Water Research 171 (2020) 115400

Contents lists available at ScienceDirect

Water Research

journal homepage: www.elsevier.com/locate/watres

A close relationship between microplastic contamination and coastal area use pattern

Mi Jang [a, b], Won Joon Shim [a, b], Youna Cho [a, b], Gi Myung Han [a], Young Kyoung Song [a, b], Sang Hee Hong [a, b, *]

[a] Oil and POPs Research Group, Korea Institute of Ocean Science and Technology, Geoje, 53201, Republic of Korea
[b] Ocean Science, Korea University of Science and Technology, Daejeon, 34113, Republic of Korea

ARTICLE INFO

Article history:
Received 24 August 2019
Received in revised form
9 December 2019
Accepted 14 December 2019
Available online 16 December 2019

Keywords:
Microplastic
Aquatic environment
Coastal area use pattern
Polymer composition
Source
Environmental matrix

ABSTRACT

Human activity is thought to affect the abundance and contamination characteristics of microplastics (MPs) in the environment, which may in turn affect aquatic species. However, few studies have examined the impact of coastal area use pattern on characteristics of MPs in coastal regions. In this study, we investigated MP contamination of abiotic matrices (seawater and sediment) and biotic matrices (bivalves and polychaetes) in three coastal regions characterized by different types of human activity, covering urban, aquafarm, and rural areas. MP abundance was higher in sediment from the urban site than in that from the rural site, but similar to that from the aquafarm site. In the abiotic matrices, different MP polymer compositions were observed among the three sites. Diverse polymers were found in marine matrices from the urban site, implying diverse MP sources in highly populated and industrialized areas. Polystyrene was more abundant in the aquafarm site, reflecting the wide use of expanded polystyrene aquaculture buoys. Polypropylene was more abundant at the rural site, probably due to the use of polypropylene ropes and nets in fishing activity. MP accumulation profiles in marine invertebrates showed trends similar to those exhibited by abiotic matrices, reflecting coastal area use patterns. These results indicate that marine MPs are generated from both land- and marine-based sources, and that the abiotic and biotic marine matrices reflect the MP characteristics.

1. Introduction

Microplastics (MPs) are a form of marine debris measuring ≤ 5 mm in size (Arthur et al., 2009; GESAMP, 2019). MPs are classified as either primary or secondary according to their production method, such that primary MPs are intentionally manufactured and added to products such as cosmetic products and abrasive materials, and secondary MPs are formed through the breakdown of larger plastics during use or after disposal to the environment (Arthur et al., 2009). Contamination by MPs has become a global environmental concern due to their ubiquitous distribution in the ocean, from pole to pole, from the surface to the seafloor (Sul and Costa, 2014; Auta et al., 2017), and within marine species at every level of the food web (Gall and Thompson, 2015; Rochman, 2018). Ingested MPs can cause physical damage such as intestinal obstruction and stomach ulcers (Carpenter et al., 1972) and can transfer contaminants such as plastic additives and sorbed chemicals from seawater to marine organisms (Jang et al., 2016; Ziccardi et al., 2016), inducing adverse biological effects (von Moos et al., 2012; Browne et al., 2013; Rochman et al., 2013).

Like macro debris, MPs in the ocean can come from a variety of land- and marine-based sources. Roughly 80% of marine debris (of which 60–80% is plastic) has been estimated to originate from land-based sources, and the remainder from marine-based activity (Sheavly, 2005). Therefore, land-based input is considered a major source of microplastics in the ocean. Some studies have attempted to identify the sources of MPs on land (e.g., clothes washing and cosmetics; Fendall and Sewell, 2009; Browne et al., 2011) and input pathways into the ocean (e.g., sewage treatment plant effluents and riverine input; Leslie et al., 2017; Hurley et al., 2018). Previous studies have reported increasing MP levels in coastal sediments and seawater near populated areas (Frère et al., 2017; Vianello et al., 2013; Yonkos et al., 2014; Song et al., 2018), implying a close relationship between land-based human activities and MP pollution of

* Corresponding author. Oil and POPs Research Group, Korea Institute of Ocean Science and Technology, Jangmok-1-gil, Geoje, 53201, Republic of Korea.
E-mail address: shhong@kiost.ac.kr (S.H. Hong).

https://doi.org/10.1016/j.watres.2019.115400
0043-1354/

marine environments. Not only land-based but also marine-based human activities (e.g. shipping, fisheries, and aquaculture) can be important sources of marine MPs. However, our understanding of these sources, their emission amounts, their pathway to the environment and the fragmentation of plastic debris after disposal remain minimal.

MP physicochemical properties such as shape, color, and polymer type can provide information about their sources (Shim et al., 2018). The original shape and color of plastics can be lost or changed by environmental weathering processes such as fragmentation and discoloration, but their main polymer type does not change. Therefore, polymer composition is a useful tool for identifying MP sources or origins in the environment. For example, alkyd particles on the ocean surface are derived from ship paint (Song et al., 2014). Polyester and acrylic fibers are commonly used for fabric (Browne et al., 2010), and synthetic rubber is a tire ingredient (Wagner et al., 2018). However, most studies performed polymer identification for a small portion of suspected particles in their samples to validate their optical microscopy particle count estimate (Li et al., 2016; Leslie et al., 2017; Rodrigues et al., 2018; Nel et al., 2018) because spectroscopic identification for all suspected particles on filter paper is a time-consuming job (Song et al., 2015). The Polymer composition of MP can provide additional information about chemical substances contained because additive chemicals differ among plastic products and polymer types (Lithner et al., 2011; Jang et al., 2017), and their leachates have a different toxicity to organisms (Bejgarn et al., 2015).

In this study, we hypothesize that the polymer composition of MPs in marine matrices would differ between regions according to coastal area use pattern, and that these compositions would be reflected in marine species inhabiting these regions. We analyzed MPs in abiotic matrices (seawater and sediment) and biotic matrices (mussels, oysters, and polychaetes) from three coastal sites representing urban, aquafarm, and rural areas. The polymer compositions of all plastic-like particles were identified. Road dust and creek water were also collected from the urban area to compare their polymer compositions with those of marine matrices collected nearby.

2. Materials and methods

2.1. Sampling strategy

We collected samples of abiotic matrices, including seawater and sediment, and biotic matrices, including mussels (*Mytilus edulis*), oysters (*Crassostrea gigas*), and polychaetes (*Perinereis aibuhitensis*), from the southern part of South Korea between March and April 2016 (Fig. 1 and Fig. S1). We selected three sites (urban, aquafarm, and rural) differing in land (or sea) use and pollution load (Fig. 1). The urban site is located in Masan Bay, a semi-enclosed bay surrounded by the densely populated city of Changwon (2682 people/km^2), which has petrochemical, heavy metal, electrical, and plastic industries. In 1983, Masan Bay was designated a 'special management area' in Korea due to its heavy loads of land-driven contaminants (Shim et al., 1999; Hong et al., 2003; Li et al., 2008). The aquafarm site is located in Jinhae Bay, and has relatively low population density (198 people/km^2) compared to the urban site. A number of oyster farms are distributed inside the bay, employing a suspended culture technique using expanded polystyrene (EPS) buoys. This bay is the main shellfish farming area in the Gyeongnam-do region, producing about 0.3 million tons of bivalves accounting for about 80% of the total shellfish production in Korea (GNDI , 2012). The rural site is located in the southernmost island of Geoje, and contains a small fishing village with low population density (55 people/km^2) and a small harbor for fishing boats, but no industrial facilities or aquafarms.

The abiotic and biotic matrix sampling sites and the sampling points at each site are presented in Fig. 1 and Fig. S1. More than 30 mussels (4–5.5 cm) and 30 oysters (4–9 cm) were collected from intertidal rocky shores from the urban and rural sites, and from under high-density polyethylene (HDPE) buoys floating on the sea surface at the aquafarm site. We sampled 100 L of surface seawater (top 0–10 cm) in triplicate near the bivalve sampling sites by hand-filtering through a net (mesh: 20 μm), and collected 100 L of sub-surface water (top 1 m) from the aquafarm site in January 2017. More than 30 polychaete worms (10–30 cm) were collected from the intertidal zone of aquafarm and rural sites using a shovel during low tide, and from the subtidal zone of the urban site using a Van Veen Grab Sampler. Intact polychaetes were sampled and rinsed with filtered seawater (Whatman, GF/F, 0.75 μm) to remove all external particles, and then transferred to a glass bottle containing 150 mL of filtered seawater. Three polychaetes were placed in a beaker for 3 days, and their feces were collected for MP analysis. During polychaete sampling at each site, we also collected sediment (top 0–5 cm) in duplicate. All samples except for the polychaetes were stored at −20 °C until analysis.

Creek discharge and urban runoff were expected to be important sources of MPs in the environment. To compare the MP contamination characteristics between land-based MP input sources and marine matrices in urban area, creek water and road dust were collected in this region. Road dust (surface area, 4 m^2) was collected from the road right next to the urban sampling site (bivalves and seawater) in January 2017 (Figs. 1 and 5) and creek water (100 L) was collected from Namcheon stream across the city in Masan Bay, during low tide in July 2018.

