5b further revealed the variation of microplastics with COD and TN in wastewater, runoff/overflow, and surface water. There was no consistent trend of increasing microplastic abundance with increasing concentrations of conventional indicators. The variation trend of COD and TN in wastewater was synchronized. However, the microplastics did not change with the variation of COD and TN. The microplastics abundance in runoff and overflow was significantly higher than that in wastewater and surface water. And the microplastics abundance in surface water after storms was mostly even higher than the untreated wastewater, despite the low concentrations of COD and TN.

From Fig. 5c, there are significant differences between microplastic abundance in wastewater, runoff/overflow, and surface water in relation to these variable indicators. For wastewater, there was no significant correlation between microplastics and TSS, turbidity and particle volume concentration. The correlation between microplastic abundance and particle volume concentration was more significant in both runoff/overflow and surface water. Compared with microplastics, particle volume concentration was easier to obtain. By monitoring particle volume concentrations, the following formula were derived for predicting microplastic abundance in runoff and overflow under different rainfall events (**Fig. S4**).

$$y = 0.086x + 26.622\, R^2 = 0.7321 (p < 0.01) \tag{6}$$

Where y is the microplastic abundance (items/L), x is the particle volume concentration (below 0.495 mm, μL/L). The formula was applicable to the rainfall depth of 2.4~21.0 mm/d or the rainfall intensity of 0.041~0.331 mm/min. And might be more suitable for the calculation of overflow load discharge from the sewer with sediment deposition, especially in the urban built-up areas.

4. Discussion

4.1. Distribution characteristics of microplastics in the generation-transport-treatment process of urban wastewater system

Residential activities, industrial production, and agriculture were all important output sources of terrigenous microplastic (Su et al., 2020). High microplastic emissions were concentrated in many industrial and commercial areas (Chen et al., 2020; Jang et al., 2020). In the West River downstream, in the south of China, the microplastics abundance was observed in commercial/public/recreational > residential > industrial > natural areas (Huang et al., 2021). Microplastics (5.85 ± 3.28 items/L) had even been found in rivers in slum areas (Alam et al., 2019). In addition, there were also studies revealing higher microplastics concentrations in waste from agricultural land, such as the Ofanto river in southeast Italy (Campanale et al., 2020), Swiss Seeland (Bigalke et al., 2022), a farmland in southeast Germany (Piehl et al., 2018), and coastal areas in Xiamen, China (Tang et al., 2018). This study provided direct evidence for the microplastic abundance in effluent discharging from drainage plots of different land use types. Microplastic abundance were found to be higher in direct discharges from residential areas (28.4 ± 4.0 items/L) and urban villages (30.8 ± 1.3 items/L) than other plots. And some temporal variability was found for residential wastewater, with higher microplastic discharge concentrations at 10 pm than at 6 pm and 1 pm (Table S7). Previous studies had similarly reported obvious variability in microplastic abundance between different times of the day (Blair et al., 2019; Cao et al., 2020). Fiber was the most predominant form in all land use types especially residential area. With polyester production dominated by synthetic fibers, the polymer was mainly PET and PE, similar to other studies (Baldwin et al., 2016; Dris et al., 2015; Ross et al., 2021). Fibers mainly originated from textile clothing, care products, cosmetics, etc. Higher microplastic abundance in the evening also provided direct evidence that laundry and washing processes were the main source of household fibers (Choi et al., 2021; Hernandez et al., 2017; Zhang et al., 2019).

