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Baseline

Distribution, compositional characteristics, and historical pollution records of microplastics in tidal flats of South Korea

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ABSTRACT

Studies on distribution of microplastics (MPs) in sediments of tidal flats are relatively scarce compared to other coastal areas. In this study, spatial and vertical distributions and compositions of MPs in tidal flat sediments along the west coast of Korea were investigated. The abundance of MPs in surface and core sediments ranged from 20 to 325 and 14 to 483 particles per 50 g dry weight, respectively. Polypropylene (51%) and polyethylene (36%) were the most dominant MPs; the size was <0.3 mm, and the shape was mostly fragments followed by fibers. The abundance of MPs in sediments has increased rapidly since the 1970s, and recently showed a slight decrease. Surface morphology of MPs analyzed using a scanning electron microscope revealed that the MPs in tidal flats were highly weathered mechanically and/or oxidatively. The results of this study provide valid baseline data on distributions of MPs in tidal flats.

Plastic usage has been steadily increasing since the 1950s, with production reaching 370 million tons in 2019 (PlasticsEurope, 2020). However, out of approximately 6300 million metric tons of plastic waste in 2015, only 9% was recycled, whereas 12% was incinerated, and 79% was accumulated in the natural environment (Geyer et al., 2017). Plastic waste properly enters the ocean via various pathways, such as wind, sewage discharge, rivers, and maritime activities (Moore et al., 2001; Murphy et al., 2016; Ryan et al., 2009). These plastics degrade into millions of small invisible particles, termed "microplastics" (MPs), which are defined as particles <5 mm in size (Law and Thompson, 2014). They are divided into primary and secondary MPs according to the generation process (Cole et al., 2011). Primary MPs refer to products manufactured in a microscopic size in industrial or domestic applications, including facial cleansers, toothpaste, resin pellets, and cosmetics (Auta et al., 2017). Secondary MPs are generated by the gradual decomposition of plastics through biodegradation, photolysis, and thermal oxidative degradation processes (Andrady, 2011). In addition, it is produced by weathering of large plastic debris, and they are considered the major source of marine MPs (Browne et al., 2010).

MPs are widely distributed in the world's ocean, including estuarine and coastal areas, as well as deep sea and polar regions (Aguilera et al., 2016; Desforges et al., 2014; Lusher et al., 2015; van Cauwenberghe et al., 2013). These small-sized MPs are likely to be ingested by various marine organisms, including filter feeders, deposit feeders, and detritivores (Thompson et al., 2004). Ingested MPs cause not only physical effects of structural damage and inflammatory responses but also cause toxic effects on organisms, such as lethality and genetic damage (Franzellitti et al., 2019). MPs adsorb organic compounds in the water column and can be concentrated to several orders of magnitude greater than those of the surrounding environment. (Rodrigues et al., 2019). Consequently, MPs could cause adverse effects on organisms due to adsorbed organic pollutants and potentially transport toxic substances through the food chain (Teuten et al., 2009). Humans are exposed to MPs by various routes (Thompson et al., 2009), and MPs have been detected in human feces, blood, and lungs (Jenner et al., 2022; Leslie et al., 2022; Schwabl et al., 2019). Thus, it is very important to investigate the distribution, fate, and potential risk of MPs in coastal environments.

Coastal sediments are considered to be a major sink of MPs; MPs

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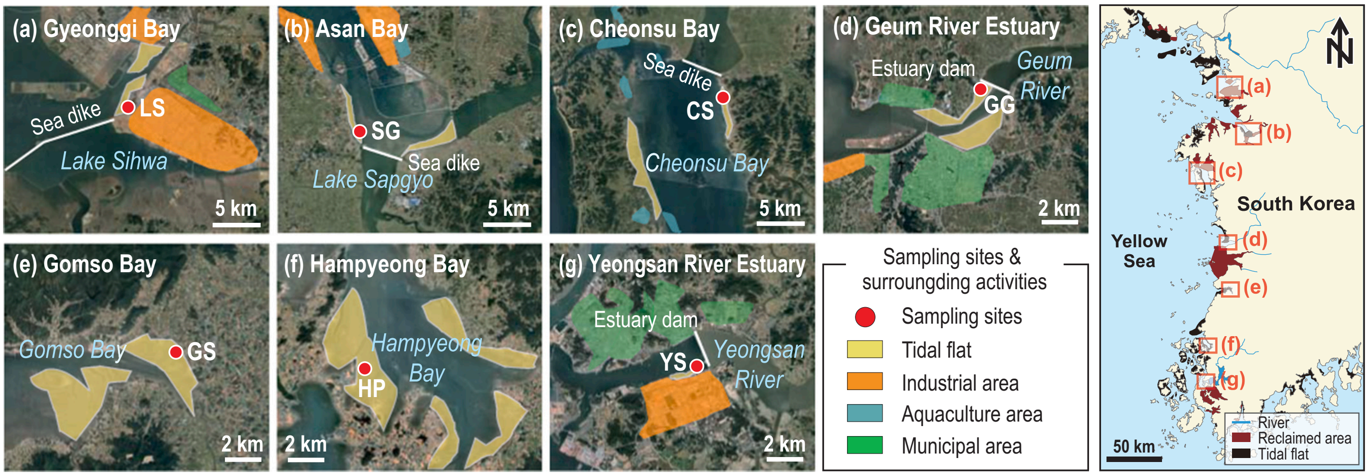

Fig. 1. Map showing the sampling sites and surrounding anthropogenic activities in tidal flats along the west coast of Korea.

introduced into the coastal environment become denser through biofouling, etc., and denser MPs can settle down to the seabed and accumulate in sediments (Wright et al., 2013). The behavior of MPs in the coastal environment seems to be similar to that of suspended sediments with similar physicochemical properties (Kane and Clare, 2019). Large amounts of MPs have been dispersed in marine environments over the past few decades (Harris, 2020). Assuming that the MPs deposited over time do not degrade, the temporal trend of MPs contamination in the core sediments can be recorded in the core sediments (Turner et al., 2019). Previous studies have successfully determined the pollution history of MPs recorded in core sediments in the marine environment, and sediment dating has been used to measure MPs inventory (Phuong et al., 2021; Uddin et al., 2021). For example, in the Belgian coast, MPs tended to continuously increase from 1993 to 2008 (Claessens et al., 2011), and MPs in Hangzhou Bay, China, increased from the 1980s to the 2000s, and thereafter no further increase (Li et al., 2020). Until now, studies on the distribution and composition of MPs in sediments have been mainly conducted in estuaries, beaches, and coastal areas, and relatively little research has been conducted on MPs in tidal flat sediments.

The west coast of Korea supports vast, well-developed tidal flats covering an estimated 2100 km^2 area (Koh and Khim, 2014). Tidal flats play an important role in natural purification, carbon accumulation, and seafood production (Kim et al., 2020; Koh and Khim, 2014; Lee et al., 2021). In addition, diverse marine benthic animals inhabit the tidal flats of Korea, with the biodiversity of this region being internationally recognized (Costello et al., 2010). The tidal flats of the west coast of Korea are naturally affected by the estuaries of large rivers, such as the Han River, Geum River, and Yeongsan River, and are also affected by artificial coastal developments, such as reclamation and embankment construction (Choi, 2014; Hong et al., 2010). Studies on the distribution and composition of MPs and the history of pollution in the tidal flats of Korea are very scarce. The objectives of the present study were to determine the spatial and vertical distribution and compositional characteristics of MPs in tidal flats. In addition, the surface morphology of MPs found in tidal flats was observed using a scanning electron microscope (SEM).

