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Microplastic emission characteristics of stormwater runoff in an urban area: Intra-event variability and influencing factors

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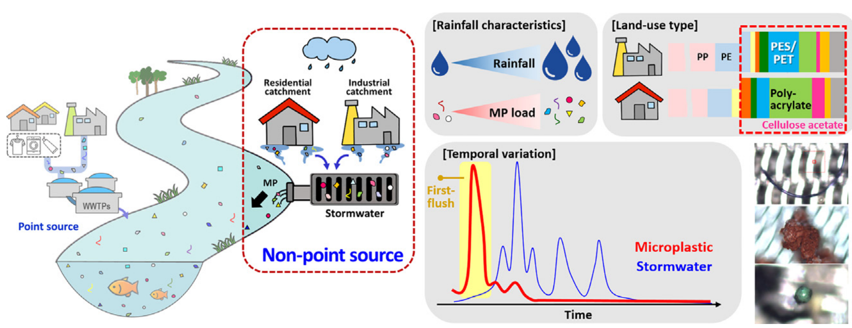
HIGHLIGHTS

- Stormwater runoff is a key transporter of microplastics (MPs) to aquatic systems.
- Most MPs were transported in the early stage of runoff (within 40 %).
- Total rainfall depth heavily influenced the load of MPs.
- The coarser and heavier MPs washed out in the later runoff period.
- Polymer compositions of MPs reflect the land-use pattern of the drainage area.

GRAPHICAL ABSTRACT



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ABSTRACT

Stormwater runoff is considered a major pathway for land-based microplastic transportation to aquatic environments. By applying time-weighted stormwater sampling at stormwater outlets from industrial and residential catchments, we investigated the emission characteristics and loads (number- and mass-based) of microplastics to aquatic environments through urban stormwater runoff during rainfall events. Microplastics were detected in stormwater runoff from industrial and residential areas in the concentration range of 68–568 n/L and 54–639 n/L, respectively. Polypropylene and polyethylene were found as major polymers accounting for around 60 % of total microplastics. The fragment was the dominant shape of microplastics, and the most common size class was 20–100 μm or 100–200 μm. The microplastic load emitted from industrial and residential areas were estimated to be 1.54–46.1 $\times$ 10^8 and 0.63–28.5 $\times$ 10^8 particles, respectively. The discharge characteristics of microplastics inter– and intra–event were affected by the land-use pattern and rainfall characteristics. The concentration of microplastics did not significantly differ between industrial and residential catchments, but the composition of polymer types reflected the land-use pattern. The microplastics in stormwater were more concentrated when the number of antecedent dry days (ADDs) was higher; the concentration of microplastics was generally peaked in the early stage of runoff and varied according to rainfall intensity during a rainfall event. The contamination level and load of microplastics were heavily affected by the total rainfall depth. Most microplastics were transported in the early stage of runoff (19–37 % of total runoff time), but the proportion of larger and heavier particles increased in the later period of runoff. The microplastic emission via stormwater runoff was significantly higher than that through the discharge of wastewater treatment plant effluent in the same area, implying that stormwater runoff is the dominant pathway for transporting microplastics to aquatic environments.

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1. Introduction

Since plastics were invented in the 1860s, they have become popular worldwide because of their light weight, durability, and low cost (Thompson et al., 2009). The widespread use of plastics in various sectors, including the packaging, building and construction, and automotive sectors, has caused extensive pollution of natural environments with plastic litter (Geyer et al., 2017; Senathirajah et al., 2022). The cumulative plastic production between 1950 and 2017 was estimated to be 9.2 billion tons (Geyer, 2020), and the worldwide annual plastic production has reached approximately 370 million tons (Plastic Europe, 2021). Geyer (2020) estimated that 76 % of all plastics produced were discarded or accumulated in natural environments. Thus, plastic constitutes 50–80 % of the waste present in the environment (Barnes et al., 2009; Ebere et al., 2019). As littered plastic waste undergoes environmental weathering, it breaks down into microplastics (<5 mm) (Andrady, 2011), which are widely present in both terrestrial and marine environments, including air, water, soil, sediment, and biota (GESAMP, 2015; Mai et al., 2018a).

Terrestrial environments, especially urbanized areas, are important contributors of microplastics to marine environments (Mai et al., 2018b). Various human activities on land, such as landfilling, dumping, residency, industry, transportation, and agriculture, generate microplastics, which are discharged to the environment via point sources (e.g., wastewater treatment plant [WWTP]) and non-point sources (e.g., stormwater runoff, atmospheric deposition, and drainage) (Su et al., 2020). Because point sources discharge microplastics directly into the environment from specific discharge points, their emissions are predictable and generally easy to manage. In contrast, non-point sources naturally and artificially discharge microplastics from unspecified discharge points; these microplastics can be diffused and dispersed over a wide area. Furthermore, it is difficult to estimate the emissions of such microplastics because they are greatly affected by weather. These characteristics lead to difficulties in monitoring, controlling, and managing non-point source pollution. Pollution from non-point sources has increased because of the increasing impervious surface area generated by urbanization, and it has become a major threat to watershed health (Shen et al., 2012). Previous studies have focused mainly on point sources of microplastic pollution, especially WWTPs (Carr et al., 2016; Murphy et al., 2016), and understanding of non-point sources remains very limited.

Among non-point sources, stormwater runoff is potentially an important pathway for microplastics to enter aquatic systems (Liu et al., 2019). Some previous studies observed a significant increase in microplastic abundance in riverine and estuarine waters (Yonkos et al., 2014; Hitchcock, 2020), and a significant change in the characteristics of microplastics in seawater after rain events (Gündoğdu et al., 2018). The characteristics of pollutants discharged into aquatic systems through stormwater runoff are expected to vary according to the land-use patterns and rainfall characteristics of the watershed (Goonetilleke et al., 2005; Ki et al., 2011); they are also expected to demonstrate large temporal variability within and between rain events (Ballo et al., 2009). However, most studies of microplastics pollution in stormwater have applied grab sampling (Liu et al., 2019; Mak et al., 2020; Ziajahromi et al., 2020; Sang et al., 2021) or a composite sampling method that includes only a small number of stormwater samples (Piñon-Colin et al., 2020; Boni et al., 2022). Thus, there is minimal understanding of the temporal variations in microplastic abundance and characteristics during a rainfall event, as well as the discharge characteristics of microplastics according to rainfall characteristics and land-use patterns.

To bridge the knowledge gap on characteristics of microplastics discharged from urban non-point sources with different land-use patterns, especially their temporal dynamics, we investigated microplastics in stormwater runoff from an urban area during three rain events. We performed time-weighted stormwater sampling at two stormwater outlets that drain runoff from residential and industrial catchments. This study aimed (i) to identify the intra- and inter-event variations in microplastic emission load and characteristics (including abundance, shape, size, and polymer type) according to urban land-use pattern and rainfall characteristics, and (ii) to evaluate the relative importance of non-point source in microplastic emissions to the aquatic system by comparison with the annual microplastic load from a WWTP that treats wastewater generated in the same area (Kim et al., 2022).