The microplastics characteristics (composition, shape and size, etc.), physicochemical properties of the water column (pH, salinity, ionic species and strength, etc.), sediment properties (particle size distribution, composition and organic matter content, etc.), disturbance time and intensity are all the key influencing factors on the migration of microplastics in the water column (Kooi et al., 2017; Jin et al., 2018; Wagner et al., 2019; Gao et al., 2023). Because of the significant differences in shape, size, and density of microplastics, microplastics differed significantly from particulate matter in its behavior (Waldschläger and Schüttrumpf, 2019; Cowger et al., 2021). In general, the surface area of a microplastic particle and its texture were practically important parameters, since they might affect the time of fouling up (Chubarenko et al., 2016). The fibers appeared to have the largest surface area for the given mass, immediately followed by films (Zhang and Choi, 2022). The settling velocity of fibers and films in still water is generally lower than that of particles and debris (Khatmullina and Isachenko, 2017; Wang et al., 2021). Meanwhile, daily wastewater was mainly fibers, which should sink due to bio-fouling by the bio-films attached to the sewer walls (Chubarenko et al., 2016). In this study, the direct discharge wastewater from the drainage plots was mainly PET (1.38 g/cm^3), PE (0.92–0.97 g/cm^3), and PS (0.96- 1.05 g/cm^3). Except for PET, the densities of the other two polymers were smaller or approximate to that of daily wastewater (0.9988–1.0041 g/cm^3) (Zhang et al., 2019). However, when the velocity in the sewers increased, the shear stress exceeded the critical value. Microplastics might re-suspend into the wastewater, similar to the composite effect of sediment and epiphytes at the riverbed (Hurley et al., 2018). As a corollary, in addition to being directly influenced by land use types, microplastic abundance in sewage sewers might have physical sedimentation, biosorption, and erosion characteristics as conventional organic matters and nutrients (Shi et al., 2018). While few studies have yet revealed the physical and dynamical properties of microplastics in sewage sewers.

Compared with previous studies of WWTPs in different countries, the microplastic abundance measured in the influent and effluent herein was at a moderate level (Table 1). The removal rates of WWTPs (53.6~70.8%) in this study was much lower than the above studies (basically above 85%). Previous studies have shown that the removal of microplastics closely related to the treatment process, physicochemical properties of microplastics, etc. The removal rates varied widely among different WWTPs (Carr et al., 2016; Long et al., 2019). The microplastic removal rates of tertiary treatment in the three wastewater plants in this study was higher than that of secondary treatment (Schernewski et al., 2021; Talvitie et al., 2017). The treated effluent in this study consisted mainly of fibers, in agreement with previous studies (Gies et al., 2018). Capturing all polyester fibers in WWTPs was a major technical challenge requiring urgent attention (Hurley et al., 2018; Ziajahromi et al., 2021), especially in densely populated older urban areas just like the present study area. In addition, it had been claimed that as the operating load of the WWTP increased, the microplastic abundance subsequently increased while the removal rates decreased significantly (Long et al., 2019). Combined drainage systems remained in some regions and stormwater and wastewater misconnection was difficult to avoid. The operational control of WWTPs during the wet weather needed sufficient attention.

4.2. Contribution of sewer overflow to microplastics pollution in urban receiving rivers

Overflow pollution is an important factor that exacerbated microplastic load in the aquatic environment under high flow rates and dispersed inputs (Li et al., 2015)(Hitchcock, 2020). In this study, microplastic discharge characteristics from sewer outlets in urban

Table 1
Overview of previous studies of microplastic abundance from WWTP in different countries for comparison to this study.

