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A Study on the Distribution of Microplastics in the South Coast of Korea and Gwangyang Bay

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Abstract: Microplastic distribution surveys centered on Korea's Gwangyang Bay and southern coastal waters. Gwangyang Bay seawater averaged 3.17 ± 1.23 particles/L, and sediments averaged 462.4 ± 143.9 particles/kg. The southern coastal seawater averaged 0.10 ± 0.09 particles/L, and the sediments averaged 50.6 ± 29.7 particles/kg. Microplastics flowing from land, through physical modeling of ocean currents in Gwangyang Bay and southern coastal waters, pass through the Yeosu Strait and flow into the southern coastal waters. At the same time, it is judged that the southern coastal waters showed somewhat lower abundance than the Gwangyang Bay waters because they move toward the Korean Strait due to the Jeju warm current water and Tsushima current water, strongly generated in summer. In addition, the seawater microplastic abundance showed a higher abundance than that on the site adjacent to the land in the southern coastal waters, which is the study area. On the other hand, the results for sediment microplastic abundance were opposite to the surface seawater microplastic results. Therefore, it is judged that entering one source of pollution does not affect the distribution of microplastics in Gwangyang Bay and southern coastal waters, but rather this occurs in different forms.

Keywords: microplastics; Korean Southwest Sea; seawater; sediment; microplastics distribution

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1. Introduction

Global plastic production is 360 million tons [1,2], and global waste products reached 90 million tons, from 60 million tons in 2015. It is reported that this is expected to increase from 155 million to 260 million tons by 2060 [2,3]. It is known that about 8 million tons of plastic used in this way flows into the ocean annually due to waste or dumping [4]. Finally, the plastic introduced into the ocean is fine-grained in the form of microplastics (0.2 to 5 mm in size, [5]), mainly through photochemical processes [6]. Microplastics that exist in this way are called 'secondary microplastics', and plastics that are intentionally manufactured in small sizes (0.2–5 mm) can be classified as primary microplastics [7–10]. Microplastics introduced into the ocean are not well distinguished in size from phytoplankton and organic detritus, and some are used as the basis for the attachment of algae [11], then misfed to zooplankton and fry [12]. The microplastics that are fed are not digested [13]. They may remain in the body and affect the physiological action of zooplankton, affecting their survival [14].

In addition, microplastics may be bioaccumulated in upper predators, such as humans, fish, and mammals, affecting human health via the food chain of the marine ecosystem [15]. Although less than 130 µm of ingested microplastics are mainly excreted, some can bind to intestinal cells and cause local immune responses [16]. In addition, there is the possibility that harmful additives, heavy metals, and adsorbed hydrophobic chemicals contained in microplastics are eluted from the body [16]. Microplastics inhaled into respiratory organs may be removed via the mucous membrane clearance mechanism of the lungs. However, very adverse effects are expected in the event of long-term exposure in individuals with poor health [17]. In addition, microscopic nanoparticles (less than 20 nm in size) have been reported to affect the fertility, metabolism, and mortality of organisms such as animal plankton, bivalves, shrimp, oysters, copepods, and lugworms [18].

Therefore, indirect studies are being conducted to determine whether microplastics will eventually enter the ocean and ultimately affect humans. In many countries, studies on the characteristics of microplastic distribution on the coasts of each country's territorial waters are being conducted to find systematically effective ways to deal with microplastic pollution [19–24]. For this purpose, studies were conducted on microplastic distribution characteristics, focusing on Korea's bays and coasts. In the results for microplastics among seawater studied in the previous study, Bay showed high results in Gwangyang Bay (0.02–5.0 mm: 2.362 particles/ L [21]/0.33–5.0 mm: 0.00165 particles/L [25]) and Yongil Bay (0.02–5.0 mm: 1.688 particles/L [21]/0.33–5.0 mm: 0.00454 particles/L [25]), areas with industrial facilities. Rural areas showed lower results than Gwangyang Bay and Yongil Bay, in the following order: Hampyeong Bay (0.02-5.0 mm: 1.548 particles/L [21]/0.33–5.0 mm: 0.00170 particles/L [25]) > Deukryang Bay (0.02–5.0 mm: 1.146 particles/L [21]/0.33–5.0 mm: 0.00112 particles/L [25]) > Cheonsu Bay (0.02–5.0 mm: 0.784 particles/L [21]/0.33–5.0 mm: 0.00279 particles/L [25]). The highest results were found in coastal areas, like the Incheon Coast (0.02–5.0mm: 4.064 particles/L [21]/0.33–5.0 mm: 0.00196 particles/L [25]). The results were in the following order: Busan Coast (0.02–5.0 mm: 2.362 particles/L [21]/0.33–5.0 mm: 0.00135 particles/L [25]) > Ulsan Coast (0.02–5.0 mm: 1.764 particles/L [21]/0.33–5.0 mm: 0.00473 particles/L [25]). In addition, the Yellow Sea/the Southern Sea (0.02–5.0 mm: 0.266 particles/L [26]), the East Sea (0.02–5.0 mm: 0.289 particles/L [26]), and the Southwest Sea (0.02–5.0 mm: 0.460 particles/L [27]) showed much lower results than the coastal results. Among the sediments, microplastic results showed an average of 211 particles/kg dw in seven regions on the West Coast in Korea. Since only one site was selected from each of the seven regions, evaluating the microplastic distribution was difficult.