2. Materials and methods

2.1. Selection of study area

To assess microplastic emission from stormwater runoff according to urban land-use pattern, we selected the city of Gumi, where a separate sewer system is well-established, as the study area. Gumi is an industrial city located in the middle of the Nakdong River. It includes national industrial complexes that specialize in areas such as electronics and semiconductors, and covers an area of approximately 38 km^2, with 2300 companies and 95,000 employees (www.gumi.go.kr). Because Gumi was a planned city, separate sewers were constructed throughout the city in accordance with the national sewerage system plan (MOE, 2018), and districts were clearly divided for each land-use pattern (e.g., industrial, residential, or commercial). These characteristics facilitate comparative studies of pollution between point and non-point sources, and among land-use patterns.

We examined the stormwater drainage networks and land-use map of Gumi, then decided on two stormwater catchments for sample collection according to the following criteria: each catchment represents residential or industrial use, and each catchment discharges stormwater through a single drainage outlet. Finally, two stormwater outlets within the Gumi National Industrial Complex IV (hereafter referred to as Complex IV), one for a residential catchment and the other for an industrial catchment, were selected as study sites. The stormwaters are discharged directly into a stream, Han Cheon (a tributary of the Nakdong River) (Fig. 1).

2.2. Sample collection

Stormwater was sampled during three rainfall events in June 2021 and July 2021. A rain gauge (RG50, Environdata, Australia; Fig. S1) was installed on the rooftop of a building near the stream, and rainfall depth was measured during each rain event. The total rainfall per event was 2.2–61.6 mm. Detailed information regarding the drainage area and rainfall characteristics is provided in Table 1. At each site, an automatic water sampler (AS950, HACH, USA) with a flow meter (AV9000, HACH, USA) was installed on the day before the rain (Fig. S1). The velocity sensor of flow meter was installed on the bottom of each drain, and the flow rate data of stormwater calculated using flow velocity, depth of stormwater and width of drains was collected every second. Stormwater runoff samples were collected from the initial runoff at intervals of 0, 15, 30, 60, 120, 240, and 360 min, in accordance with the National Institute of Environmental Research method (NIER, 2012) (Table S1, Fig. 2). The sampling interval was determined according to the predicted rainfall time and field condition including rainfall intensity, stormwater flow rate, and water level in the drains. In total, 18, 12, and 14 stormwater samples were obtained in situ during Events 1, 2, and 3, respectively. Some of these samples were used for microplastics analysis in the lab. We attempted to collect the initial runoff at short time intervals until the peak flow was reached. Samples were collected to a total volume of 6 L, filtered through a 20-μm mesh hand net in situ, and transferred to pre-cleaned glass bottles. In addition, 1 L of stormwater was collected for analysis of suspended solids (SS) to compare the emission characteristics between particulate matter and microplastics during stormwater runoff. Since stormwater runoff samples were collected from stormwater drain outlets, microplastics in the runoff samples in this study included those that were deposited on ground surfaces and also retained in storm drains.

2.3. Microplastic analyses of stormwater samples

Microplastics in stormwater were analyzed with modification to the method suggested by Eo et al. (2019) and Song et al. (2021). Stormwater

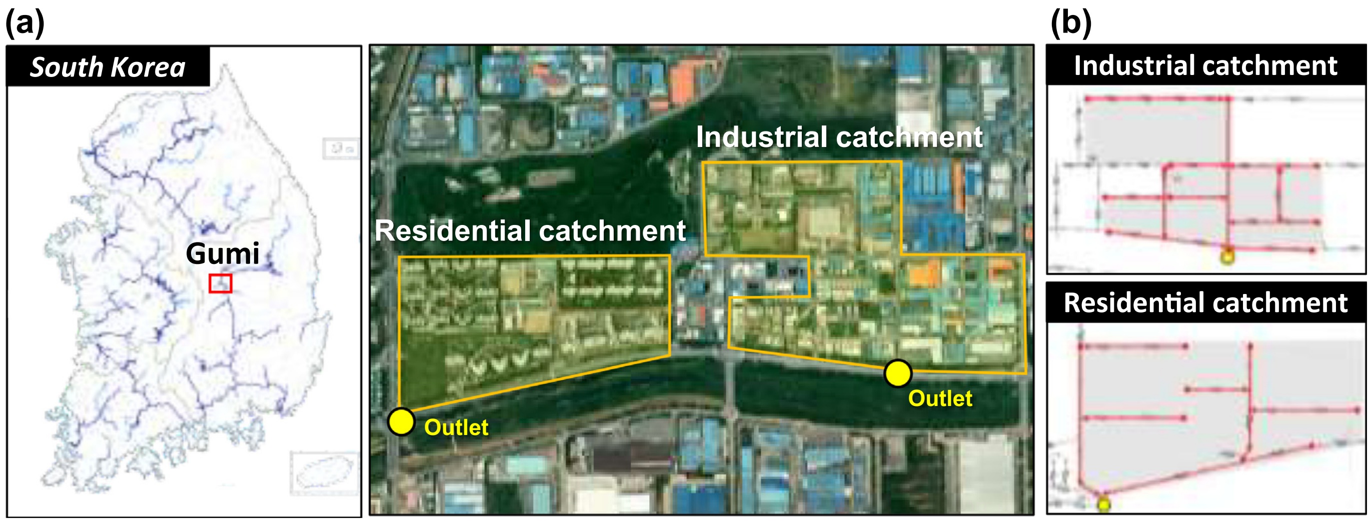

Fig. 1. (a) Map of the study area and sampling sites (yellow circles) of stormwaters in Gumi, Korea. Yellow shaded areas are industrial and residential catchments. (b) Stormwater drainage networks of the catchments.

samples were passed through a 20-μm stainless steel sieve and transferred to a glass beaker with HPLC-grade water. The samples were dried in an oven at 60 °C until all water was removed. To oxidize organic matter, wet peroxide oxidation was carried out by the addition of 20 mL of Fe (II) solution (0.05 mol/L) and 20 mL of 30 % hydrogen peroxide (H_2O_2; 35 %) at 60 °C for 30 min. This step was repeated until no organic matter was visible. The contents of the mixture were rinsed through a 20-μm sieve with HPLC-grade water, then transferred to a density separation funnel using lithium metatungstate (1.6 g/cm^3). After a settling time of >12 h, the precipitated particles were drained; particles in the supernatant were filtered onto stainless-steel filter paper with a pore size of 10 μm (13 or 25 mm Ø).

Microplastics larger than 20 μm were identified using a Fourier transform infrared microscope (Nicolet iN10 MX; Thermo Fisher Scientific, USA) with a linear array detector. Infrared mapping images were obtained using ultrafast mapping in reflection mode with a 25 μm × 25 μm step size. Images were obtained from one section (<12 mm × 12 mm) of 13-mm Ø filter paper and from three sections of 25-mm Ø filter paper. Each section required a time of approximately 30 min (0.113 s/scan). The infrared spectrum range was 4000–715 cm^{-1}, and the carbon dioxide peak in the range of 2200–2400 cm^{-1} was eliminated. The collected spectra were collated with the reference spectra by using the spectra profiling function in Picta software (Thermo Scientific, USA). The spectra of 13 polymer types collected from commercial products were used for profiling. These types included polypropylene (PP), polyethylene (PE), polyethylene vinyl acetate (PEVA), polyethylene terephthalate/polyester (PET/PES), polyamide, alkyd, polyepoxides, polyacrylate, polystyrene, polyurethane, polyvinyl chloride, polystyrene butadiene, and cellulose acetate (CA). The spectra of other polymers detected during profiling were added to the profiling. After spectra profiling, the polymer type, size (including the longest and shortest axes), and shape of the microplastics were manually identified and recorded. There was >70 % agreement with the library spectrum (Thermo Scientific, USA). The microplastic abundance was expressed as number of particles per 1 L of stormwater (n/L). The analysis of tire wear particles with large amount of carbon black using FT-IR is challenging (Werbowski et al., 2021), and then detailed results on these particles were limited in this study.