Country	Mesh sizes (mm)	Major polymer types	Average concentration of MPs (items/L)		Proportion of fibers	Removal rates	Refs.
			Influent	Effluent			
USA	>0.043	/	136.73±48.8	13.65±8.80	>60%	85.2%–97.6%	Conley et al. (2019)
Finland	>0.250	PET	57.6 ± 12.4	0.4 ± 0.1	62.0%	99.4%	Lares et al. (2018)
Australia	>0.025	PET/PE/PP	92.0	0.18	83.7%	99.8%	Ziajahromi et al. (2021)
Canada	>0.001	PET/PS	31.1 ± 6.7	0.5 ± 0.2	60.0%	98.3%	Gies et al. (2018)
Italy	>0.063	PET/PA	2.5 ± 0.3	0.4 ± 0.1	/	84.0%	Magni et al. (2019)
Spain	>0.025	PET/PE/PP	/	10.7 ± 5.2	28.0%	93.7%	Edo et al. (2020)
Morocco	>0.063	PE/PP/PS	188.0	50.0	70.0%	73.4%	Hajji et al. (2023)
Thailand	>0.050	PET/PE/PP	77±7.21	10.67±3.51	>60%	86.1%	Tadsuwan and Babel (2022)
Korea	>0.100	PP/PE/PET	10–470	0.004–0.51	31.8%	98.7%–99.99%	Park et al. (2020)
China	>0.025	PP/PE/PET	0.70–8.72	0.07–0.78	86.5%	89.2%–93.6%	Zhang et al. (2021)
China	>0.095	PET/PU/PE	65.0 ± 4.3–105.0 ± 5.3	3.0 ± 1.6–6.0 ± 2.8	76.7%–90.0%	83.7%- 96.3%	Jiang et al. (2022)
China	>0.125	Cellulose/PET/PP	117.0	52.0	74.4%	55.6%	Bai et al. (2018)
This study	>0.030	PET/PE/PS	42.3 ± 9.2	15.8 ± 5.1	>75%	53.6%–70.8%	

catchments were investigated for the first time. The results showed that the highest peak abundance being about 10 times higher than that in treated effluent from WWTP. The microplastic abundance from overflow outlets obtained in this study (83.1 ± 40.2 items/L) were significantly higher than those of stormwater sewers in Hong Kong (1.4 to 6.8 items/L) (Mak et al., 2020) and in Wuhan 2.75 ± 0.76 to 19.04 ± 2.96 items/L (Sang et al., 2021). The main reason might be the presence of misconnected wastewater in the drainage area of this study. However, the results of the study were lower than that in the combined system overflow in Shanghai (130±30 to 8500±1241 items\/L) (Chen et al., 2020) and Paris (190–1046 fibers/L) (Dris et al., 2015). The possible reason might be attributed to a combination of urbanization, population density, or sampling and analytical methods. The high percentage of polymer that appeared were PET, PE and PS, and toxic plastics further harmed the ecology of the receiving urban water bodies.

The first flush effect was an important indicator to characterize the pollution process of overflow pollution (Peng et al., 2016; Peter et al., 2020). In this study, microplastic abundance in a single overflow event also showed first flush effect (Supplementary Material), and was similar to COD and TSS. In the early stage of rainfall, the scouring effect of surface runoff and wastewater in sewers carried the microplastic particles accumulated in dry weather to the receiving water bodies. However, the dilution effect of rainwater increased in the later stage of rainfall, resulting in a decrease of microplastics concentration. The overflow effluent in this study was dominated by fibers, which tend to settle on the surface (Sutton et al., 2016). Also, they could be resuspended and discharged during heavy rainfall (Chen et al., 2020). Numerous studies had revealed the significant effects of rainfall, intensity, and antecedent dry period on the concentration of pollutants such as organic matter, nutrients, and heavy metals in the overflow (Yu et al., 2022; Zhang et al., 2018). The significant differences in microplastic abundance among the four overflow events revealed that the response of microplastic abundance to rainfall conditions was basically consistent with other pollutants. The results suggested that prolonged antecedent dry period would cause pollutants on the surface and sediment in the sewer to accumulate, increasing the concentration of effluent when rainfall started (Barone et al., 2019). While the rainfall depth and intensity increased, the dilution effect became more noticeable. In addition, heavy rainfall would increase the scouring on the surface, increasing the microplastics concentration of runoff, and enhance the erosion of sediment in the sewer (Semadeni-Davies et al., 2008; Sandoval et al., 2013).