As a result, many studies have been conducted on Korea's bays and coasts. Still, studies on the characteristics of microplastic distribution connecting the bays and coasts have yet to be conducted. Therefore, it is essential to study microplastic distribution characteristics that connect the bays and coasts adjacent to cities and industrial facilities in Korean waters. Gwangyang Bay has a national industrial complex with several plastic manufacturing factories. It is also divided into five regions (Yeoseo, Suncheon, Gwangyang in Jeollanam-do, and Hadong and Namhae in Gyeongsangnam-do), making it a densely populated area. Depending on the environmental characteristics around Gwangyang Bay, many microplastics are expected to flow into the southern coastal waters of Korea according to the ocean current after entering Gwangyang Bay. Therefore, surface seawater and surface sediments are collected from the southern coast connected to Gwangyang Bay and Gwangyang Bay and analyzed using FT–IR equipped with array detectors. The analysis results identify the microplastic distribution characteristics (abundance, polymer type, size, shape, etc.).

2. Materials and Methods

2.1. Sampling Method

On 6 September 2021, the Gwangyang Bay area collected samples from five vertices using a small vessel. In addition, the southern coastal waters (SS area) collected samples from 15 vertices from September 1st to 3rd, 2021, using the Chonnam National University training ship Sae Dong-baek (2996 tons) (Figure 1). Water temperatures and salinity were measured on site at 0.5 m below the surface at individual sampling points using CTD (SBE 19, Sea-bird Electronic, Bellevue, Washington, DC, USA). To minimize plastic contamination during sample collection, all samplers, such as buckets, sieves, and wires, were made of stainless steel, excluding plastic materials.

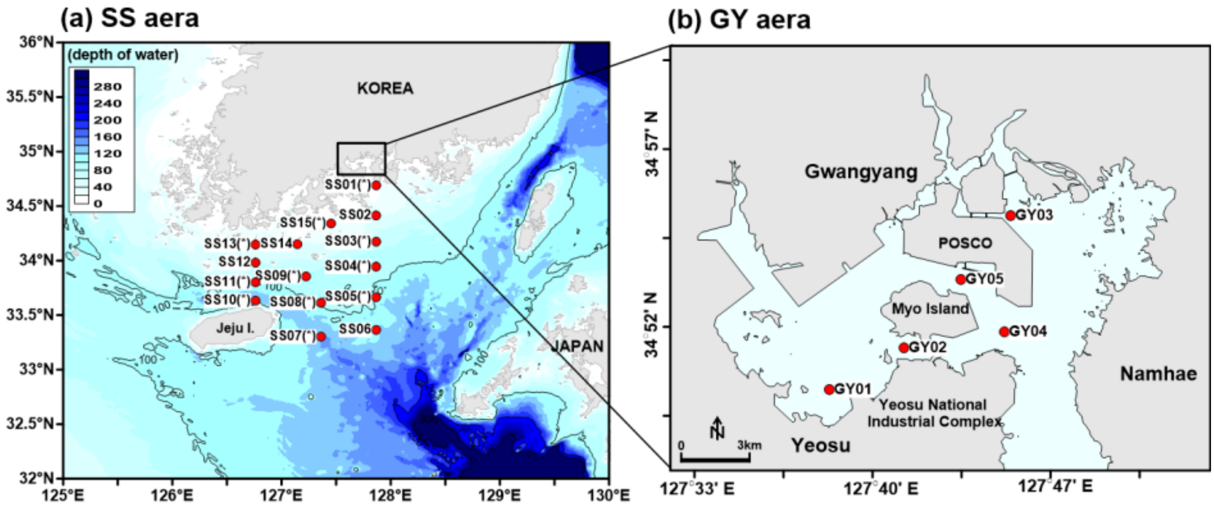

Figure 1. The study areas and sampling stations. (**a**): South Sea Coast of Korea, SS area-(*): sediment site; (**b**): Gwangyang Bay of Korea, GY area.