Microplastic analysis was performed on all stormwater samples (18 samples) collected during Event 1. In the consideration of the rainfall characteristics of each event and the results of Event 1 such as time variation in microplastic abundance, SS concentration and flow rate of stormwater, we selected samples from Event 2 (6 of 12 samples) and Event 3 (9 of 14 samples) for microplastic analysis. Detailed information about the recovery test, quality control, and statistical analyses is provided in the Supplementary material (S).

2.4. Mass concentration of microplastics

The microplastic abundance in stormwater was converted to mass concentration using the shape, size, and polymer density of each particle. It was

Table 1
Characteristics of (a) catchments studied and (b) rainfall events.

(a) Characteristics of catchments

	Latitude	Longitude	Drainage area (m^2)	Impermeable area (%)	Land slope (°)	Facility type (composition of area; %)	Population density (n/km^2)
Industrial catchment	36°8′ 15.34″N	128°26′ 39.36″E	363,359	84	1.68	Manufacturing (51 %; machine, electronic components, metals, etc.) > research institute (27 %) > commercial (9 %) > green area (2.9 %)	2,200
Residential catchment	36°8′ 12.32″N	128°25′ 45.74″E	257,700	60	4.51	Apartment (79 %) > school (12 %) > green area (8.5 %)	35,000

(b) Characteristics of rainfall events

Event	Rainfall date and time		Antecedent dry days (d)	Total rainfall (mm)	Rainfall duration (h)	Rainfall intensity (mm/h)	Runoff volume (m^3)		Runoff coefficient	
	Start	End					Industrial	Residential	Industrial	Residential
Event 1	21-06-03 10:00	21-06-04 3:00	2	27	17	1.59	6431	2974	0.66	0.43
Event 2	21-06-22 17:00	21-06-22 18:00	18	2.2	1	2.20	267	78	0.33	0.14
Event 3	21-07-05 20:00	21-07-06 8:00	1	61.6	12	5.13	16924	8918	0.76	0.56

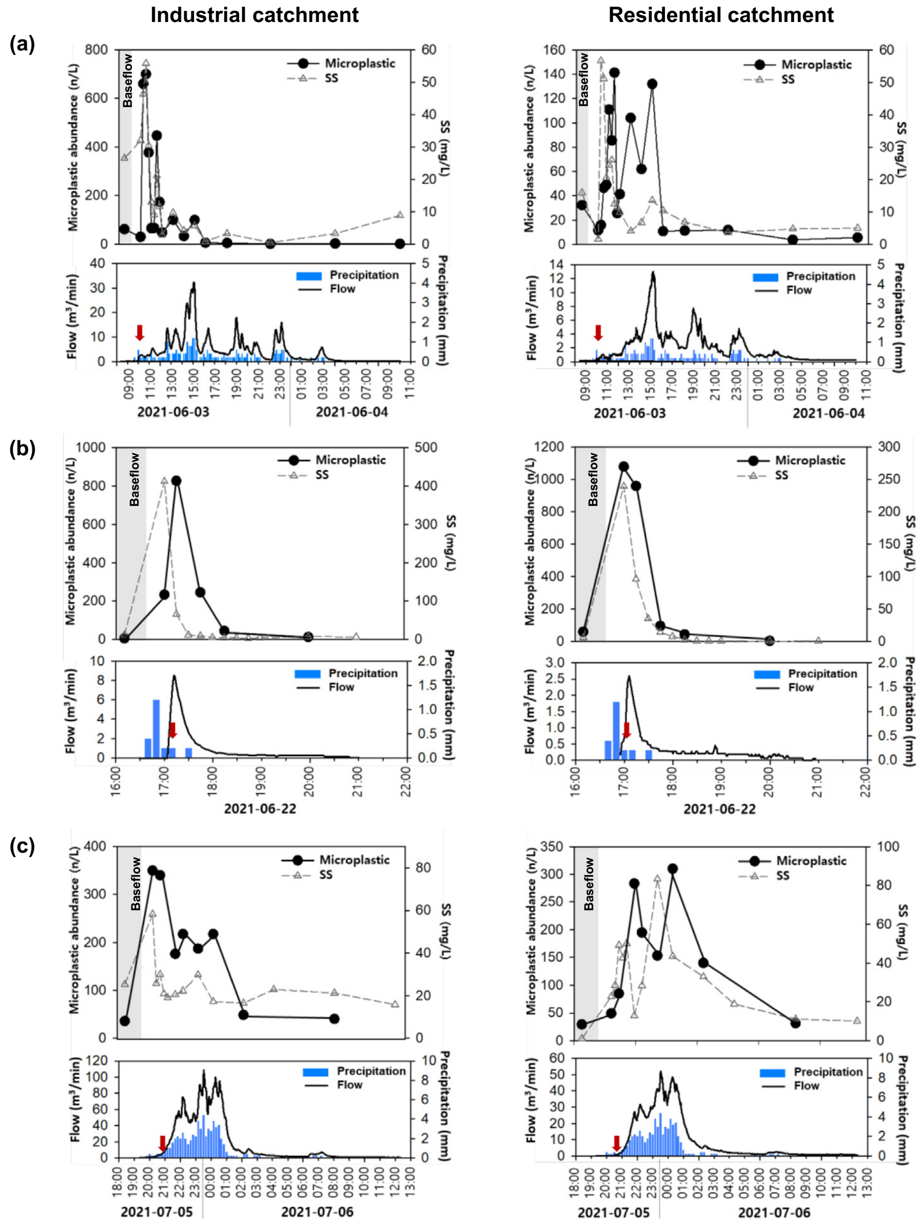

Fig. 2. Temporal changes in the concentrations of microplastics and suspended solids (SS) in stormwater, the flow rate, and the precipitation for industrial and residential catchments during Event 1 (a), Event 2 (b), and Event 3 (c). Light-gray section indicates baseflow before rainfall; red arrow indicates initiation point of stormwater runoff.

assumed that fragments and films were cuboid, fibers were cylindrical, and spheres were spherical.

$$\text{Mass of fragment and film} = \boldsymbol{l} \times \boldsymbol{w} \times \boldsymbol{h} \times \boldsymbol{d}.$$

$$\text{Mass of fiber} = \pi r^2 l \times d$$

$$\text{Mass of sphere} = \frac{3}{4}\pi r^3 \times d$$

where l and w are the length and width, respectively: the measured value of longest and shortest axes of particles; h is the height, and the minimum and maximum heights were assumed to be one-tenth and one-third of the longest axis, respectively (Poulain et al., 2018); d is density of polymers (Eo et al., 2021); r is the radius of fiber and sphere: the measured value.

2.5. Estimation of microplastic load through stormwater runoff

The microplastic load through stormwater runoff during an event was calculated using the following equation:

$$\sum_{n=1}^{N} (Q_n \times \Delta t_n \times A_n)$$

where Q_n, Δt_n, and A_n are the flow rate, sampling time interval, and microplastic concentration, respectively; N is the total number of samples. The event mean concentration (EMC) and microplastic load per drainage area were calculated through division of the microplastic load by the runoff volume and the drainage area, respectively.