2.2. Microplastic analysis

The information on the volume and number of samples subjected to microplastic analysis is summarized in Table S1. Sediment samples were analyzed following the method of Eo et al. (2018). About 50 mL of wet sediment was weighed in a polyethylene (PE) bottle, and then mixed with diluted lithium metatungstate solution (LMT; 50 mL, 1.6 g/mL), and shaken for 1 min. After settling, the supernatant was transferred to a glass beaker. This process was repeated three times. The supernatant was passed through a 20-μm metal sieve. The isolated particles were treated with hydrogen peroxide (H_2O_2; 35%) and Fe (II) solution (75 °C, 100 rpm) to remove organic matter. Secondary density separation was then conducted using sodium chloride (NaCl; 1.2 g/mL), and supernatant was filtered through filter paper (20 μm, polycarbonate, Sterlitech) using a vacuum system. Road dust samples (50 mL, 228 g) were treated in the same manner as sediment samples.

The analysis of MPs in seawater and creek water was performed following the method of Song et al. (2018). The water sample was passed through a 20-μm metal sieve to reduce the volume. The remaining sample in the sieve was transferred to a glass beaker, and wet peroxide oxidation with 35% H_2O_2 and Fe (II) solution added at 75 °C. After removing organic matter, 6 g of NaCl was added per 20 mL of sample and stirred at 100 rpm. The sample was transferred to glass funnel for density separation. After one day, the settled particles were drained and the supernatant was filtered through filter paper (20 μm, polycarbonate, Sterlitech).

Bivalve samples were analyzed following the method of Jang et al. (2016) and Cho et al. (2019). After removing the shell, the soft tissue was rinsed with commercial ultrapure water (HPLC grade, Daejung Chemicals and Metals Co.) to remove remaining particles. The sample was placed in a glass beaker, and then 10 mL lipase (Lipex 100L; Novozymes, Bagsværd Denmark), 5 mL of protease (Savinase 16L; Novozymes) and 185 mL of distilled water was

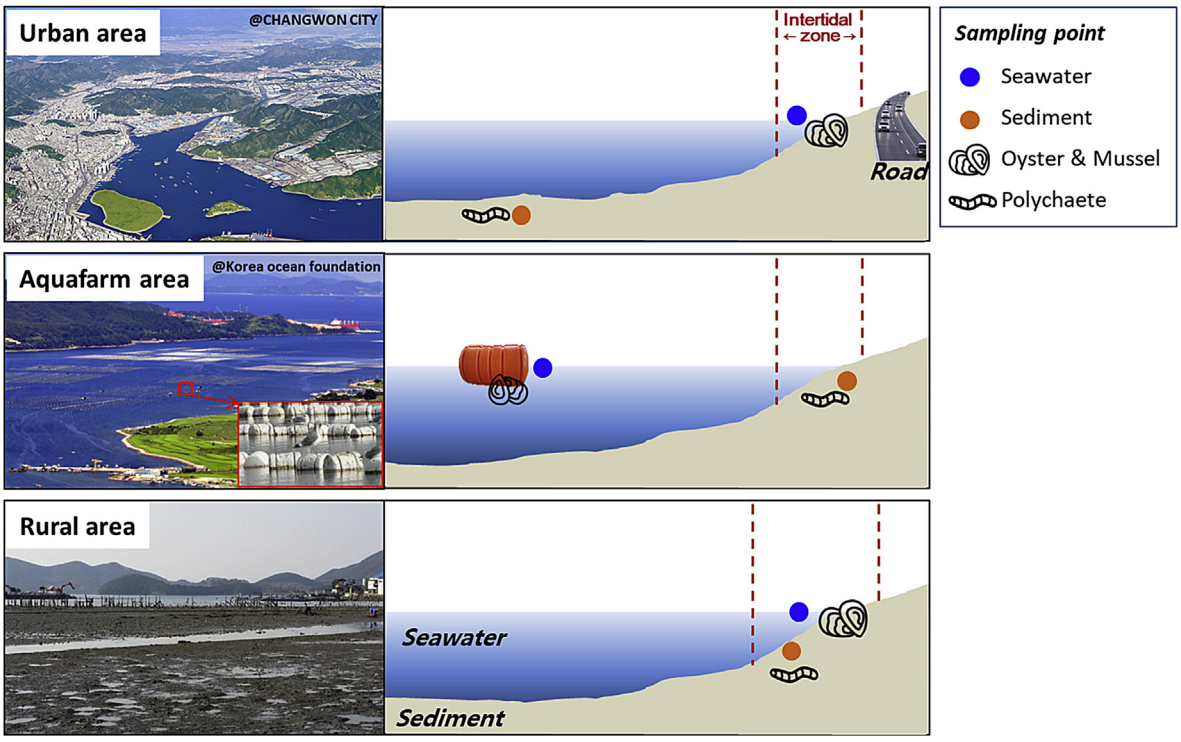

Fig. 1. Sampling sites (left) and sampling points of marine matrices (right). Photographs show the landscapes of the urban, aquafarm, and rural sites. Blue and orange dots indicate seawater and sediment sampling points, respectively; oyster/mussel and polychaete sampling points are indicated by representative symbols; blue and black stars indicate creek water and road dust sampling points, respectively.

added to digest organic matter. Using a magnetic stirrer, the sample solution was stirred overnight at 200 rpm and 60 °C. After this digestion step, the digested solution was sieved through a 20 μm sieve and rinsed with ultrapure water. The particles remaining on the sieve were transferred to a glass funnel with NaCl (1.2 g/mL), and left overnight. The precipitate was removed twice. The supernatant was filtered through filter papers (20 μm, polycarbonate, Sterlitech).

The poychaete feces were prepared for microplastic analysis in accordance with the methods described in Jang et al. (2018). The fecal samples were treated with hydrogen peroxide to remove organic material, and left 3 days. The samples were transferred to a glass funnel with LMT solution for density separation. The supernatant was filtered through filter papers (20 μm, polycarbonate, Sterlitech).

Filters were dried at room temperature and stored in glass Petri dishes. All suspected MP particles (to 50 μm) found on filter paper were identified using a Fourier-transform infrared spectroscopy (FTIR) microscope (Thermo Nicolet 6700, Continuum; Thermo Scientific, Waltham, MA, USA; Song et al., 2015). μFT-IR spectra of all particles were collected with an average of 128 scans over the spectral range of 650–4000 cm^{-1} in attenuated total reflectance (ATR) mode, and their polymer types were confirmed based on comparison with a spectral library database (Thermo Scientific) and recorded. Spectral matching with a hit index ≥70% was considered acceptable. All MPs were imaged, and the longest axis was recorded. According to their shape, MPs were categorized as fragment, fiber, film or sphere by the following criteria. Fragment is the irregularly shaped particle that appears to be broken down from a large object. Fiber is a thread-shaped item with aspect ratio (length-to-width) > 5:1. Film is a flat and flexible particle with smooth or angular edges. Sphere is a particle with spherical, smooth or granular shape.

To prevent contamination, cotton lab coats and latex gloves were worn during the experiment, and samples were kept covered with aluminum foil or glass. The apparatus made of glass or metal was used in all processes except for PE bottle in sediment analysis. All equipment was rinsed twice with filtered Milli-Q water (Millipore, Tokyo, Japan) and all liquid solutions were filtered through glass fiber filters (GF/F, 0.75 μm, Whatman, Maidstone, United Kingdom (UK)). The contamination of PE particle originated from the PE bottle was checked by analyzing water samples rinsed the inside of the bottles with filtered Milli-Q water (100 ml × 3 times, n = 3) before use and also by running a procedural blank sample. As a result, three, zero and zero PE particles were detected in three bottle-washed samples, which is very limited compared to MP numbers found in sediment samples. In addition, no PE particle was found in the procedural blank sample. This result indicates that microplastic contamination by PE bottle was negligible.

Method validation was performed for organic matter digestion (H_2O_2 or enzyme mixture) and density separation (LMT or NaCl) processes through spike tests. PE (specific density = 0.95 g/cm^3), polypropylene (PP, 0.94 g/cm^3), EPS (0.015 g/cm^3), and polyester (1.35 g/cm^3) particles (<1 mm) were made in the lab by grinding large plastic products and used as test materials. Ten particles of each polymer were spiked for each test in triplicate. In both digestion processes (H_2O_2 and enzyme mixture), all test particles were recovered without physical damage or discoloration of polymers. The recoveries of microplastics in density separation process were as follows: PE (100%), PP (97 ± 6%), EPS (90 ± 10%), and polyester (60 ± 10%) for the NaCl treatment group, and PP (100%), PE (100%), EPS (97 ± 6%), and polyester (97 ± 6%) for the LMT treatment group.

Procedural blank samples were analyzed for every matrix. The background MP levels were 0.7 ± 0.5 particles/filter paper in bivalve samples (number of procedural blank samples, n = 18),

0 particles/filter paper in polychaete samples (n = 1), 2.3 ± 1.9 particles/filter paper in seawater samples (n = 4) and 0 particles/filter paper in sediment samples (n = 1), accounting for less than 10% of the MPs in all samples (Table S2). It is difficult to completely prevent contamination when detecting particles in the micron range; however, the background levels should be <10% of the average values determined from the samples (Hanke et al., 2013). A matrix spike test was performed for bivalve samples (ten particles of each PE, PP, EPS, and polyester, 1 mm length, n = 3). The mean recoveries (n = 3) of the added MPs were 90 ± 17% for PE, 97 ± 6% for PP, 97 ± 6% for EPS, and 87 ± 6% for polyester.