In this study, for the first time, simultaneous sampling of surface runoff, misconnected wastewater in stormwater sewer, sewer sediment and final overflow discharge in a specific catchment area. The highest microplastic abundance were found in road runoff, which was consistent with previous study (Chen et al., 2022b). Granular and film plastics found in road runoff were different with wastewater in this study, which might be tire and road wear particles, vehicle wear, etc. (Yukioka et al., 2020). Fibers were still the dominant microplastic form in surface runoff, which might originate from synthetic clothing, plastic waste, paper towels, packaging materials, etc. The higher proportion of PET, PE and PS confirmed this speculation (Talbot et al., 2022; Wang et al., 2019; Yan et al., 2019). Studies have also revealed that cigarette butts composed of cellulose were one of the most common microplastics in the environment (Shen et al., 2021). The large number of masks were also used during the COVID-19 (Dissanayake et al., 2021). Meanwhile, atmospheric deposition was also an important source of microplastics. The microplastic abundance in the rainwater background monitored in this study was 8.8 ± 2.9 items/L, mainly fibers (Liu et al., 2019b). Through previous studies and the results of our research group's existing studies, sewer sediments had been proved to be a key source of overflow pollutants, such as COD, nutrients, and heavy metals (Gasperi et al., 2010; Li et al., 2022).

Storm and flood events were the primary periods when microplastic entered water bodies (Hitchcock, 2020). Microplastic abundance increased seven times after rainfall along the California coast of the United States (Moore et al., 2011). And microplastic load in the Clackamas River and the Johnson Creek in Poland was significantly higher in August than in February (Talbot et al., 2022). The same results also occurred in the Brisbane River, Australia (He et al., 2020), Nakdong River, Korea (Eo et al., 2019), Baltic Sea (Schernewski et al., 2021), and in northwest England (Hurley et al., 2018). In this study, the overflow effluent and the dense distribution of outlets directly influenced microplastic abundance in the water column. High microplastic abundance lasted for nearly 1 day after the storm. Low-density microplastics (PE and PP) were more likely to float on the water surface or be suspended in the water column (Kooi et al., 2017). The tendency of microplastics to accumulate in the sediment also because urban rivers were mostly rich in humic substances and microbial communities (Eo et al., 2019) Meanwhile, rubber dams or interception measures in urban streams might exacerbate microplastic abundance in the substrate. During the wet weather, increased river flows could cause microplastics deposited in river substrates to resuspend in the water column, transforming temporarily from a microplastic sink to a source of pollution (Woodward et al., 2021). In this study, microbeads or regular particles were only considered. Also, particles below 0.03 mm were not considered due to the limitations of the detection method. The amount of microplastics reported in this study might be underestimated. Therefore, rainfall and storm occurrence were critical periods for riverine input of microplastics to the ocean, which were possibly more harmful than direct discharges from WWTP or other point sources pollution.

4.3. Microplastics emission in the urban drainage systems: characteristic pollutants and total amount control

Many urban drainage systems currently suffer from breakage, blockage, and misconnection due to a variety of factors such as unqualified construction and improper maintenance and management (Sun, 2020; Xing et al., 2020). In areas where the sewer invert lies below the ground water table, sewer infiltration commonly occurs through sewer cracks and joints, increased hydraulic loading and reduced wastewater treatment efficiency. Marker species approach was a commonly method to account for the infiltration in the sewer, which mainly utilized the difference in concentration of characteristic factors and was more appropriate to use conservative substances (Zhao et al., 2020). Some studies claimed that microplastics had also been detected in groundwater (0–7 $\times$ 10^{-3} items/L) (Mintenig et al., 2019), but at much lower concentrations than in daily wastewater (13.6–30.8 items/L). Similarly, microplastic characteristics in surface runoff also differ significantly from those of daily wastewater, especially characteristic plastics such as rubber particles from tire wear and road coatings. Subsequent studies could explore the operability of microplastics as a marker specie for groundwater infiltration or rainwater inflow in sewage sewers.