The monthly and annual microplastic loads discharged from the entire area of Complex IV into the aquatic environment (Han Cheon) through stormwater runoff were estimated. The total rainfall was categorized into four classes (<10 mm, 10–30 mm, 30–50 mm, and >50 mm) in accordance with the National Institute of Environmental Research method (NIER, 2012). The microplastic load per rainfall event of each rainfall class was calculated through multiplication of the surface areas of industrial (2.57 km^2), residential (0.76 km^2), and commercial (2.16 km^2) catchments within Complex IV by the microplastic load per drainage area of each rainfall class (Table S2). The unobserved microplastic load per drainage area of the 30–50 mm rainfall class was extrapolated as the value calculated by applying the mean precipitation (39 mm) over the past 10 years to the quadratic regression equation that related microplastic load per drainage area to precipitation (Fig. S2). The microplastic load per unit area for the commercial area was extrapolated from the value of the residential area, and it was assumed that no microplastics were emitted from the green area. Finally, the monthly microplastic load was calculated through multiplication of the total microplastic load of each rainfall class by the mean annual rainfall frequency of each class and each month for 10 years (2011–2020; www.wamis.go.kr; Table S3). The microplastic load through baseflow was excluded in the calculation of microplastic load and estimation of monthly and annual microplastic load, since it was insignificant; mostly <0.0005 % of the total load with the exception of 0.2 % at the event 3 (with the fewest number of antecedent dry days) in the industrial catchment.

2.6. Analysis of SS in stormwater samples

Known volumes of stormwater samples (50–500 mL) were filtered through pre-dried and pre-weighed GF/F glass microfiber filters (47 mm; Whatman, UK). The filters were oven-dried at 60 °C for >24 h, then weighed. The SS value was calculated as the change in filter weight after drying (Eo et al., 2019).

3. Results

3.1. Microplastic abundance in stormwater

A total of 6316 microplastics were detected in stormwater runoff samples. The microplastic abundances in samples from industrial and residential catchments ranged from 1 to 827 n/L and 2 to 1080 n/L, respectively (Fig. 2, Table S1). During the three events, the EMC of microplastics from the industrial catchment ranged from 68 to 568 n/L, while the EMC of microplastics from the residential catchment ranged from 54 to 639 n/L (Table 2). In both catchments, the EMC of microplastics in stormwater was highest during Event 2, but there was no significant difference among the three rain events (Kruskal–Wallis test, $p > 0.05$). The highest microplastic abundance in stormwater from the industrial catchment was observed within 1 h (sample #1–5) from the beginning of the runoff; the highest microplastic abundances in stormwater from the residential catchment were observed immediately after the beginning of runoff and after the flow rate had increased.

3.2. Concentration of SS in stormwater

The concentration of SS was also highest during Event 2, especially immediately after the initiation of runoff (#1–3; Fig. 2, Table S4). A significant correlation between the concentration of SS and the abundance of microplastics was observed only during Event 1 from the industrial catchment ($r = 0.84$, $p < 0.001$) and during Event 2 from the residential catchment ($r = 0.91$, $p < 0.05$) (Fig. S3).

3.3. Characteristics of microplastics in stormwater

Microplastics with various characteristics were observed in the stormwater samples (Fig. S4). The longest dimension of the microplastics ranged from 20 to 6904 μm. The most common size class of non-fiber particles (including fragments, films, and spheres) was 20–100 μm, and the distribution showed decreasing composition with increasing size (Fig. 3(a)). The fiber particles had peaks in the size classes of 100–200 and 200–300 μm (Fig. 3(b)). All shapes, including fragments, fibers, spheres, and films, were observed in the stormwater samples (Fig. 3(c), Table S6). Fragments were the most abundant shape, followed by fibers, in both catchments. Spheres and films were found rarely. In total, 27 types of polymers were detected in the stormwater samples (Fig. 3(d)). More various polymer types were observed in industrial catchment samples (26 types) than in residential catchment samples (20 types). PP and PE were major polymer types, constituting >60 % of the total microplastics. The polymer composition of fragments in the industrial catchment was PP (52 %) > PE (20 %) > PET/PES (4 %) = polyacrylate (4 %); the composition in the residential catchment was PP (49 %) > PE (17 %) > polyacrylate (12 %) (Table S7). The polymer composition of fibers in the industrial catchment was PP (49 %) > PET/PES (34 %) > polyacrylate (4 %); the composition of fibers in the residential catchment was PP (56 %) > CA (14 %) > PET/PES (12 %).

Table 2
Number- and mass-based loads, event mean concentrations (EMCs), and load per drainage area values of microplastics from stormwater runoff in industrial and residential catchments. Microplastic loads were estimated using microplastic abundance and stormwater flow rate during runoff. Mass-based microplastic load was estimated considering the shape, size, and density of microplastics. EMC and microplastic load per drainage area were calculated through division of the microplastic load by the flow rate and the drainage area, respectively. Details are provided in the Materials and methods.

	Industrial catchment	Residential catchment
Number-based microplastic load (n)		
Event 1	3.29×10^8	1.33×10^8
Event 2	1.54×10^8	0.63×10^8
Event 3	4.61×10^9	2.85×10^9
Mass-based microplastic load (kg)		
Event 1	0.25–0.67[a]	0.10–0.29
Event 2	0.07–0.21	0.004–0.01
Event 3	0.87–2.47	1.91–5.71 (0.78–1.95)[b]
Number-based EMC (n/L)		
Event 1	68	54
Event 2	568	639
Event 3	188	226
Mass-based EMC (μg/L)		
Event 1	51–137	40–118
Event 2	243–776	44–115
Event 3	35–100	152–454
Number-based microplastic load per drainage area (n/m^2)		
Event 1	905	516
Event 2	424	244
Event 3	12,687	11,059
Mass-based microplastic load per drainage area (mg/m^2)		
Event 1	0.69–1.84	0.28–0.80
Event 2	0.16–0.58	0.01–0.03
Event 3	2.39–6.80	5.26–15.71 (2.15–5.37)[b]

[a] Minimum–maximum.
[b] Value excluding two particles with diameter >1 mm that constituted 60–70 % of the total mass-based load.

3.4. Microplastic load through stormwater runoff

The calculated total load of microplastics through stormwater runoff is shown in Table 2. The number-based microplastic load discharged during the three events from the industrial catchment was 154 to 4610 million particles, while the load from the residential catchment ranged from 63 to 2850 million particles. The mass-based microplastic load from the industrial catchment ranged from 0.07 to 2.47 kg, while the load from the residential catchment ranged from 0.004 to 5.71 kg. The number-based microplastic load per drainage area during the three events was slightly higher in industrial catchment (424–12,687 n/m^2) than in the residential catchment (244–11,059 n/m^2), but the difference was not statistically significant (Wilcoxon signed-rank test, $p > 0.05$). The mass-based microplastic load per drainage area also tended to be higher in the industrial catchment than in the residential catchment, except during Event 3. In the residential