Seven representative tidal flats along the west coast of Korea were selected (Fig. 1). Study sites included Gyeonggi Bay (outer region of Lake Sihwa, LS), Asan Bay (outer region of Lake Sapgyo, SG), Cheonsu Bay (CS), Geum River Estuary (GG), Gomso Bay (GS), Hampyeong Bay (HP), and Yeongsan River Estuary (YS) (details in Table S1 of the Supplementary Materials). Surface and core sediments were collected in July 2020 and January 2021, respectively. The top 5 cm of surface sediment was collected using a stainless-steel spoon in triplicate at all sites during low tide. The samples were transferred to a 125 mL glass jar. Sediment cores were collected at three stations (Sites LS, GG, and HP). The cores extended from the surface to 40 cm depth and were obtained using a hand piston core sampler with a 4 cm diameter. Each core was divided into 5 cm intervals and was stored in a glass bottle.

Sample preparation and density separation for analysis of MPs in sediments were conducted following the previously reported method with some modifications (Masura et al., 2015). The 50 g of wet sediments were passed through a sieve with 20 μm mesh (Eo et al., 2018), which is almost the detection limit of Fourier transform infrared microscopy (FT-IR, iN10, Thermo Fisher Scientific, Madison, WI) (Song et al., 2018), to reduce the time for density separation by removing small-grained sediments that float easily in the case of tidal flat samples. NaCl is the representative density separation solution, as it is highly available, cheap, and eco-friendly (Nuelle et al., 2014). Saturated NaCl (200 mL, 1.2 g/cm^3) was added to the sample and mixed well. After 15 min, to allow sediments to settle, the supernatant was transferred to another beaker. The first separation was completed by repeating this process three times. Separated and retrieved samples were then placed in an oven at 60 °C until they were completely dried. To remove organic matter, 20 mL of 35% hydrogen peroxide and 20 mL Fe(II) solution were added to samples. Then, the samples were placed on a hot plate multi-stirrer at 75 °C and 180 rpm and were reacted. Treated samples were sieved and washed with deionized water. For the final density separation, the samples were transferred to a funnel, and NaCl was added. After 24 h, each supernatant was filtered through stainless mesh filter paper (pore size 20 μm, Ø 25 mm).

The entire surface of the filter paper was measured to identify and count MPs using FT-IR (Table S2). The blank of the mapping image was measured as an empty spot on the filter paper. Images were collected in transmission mode using ultrafast mapping with 25 μm × 25 μm step size and 100 μm × 100 μm aperture size at a resolution of 16 cm^{-1}. The spectra were collected across the wavelength range of 715–4000 cm^{-1} with a single scan lasting 0.113 s. The collected spectra data were compared with the spectrum library using OMNIC Picta (Ver. 1.7, Thermo Fisher Scientific) (Fig. S1). When the matching rate between the reference spectrum and sample spectrum exceeded 70%, it was considered acceptable (Lusher et al., 2015). The shape of MPs was classified into fragments, fibers, and films. A thread-shaped particle with a ratio of length to width > 5:1 was fiber; flat or flexible particle was film; and irregular shapes or particle which were not clearly defined other shapes were classified as fragments (Jang et al., 2020a). MPs were also classified by size (0.1–0.3 mm, 0.3–0.6 mm, 0.6–1.0 mm, 1.0–5.0 mm) based on their maximum length. To prevent external contamination, stainless steel and glass materials were used during the entire laboratory analysis. A laboratory cotton coat and nitrile gloves were always worn. All experimental instruments were sealed with aluminum foil to block

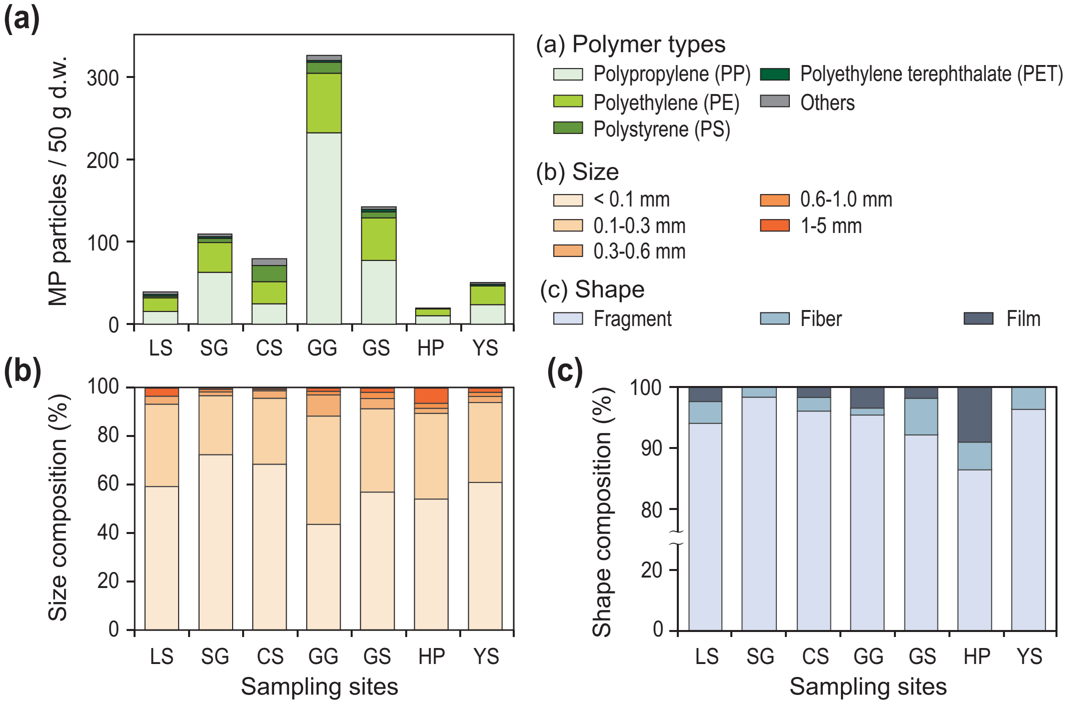

Fig. 2. (a) Spatial distributions of microplastics with polymer type in tidal flats along the west coast of Korea, and their (b) size and (c) shape compositions.

airborne contamination. To reduce error, we repeated the analysis in triplicate with blank samples. The samples were analyzed by FT-IR after performing identical pretreatment processes to the other samples with 1 L of deionized water.

Water, mud, and organic contents were measured to identify the characteristics and sediment samples. Water content (WC) was determined by calculating the dry weight of 10 g of wet sediment, and mud content (MC) was determined by measuring the mass change of sediment after 63 μm wet sieving. Each sediment sample was calculated as follows suggested by Buchanan (1984) (Eqs. (1) and (2));

$$\text{WC}\ (\%) = ((\text{Wet weight} - \text{Dry weight})/\text{Wet weight}) \times 100 \tag{1}$$

$$\text{MC}\ (\%) = ((\text{Dry weight} - \text{Sieved weight})/\text{Dry weight}) \times 100 \tag{2}$$

Loss on ignition (LOI) was analyzed to measure organic matter content. Wet sediment samples were dried at 105 °C for 24 h (DW105) and pulverized into small particles. Then, sediments of constant weight were placed in a muffle furnace. The organic matter was combusted at 550 °C (DW550), and LOI was then calculated using the following equation by Heiri et al. (2001) (Eq. (3)).

$$\text{LOI}\ (\%) = ((\text{DW105} - \text{DW550})/\text{DW105}) \times 100 \tag{3}$$

The sedimentation rates (cm y^{-1}) of core sediments were calculated using a constant rate of supply (CRS) model for the vertical profiles of excess ^{210}Pb activity (Lee et al., 2019). The ^{210}Pb and ^{226}Ra in sediments were carried out by gamma spectroscopy attached to a well-type HPGe detector of the Korea Basic Science Institute (KBSI, Korea). The age of each sediment portion of the core was calculated by multiplying the average sedimentation rate with the average depth of each portion (Li et al., 2020). The average sedimentation rate in Site GG was 0.46 cm y^{-1} (Fig. S2). This value was comparable to the sedimentation rate (0.49 cm y^{-1}) in the tidal flat of Jeonbuk, South Korea, reported previously (Lee et al., 2021). Unfortunately, in Sites HP and LS, the calculated vertical profiles of excess ^{210}Pb activity indicated infeasible results for sediment dating. Bioturbation by various activities of tidal flat organisms was one of the important factors that change the sediment of the tidal flat; the sediments could be disturbed by sediment-dwelling organisms, such as mud crabs and lugworms (Kristensen et al., 2012; Reineck and Singh, 1980). Thus, the results of the age determination of sediment cores of Sites HP and LS were excluded because the excess ^{210}Pb profile was not properly interpreted (Crusius and Kenna, 2007).