The total annual discharge load of microplastics have been directly accounted in different regions using the effluent from WWTPs or the discharge from pumping stations (Bollmann et al., 2019; Chen et al., 2022a). Qi et al. (2022) stated that the average microplastic discharge load in personal care and cosmetics was 2.18 million particles per capita per year. Kole et al. (2017) estimated per capita emission ranged from 0.23 to 4.7 kg/year, with a global average of 0.81 kg/year from wear and tear tyres. However, there was a lack of microplastic production coefficient that correspond to the actual level of social development in each region. This study aimed to account for the first time the per capita generation equivalents of microplastics in drainage plots, combined with population and actual water usage. The results showed that the average microplastics per day emitted per capita in household wastewater was 3461.5 items. The per capita production coefficient closely related to the level of urbanization and water consumption habits of the region. It was an important basic index for statistical calculation and evaluation of the effectiveness of the wastewater treatment industry.

The average microplastic discharge from the four overflow events in catchment A monitored in this study was about 1.85×10^{7} items. Based on the per capita production coefficient and the removal rates of the WWTP (70.8%), the microplastic emissions in the catchment A was about 4.40×10^{6} items/day. Microplastic emissions from the single overflow event in wet weather was about 4–5 times higher than treated effluent discharge in dry weather. A quantitative estimation method was proposed to show that the annual emissions load of microplastics via urban drainage system in this research area was 5.83×10^{10} items/km^{2}, of which the proportion of emissions in wet weather accounted for about 60%. This study confirmed the non-negligible microplastic emissions during rainfall or storm events. At present, similar with other particulate pollutants, the increase of rainfall and intensity would intensify the microplastics emissions. However, there was a lack of research on the microplastic discharge load under different rainfall conditions, which also made the operational design of interception and precipitation facilities more difficult.

Turbulent flow in the sewers might exacerbate the secondary movement of microplastic fibers (Choi et al., 2022). The scientific definition of microplastic morphology in wastewater might provide a basis for reasonable prediction of resistance models (Zhang and Choi, 2022). In addition, a range of other anthropogenic stressors in the sewers interact with microplastics, such as sediments, nutrients, pathogens, and anoxic or anaerobic environment, which might pose a higher risk after storms and floods (Eckert et al., 2018; McCormick et al., 2014). The results of this study might provide additional data to support the establishment of a database on microplastic abundance in overflow pollution. Meanwhile, the importance of implementing measures to reduce the output of microplastics in wet weather was emphasized. The hydraulic and dynamical properties of specific microplastics should also be considered when selecting and operating facilities such as overflow registers, stormwater retention ponds, and constructed wetlands. The effective capture of microplastics along with the effective removal of traditional pollutants should also considered.

5. Conclusion

This study determined the migration and emission of microplastics in urban drainage systems during normal and storm flows. The impacts of meteorological conditions and land use were explored. Microplastics were detected at the sewage discharge from drainage plots with different land use types. The highest and lowest abundance were residential and commercial areas, respectively. The average microplastics per day emitted per capita in household wastewater was 3461.5 items. The main microplastic forms in the wastewater were fibers, with particle size less than 1 mm. PET and PE were the two main microplastic polymers. The microplastic removal rates of the three WWTPs in the study area ranged from 53.6 to 70.8%, and the treatment facilities had limited removal of fibers.

Sewer overflow pollution in wet weather was an important source of microplastics in urban receiving river. Sewer overflow discharge can greatly aggravate microplastic abundance to 83.1 ± 40.2 items/L. This study confirmed the major contribution of road runoff and sewer sediments scouring to microplastic emission load. In addition, microplastic abundance in runoff/overflow and surface water showed a more significant positive correlation with particle volume concentrations. A quantitative estimation method was proposed to show that the annual emissions load of microplastics via urban drainage system in this research area was 5.83×10^{10} items/km^{2}, of which the proportion of emissions in wet weather accounted for about 60%. More attention needs to be paid to control and reduce the microplastics discharge from urban drainage systems to waterways during wet weather.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Supplementary materials

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