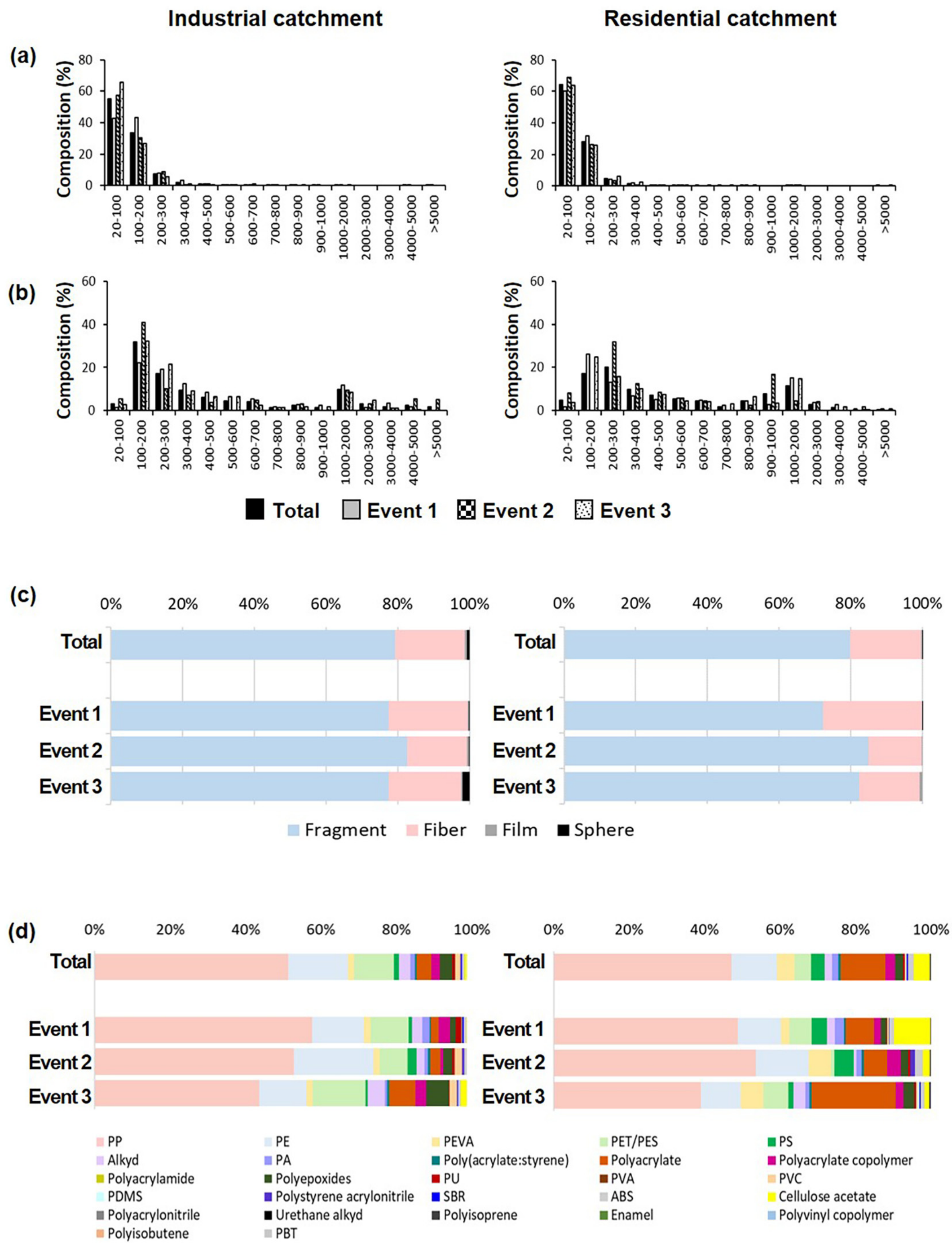

Fig. 3. Characteristics of microplastics in stormwater from industrial (left) and residential (right) catchments. Size distributions of non-fibers (a) and fibers (b). Compositions of shape (c) and polymer type (d).

catchment during Event 3, two polyarylate particles with longest and shortest diameter > 1 mm were detected; these constituted 60–70 % of the total weight-based load. Excluding these two particles, the microplastic load per unit area in the residential catchment was 2.15–5.37 mg/m^2, which was generally lower than the value in the industrial catchment (2.39–6.80 mg/m^2). The annual load of microplastics discharged into the Han Cheon from Complex IV through stormwater runoff was estimated to be 287 billion particles (Table S5). Approximately 90 % of the total microplastic load was emitted at the beginning of rainfall, within 25 %, 19 %, and 37 % of the storm duration during Events 1, 2, and 3, respectively (Fig. 4).

4. Discussion

4.1. Factors influencing microplastic emission through stormwater runoff in urban catchments

4.1.1. Urban land-use pattern

The level of microplastic contamination in stormwater is expected to vary according to land-use patterns and human activities. This study investigated microplastic contamination in stormwater runoff from industrial and residential catchments in one city to identify differences in the amount and characteristics of microplastic contamination according to urban land-

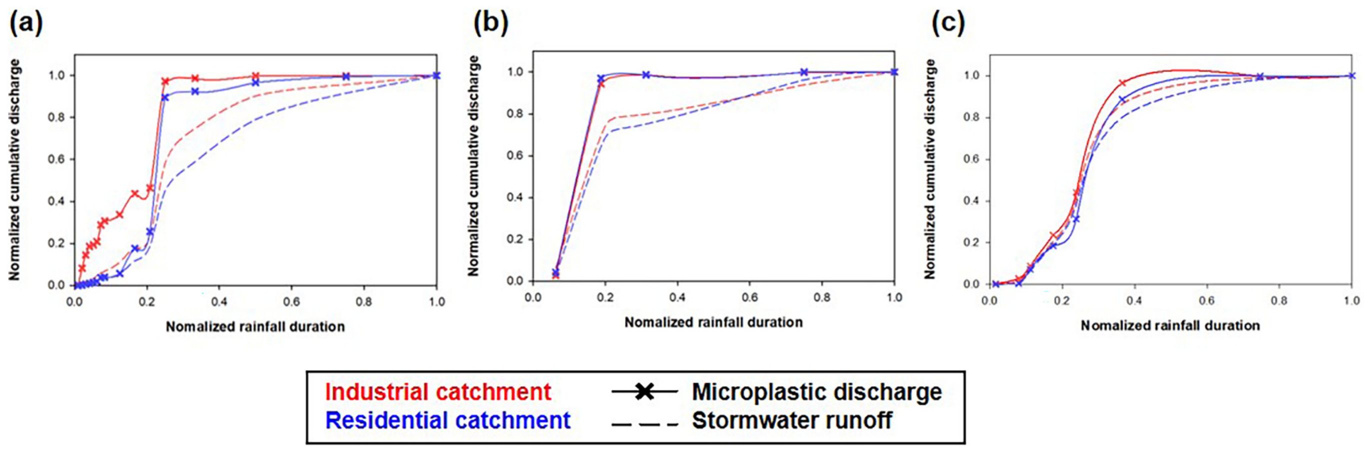

Fig. 4. Cumulative distributions of microplastic discharge and stormwater runoff during (a) Event 1, (b) Event 2, and (c) Event 3. Red and blue lines indicate industrial and residential catchments, respectively.

use patterns. In terms of the emission load, the microplastic load per unit area did not significantly differ between industrial and residential catchments (Wilcoxon signed-rank test, $p > 0.05$; Table 2). Previous studies have shown that the level of microplastic contamination is positively correlated with the population density of the catchment area (Browne et al., 2011; Yonkos et al., 2014). A large floating population during working hours in an industrial catchment that has lower a population density (2200 people/km^2) than a residential catchment (35,000 people/km^2) can result in comparatively greater microplastic emission from the industrial catchment. The load per unit area of PET/PES in both catchments, which mainly originates from human-worn items such as clothing, backpacks, and bags, was comparable (industrial catchment (I): 23–660 n/m^2, residential catchment (R): 16–394 n/m^2). De Falco et al. (2018) found that the number of fibers released into the air from clothes for everyday wear was comparable to the number discharged into wastewater after laundering.