The surface morphology of detected MPs in sediment samples was analyzed using Field Emission SEM (MIRA 3 FE, Tescan, Brno, Czech Republic) (Table S2). The samples were mounted on aluminum stubs with carbon tape and coated with platinum, then the surface features of each MP particle were observed by SEM.

Statistical analysis was performed using SPSS 18.0 software (PASW Statistics 18, SPSS Inc., Chicago, IL). The Kruskal-Wallis test and multi-comparison analysis were used to test for individual differences between sampling sites. Spearman rank correlation was used to determine correlations of MPs with organic materials and mud contents. A mean of 0.6 MPs particles was detected in the blank samples. The identified polymer types were polypropylene (PP) and polyethylene (PE). Procedural contamination should be <10% of the average values detected in samples (Galgani et al., 2013). Thus, it was considered that external contamination of MPs did not affect the data obtained from the present study.

MPs were detected in all sediment samples of the tidal flats along the west coast of Korea, with a mean 109 particles per 50 g sediment dry weight (d.w.) (Fig. 2 and Table S3). The MPs in tidal flat sediments were of various sizes, shapes, and colors (Fig. S3). Great number of MPs was found in Site GG (322 particles per 50 g d.w.), followed by GS (142 particles per 50 g d.w.) and SG (109 particles per 50 g d.w.). Although it is difficult to compare with other studies due to their different sampling methods, the results of the present study were compared with those using similar separation methods and quantification units (Table 1). MPs abundance in the lagoon of Venice (Italy) (Vianello et al., 2013) and mangroves of Beibu Gulf (adjoining China-Vietnam) (Zhang et al., 2020) was similar to the current study. In contrast, the abundance of MPs recorded on sediments of tidal flats in the present study was generally greater than those of sandy beaches reported previously (Table 1). The greater concentrations of MPs in sediments of tidal flats than those of sandy beaches seemed to be attributed to sedimentation characteristics. Land-derived MPs behave similarly to suspended sediments, which are

Table 1
Distributions of microplastics in coastal sediments reported in previous studies and obtained from the present study.

Regions	Location and country	Range (min.–max., number of MPs/kg dw)	Mean (number of MPs/kg dw)	References
Tidal flat	East Frisian Island, Germany	3600–13,600	–	Liebezeit and Dubaish (2012)
	Western coastline, Korea	405–6498	2191	This study
	Halifax Harbor, Canada	2200–6000	3567	Mathalon and Hill (2014)
	Hong Kong	0.99–2116	268	Lo et al. (2018)
Estuarine area	Dutch estuary	3010–3600	3300	Leslie et al. (2013)
	Dutch coast	100–720	440	Leslie et al. (2013)
	Changjiang Estuary	20–340	–	Peng et al. (2017)
Coastal area	Scapa flow, Orkney	730–2300	–	Blumenröder et al. (2017)
	Khark Island, Iran	295–1085	–	Akhbarizadeh et al. (2017)
	Oman Sea	138.3–930.3	–	Kor et al. (2020)
	Hong Kong	49–279	–	Tsang et al. (2017)
Mangrove	Beibu Gulf, China	273–3520	–	Zhang et al. (2020)
	Singapore	12–62.7	36.8	Mohamed Nor and Obbard (2014)
Lagoon	Venice, Italy	672–2175	1445.2	Vianello et al. (2013)
	North Tunisian coast	141.2–461.3	316 ± 124	Abidli et al. (2018)
Beaches	Europe	72–1512	–	Lots et al. (2017)
	Hiroshima Bay, Japan	5–1245	–	Sagawa et al. (2018)
	Lesser Antilles, Caribbean Sea	68–620	261 ± 6	Bosker et al. (2018)
	Biscay Bay	145–382	–	Masiá et al. (2019)
	Hengchun Peninsula, Taiwan	80–480	200	Chen and Chen (2020)
	Baja California Peninsula, Mexico	16–312	135 ± 92	Piñon-Colin et al. (2018)
	Baltic Sea, Poland	76–295	–	Urban-Malinga et al. (2020)
	Belgium	48.7–156.2	92.8	Claessens et al. (2011)
	Slovenia	–	177.8	Laglbauer et al. (2014)
	Southern Baltic Sea	25–53	39 ± 10	Graca et al. (2017)

-: Unable to calculate due to the absence of raw data.

accumulated in tidal flats. Previous studies also reported that tidal flats were more polluted by MPs (Liebezeit and Dubaish, 2012; Lo et al., 2018). Overall, the pollution level of MPs in the tidal flat sediments along the west coast of Korea was greater than in other countries and regions. This result is likely due to various plastic sources on the west coast of Korea (Kim and Lee, 2020) and the low removal efficiency in wastewater treatment systems. The recovery and recycling rates of plastic packaging waste from households in Korea are very low compared to other developed countries (Jang et al., 2020b). Plastic waste that is not properly disposed of can leak into marine environments through various routes (Rochman, 2018).

The Site GG, where the largest number of MPs was detected (significantly greater than other sites, $p < 0.05$), is located in the estuary of the Geum River (Fig. 2a). MPs transported through rivers and entering estuaries can aggregate with suspended particles, become heavy and accumulate in tidal flats like other suspended sediments (Lebreton et al., 2017; Nel et al., 2017). The Geum Estuary is also an active fishing area (Lee and Rahimi Midani, 2015). Fishing activities can generate large amounts of plastic waste, such as nets and ropes, which can be an important source of MPs (Montarsolo et al., 2018). In Gunsan City, where Site GG is located, there were 19 national, regional, and small fishing ports, the most among the study areas (Korea Maritime Institute, 2020) (Table S4 and Fig. 2a). There were 20 fishing ports in Buan-gun, where Site GS was located, and the concentration of MPs was the second highest. On the other hand, a low number of fishing ports are located near Site HP (Kang et al., 2009; Korea Maritime Institute, 2020), the site of low MPs concentration. Hampyeong Bay is directly adjacent to rural areas and has the lowest urbanization and industrialization of the seven sampling sites. In addition, it has a semi-enclosed bay without the influx of other significant pollutants. Overall, the results of the present study indicated that MPs in tidal flat sediments of the west coast of Korea are affected by both estuaries and fishing activities. Previous studies also suggested that MPs in coastal areas originate mainly from domestic, industrial, and fishing activities and are introduced from freshwater, marine, and terrestrial sources (Kim et al., 2015; Li et al., 2016; Zhang, 2017).