2.3. Statistical analyses

Statistical analyses were conducted using SPSS v. 19 and Sigma Plot v. 12.0 software. Statistical differences in MP abundance among the matrices and/or study sites were assessed using analysis of variance (ANOVA) followed by Tukey's test if the data were normally distributed; otherwise, the Kruskal–Wallis test was used, followed by multiple comparisons. Normality was tested using the Shapiro–Wilk W test. Differences in polymer types between the sites were assessed using principal component analysis (PCA).

3. Results

3.1. Regional MP contamination characteristics

3.1.1. Abundance, shape, color, and size of MPs

The mean MP abundances in seawater, sediment, oysters, mussels, and polychaetes were 0.77 ± 0.88 (median: 0.5) particles/L, 0.94 ± 0.69 (0.91) particles/g wet weight (w.w.), 1.13 ± 0.84 (0.84) particles/g w.w., 1.43 ± 1.45 (1.12) particles/g w.w., and 0.71 ± 1 (0.44) particles/g w.w., respectively. The MP levels in each matrix (except for sediment) were similar among the sites ($p > 0.5$, ANOVA or Kruskal–Wallis, Fig. 2). The mean MP levels in seawater and polychaetes from the aquafarm site were higher than those from other sites, but not significantly ($p > 0.5$, ANOVA or Kruskal–Wallis). The MP concentration in sediment from the urban site was significantly higher than that from the rural site ($p < 0.05$, ANOVA), but not different from that of the aquafarm site (Fig. 2).

MPs were found in various sizes, shapes, and colors in all matrices. We determined the frequency of MPs in various size ranges, with results as follows: 50–100 μm (10.8%), 100–200 μm (38.4%), 200–300 μm (18.1%), 300–400 μm (8.3%), 400–500 μm (8.1%), 500–600 μm (4.8%), 600–700 μm (3.9%), 700–800 μm (4.1%), 800–900 μm (1.8%), and 900–1000 μm (1.7%) (Fig. S2a). The most common size class was 100–200 μm in all matrices. Greater proportions of MP particles <300 μm were found in biotic matrices, including mussel (83%), oyster (78%), and polychaete (70%), than in abiotic matrices, including seawater (59%) and sediment (47%; Fig. S2a). Fragments were the most dominant component across all matrices, accounting for 89% of MP particles (Fig. S2b and Table S5). Fibers were more abundant in polychaetes (23%) and sediments (19%) than in bivalves (7%) and seawater (10%), but the difference was statistically insignificant. Film and sphere type of MP were not found in all samples. Various colors were detected, including blue, black, pink, yellow, red, green, and purple; however, colorless or transparent MP was dominant in both the abiotic and biotic matrices, accounting for 95% of all MPs (Fig. S2c). More colored MP was found in polychaete matrices than in other matrices due to the high proportion of black fibers (28%) in polychaete samples. There was no significant difference in MP size, shape, or color among the matrices or sampling sites.

3.1.2. MP polymer types

A total of 12 polymer types were identified, including PE, polypropylene (PP), polystyrene (PS), nylon, polyester, acrylic, polyvinyl acetate (PVA), polyurethane (PU), alkyd,

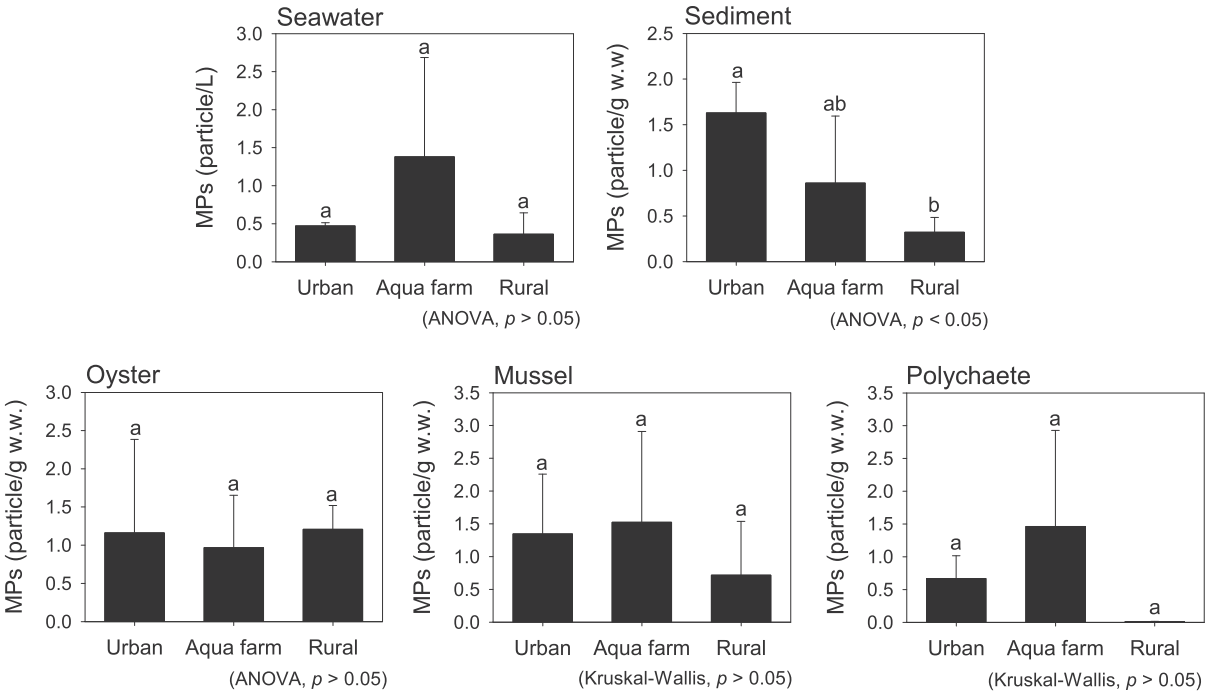

Fig. 2. Mean concentrations of microplastics in seawater, sediments, oysters, mussels, and polychaetes from urban, aquafarm, and rural sites.

polyoxymethylene (POM), polyvinyl chloride (PVC), and styrene butadiene (SB) copolymer (Fig. 3 and Table S4). Most PS MPs were composed of white EPS (density, 0.02–06 g/cm^3), with a glossy fluffy surface. Among all sites and matrices, low-density (compared to seawater, specific density = 1.02 g/cm^3) polymers (PE, PP, and EPS) were more common than high-density polymers (nylon, polyester, acrylic, PVA, PU, alkyd, POM, PVC, and SB copolymer; Fig. 3 and Table S3).

In abiotic matrices (seawater and sediment), we found nine polymer types in the urban site samples. The percentage of polymer type to the total microplastic found in abiotic matrices from urban site is as in the following: PP (33%) > PE (28%) > polyester (22%) > PU (20%) > PS (10%) > acrylic (6%) > PVA (4%) > alkyd (4%) > POM (2%; Fig. 3a). The proportion of high-density polymers was significantly higher (33.7%) in abiotic matrices from the urban site than in those from the aquafarm (1.9%) and rural sites (6.7%) ($p < 0.05$, Kruskal–Wallis). At the aquafarm site, seven polymer types were found in abiotic matrices, with the following composition: PS (49%) > PP (26%) > PE (23%) > PVA (0.8%) > polyester (0.4%) > acrylic (0.4%) > PU (0.4%; Fig. 3a). The proportion of PS was significantly higher in abiotic matrices from the aquafarm site (49.3%) than those from the urban (5.8%) and rural (1.3%) sites ($p < 0.05$, ANOVA; Fig. 3a), reflecting the widespread use of EPS buoys in the aquaculture industries (MOE, 2012; Lee et al., 2015). At the rural site, seven polymer types were found in abiotic matrices, with the following composition: PP (64%) > PE (28%) > polyester (2.7%) > PS (1.3%) > PVA (1.3%) > PU (1.3%) > POM (1.3%; Fig. 3a). PP MPs were abundant, probably due to the common use of PP rope in fishing activity (Jang et al., 2014a).

PCA was performed to identify regional MP contamination characteristics by examining the polymer compositions of abiotic samples from the three regions (Fig. 4). Two principal components explained >75% of the total variance in the data matrix. On the loading plot, PP, PS, and other polymers were well separated. The major negative loading polymer of component 1 was PP, and the major positive loading polymer was PS; PE and various other polymers were loaded between PP and PS. The distribution pattern of samples on the score plot is based on regional differences. Abiotic samples from the urban site are located at the middle of the plot, whereas those from rural and aquafarm sites are at the left and right sides, respectively. Thus, our PCA analysis indicates high polymer diversity in the urban site, with PS and PP polymers mainly occurring in the aquafarm and rural sites, respectively. Thus, within abiotic matrices, MP contamination was closely related to regional human activities.