However, microplastics can also be generated from industrial activities. Because Complex IV specializes in components such as electronics, machines, and semiconductors (Table 1), there are no typical plastic pollution sources (e.g., plastic, textile, and rubber manufacturing facilities) within the study site. Nevertheless, the composition and EMC of each polymer differed according to land-use pattern, except for major polymers such as PP and PE (Fig. 3(a), Table S7). Green spherical polyepoxide particles of 20–30 μm were only detected in the industrial catchment; they were particularly abundant during Event 3 (1.50 %, EMC: 3.35 n/L) (Fig. S4). Spherical particles are classified as primary microplastics, which are intentionally manufactured for use in industrial and medical products, as well as personal care products (Yurtsever and Yurtsever, 2019). They may also originate from the epoxy powder coating process, which is widely used in mechanical, electronic, and chemical engineering (Van Horn, 2011; Du et al., 2016). Although these particles are typically controlled as industrial waste after use, mismanaged particles can enter the environment.

In the residential catchment, polyacrylate was the most abundant polymer after PP and PE. Fragment-type polyacrylate was more abundant in the residential catchment (12.25 %, EMC: 8.88–51.14 n/L) than in the industrial catchment (3.75 %, EMC: 0.07–21.10 n/L). Polyacrylate is commonly used in paint and coating formulations (Khataee et al., 2016; Ajekwene, 2020); it can originate from road markings, building coatings, and roof waterproofing (Gaylarde et al., 2021). All of paint particles (white, $n = 5$) collected from roadside and exterior wall of a building in study area were identified as polyacrylate. The paint and coating layers can be degraded and released under the influences of surface texture, material quality, and weather (Chai et al., 2014). Notably, the abundant and wide roads in school zones in the residential catchment are paved with dark red paint (Fig. 5(a)). These road pavements can weather easily and release particles during the frequent friction caused by vehicles (Vijayan et al., 2019). The polyacrylate detected in the residential catchment was mainly red (red 83 % ≫ white 4 % = orange 4 % > translucent 2 %), whereas the polyacrylate in the industrial catchment was more evenly distributed among red (35 %), white (16 %), translucent (13 %), and blue (11 %). The spectra of the red polyacrylate particles were consistent between stormwater samples and roadside area; they demonstrated a >75 % match with the reference spectrum in the library. These results provide strong evidence that microplastic contamination in stormwater can directly reflect contamination characteristics in the drainage area.

4.1.2. Topographic characteristics

The physical characteristics of a drainage area, such as surface imperviousness and land slope, can affect the discharge of stormwater that transports microplastics to an aquatic system. More impervious surfaces and steeper slopes can increase the stormwater runoff volume and pollutant load (Chithra et al., 2015; Zhang et al., 2019); they can also cause a more intense "first flush" of stormwater and pollutants (Lee and Bang, 2000). In this study, a slightly higher microplastic load per unit area and a stronger "first flush" effect were observed in the industrial catchment (higher impermeability), compared with the residential catchment (greater land slope) (Tables 1 and 2, Fig. 4), implying that surface imperviousness had a greater effect on the microplastic load, compared with land slope. However, as observed during Event 3, the impacts of physical properties on the microplastic load per unit area tended to decrease with increasing total rainfall and increasing rainfall intensity (Table 1 and 2). Zhang et al. (2019) also reported that the effect of rainfall intensity on pollutant load was greater than the effect of topographic condition.

4.1.3. Rainfall characteristics

The highest EMC and concentration of microplastics were observed during Event 2, which had the highest number of antecedent dry days (ADDs). Among rainfall characteristics (total rainfall, ADDs, and hourly rainfall intensity), only the number of ADDs was significantly correlated with the EMC of microplastics ($r = 0.94, p < 0.05$). Pollutant accumulation on catchment surfaces during the dry period increases with the number of ADDs (Vaze and Chiew, 2002). Therefore, more ADDs can lead to higher concentrations of microplastics. The microplastic abundance in the stormwater runoff was high during the early part of each rainfall event, which is regarded as the "first flush" phenomenon (Figs. 2 and 4). The flushing of accumulated pollutants from road surfaces, structures, and sewers can occur during the early runoff period (Lee and Bang, 2000). Additionally, airborne microplastics can be washed to the surface by rainfall, especially at the beginning of a rainfall event (Abbasi, 2021).

The microplastic abundance in stormwater during a rainfall event also tended to vary according to rainfall intensity and flow rate (Fig. 2). In Event 2, a single-peak rainfall event, the microplastic abundance also