Meanwhile, the correlation between sediment properties and MPs in the present study cannot be neglected. Organic contents ($R = 0.791$, $p < 0.001$) and mud contents ($R = 0.814$, $p < 0.001$) were also positively correlated with the concentrations of MPs. Mud contents were as high as 98.1% and 94.8% at the sites of GG with the highest MP concentration and GS with the second highest concentration, respectively (Table S1). A correlation between mud content and abundance of MPs was also found in Venice lagoon sediments (Vianello et al., 2013). Finer sediments generally produce lower wave energy; thus, they more easily accumulate MPs (Kowalski et al., 2016). Several studies have shown that organic matter content was correlated with MPs (Haave et al., 2019; Maes et al., 2017). Both MP and particulate organic carbon tend to accumulate well in low-energy sedimentary environments. A large number of MPs appear in low-energy sedimentary environments with high trapping efficiency, especially in muddy, low-energy estuaries, fjords, and lagoons (Harris, 2020). Furthermore, a high organic content can increase the probability of capturing buoyant MPs, as plastics have a hydrophobic surface that promotes microbial community and biofilm formation (Zettler et al., 2013). Thus, it means that the abundance of MPs for the same dry weight can be affected by the sediment properties, such as mud and organic contents.

Fourteen types of polymers were detected in sediments of tidal flats (Table S5). Polypropylene (PP) and polyethylene (PE) were the most abundant types of polymers (51% and 36%, respectively), followed by polystyrene (PS, 6.1%) and polyethylene terephthalate (PET, 3.3%) (Fig. 2a). PP and PE are the most commonly used types of plastic polymers globally (PlasticsEurope, 2020). Other types of polymers accounted for $<1\%$ of the total, and included polyamide, polyurethane, styrene-acrylonitrile copolymer, polyacrylate, poly(acrylate-styrene), polyvinyl copolymer, polyethylene vinyl acetate, alkyd, polyepoxides, and acrylonitrile butadiene styrene. MPs detected in tidal flats were similar to those of MPs polymers on sandy beaches on the west coast (Table S6), and PP accounted for 67% of MPs < 1 mm (Eo et al., 2018). PS showed a relatively greater in Site CS compared with other sites (Fig. 2a). These results are attributed to the widespread use of seaweed and oyster farms in this region and the widespread use of expanded polystyrene (EPS) buoys (Cho et al., 2019). A previous study investigating surface seawater MPs along the coast of Korea reported that the percentage of EPS was the highest in Cheonsu Bay (Kwon et al., 2020).

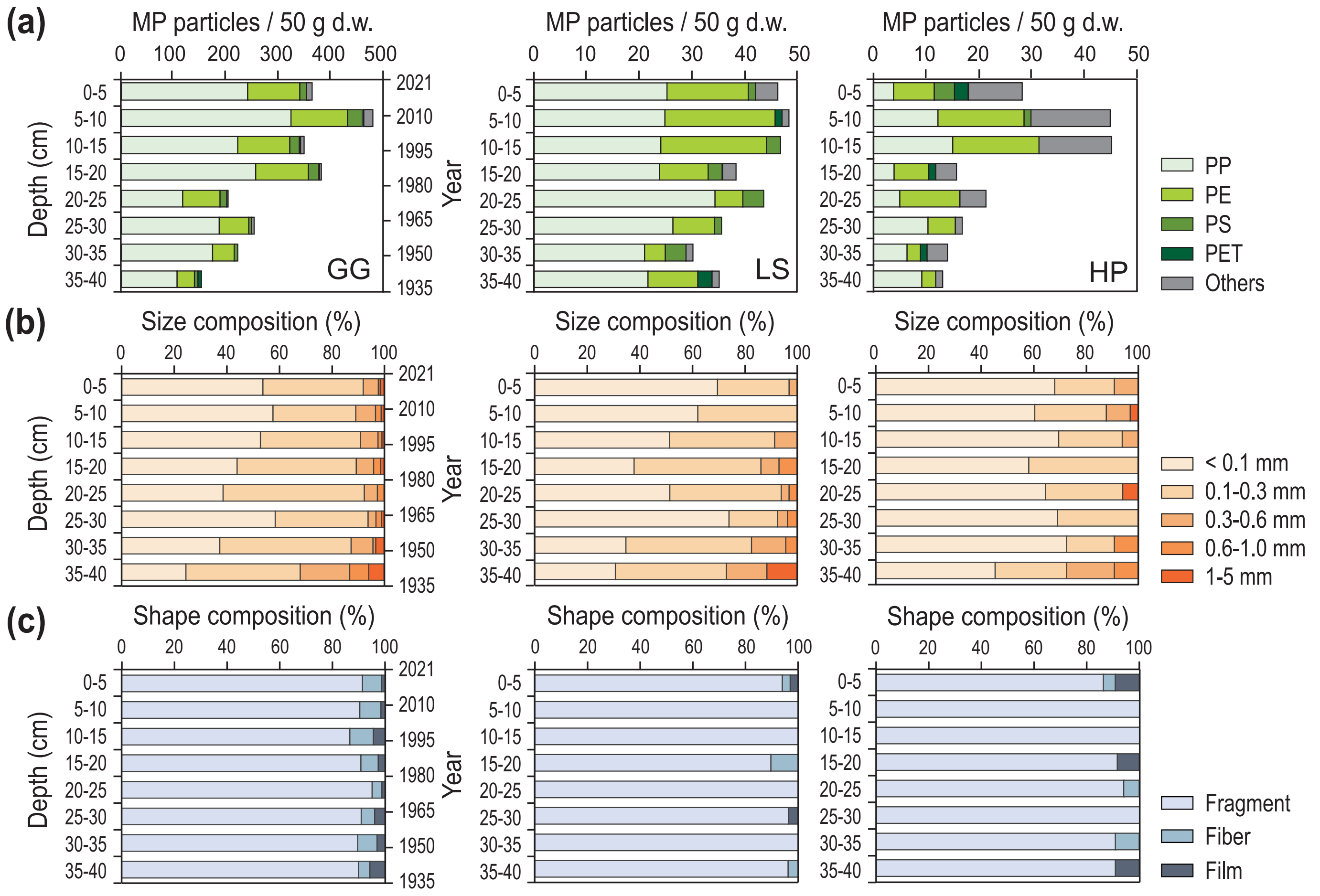

Fig. 3. (a) Vertical distributions of microplastics with polymer type in Sites GG, LS, and HP in the west coast of Korea, and their (b) size and (c) shape compositions.

Meanwhile, the size of MPs in surface sediments ranged from 25 to 2420 μm. More than 90% of the detected MPs were <1 mm in size, with MPs < 100 μm being the most abundant. (Fig. 2b). This result is comparable to the previous studies (Lots et al., 2017; Mohamed Nor and Obbard, 2014). Due to the high bioaccessibility of small-sized MPs, adverse effects on coastal ecosystems are of great concern (Moore et al., 2001). In terms of the shape of MPs, fragments that were secondarily formed by the breakdown of large plastic particles were predominant at all sites (Fig. 2c). A previous study of coastal waters in Korea also showed that fragmented MP was dominant, accounting for 81% of the total detected MPs (Song et al., 2018). The second most abundant form of MPs was fiber, which is often reported to originate from sewage discharge (Browne et al., 2011). The film was the third most abundant form of MPs, generally originating from plastic packaging and mulching (Huang et al., 2020). In Site HP, the relative contribution of the film was greater than those of other sites (Fig. 2c). PE film mulching is widely used in agricultural activities, and film-type MPs detected in Site HP were found to be PE, indicating that it might be associated with local agricultural activities.

MPs were found in all core sediment samples from surface to 40 cm depth at Sites LS, GG, and HP, ranging from 30 to 48 particles per 50 g d.w. (LS), from 152 to 483 particles per 50 g d.w. (GG), and from 14 to 45 particles per 50 g d.w. (HP) (Fig. 3 and Table S7). The vertical distributions of MPs in all three sediment cores commonly showed a decreasing trend as the depth increased. This result was consistent with increasing plastic production and usage over time (PlasticsEurope, 2010). A similar trend was found in previous studies (Brandon et al., 2019; Mao et al., 2021; Willis et al., 2017). Interestingly, the highest MP concentrations were found in the sub-surface layers (5–10 cm depth) of all sediment cores. Many species of macrozoobenthos, such as Annelida, Mollusca, and Arthropoda, inhabit the tidal flats along the west coast of Korea (Park et al., 2014). It is possible that sediment disturbances of these tidal flat organisms may have affected the vertical distribution of MPs (Näkki et al., 2017). Another possibility is that MPs in surface sediments were resuspended and moved by tidal currents. It may also reflect the recent efforts to reduce MPs and the effectiveness of environmental policies. Further investigation of recent inflows of MPs and their distributions in the environments is needed.