We then compared the MP polymer compositions between biotic and abiotic matrices (Fig. 3b and Table S4). As in abiotic matrices, we observed high polymer composition diversity (ten polymer types) in biotic matrices at the urban site. There was a significantly higher proportion of high-density polymers in bivalves at the urban site (20.5%) than at the aquafarm (7.3%) and rural (0%) sites ($p < 0.05$, Kruskal–Wallis). SB copolymers were found in bivalve samples from the urban site (Fig. 3b). The proportion of high-density polymers found in polychaetes at the urban site (25%) was similar to that at the aquafarm site (35.7%); however, the diversity of polymer types was higher in samples from the urban site (n = 5; nylon, polyester, PVA, PU, and PVC) than in those from the aquafarm site (n = 1; polyester). At the aquafarm site, PS

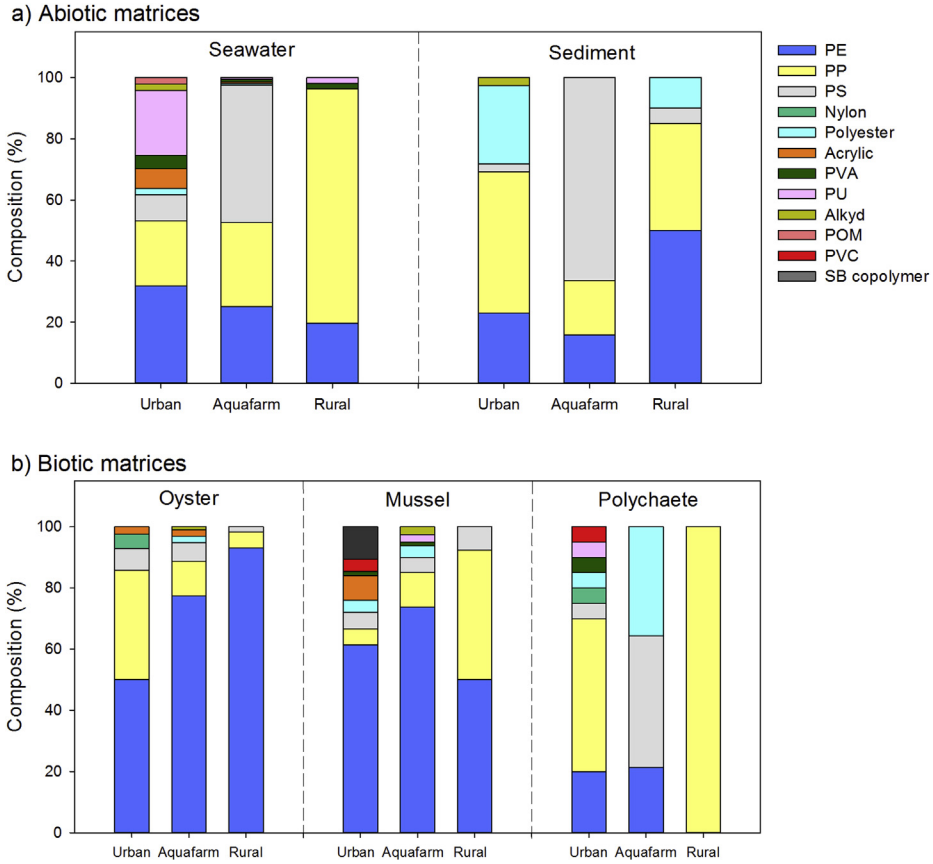

Fig. 3. Compositions of polymer types in (a) abiotic matrices (seawater and sediment) and (b) biotic matrices (oysters, mussels, and polychaetes) from urban, aquafarm, and rural sites.

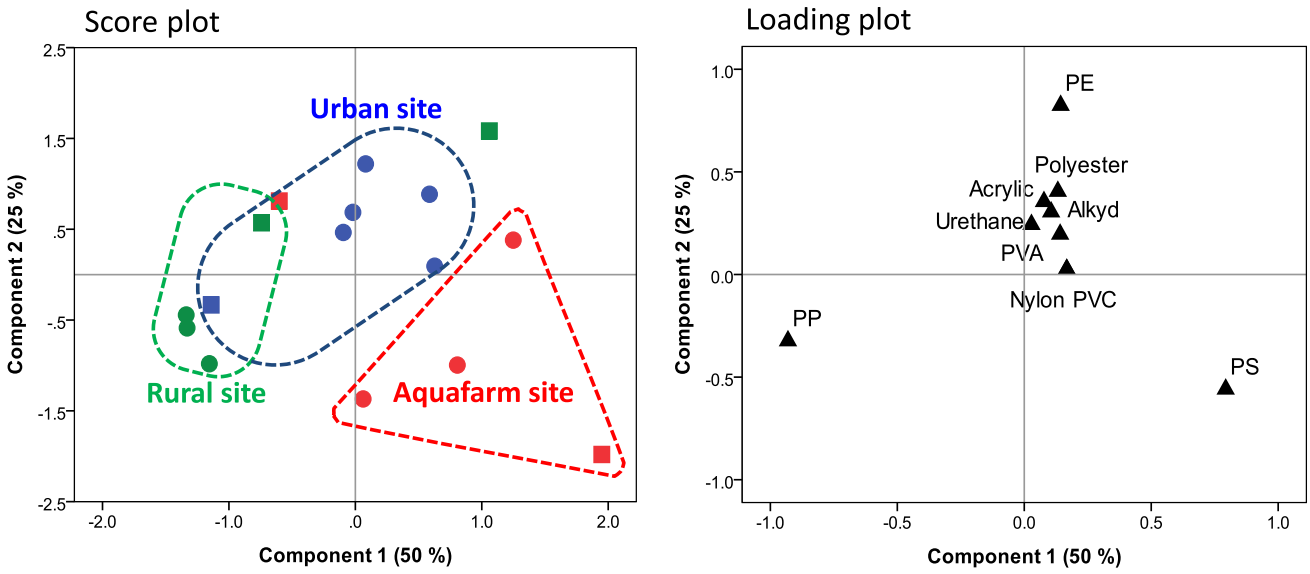

Fig. 4. Results of principal component analysis identifying the regional microplastic contamination characteristics. Polymer composition data for sediment (■) and seawater (●) samples from urban, aquafarm, and rural sites were used in the analysis.

was dominant among polychaetes (42.9%), like as in sediment (66.4%) and seawater (44.9%; Fig. 3b). The proportion of PS was low among bivalves (5.6%) living in the sub-surface water column, reflecting its reduced composition in the sub-surface layer of water column (5.3%; Fig. S3). Only three polymer types were found in biotic matrices from the rural site (Fig. 3b). PP and PE were abundant in both bivalves (95.4%) and polychaetes (100%), and in abiotic matrices (91.9%; Fig. 3). These results show that the bioaccumulation profiles of MPs in marine invertebrates reflected those of the abiotic matrices.

3.2. MPs in urban creek water and road dust

Eight polymer types, including PE (5%), PP (29%), PS (5%), polyester (24%), acrylic (5%), PVA (5%), PVC (24%), and SB copolymer (5%), were found in creek water from the urban site, indicating that diverse polymers are discharged from the city to the ocean via river streams. The proportion of high-density polymers in creek water (63%) was higher than those in seawater (37%) and sediment (29%; Table S3) from the urban site. The high diversity of polymer types and large proportion of high-density polymers in both creek water and marine matrices suggest that MPs within Masan Bay come from inland sources (Table S3).

Within the road dust samples, we found five polymer types in the following proportions: PP (41%) > PE (31%) > PS (17%) > SB copolymer (7%) > PVA (5%) (Table S3). Black MPs were dominant (52%; Fig. S4), representing 36% of PE, 46% of PP, 14% of PS, 9% of PVA, and 9% of SB copolymer particles. SB copolymer, widely used as a tire ingredient, was found in both road dust and bivalve samples from the urban site, implying that tire particles are transported from roads to marine environments, eventually accumulating in marine biota (Fig. 5).

4. Discussion

MPs collected from marine matrices in this study were mainly identified as PE and PP (Fig. 3), which are the most common polymer types in seawater, marine sediment, and marine

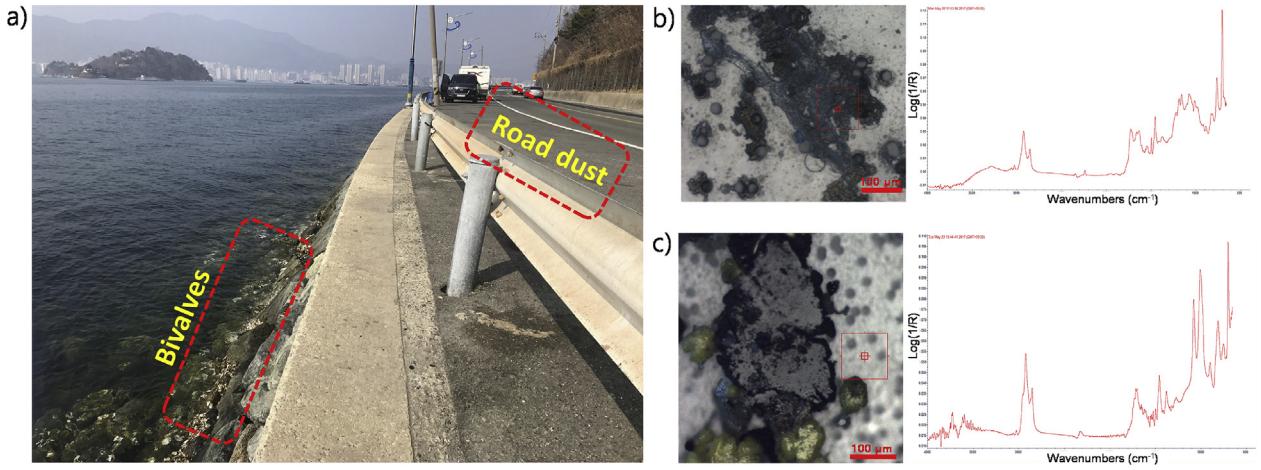

Fig. 5. (a) Sampling locations for bivalves and road dust from the urban site. Spectra of polystyrene butadiene copolymer for b) bivalve and c) road dust samples.

organisms worldwide (Frère et al., 2017; Zhang et al., 2017; Phuong et al., 2018). PE and PP are produced in large quantities globally and are widely used (Andrady, 2015); their low specific density (PP, 0.95 g/cm^3; PE, 0.91–0.97 g/cm^3) allow them to float on water surfaces and travel long distances via ocean currents, resulting in wide distribution, even to non-urbanized or remote areas.