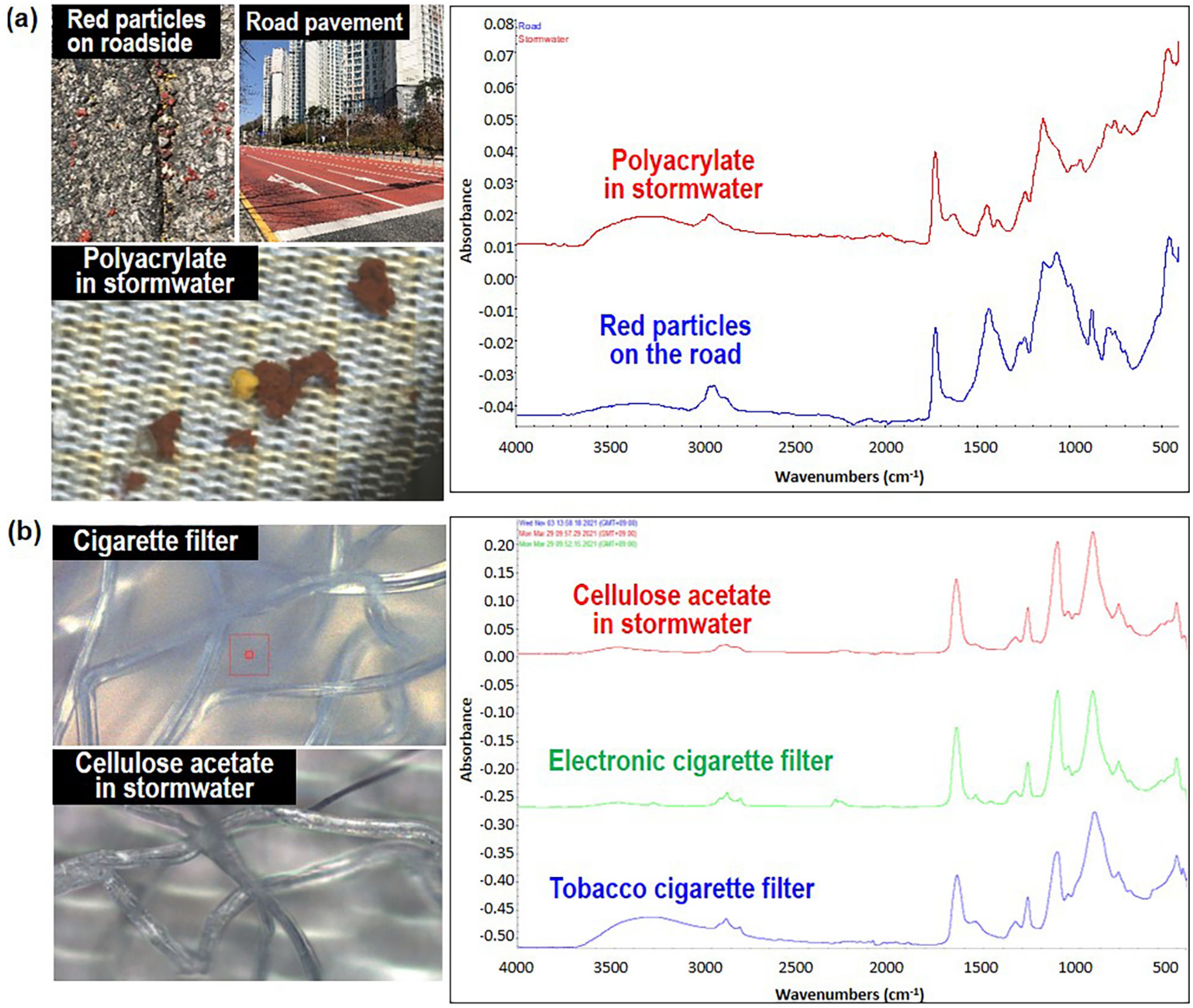

Fig. 5. Photographs of microplastics and their transform-infrared spectra. (a) Red particles collected from the roadside of red pavement and red polyacrylate in stormwater samples. (b) Cigarette filter and cellulose acetate (CA) fiber in stormwater samples.

showed a single peak, and most microplastics were discharged within the early runoff period (Fig. 4). In Events 1 and 3, a secondary flush effect occurred because of the bimodal distribution of the rainfall. Because the rainfall intensity and flow rate increased sequentially, the microplastic abundance showed a tendency for further increase and an additional peak in the residential catchment. Significant correlations between microplastic abundance and rainfall intensity ($r = 0.83$, $p < 0.05$) and flow rate ($r = 0.80$, $p < 0.05$) were only observed in the residential catchment during Event 3. Along with microplastics, the SS concentration in stormwater also varied according to rainfall intensity and flow rate during a rainfall event (Fig. 2). However, only a partial a correlation between microplastics and SS was observed (Fig. S3). In general, SS eluted prior to microplastics (Fig. 2). SS was sampled with a smaller cut-off size (0.7 μm) than microplastics (20 μm); therefore, the fast-moving nature of the fine particles may have caused SS to show a faster elution peak, compared with microplastics. Among the three events, the largest number- and mass-based loads of microplastics were observed during Event 3, which had the largest runoff volume and rainfall (Table 1 and 2). The microplastic load was positively correlated with total rainfall (I: $r = 0.92$, $p > 0.05$; R: r = 0.92, $p > 0.05$) and flow rate of stormwater (I: $r = 0.94$, $p > 0.05$; R: $r = 0.95$, $p < 0.05$).

The compositions of microplastic characteristics (size, polymer type, and shape) changed according to runoff time (baseflow, initial 30 % of the runoff period, and subsequent 70 % of the runoff period) (Fig. S5). The low-density polymers (density lower than the density of fresh water [<1.00 g/cm^3]), including PP, PE, PEVA, and polydimethylsiloxane (Eo et al., 2021), were most abundant in stormwater from baseflow (83 %), followed by initial runoff (71 %) and subsequent runoff (58 %). The size composition of fiber particles showed that larger fibers were more abundant in later stormwater runoff (<300 μm: 41.48 %, 300–1000 μm: 35.76 %, >1000 μm: 22.76 %) than in baseflow (<300 μm: 49.54 %, 300–1000 μm: 37.39 %, >1000 μm: 13.07 %) or earlier stormwater runoff (<300 μm: 56.01 %, 300–1000 μm: 30.15 %, >1000 μm: 13.83 %). Thus, larger and heavier particles that require higher flow for transport can be released during the later stage of a rainfall event. This is consistent with the finding by Morgan et al. (2017) that finer SS was transported during the initial part of the rain event. This result implies that higher rainfall energy is needed to wash out coarser and heavier microplastics.

To summarize the discharge characteristics of microplastics, first, the level of microplastic contamination did not significantly differ between residential and industrial catchments in Gumi, but the polymer-type composition reflected the contamination characteristics of each land-use pattern. Second, the rainfall characteristics, rather than the topographic characteristics, were the critical factors that influenced microplastic loading. Third, the abundance of microplastics in stormwater was correlated with the number of ADDs, which increased the accumulation of microplastics on surfaces. Fourth, the microplastic abundance in stormwater was generally high in the early runoff period and co-varied according to the rainfall intensity during a rainfall event. Fifth, the total microplastic load was strongly affected by rainfall and runoff volume; most of the load was discharged in the initial runoff period. The results of this study imply that the abundance and characteristics of microplastics varied according to land-use pattern, rainfall characteristics, and runoff time. To understand the behavior of microplastics discharged through stormwater runoff, there is a need to consider time-weighted sampling, which necessarily includes the initial stormwater runoff, and to monitor microplastic contamination according to the land-use characteristics and rainfall.

4.2. Comparison with the point source and aquatic environment

Kim et al. (2022) investigated microplastic contamination in the effluent from a WWTP located in Complex IV, Gumi. The WWTP treats wastewater (approximately 50 million L/day) from households (~20 %) and industries (~80 %, Complex IV) located around the Han Cheon watershed. The microplastic concentration in the effluent of the WWTP was 0.39 ± 0.16 n/L, which is significantly lower than the values observed in stormwater samples in the present study (*t*-test, $p > 0.05$; Fig. 2, Table 2). They estimated that the annual microplastic load from the WWTP into the stream was approximately 2.9 billion particles. These result explains that stormwater runoff (287 billion particles) contributes 99 % of the total microplastic load, and WWTP effluent contributes 1 % of them (Table S5). This result clarifies that non-point source emission serves as the dominant pathway for transporting microplastics into aquatic environments. In a study of the Baltic Sea, Schernewski et al. (2021) found that a significant amount of microplastics enters through non-point sources (62 %) among urban sources (WWTPs: 25 %). According to our calculation (Table S5), the annual microplastic load entering Han Cheon was estimated to be higher in the wet season, which constituted 68 % of the annual load, because the total precipitation was concentrated in the wet season (64 % of annual precipitation). The sampling in this study was only carried out during the wet season, with short ADDs; thus, the microplastic concentration during the dry season, with long ADDs, might have been underestimated. Nevertheless, the seasonal microplastic loading pattern concentrated in the wet season would not change because the total microplastic load is heavily influenced by total rainfall. Eo et al. (2019) reported that the highest microplastic abundance in the Nakdong River, of which Han Cheon is a tributary, was observed during the wet season; the microplastic load discharged from the Nakdong River into the ocean was also concentrated during the wet season (70–80 %). These results imply that non-point sources, rather than point sources, can be the major input source of microplastics, especially during the wet season.