To quantify temporal trends in MPs, age data for the GG sediment core were analyzed (Fig. 3a). A minor increase in MPs occurred from the 1930s to 1970s, and MP abundance sharply increased from the 1980s to 2010s. This phenomenon might reflect the rapid economic growth in South Korea, with the gross domestic product (GDP) noticeably increasing from 1970 ($70,000) to 2010 ($1.2 million), due to extensive industrialization and urbanization (Jung, 2011). Of note, the temporal decrease in MPs during the 1990s might be related to dike construction in the Geum River Estuary, which was completed in 1990. Sediment fluxes after estuary dam construction showed that sediment transported by the Geum River was completely enclosed to the estuary (Kim et al., 2006). Consequently, large quantities of MPs from upstream areas might have been deposited in the inner part of the Geum River dike. Accordingly, the number of MPs may be influenced by changes in the surrounding land uses and activities.

Regarding MP size and shape, similar results with surface sediments were obtained for all depths in core sediments (Fig. 3b and c). Fragments were found in large proportion at all depths. MPs with relatively smaller sizes were found in surface sediments compared to the deep layer. This result seems to be because MPs are continuously decomposed by

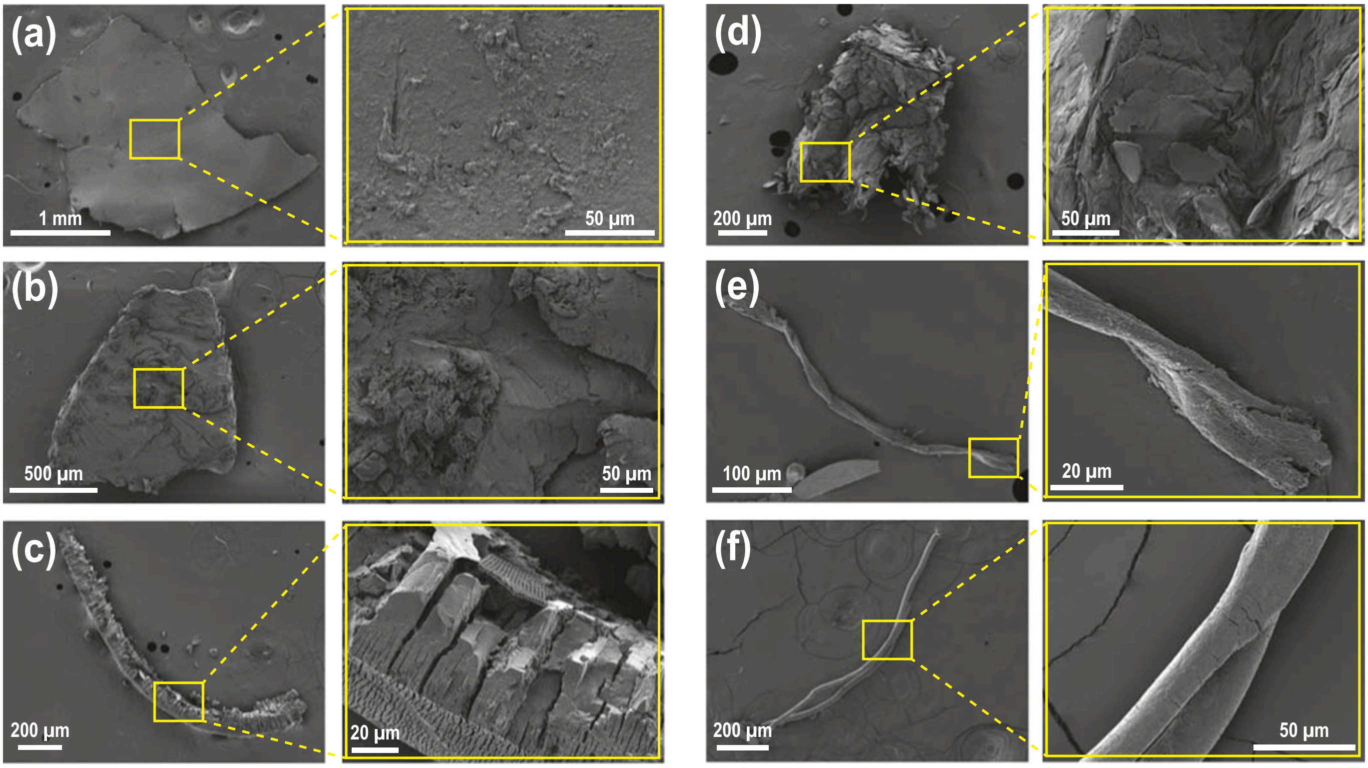

Fig. 4. Scanning electron microscope images of microplastic particles obtained from the present study. (a) Polyethylene film, (b-c) polypropylene fragment, (d) polystyrene fragment, (e) polypropylene fiber, and (f) polyethylene terephthalate fiber.

ultraviolet radiation in the surface layer of tidal flat. Degradation of plastics occurs primarily through photolysis in plastics exposed to air (Andrady, 2011). The types of polymers in sediment cores were also investigated (Table S8). PP and PE exceed 80% at all depths in Sites LS and GG, similar to the surface sediments. However, polymethyl methacrylate (PMMA), which had not been previously found in surface sediment (July 2020), was abundant in Site HP (January 2021) (Table S8). PMMA is often used in photoselective films for greenhouse farming, particularly during winter in Korea (Rasheed et al., 2018). Thus, the seasonal difference in sampling time might explain the absence of PMMA in surface sediment samples.

The surface morphology of detected MPs in tidal flat sediments was examined in detail using SEM (Fig. 4). Film-type MPs had some cracks and grooves on the surface, but showed a smoother surface compared to other types (Fig. 4a). Fragmented MPs exhibit irregular and relatively thick and hard surfaces (Fig. 4b-c). The MP of the PS polymer appeared torn with elongation rather than breakage due to fragmentation of the surface (Fig. 4d). Fiber-shaped MPs were observed twisted or agglomerated forms, and cracks appeared on the surface (Fig. 4e-f). Morphological features indicate that the MPs in tidal flat sediments were influenced by mechanical and oxidative weathering associated with continuous environmental exposures (Yaranal et al., 2021). Since the tidal flat is a dynamic environment where strong tidal energy exists and is periodically exposed to the air, it has suitable conditions for weathering of MPs. The abrasion MPs surface can produce smaller MP particles, and this process can exponentially increase the number of MPs in the natural environment.

Overall, the present study revealed the widespread distribution of a large number of MPs in tidal flats along the west coast of Korea. The present study advances our current understanding of the status of MPs pollution in tidal flats. The presence of high concentrations of MPs in tidal flats can cause serious environmental and ecological problems. This is because a variety of animals live in tidal flats, and they are the main food sources for upper predators, such as seabirds, and are valuable fishery resources for humans. Further research is needed on the accumulation and risk of MPs in tidal flat organisms and their transport to higher-level predators.

CRediT authorship contribution statement

Jaeyeon Park: Conceptualization, Investigation, Formal analysis, Data curation, Visualization, Writing - original draft. Seongjin Hong: Conceptualization, Data curation, Visualization, Writing - original draft, Writing - review & editing. Won Joon Shim: Conceptualization, Methods development, Writing - review & editing. Jong Seong Khim: Conceptualization, Writing - review & editing, Project administration, Funding acquisition. Jinsoon Park: Conceptualization, Investigation, Formal analysis, Data curation, Writing - review & editing, Funding acquisition, Supervision.