In the current study, the lowest polymer diversity was observed at the rural site, which mainly containing PE and PP (Figs. 3 and 4); the MP abundance was slightly lower at the rural site than at the urban or aquafarm sites. This result raises the possibility of the existence of a local source for MPs at the rural site. In rural coastal areas, small-scale capture fisheries widely employ fishing nets and ropes; the weathering and fragmentation of fishing gear can be a potential MP source for marine environments. Fishing rope and rope debris from Korean coasts mainly consist of PP and PP/PE copolymer (polypropylene + poly[propylene:ethylene]) (Jang et al., 2014a). Interestingly, many PP MPs found in samples from the rural site had line-patterned surfaces (Fig. 6a), resembling those of PP rope (Fig. 6c). In contrast, many PP MPs from the urban site had smooth or solid surfaces (Fig. 6b), which are consistent with PP baskets and containers (Fig. 6d). PP MPs with line patterns were most prevalent at the rural site (67%), followed by the aquafarm (37%) and urban sites (19%). This result indicates that the surface characteristics of MPs can provide information about their origins.

PS MP was more abundant than other polymer types in seawater, sediment, and polychaetes from the aquafarm site (Figs. 3 and 4), reflecting the widespread use of EPS buoys in this region. The suspended long-line culture system, supported by numerous floating EPS buoys, is commonly used for oyster production in South Korea. About 2 million EPS buoys are newly deployed every year, but only 28% of these are retrieved by the government for recycling (MOE, 2012). It is estimated that 990,000 EPS buoys were lost or disposed of inappropriately in 2012 (Jang et al., 2014b), resulting in the prevalence of EPS debris and its fragments on the Korean coasts (Lee et al., 2013; Heo et al., 2013; Kang et al., 2015). EPS debris is easily fragmented by mechanical abrasion because it is a cellular plastic material consisting of numerous small spherical balls 2–4 mm in diameter. Ultraviolet (UV) radiation can produce micro- and nano-sized EPS particles directly on the surface of EPS debris (Song et al., 2017). Polychaetes inhabiting EPS debris can also create numerous micro-sized EPS particles (Jang et al., 2018). In the current study, we expected high ingestion of EPS particles by bivalves from the aquafarm site; however, less PS MP was found at this site than PE or PP. This finding is inconsistent with the relatively high proportion of PS observed in surface seawater. This difference may be related to exposure conditions in the water column. Bivalve samples were collected under floating PE buoys at the aquafarm site (Fig. S3). Therefore, these bivalves should not have been exposed to the top surface layer (including microlayer) of the water column, but instead to the sub-surface layer, where EPS particles were less abundant due to their high buoyancy (0.02–06 g/cm^3; Fig. S3). In contrast, polychaetes inhabiting the intertidal zone at the aquafarm site showed high proportions of PS MPs. Nevertheless, the concentration of PS MPs in bivalves at the aquafarm site (0.67 ± 1.48 particles/individual) was higher than those from the urban (0.36 ± 0.72 particles/individual) and rural (0.11 ± 0.24 particles/individual) sites, but the difference was statistically insignificant. The unexpectedly high PE particle abundance (7.21 ± 6.05 particles/individual) among aquafarm bivalve samples was responsible for the low proportion of PS particles. The high ingestion of PE particles by oysters and mussels at this site may be due to the wide use of PP/PE copolymer ropes and HDPE buoys along with EPS buoys for aquafarming in this region (Fig. S5). The aquafarms use the hanging-long line culture system, where the oyster strings are suspended on submerged PP/PE ropes, supported by numerous EPS and HDPE floating buoys (Fig. S5). Each long line

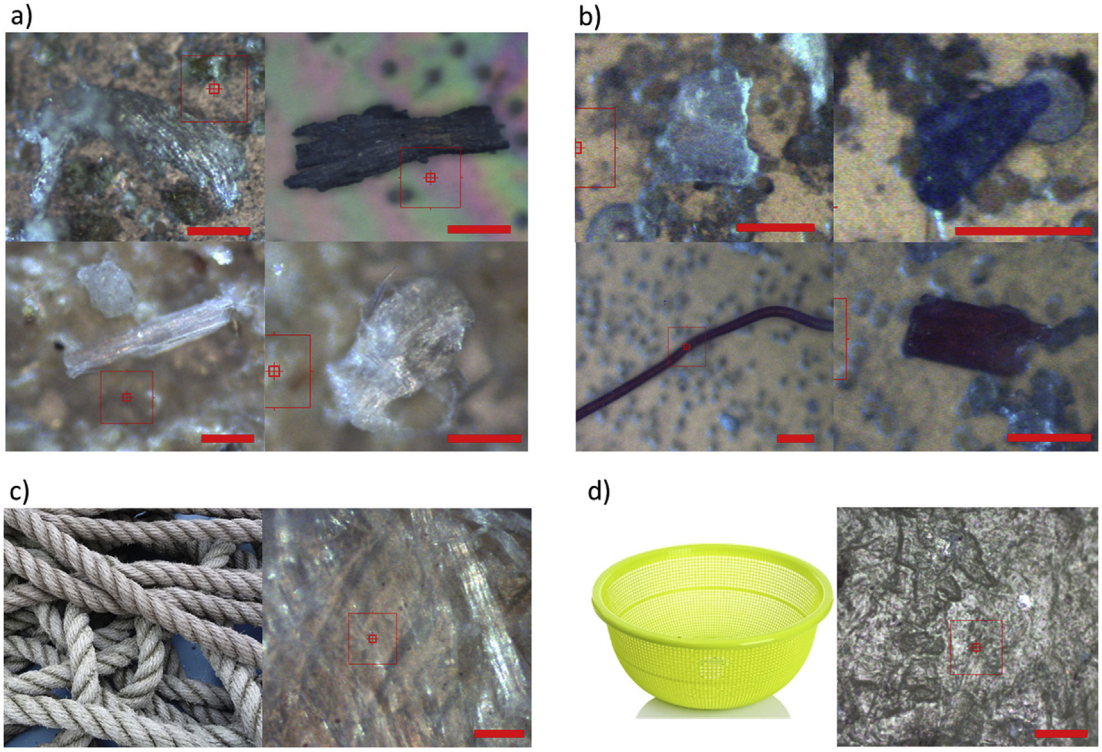

Fig. 6. Microscope images of PP MPs in marine matrices from a) rural and b) urban sites. Pictures of PP type of a) fishing rope and b) plastic try, and their surface images (red bar: 100 μm).

is around 100 m of rope. Jang et al., 2014a reported that polymer type of ropes (n = 9) currently employed in aquacultures, fishing nets and ships was PP/PE copolymer (78%) and PE (22%). HDPE buoys, an alternative to EPS buoys, are constructed with roto-moulded polyethylene hulls filled with EPS foam. Since 2009, the Korean government has encouraged the replacement of EPS buoys with more durable buoys to reduce EPS pollution in the Korean coastal environment (Lee et al., 2015). PE buoys are now frequently found in oyster aquaculture farms (Fig. S5). PE MPs are likely to be produced from PE buoys, ropes and their debris via weathering processes (Song et al., 2017), providing a PE source in marine environments.

Diverse polymers were found in marine matrices from the urban site (Figs. 3 and 4). Similarly, a high diversity of polymer types (PE, PP, PS, polyester, acrylic, PVA, SB copolymer, and PVC) was observed in creek water that had passed through the city, indicating that MPs in marine matrices may have come from inland sources (Table S3). Song et al. (2018) also reported that high-density polymers are frequently detected in seawater from urban area compared to that from rural area. Sewage treatment plant effluents are the major discharge route of domestic and industrial wastewater (about 260,000 tons/day) into the bay. Some untreated sewage and wastewater also enters the bay through streams (MOMAF, 2004; Lee et al., 2011; Eo et al., 2019). Lebreton et al. (2017) estimated that 1.15–2.41 million tons of plastic waste enters the ocean yearly from global rivers. Hardesty et al. (2016) reported that plastic transport by water (e.g., by storm drains and creeks) is the second most important factor in plastic load to coastal regions. In road dust samples, SB copolymer (a known tire ingredient) was found, along with PE, PP, PS, and PVA (Table S3 and Fig. 5). Tire materials are complex mixtures comprising natural and synthetic petroleum-based rubber and other materials such as carbon black, mineral oils, ZnO, and sulfur (Wagner et al., 2018). Styrene-butadiene rubber (SBR) is a component of synthetic rubber. The interaction of tires with pavement generates numerous MP particles. Kole et al. (2017) estimated that 5.9 million tons of tire particles are emitted globally each year. About 90% of these particles are deposited on or beside roads, to eventually be carried away by surface runoff (Wicke et al., 2012; Huber et al., 2016). We detected SB copolymers in road dust (9%) and in bivalves (7%) living near roads (Fig. 5 and Table S3). This result indicates that tire particles are transported from roads to the marine environment. To our knowledge, this is the first field report of tire particle ingestion by marine biota. In a recent study, individual-level adverse effects, such as on survival, growth, and feeding rate, were not found in freshwater benthic invertebrates following ingestion of tire particles (Redondo-Hasselerharm et al., 2018). However, tire particle ingestion by low-trophic-level marine organisms provides new insight into their potential ecological risks to marine communities. These organisms are likely to carry tire particles and their associated chemicals into the marine food web (Wright et al., 2013). Humans can also ingest micro-sized tire particles via seafood consumption.