The characteristics (size distribution and polymer composition) of microplastics in stormwater was similar with those in effluent of the WWTP in the study area (Kim et al., 2022). The particles smaller than 200 μm were predominant (stormwater: 77 %; WWTP: 88 %), and PP, PE, and PES/PET were major polymer types (stormwater: 64–78 %; WWTP: around 80 %, excluding winter). Interestingly, the composition of fiber was higher in WWTP effluent (22–60 %) than stormwater (15–28 %). This result might be due to the inflow of fiber-enriched sewage to the WWTP (fiber composition in influent: 25–37 %) and the lower removal efficiency of fibers than fragments in WWTP (Kim et al., 2022). The major characteristics of microplastics in stormwater were also similar with fresh water (Nakdong River) and coastal seawater of Korea (Eo et al., 2018; Song et al., 2018; Cho et al., 2021). The fragment is the predominant shape; PP, PE, and PET/PES are the major polymers, constituting 60 % of the types. However, the composition of specific microplastics differed with increasing distance from land to sea. The sphere was more abundant in stormwater (0.35 % overall; 0.73 % in the industrial catchment) than in seawater (0.15 %; Cho et al., 2021). Spherical microplastics are mainly used on land (IUCN, 2017); these usage patterns were closely reflected in the water column. In addition, more diverse polymer types were detected in stormwater (27 types) than in seawater (16–22 types; Song et al., 2018; Cho et al., 2021). Among the polymer types, CA was more abundant in stormwater from the residential catchment (4.25 %) than in seawater (<1 %; Song et al., 2018). Although there is ongoing debate concerning whether to classify CA as synthetic polymer, which is used primarily in the production of cigarette filters (84 % of total CA production; Puls et al., 2011), it (UNEP, 2021) may be meaningful as a man-made fiber. Most of the CA fiber in stormwater was transparent and had diameters of 20–30 μm, which is similar to the characteristics of CA in commercial cigarette filters (Fig. 5(b)). Moreover, the spectra of CA fibers in stormwater samples and commercial products were very similar.

4.3. Comparison with other stormwater studies

The abundances and characteristics of microplastics that we observed in stormwater were compared with the findings in other studies (Table S8). Because most studies of microplastic contamination in stormwater did not present EMC in terms of the total flow rate, simple mean abundances of microplastics were used for comparisons among studies (I: 160–228 n/L, R: 50–373 n/L, Table S1). Irrespective of land-use pattern and sample collection time, the microplastic abundances (50–373 n/L) in stormwater from Korea were comparable to values from Mexico (88–289 n/L; Piñon-Colin et al., 2020), Canada (186 n/L; Smyth et al., 2021), and France (3–129 n/L; Treilles et al., 2021); they were higher than values from China (2.75–19.04 n/L; Sang et al., 2021), Hong Kong (1.4–6.8 n/L; Mak et al., 2020), and the United States (11–24.6 n/L; Werbowski et al., 2021, 0.3–0.37 n/L; Boni et al., 2022). In comparisons of studies that analyzed stormwater samples from industrial and residential areas, the microplastic abundance in stormwater runoff within the initial 30 min in Mexico (I: 180 n/L, R: 128–289 n/L; Piñon-Colin et al., 2020) was lower than the abundance from the same runoff period in the present study (I: 449 ± 287 n/L, R: 321 ± 479 n/L). The abundances from China (I: 16.54–19.04 n/L, R: 10.46–12.17 n/L; Sang et al., 2021) and from retention ponds and wetlands in Denmark (I: 5.25–11.35 n/L, R: 0.49–1.4 n/L; Liu et al., 2019) and Australia (I: 26 n/L, R: 17 n/L; Monira et al., 2022) were comparable to abundances from the later runoff period in the present study (Table S8). The microplastic abundances in stormwater from industrial and residential catchments in Korea, China, and Denmark were higher than the abundances in samples from roads and highways in each country (Korea: 8.6–24.3 n/L, Yano et al., 2021; China: 5.08–6.29 n/L, Sang et al., 2021; Denmark: 0.49 n/L, Liu et al., 2019). PP, PE, and PET/PES were generally abundant polymer types, regardless of land-use pattern (this study; Liu et al., 2019; Olesen et al., 2019; Mak et al., 2020; Ziajahromi et al., 2020; Sang et al., 2021; Smyth et al., 2021; Treilles et al., 2021; Werbowski et al., 2021; Monira et al., 2022; Herath et al., 2022), but rubber was a dominant polymer type in samples from highways (Lange et al., 2021; Yano et al., 2021). The most abundant microplastic shape was the fragment or fiber, but no clear patterns were observed according to region, sampling time, or land-use pattern. The number of studies concerning microplastic contamination in stormwater has increased in recent years; however, comparisons among these studies are limited because no standardized protocol has been used. The cut-off size during sample collection and analysis, which can directly affect the final result because of the potential for missing small particles (Eo et al., 2019), varied from 6 to 300 μm (Yano et al., 2021; Monira et al., 2022). The application of a density separation solution with low density cannot cover a high-density polymer; however, various solutions with densities ranging from 1.2 to 1.9 g/cm^3 were used (Lange et al., 2021; Boni et al., 2022). In addition, the proportion of chemically identified particles among the total particles on a filter can influence the final result (Song et al., 2015; Cho et al., 2021). Although most studies have analyzed microplastics using chemical identification and attempted to apply it to a large proportion of the total suspected particles, the proportion of identified particles has varied among studies; in some studies, it has been unclear whether chemical analysis results were described. Therefore, comparisons among studies should consider the analytical method used in each study. Additionally, differences in sampling times and intervals, as well as insufficient information concerning sample collection, limit comparisons of data. Because microplastic abundance considerably varies during a rainfall event, protocols from sampling to presentation of results should be developed to reduce data gaps and present reliable data. In particular, a detailed method for sample collection should be proposed that considers the microplastic discharge characteristics in stormwater runoff.

5. Conclusion

The continuous input of microplastics into aquatic systems can present risks and harmful impacts to ecosystems and human health. There is a growing body of research concerning the sources, pathways, behaviors, and fates of microplastics. However, information on microplastics entering aquatic environments through non-point sources remains limited, especially temporal dynamics of microplastic discharge through stormwater runoff. This study provides understanding of the intra- and inter-event variations in microplastic emission characteristics (abundance, shape, size and polymer types) from stormwater runoff during rainfall events in urban area with different land-use patterns (industrial and residential catchments). The findings of this study indicated that the rainfall characteristics (e.g., total rainfall, ADDs, and rainfall intensity) and land-use pattern were important factors influencing discharge loads or characteristics of microplastics between and within rainfall events, and that the initial stormwater runoff during a rainfall event carries a considerable portion of the microplastic load. Surprisingly, stormwater runoff contributed a significant portion (99 %) of microplastic load to the stream, while that of WWTP effluent (1 %) was negligible. This finding means that non-point source is a key pathway for transporting microplastics into aquatic environments. Because most stormwater is discharged into the environment without any treatment, management strategies should be developed to reduce microplastic emission. Furthermore, continuous monitoring of microplastics from non-point sources, considering land-use patterns and rainfall characteristics, is needed to understand the behavior of microplastics during movement from terrestrial environments to aquatic environments. Finally, detailed guidelines for monitoring microplastics in stormwater runoff—especially for sample collection, considering the temporal variability of microplastic elution during stormwater runoff—should be developed to ensure data compatibility among studies.

CRediT authorship contribution statement

Youna Cho: Formal analysis, Investigation, Visualization, Writing – original draft. **Won Joon Shim:** Conceptualization, Investigation, Supervision. **Sung Yong Ha:** Investigation, Methodology. **Gi Myung Han:** Investigation, Methodology. **Mi Jang:** Investigation, Methodology. **Sang Hee Hong:** Conceptualization, Methodology, Investigation, Funding acquisition, Project administration, Supervision, Writing – review & editing.

Data availability

No data was used for the research described in the article.

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.

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Appendix A. Supplementary data

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