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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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.marpolbul.2023.114741.

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Supplementary materials for

Distribution, compositional characteristics, and historical pollution records of microplastics in tidal flats of South Korea

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Supplementary Tables

Table S1. Locations of sampling sites and sediment characteristics in tidal flats along the west coast of South Korea.

Study sites	**Location**		**Mud contents (%)**	**Loss of ignition (%)**
	Latitude (ºN)	**Longitude (ºE)**		
Gyeonggi Bay (Lake Sihwa, LS)	37.3362	126.6901	65.4	2.4
Asan Bay (Lake Sapgyo, SG)	36.8939	126.8224	88.2	5.6
Cheonsu Bay (CS)	36.5869	126.4584	26.2	1.9
Geum River Estuary (GG)	36.0176	126.7398	98.1	6.9
Gomso Bay (GS)	35.5778	126.6644	94.8	6.5
Hampyeong Bay (HP)	35.0889	126.3531	21.2	1.9
Yeongsan River Estuary (YS)	34.7801	126.4431	40.7	2.0

Table S2. Instrumental conditions for analysis of microplastics using FT-IR and FE-SEM.

Instrument and model	**FT-IR iN10**
Manufacturer	Thermo Fisher
Mode	Transmission
Step size	25 µm x 25 µm
Aperture size	100 µm x 100 µm
Resolution	16 cm^{-1}
Spectral range	715–4000 cm^{-1}
Number of scans	Single
Scan time	0.113 s
Software	OMNIC^{TM} Picta^{TM}
Instrument and model	**MIRA 3 FE-SEM**
Manufacturer	Tescan
Electron Gun type	Schottky type
Accelerating Voltage	3.0 kV
Aperture size	126.4584
Resolution SE	1.2 nm–3.0 nm
Resolution BSE	2.0 nm
Probe Current	1pA–200nA
Coating	Platinum
Software	MIRA TC

Table S3. The number of microplastics per 50 g of dry sediments in tidal flats along the west coast of South Korea.

Replicates	**LS**	**SG**	**CS**	**GG**	**GS**	**HP**	**YS**	**Blank**
1	26.44	128.99	50.58	292.40	63.33	11.20	51.06	0
2	40.65	140.15	111.71	321.68	165.38	18.76	42.79	2
3	52.00	57.48	77.06	360.57	198.60	30.78	59.03	0
SUM	119.09	326.62	239.34	974.65	427.31	60.75	152.87	2
Mean	39.70	108.87	79.78	324.88	142.44	20.25	50.96	0.67
SD	12.80	44.86	30.66	34.20	70.49	9.88	8.12	

Table S4. The number of fishing ports along the west coast of South Korea (data refers from Korea Maritime Institute (2020)).

Location Province	City	Number of fishing port[a] National fishing port	Regional fishing port	Fishing village port	Small-scale fishing ports	Sum	Adjacent Sites
Gyeonggi	Ansan	0	1	3	8	12	LS
	Shiheung	0	1	0	3	4	
Chungnam	Dangjin	1	1	4	1	7	SG
	Hongsung	1	2	0	2	5	CS
	Seocheon	1	3	3	2	9	GG
Jeonbuk	Gunsan	4	4	11	0	19	
	Buan	2	5	5	8	20	GS
Jeonnam	Hampyung	0	0	1	4	5	HP
	Mokpo	0	0	0	8	8	YS

[a] Korea Maritime Institute (2020). Fisheries and Marine Environment Statistics.

Table S5. The number of microplastics by polymer type in sediments in tidal flats along the west coast of South Korea (Number of microplastics per 50 g of dry sediments).

Polymer type	**LS**	**SG**	**CS**	**GG**	**GS**	**HP**	**YS**
Polypropylene	16.14	62.61	25.06	232.06	76.98	10.55	24.18
Polyethylene	16.58	36.1	27.15	72.04	52.13	8.01	22.03
Polystyrene	1.3	5.64	18.79	13.16	6.67	0	0.86
Polyethylene terephthalate	2.18	2.26	0.41	1.38	3.64	1.69	1.74
Polyamide	0	0	3.34	0	0.6	0	0
Polyurethane	0.43	0	1.67	0.69	0	0	0
Styrene acrylonitrile copolymer	0.87	0.57	0	2.77	1.21	0	1.72
Poly(acrylate-styrene)	0.44	0	0	0	0	0	0.43
Polyacrylate	0.87	0.56	0	0	0	0	0
Polyvinyl copolymer	0.45	0	0	0	0	0	0
Polyethylene vinyl acetate	0	0	1.25	0	0	0	0
Alkyd	0	1.13	1.26	0	0.6	0	0

Table S6. Distributions of microplastics in sediments in estuarine and coastal areas of South Korea.

Location	Type of sample	MP abundances	Size of MPs	Shape of MPs	Polymer type	Main sources	References
Six semi-enclosed bay sand two nearshore areas	Coastal waters	1736 ± 1179 #/m^3 (surface) 42 ± 342 #/m^3 (middle) 394 ± 443 #/m^3 (bottom)	752 ± 711 (fibers) 197 ± 168 (non-fibers)	Fragments 81% Fibers 18% Spheres and film 1%	PP 41 ± 17% PE 21 ± 15% EVA 19 ± 20% (fibers) PP 92 ± 10% PES 4.7 ± 7.7% (non-fibers)	Populated areas, river input (horizontal) Biofouling, aggregation, and fecal pellets (vertical)	Song et al. (2018)
Sandy beaches along the coast	Beach sediments	0–2088 #/m^2 (L-MP, 1–5 mm) 1400–62800 #/m^2 (S-MP, <1 mm)	<300 µm 81%	EPS 94.8% Fragments 2.6% Pellets 1.9% (L-MPs) Fragments 96.7% Fiber 3.1% Spheres 0.2% (S-MPs)	PS 95% (L-MPs) PE 49.2% PP 37.8% PS 9.6% (S-MPs)	Aquaculture industry (L-MPs) Plastic debris by weathering (S-MPs)	Eo et al. (2018)
Nakdong River	Surface, mid waters and sediments	293 ± 83 #/m^3 (upstream) 4760 ± 5242 #/m^3 (downstream) 1970 ± 62 #/kg (sediment)	<300 µm 74% (waters) <300 µm 81% (sediments)	Fragments 69% Fibers 30% Spheres and film <1% (waters) Fragments 84% Fibers 15% Spheres 1% (sediments)	PP 41.8% PES 23.1% (waters) PP and PE 50% (sediments)	Population density, sewage, and wastewater treatment facilities	Eo et al. (2019)
Nakdong River Estuary and beaches on Geoje Island	Beached debris	8205 #/m^2 (May) 27606 #/m^2 (September)	MP more abundant than larger plastics	Styrofoam 99% (May) Styrofoam 96% Pellet 3% (September)	–	Oyster aquaculture facilities	Lee et al. (2015)
Near the Nakdong River mouth on the southeastern coast	Neustonic debris	0.62–57 #/m^3 (trawl, May) 0.64–860 #/m^3 (trawl, July) 260–1410 #/m^3 (hand-net, May) 210–15560 n/m^3 (hand-net, July)	<2 mm 17–51%	Paint 49% (trawl, May) Styrofoam 51% (trawl, July) Paint 39% (hand-net, May) Styrofoam 31% (hand-net, July)	PE (hard plastic) Alkyd (paint particles) PES (fibers)	Riverine discharge, aquaculture farms, and paint flaking off ships	Kang et al. (2015)