In many studies, fibers have been reported as the predominant MP shape among marine matrices (>90%) (Cesa et al., 2017; Leslie et al., 2017). Much attention has been paid to synthetic fiber pollution; washing machine effluents are a major source of MP fibers in the ocean (Hartline et al., 2016; Napper and Thompson, 2016). However, in this study, fragments were the most common type of MP found, perhaps due to different plastic usage patterns between countries. Differing MP analysis methods, especially size range and chemical characterization, may also have contributed to this difference. Eo et al. (2019) observed that fragment type was predominant in MP < 300 μm in river water, but fiber type was dominant in MP > 300 μm. In a number of recent European studies, MP fragments were found to be the dominant shape in bivalves when spectroscopy was used to identify suspected plastics (Digka et al., 2018; Phuong et al., 2018), whereas fibers were dominant when visual identification was conducted along with spectroscopy identification for sub-samples (commonly < 10% of total particles; Catarino et al., 2017; Leslie et al., 2017; Renzi et al., 2018). Cho et al. (2019) reported that > 80% of microfibers on filter papers was identified as natural fibers such as cotton and paper. Visual identification cannot successfully distinguish between synthetic and natural fibers because many synthetic fibers are colorless, and natural fibers can contain various colors that appear in synthetic fibers, such as yellow and red (Song et al., 2015), resulting in overestimations of microfiber concentration. In this study, the polymer composition of fiber MPs was 63% for PP, 16% for PE, 13% for polyester, and 6% for PU, implying that most fibers originated from fishing activities.

It is commonly thought that 80% of marine debris originates from land-based sources (Andrady, 2011; Nelms et al., 2017; Wang et al., 2017). Similarly high MP abundances have been reported in seawater, sediment, and bivalves near populated regions (Frère et al., 2017; Antunes et al., 2018; Li et al., 2018; Eo et al., 2018). However, in this study, the mean MP concentrations at the aquafarm site (population density: 198 people/km^2) were similar to or higher than those at the urban site (2683 people/km^2; Fig. 2). Moreover, although the population density was much lower at the rural site (55 people/km^2) than at the urban site, the MP abundances in seawater and marine invertebrates were not significantly different between these sites (Fig. 2). This result indicates a wide dispersion of MPs in a large region along the coast and the existence of regional sources for MPs in both the aquafarm and rural sites; the polymeric composition also supports regional marine-based human activities as a source of MPs. Hong et al. (2014) reported that EPS buoys (12.8% of marine debris) and fishing ropes (8.2%) account for most of the beach macro-debris along the coasts of Korea, meaning that the main sources of beach marine debris in Korea being fisheries, including commercial fisheries and marine aquaculture. Due to its high salt content, the recycling rate of plastic waste is much lower for plastics used for marine-based activities than for land-based activities, resulting in a higher abundance of singular fishery items debris in coastal regions (Jang et al., 2014a). Similar to Korea, marine debris originating from marine-based sources has been reported to be dominant in Australia, Brazil, Japan, and Chile, where fisheries are active (Edyvane et al., 2004; Santos et al., 2005; Hinojosa and Thiel, 2009; Goto and Shibata, 2015). Jambeck et al. (2015) estimated that 4.8–12.7 million metric tons of plastic waste generated on land enters the ocean every year. Similarly, Lebreton et al. (2017) estimated that 1.15–2.41 million tons of plastic waste currently enters the ocean from land via rivers every year. However, there is no estimation of the mass of plastic marine debris generated from marine-based sources. The results of this study highlight need for research on plastic waste generated from marine-based human activities. Such research is even more important for countries with developed marine industries.

5. Conclusion

MPs are being introduced into the ocean at an unprecedented rate. Several studies have been conducted on the impact of human activities on the MP abundance in the surrounding marine environment. However, limited information is available on the impact of different types of human activity on the characteristics (e.g., shape and polymer type) of MPs in the marine environment. In this study, MP contamination was studied in abiotic and biotic marine matrices from three coastal regions with different types of local human activities. In samples from the urban site, MPs of diverse

polymer types were found in marine matrices, demonstrating a close relationship with inland sources such as road dust and creek water. MP polymer types and shapes revealed that marine-based human activities such as fisheries and marine aquaculture have become important sources of MPs in aquafarm and rural sites. Our results indicate that regional human activities affect the contamination characteristics of MPs in the marine environment. Further research should be performed to evaluate the MP contamination caused by marine-based human activities, especially in countries with developed marine industries.

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.

Acknowledgements

This study was supported by a research project titled “Environmental Risk Assessment of Microplastics in the Marine Environment” from the Ministry of Oceans and Fisheries, South Korea.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.watres.2019.115400.

References

Andrady, A.L., 2011. Microplastics in the marine environment. Mar. Pollut. Bull. 62, 1596–1605. https://doi.org/10.1016/j.marpolbul.2011.05.030.

Andrady, A.L., 2015. Persistence of plastic litter in the oceans. In: Bergmann, M., Gutow, L., Klages, M. (Eds.), Marine Anthropogenic Litter. Springer International Publishing, Cham, pp. 58–60, 2015.

Antunes, J., Frias, J., Sobral, P., 2018. Microplastics on the Portuguese coast. Mar. Pollut. Bull. 131, 294–302. https://doi.org/10.1016/j.marpolbul.2018.04.025.

Arthur, C., Baker, J., Bamford, H., 2009. Proceedings of the International Research Workshop on the Occurrence, Effects and Fate of Microplastic Marine Debris, p. 49 (NOAA Technical Memorandum NOS-OR&R-30).

Auta, H.S., Emenike, C.U., Fauziah, S.H., 2017. Distribution and importance of microplastics in the marine environment: a review of the sources, fate, effects, and potential solutions. Environ. Int. 102, 165–176. https://doi.org/10.1016/j.envint.2017.02.013.

Bejgarn, S., MacLeod, M., Bogdal, C., Breitholtz, M., 2015. Toxicity of leachate from weathering plastics: an exploratory screening study with Nitocra spinipes. Chemosphere 132, 114–119. https://doi.org/10.1016/j.chemosphere.2015.03.010.

Browne, M.A., Galloway, T.S., Thompson, R.C., 2010. Spatial patterns of plastic debris along Estuarine shorelines. Environ. Sci. Technol. 44, 3404–3409. https://doi.org/10.1021/es903784e.

Browne, M.A., Crump, P., Niven, S.J., Teuten, E., Tonkin, A., Galloway, T., Thompson, R., 2011. Accumulation of microplastic on shorelines woldwide: sources and sinks. Environ. Sci. Technol. 21, 9175–9179. https://doi.org/10.1021/es201811s.

Browne, M.A., Niven, S.J., Galloway, T.S., Rowland, S.J., Thompson, R.C., 2013. Microplastic moves pollutants and additives to worms, reducing functions linked to health and biodiversity. Curr. Biol. 23, 2388–2392. https://doi.org/10.1016/j.cub.2013.10.012.

Carpenter, E.J., Anderson, S.J., Harvey, G.R., Miklas, H.P., Peck, B.B., 1972. Polystyrene spherules in coastal waters. Science 178, 749–750.

Catarino, A.I., Thompson, R., Sanderson, W., Henry, T.B., 2017. Development and optimization of a standard method for extraction of microplastics in mussels by enzyme digestion of soft tissues. Environ. Toxicol. Chem. 4, 947–951. https://doi.org/10.1002/etc.3608.

Cesa, F.S., Turra, A., Baruque-Ramos, J., 2017. Synthetic fibers as microplastics in the marine environment: a review from textile perspective with a focus on domestic washings. Sci. Total Environ. 598, 1116–1129. https://doi.org/10.1016/j.scitotenv.2017.04.172.

Cho, Y., Shim, W.J., Jang, M., Han, G.M., Hong, S.H., 2019. Abundance and characteristics of microplastics in market bivalves from South Korea. Environ. Pollut. 245, 1107–1116. https://doi.org/10.1016/j.envpol.2018.11.091.

Digka, K., Tsangaris, C., Torre, M., Anastasopoulou, A., Zeri, C., 2018. Microplastics in mussels and fish from the Northern Ionian Sea. Mar. Pollut. Bull. 135, 30–40. https://doi.org/10.1016/j.marpolbul.2018.06.063.

do Sul, J.A.I., Costa, M.F., 2014. The present and future of microplastic pollution in the marine environment. Environ. Pollut. 185, 352–364. https://doi.org/10.1016/j.envpol.2013.10.036.

Edyvane, K.S., Dalgetty, A., Hone, P.W., Higham, J.S., Wace, M.N., 2004. Long-term marine litter monitoring in the remote great Australian Bight, South Australia. Mar. Pollut. Bull. 48, 1060–1075. https://doi.org/10.1016/j.marpolbul.2003.12.012.