Jinhae Bay	Sea surface microlayer	94 ± 68 #/L (paint particles) 88 ± 68 #/L (non-paint)	50–100 µm (fragments peak) <50 µm (spherules peak)	Fragments 75% Spherules 14% Fibers 5.8% EPS 4.6%	Alkyd 35% poly(acrylate-styrene) 16%	River input, fishing vessels, and industrial complexes	Song et al. (2015)
Six semi-enclosed bays and two nearshore areas	Surface waters (0.33–5 mm)	1.12–4.74 #/m^3	0.33–0.5 mm 46% 0.5–1 mm 31% 1–5 mm 22% (urban) 0.33–0.5 mm 34% 0.5–1 mm 37% 1–5 mm 28% (rural)	Fiber 36.8% (urban) Fiber 31.3% (rural)	PP 62.4% (rural) PP 53.2% (urban)	Populated areas and river input	Kwon et al. (2020)
Han River and its tributaries	Surface and 2 m depth waters	7.0 ± 12.9 #/m^3 (0 m) 102.0 ± 50.3 #/m^3 (2 m) 1.2 ± 234.5 #/m^3 (tributaries)	<1 mm more than 90%	Fragments >73% Rest were fiber (waters)	Silicone 23.1% PS 16.9% PP 15.4% PES 13.8% (waters)	Sewage treatment plant	Park et al. (2020)
Soya island, Incheon–Gyeonggi coastal sea	Surface sediment	46334 ± 71291 #/m^2	1–2 mm (EPS peak) >1 mm (non-EPS peak, southern beaches) 50–300 µm (non-EPS peak, northern beach)	–	EPS >90% at all stations	Styrofoam buoys for aquaculture	Kim et al. (2015)

Table S7. The number of microplastics per 50 g dry core sediments in tidal flats (Sites LS, GG, and HP) along the west coast of South Korea.

Depth (cm)	LS	GG	HP
0–5	46.28	367.17	28.81
5–10	48.26	483.37	44.90
10–15	46.42	350.66	44.92
15–20	38.69	387.20	15.77
20–25	43.30	201.45	21.48
25–30	35.74	252.02	16.64
30–35	30.28	225.26	14.09
35–40	35.09	152.02	14.39

Table S8. The number of microplastics by polymer type in core sediments in tidal flats along the west coast of South Korea (Number of microplastics per 50 g of dry sediments; Sites LS, GG, and HP).

Sites	**Depth (cm)**	**Polypropylene**	**Polyethylene**	**Polystyrene**	**Polyethylene terephthalate**	**Polyamide**	**Polyurethane**	**Styrene acrylonitrile copolymer**	**Polyacrylate**	**Alkyd**	**Polyepoxides**	**Acrylonitrile butadiene styrene**	**Polymethyl methacrylate**	**Polyethylene vinylacetate**	**SUM**
LS	0–5	25.25	15.43	1.4	0	1.4	0	0	0	0	0	0	2.81	0	46.28
	5–10	24.78	20.87	0	1.3	0	0	0	0	0	0	1.3	0	0	48.26
	10–15	23.87	19.89	2.65	0	0	0	0	0	0	0	0	0	0	46.42
	15–20	24.02	9.34	2.67	0	0	0	0	0	1.33	0	1.33	0	0	38.69
	20–25	34.12	5.25	3.94	0	0	0	0	0	0	0	0	0	0	43.3
	25–30	26.48	7.94	1.32	0	0	0	0	0	0	0	0	0	0	35.74
	30–35	21.07	3.95	3.95	0	0	0	1.32	0	0	0	0	0	0	30.28
	35–40	21.59	9.45	0	2.7	0	0	1.35	0	0	0	0	0	0	35.09
GG	0–5	243	100.4	13.21	0	2.64	0	0	2.64	0	0	5.28	0	0	367.17
	5–10	325.7	108.6	28.43	0	0	2.58	5.17	2.58	2.58	0	2.58	5.17	0	483.37
	10–15	222.4	99.44	18.32	2.62	0	0	0	0	0	0	5.23	0	2.62	350.66
	15–20	259.8	101.9	20.38	0	0	0	2.55	0	0	2.55	0	0	0	387.2
	20–25	115.8	70.51	12.59	0	0	0	2.52	0	0	0	0	0	0	201.45
	25–30	185.8	56.01	5.09	0	0	0	5.09	0	0	0	0	0	0	252.02
	30–35	176.5	41.8	6.97	0	0	0	0	0	0	0	0	0	0	225.26
	35–40	105.8	33.05	6.61	6.61	0	0	0	0	0	0	0	0	0	152.02
HP	0–5	3.93	7.86	3.93	2.62	0	0	0	0	1.31	0	0	9.17	0	28.81
	5–10	12.25	16.33	1.36	0	0	0	0	4.08	0	0	0	10.89	0	44.9
	10–15	14.97	16.34	0	0	0	0	0	5.45	0	0	1.36	6.81	0	44.92
	15–20	3.94	6.57	0	1.31	0	0	0	0	0	0	0	3.94	0	15.77
	20–25	5.05	11.37	0	0	0	0	0	1.26	0	1.26	0	2.53	0	21.48
	25–30	10.24	5.12	0	0	0	1.28	0	0	0	0	0	0	0	16.64
	30–35	6.41	2.56	0	1.28	0	0	0	1.28	0	0	0	2.56	0	14.09
	35–40	9.16	2.62	0	0	1.31	0	0	0	0	0	1.31	0	0	14.39

Supplementary Figures

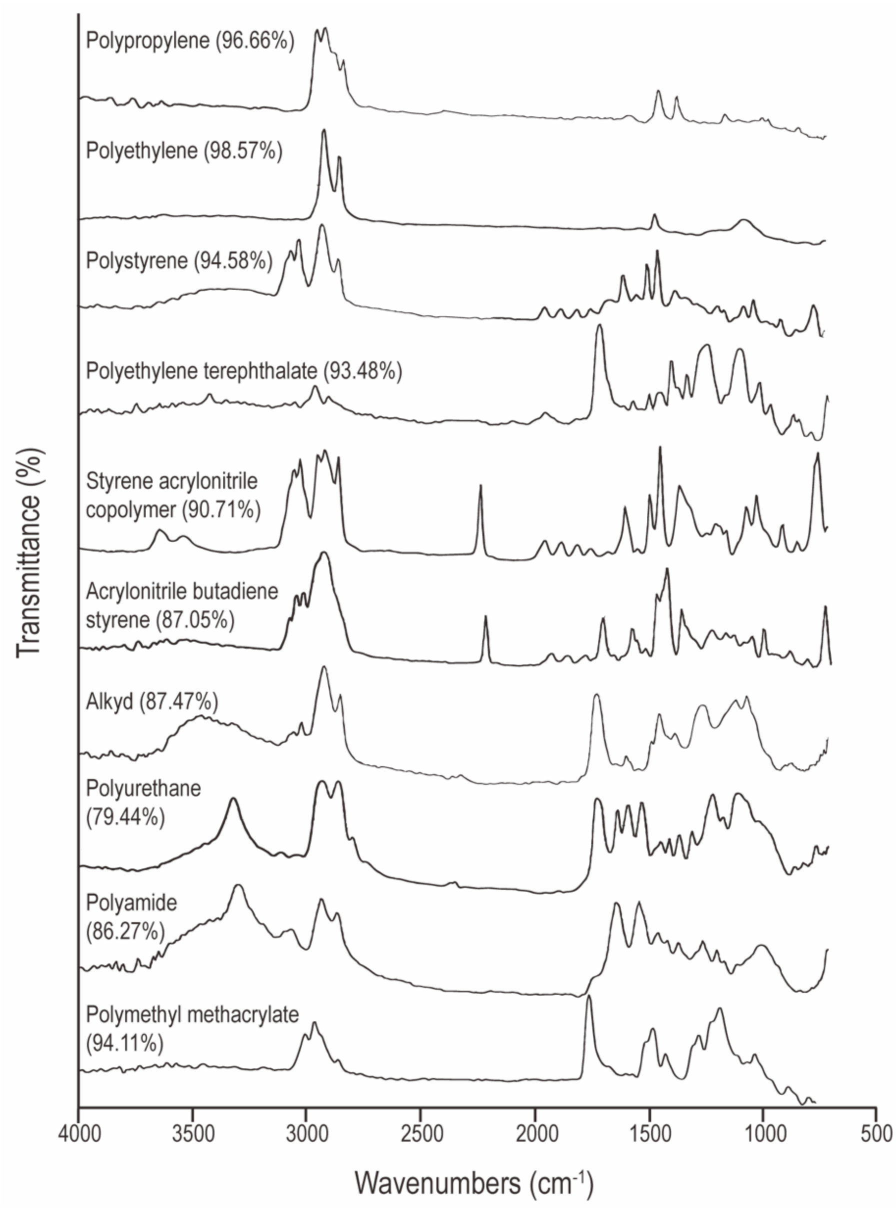

Fig. S1. The FT-IR spectral data of microplastics obtained from the present study.

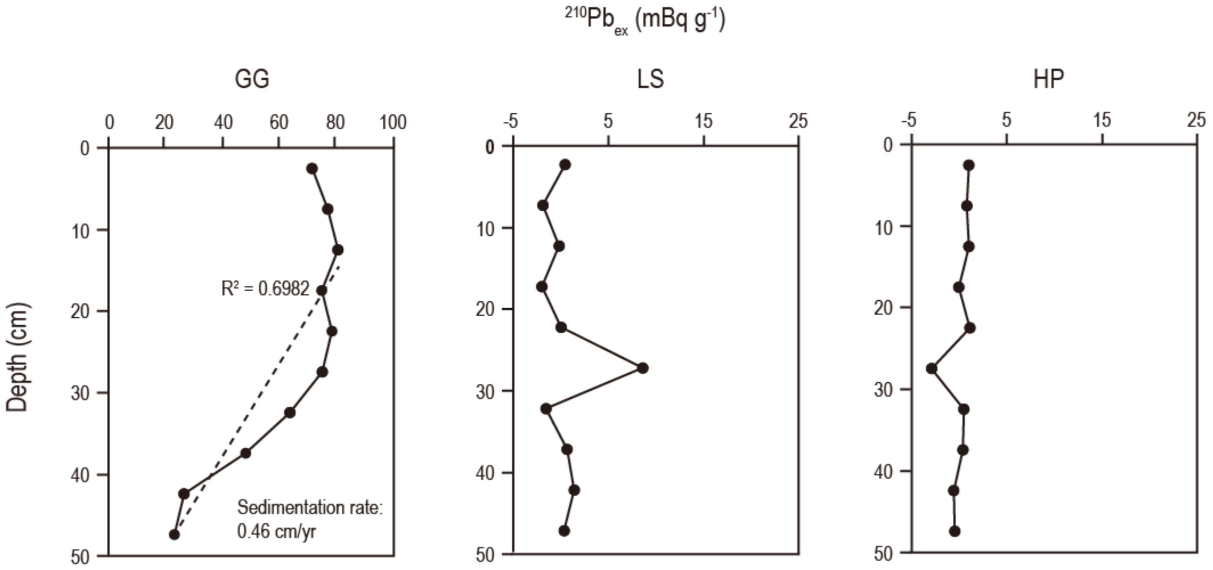

Fig. S2. Vertical profiles of unsupported ^{210}Pb ($^{210}Pb_{ex}$) and sedimentation rates in core sediments in tidal flats along the west coast of South Korea (Sites LS, GG, and HP).

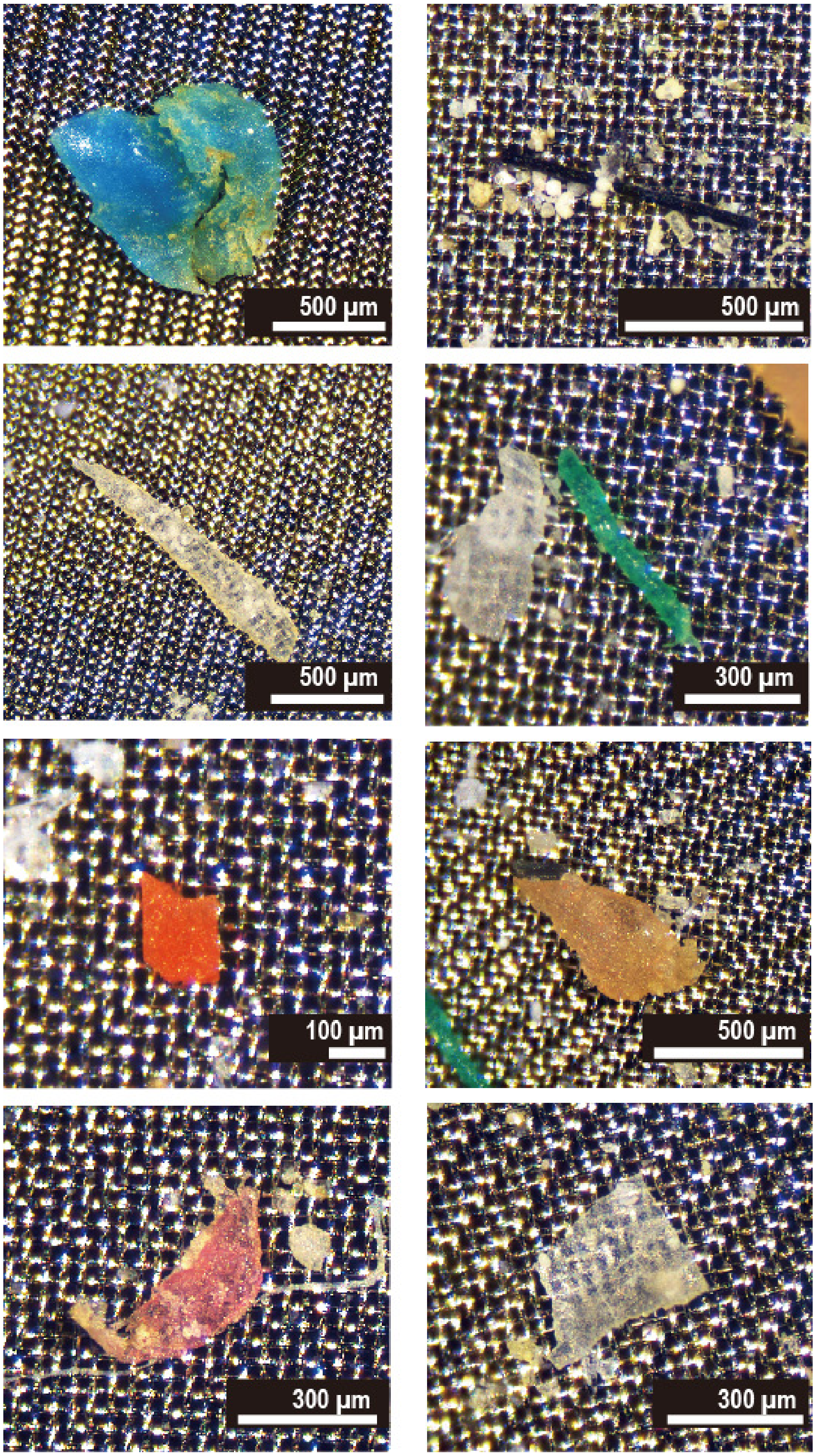

Fig. S3. Microplastics of various colors observed in Site GG core sediments using a stereomicroscope (Leica S9 D).