Eo, S., Hong, S.H., Song, Y.K., Lee, J., Lee, J., Shim, W.J., 2018. Abundance, composition, and distribution of microplastics larger than 20 μm in sand beaches of South Korea. Environ. Pollut. 238, 894–902. https://doi.org/10.1016/j.envpol.2018.03.096.

Eo, S., Hong, S.H., Song, Y.K., Han, G.M., Shim, W.J., 2019. Spatiotemporal distribution and annual load of microplastics in the Nakdong River, South Korea. Water Res. 160, 228–237.

Fendall, L.S., Sewell, M.A., 2009. Contributing to marine pollution by washing your face: microplastics in facial cleansers. Mar. Pollut. Bull. 58, 1225–1228. https://doi.org/10.1016/j.marpolbul.2009.04.025.

Frère, L., Paul-Pont, I., Rinnert, E., Petton, S., Jaffre, J., Bihannic, I., Soudant, P., Lambert, C., Huvet, A., 2017. Influence of environmental and anthropogenic factors on the composition, concentration and spatial distribution of microplastics: a case study of the Bay of Brest (Brittany, France). Environ. Pollut. 225, 211–222. https://doi.org/10.1016/j.envpol.2017.03.023.

Gall, S.C., Thompson, R.C., 2015. The impact of debris on marine life. Mar. Pollut. Bull. 92, 170–179. https://doi.org/10.1016/j.marpolbul.2014.12.041.

GESAMP, 2019. Guidelines or the monitoring and assessment of plastic litter and microplastics in the ocean. In: Kershaw, P.J., Turra, A., Galgani, F. (Eds.), Rep. Stud. GESAMP No. 99, 130p. IMO/FAO/UNESCO-IOC/UNIDO/WMO/IAEA/UN/UNEP/UNDP/ISA Joint Group of Experts on the Scientific Aspects of Marine Environmental Protection.

GNDI (Gyung Nam Development Institute), 2012. Strategy for enhancement of shellfish farming in Gyungnam province. GNDI. Retrieved from. http://gndi.re.kr/gndi2016/report/sub01.html?mode=view&cate=8&subcate=8&number=770&keyfield=keyfield=&key=&area=&page=28. On Feb 8, 2019, 1–8.

Goto, T., Shibata, H., 2015. Changes in abundance and composition of anthropogenic marine debris on the continental slope off the Pacific coast of northern Japan, after the March 2011 Tohoku earthquake. Mar. Pollut. Bull. 95, 234–241. https://doi.org/10.1016/j.marpolbul.2015.04.011.

Hanke, G., Galgani, F., Werner, S., Oosterbaan, L., Nilsson, P., Fleet, D., Kinsey, S., Thompson, R., Van Franeker, J.A., Vlachogianni, T., Palatinus, A., Scoullos, M., Veiga, J.M., Matiddi, M., Alcaro, L., Maes, T., Korpinen, S., Budziak, A., Leslie, H., Gago, J., Liebezeit, G., 2013. Guidance on Monitoring of Marine Litter in European Seas. European Commission.

Hardesty, B.D., Schuyler, Q., Lawson, T.J., Opie, K., Wilcox, C., 2016. Understanding Debris Sources and Transport from the Coastal Margin to the Ocean. CSIRO.

Hartline, N.L., Bruce, N.J., Karba, S.N., Ruff, E.O., Sonar, S.U., Holden, P.A., 2016. Microfiber masses recovered from conventional machine washing of new or aged garments. Environ. Sci. Technol. 50, 11532–11538. https://doi.org/10.1021/acs.est.6b03045.

Heo, N.W., Hong, S.H., Han, G.M., Hong, S., Lee, J., Song, Y.K., Jang, M., Shim, W.J., 2013. Distribution of small plastic debris in cross-section and high strandline on Heungnam beach, South Korea. Ocean Sci. J. 48, 225–233. https://doi.org/10.1007/s12601-013-0019-9.

Hinojosa, I.A., Thiel, M., 2009. Floating marine debris in fjords, gulfs and channels of southern Chile. Mar. Pollut. Bull. 58, 341–350. https://doi.org/10.1016/j.marpolbul.2008.10.020.

Hong, S.H., Yim, U.H., Shim, W.J., Oh, J.R., Lee, I.S., 2003. Horizontal and vertical distribution of PCBs and chlorinated pesticides in sediments from Masan Bay, Korea. Mar. Pollut. Bull. 46, 244–253. https://doi.org/10.1016/S0025-326x(02)00399-5.

Hong, S., Lee, J., Kang, D., Choi, H., Ko, S., 2014. Quantities, composition, and sources of beach debris in Korea from the results of nationwide monitoring. Mar. Pollut. Bull. 84, 27–34. https://doi.org/10.1016/j.marpolbul.2014.05.051.

Huber, M., Welker, A., Helmreich, B., 2016. Critical review of heavy metal pollution of traffic area runoff: occurrence, influencing factors, and partitioning. Sci. Total Environ. 541, 895–919. https://doi.org/10.1016/j.scitotenv.2015.09.033.

Hurley, R., Woodward, J., Rothwell, J.J., 2018. Microplastic contamination of river beds significantly reduced by catchment wide flooding. Nat. Geosci. 11, 251–257. https://doi.org/10.1038/s41561-018-0080-1.

Jambeck, J.R., Geyer, R., Wilcox, C., Siegler, T.R., Perryman, M., Andrady, A., Narayan, R., Law, K.L., 2015. Plastic waste inputs from land into the ocean. Science 347, 768–771. https://doi.org/10.1126/science.1260352.

Jang, M., Shim, W.J., Han, G.M., Rani, M., Song, Y.K., Hong, S.H., 2016. Styrofoam debris as a source of hazardous additives for marine organisms. Environ. Sci. Technol. 50, 4951–4960. https://doi.org/10.1021/acs.est.5b05485.

Jang, M., Shim, W.J., Han, G.M., Rani, M., Song, Y.K., Hong, S.H., 2017. Widespread detection of a brominated flame retardant, hexabromocyclododecane, in expanded polystyrene marine debris and microplastics from South Korea and the Asia-Pacific coastal region. Environ. Pollut. 231, 785–794. https://doi.org/10.1016/j.envpol.2017.08.066.

Jang, M., Shim, W.J., Han, G.M., Song, Y.K., Hong, S.H., 2018. Formation of microplastics by polychaetes (Marphysa sanguinea) inhabiting expanded polystyrene marine debris. Mar. Pollut. Bull. 131, 365–369. https://doi.org/10.1016/j.marpolbul.2018.04.017.

Jang, Y.C., Lee, J., Hong, S., Lee, J.S., Shim, W.J., Song, Y.K., 2014a. Sources of plastic marine debris on beaches of Korea: more from the ocean than the land. Ocean Sci. J. 49, 151–162. https://doi.org/10.1007/s12601-014-0015-8.

Jang, Y.C., Lee, J., Hong, S., Mok, J.Y., Kim, K.S., Lee, Y.J., Choi, H., Kang, H., Lee, S., 2014b. Estimation of the annual flow and stock of marine debris in South Korea for management purposes. Mar. Pollut. Bull. 86, 505–511. https://doi.org/10.1016/j.marpolbul.2014.06.021.

Kang, J.H., Kwon, O.Y., Lee, K.W., Song, Y.K., Shim, W.J., 2015. Marine neustonic microplastics around the southeastern coast of Korea. Mar. Pollut. Bull. 96, 304–312. https://doi.org/10.1016/j.marpolbul.2015.04.054.

Kole, P.J., Lohr, A.J., Van Belleghem, F.G.A.J., Ragas, A.M.J., 2017. Wear and tear of tyres: a stealthy source of microplastics in the environment. Int. J. Environ. Res. Public Health 14, 1265. https://doi.org/10.3390/ijerph14101265.

Lebreton, L.C.M., van der Zwet, J., Damsteeg, J.W., Slat, B., Andrady, A., Reisser, J., 2017. River plastic emissions to the world's oceans. Nat. Commun. 8, 15611. https://doi.org/10.1038/ncomms15611.

Lee, H.J., Hong, S.H., Kim, M., Ha, S.Y., An, S.M., Shim, W.J., 2011. Tracing origins of sewage and organic matter using dissolved sterols in masan and Haengam bay, Korea. Ocean Sci. J. 46, 95–103. https://doi.org/10.1007/s12601-011-0009-8.

Lee, J., Hong, S., Song, Y.K., Hong, S.H., Jang, Y.C., Jang, M., Heo, N.W., Han, G.M., Lee, M.J., Kang, D., Shim, W.J., 2013. Relationships among the abundances of plastic debris in different size classes on beaches in South Korea. Mar. Pollut. Bull. 77, 349–354. https://doi.org/10.1016/j.marpolbul.2013.08.013.

Lee, J., Hong, S., Jang, Y.C., Lee, M.J., Kang, D., Shim, W.J., 2015. Finding solutions for the styrofoam buoy debris problem through participatory workshops. Mar. Policy 51, 182–189. https://doi.org/10.1016/j.marpol.2014.08.008.

Leslie, H.A., Brandsma, S.H., van Velzen, M.J.M., Vethaak, A.D., 2017. Microplastics en route: field measurements in the Dutch river delta and Amsterdam canals, wastewater treatment plants, North Sea sediments and biota. Environ. Int. 101, 133–142. https://doi.org/10.1016/j.envint2017.01.018.

Li, D., Dong, M., Shim, W.J., Yim, U.H., Hong, S.H., Kannan, N., 2008. Distribution characteristics of nonylphenolic chemicals in Masan Bay environments, Korea. Chemosphere 71, 1162–1172. https://doi.org/10.1016/j.chemosphere.2007.10.023.

Li, H.X., Ma, L.S., Lin, L., Ni, Z.X., Xu, X.R., Shi, H.H., Yan, Y., Zheng, G.M., Rittschof, D., 2018. Microplastics in oysters *Saccostrea cucullata* along the Pearl river Estuary, China. Environ. Pollut. 236, 619–625. https://doi.org/10.1016/j.envpol.2018.01.083.

Li, J.N., Qu, X.Y., Su, L., Zhang, W.W., Yang, D.Q., Kolandhasamy, P., Li, D.J., Shi, H.H., 2016. Microplastics in mussels along the coastal waters of China. Environ. Pollut. 214, 177–184. https://doi.org/10.1016/j.envpol.2016.04.012.

Lithner, D., Larsson, A., Dave, G., 2011. Environmental and health hazard ranking and assessment of plastic polymers based on chemical composition. Sci. Total Environ. 409, 3309–3324. https://doi.org/10.1016/j.scitotenv.2011.04.038.

MOE, 2012. Announcement No.2012-243. Required Recycle Ratio for Each Product for the Year of 2013. Korea Ministry of Environment (In Korean).

MOMAF, 2004. Development of Integrated Environmental Management System for the Coastal Area of Korea. Ministry of Marine affairs and Fisheries, pp. 260–282 (In Korean).

Napper, I.E., Thompson, R.C., 2016. Release of synthetic microplastic plastic fibres from domestic washing machines: effects of fabric type and washing conditions. Mar. Pollut. Bull. 112, 39–45. https://doi.org/10.1016/j.marpolbul.2016.09.025.

Nel, H.A., Dalu, T., Wasserman, R.J., 2018. Sinks and sources: assessing microplastic abundance in river sediment and deposit feeders in an Austral temperate urban river system. Sci. Total Environ. 612, 950–956. https://doi.org/10.1016/j.scitotenv.2017.08.298.

Nelms, S.E., Coombes, C., Foster, L.C., Galloway, T.S., Godley, B.J., Lindeque, P.K., Witt, M.J., 2017. Marine anthropogenic litter on British beaches: a 10-year nationwide assessment using citizen science data. Sci. Total Environ. 579, 1399–1409. https://doi.org/10.1016/j.scitotenv.2016.11.137.

Phuong, N.N., Poirier, L., Pham, Q.T., Lagarde, F., Zalouk-Vergnoux, A., 2018. Factors influencing the microplastic contamination of bivalves from the French Atlantic coast: location, season and/or mode of life? Mar. Pollut. Bull. 129, 664–674. https://doi.org/10.1016/j.marpolbul.2017.10.054.

Redondo-Hasselerharm, P.E., Falahudin, D., Peeters, E.T.H.M., Koelmans, A.A., 2018. Microplastic effect thresholds for freshwater benthic macroinvertebrates. Environ. Sci. Technol. 52, 2278–2286. https://doi.org/10.1021/acs.est.7b05367.

Renzi, M., Guerranti, C., Blaslovic, A., 2018. Microplastic contents from maricultured and natural mussels. Mar. Pollut. Bull. 131, 248–251. https://doi.org/10.1016/j.marpolbul.2018.04.035.

Rochman, C.M., Hoh, E., Kurobe, T., Teh, S.J., 2013. Ingested plastic transfers hazardous chemicals to fish and induces hepatic stress. Sci. Rep. 3, 3263. https://doi.org/10.1038/srep03263.

Rochman, C.M., 2018. Microplastics research - from sink to source. Science 360, 28–29. https://doi.org/10.1126/science.aar7734.

Rodrigues, M.O., Abrantes, N., Goncalves, F.J.M., Nogueira, H., Marques, J.C., Goncalves, A.M.M., 2018. Spatial and temporal distribution of microplastics in water and sediments of a freshwater system (Antua River, Portugal). Sci. Total Environ. 633, 1549–1559. https://doi.org/10.1016/j.scitotenv.2018.03.233.

Santos, I.R., Friedrich, A.C., Barretto, F.P., 2005. Overseas garbage pollution on beaches of northeast Brazil. Mar. Pollut. Bull. 50, 782–786. https://doi.org/10.1016/j.marpolbul.2005.04.044.

Sheavly, S.B., 2005. Sixth Meeting of the UN Open-Ended Informal Consultative Process on Oceans & the Law of the Sea. In: http://www.un.org/depts/los/consultative_process/6thmeetingpanel.htm#Discussion Panel B.

Shim, W.J., Hong, S.H., Eo, S., 2018. Marine microplastics: abundance, distribution, composition. In: Zeng, E.Y. (Ed.), Microplastic Contamination in Aquatic Environments. Elsevier, pp. 1–26.

Shim, W.J., Oh, J.R., Kahng, S.H., Shim, J.H., Lee, S.H., 1999. Horizontal distribution of butyltins in surface sediments from an enclosed bay system, Korea. Environ. Pollut. 106, 351–357. https://doi.org/10.1016/S0269-7491(99)00109-8.

Song, Y.K., Hong, S.H., Jang, M., Kang, J.H., Kwon, O.Y., Han, G.M., Shim, W.J., 2014. Large accumulation of micro-sized synthetic polymer particles in the sea surface microlayer. Environ. Sci. Technol. 48, 9014–9021. https://doi.org/10.1021/es501757s.

Song, Y.K., Hong, S.H., Jang, M., Han, G.M., Rani, M., Lee, J., Shim, W.J., 2015. A comparison of microscopic and spectroscopic identification methods for analysis of microplastics in environmental samples. Mar. Pollut. Bull. 93, 202–209. https://doi.org/10.1016/j.marpolbul.2015.01.015.

Song, Y.K., Hong, S.H., Jang, M., Han, G.M., Jung, S.W., Shim, W.J., 2017. Combined effects of UV exposure duration and mechanical abrasion on microplastic fragmentation by polymer type. Environ. Sci. Technol. 51, 4368–4376. https://doi.org/10.1021/acs.est.6b06155.

Song, Y.K., Hong, S.H., Eo, S., Jang, M., Han, G.M., Isobe, A., Shim, W.J., 2018. Horizontal and vertical distribution of microplastics in Korean coastal waters. Environ. Sci. Technol. 52, 12188–12197. https://doi.org/10.1021/acs.est.8b04032.

Vianello, A., Boldrin, A., Guerriero, P., Moschino, V., Rella, R., Sturaro, A., Da Ros, L., 2013. Microplastic particles in sediments of Lagoon of Venice, Italy: first observations on occurrence, spatial patterns and identification. Estuar. Coast Shelf Sci. 130, 54–61. https://doi.org/10.1016/j.ecss.2013.03.022.

von Moos, N., Burkhardt-Holm, P., Kohler, A., 2012. Uptake and effects of microplastics on cells and tissue of the blue mussel Mytilus edulis L. After an experimental exposure. Environ. Sci. Technol. 46, 11327–11335. https://doi.org/10.1021/es302332w.

Wagner, S., Huffer, T., Klockner, P., Wehrhahn, M., Hofmann, T., Reemtsma, T., 2018. Tire wear particles in the aquatic environment - a review on generation, analysis, occurrence, fate and effects. Water Res. 139, 83–100. https://doi.org/10.1016/j.watres.2018.03.051.

Wang, W.F., Ndungu, A.W., Li, Z., Wang, J., 2017. Microplastics pollution in inland freshwaters of China: a case study in urban surface waters of Wuhan, China. Sci. Total Environ. 575, 1369–1374. https://doi.org/10.1016/j.scitotenv.2016.09.213.

Wicke, D., Cochrane, T.A., O'Sullivan, A., 2012. Build-up dynamics of heavy metals deposited on impermeable urban surfaces. J. Environ. Manag. 113, 347–354. https://doi.org/10.1016/j.jenvman.2012.09.005.

Wright, S.L., Thompson, R.C., Galloway, T.S., 2013. The physical impacts of microplastics on marine organisms: a review. Environ. Pollut. 178, 483–492. https://doi.org/10.1016/j.envpol.2013.02.031.

Yonkos, L.T., Friedel, E.A., Perez-Reyes, A.C., Ghosal, S., Arthur, C.D., 2014. Microplastics in four Estuarine rivers in the chesapeake bay, USA. Environ. Sci. Technol. 48, 14195–14202. https://doi.org/10.1021/es5036317.

Zhang, K., Xiong, X., Hu, H.J., Wu, C.X., Bi, Y.H., Wu, Y.H., Zhou, B.S., Lam, P.K.S., Liu, J.T., 2017. Occurrence and characteristics of microplastic pollution in Xiangxi bay of three Gorges reservoir, China. Environ. Sci. Technol. 51, 3794–3801. https://doi.org/10.1021/acs.est.7600369.

Ziccardi, L.M., Edgington, A., Hentz, K., Kulacki, K.J., Driscoll, S.K., 2016. Microplastics as vectors for bioaccumulation of hydrophobic organic chemicals in the marine environment: a state-of-the-science review. Environ. Toxicol. Chem. 35, 1667–1676. https://doi.org/10.1002/etc.3461.