For sample collection, 100 L of 0–20 cm of surface seawater was collected using a stainless bucket with a depth of 20 cm. Seawater raised on board was immediately filtered using a 20 μm sieve made of stainless material. Filtered samples were transferred to the laboratory in 1 L brown hard glass bottles.

Sediments were collected using grabs. A stainless-steel spoon collected about 500 g of sediment with a 0–3 cm surface area. Then, it was transferred to the laboratory in a brown hard glass bottle. The surface seawater and surface sediment sample storage container was washed with ultra-pure distilled water filtered using a filter made of 20 μm stainless steel. The collected samples were stored at 4 °C until needed.

2.2. Microplastic Pre-Treatment Method

For surface seawater, the National Institute of Fisheries Science's *Investigation Guidelines for Qualitative. Quantification of Microplastics Remaining in Seawater and Fisheries Life* [28] was used. The National Institute of Fisheries Science's *Investigation Guidelines for Qualitative. Quantification of Microplastics Residual among Sediment Deposits* [29] was used for sediment. In addition, all reagents used were from residual pesticide analysis classes. All labeled glassware used in the analysis was washed with ultra-pure distilled water filtered using a filter made of 20 μm stainless steel.

2.2.1. Surface Seawater Microplastic Pretreatment Method

After sampling, only the filtered samples (20 μm–5 mm) were transferred into a 500 mL beaker using sieves with 20 μm and 5 mm meshes, and dried in a natural convection dryer (LDO-080N, Daihan Labtech Co., Namyangju, Korea) at 90 °C for 24 h. Next, 20 mL of 35% hydrogen peroxide (CAS No.: 7722-84-1, 35% hydrogen peroxide, JUNSEI Chemical Co., Ltd., Tokyo, Japan), 20 mL of an aqueous iron sulfate solution, and a suitable amount of sulfuric acid (CAS No.: 7664-93-9, sulfuric acid, JUNSEI Chemical Co., Ltd., Tokyo, Japan) were successively added to the beaker. The aqueous iron sulfate solution was prepared using 7.5 g of iron sulfate (CAS No.: 7720-78-7, iron sulfate, JUNSEI Chemical Co., Ltd., Tokyo, Japan) in 500 mL of distilled water. The beaker was covered using aluminum foil and left in a fume hood at room temperature for 5 min. The solution was stirred at 180 rpm at 75 °C (MSH-20D, Daihan Scientific Co., Ltd., Wonju, Korea) for 30 min. If organic matter remained after digestion, 20 mL of 35% hydrogen peroxide was poured into the beaker. This process was repeated until the organic matter was completely digested.

After digesting the organic matter, the sample was filtered through the 20 μm mesh sieve, and the beaker was rinsed with ultrapure water (up to 18.3 MΩ-cm, HIQ1, Human Science Co., Ltd. Hanam, Korea) to transfer the entire particles on the sieve. The whole of the particles on the sieve were transferred to a 250 mL density separation funnel using a 6.7 M NaI solution (CAS No.: 7681-82-5, NaI solution, JUNSEI Chemical Co., Ltd., Tokyo, Japan) with a density of 1.6 g/cm^3. After that, 100 mL of NaI was added to a funnel, which was covered with aluminum foil and left for 24 h. The deposited matter in the lower layer was discarded, and the supernatant was filtered on a metal filter (pore size, 20 µm; diameter, 24 mm) and dried in a desiccator for 24 h.

2.2.2. Sediment Microplastic Pretreatment Method

250 g of the sample was transferred to a 500 mL glass bottle, and 300 mL of NaI with a 1.6 g/cm^3 density was added, sealed well with a lid, and shaken hard for 1 min. After that, the dense particles can stand for about 30 min until they subside. Afterwards, it was transferred to a 1 L glass beaker, and all the particles attached to the upper wall of the bottle were transferred using a cleaning bottle containing NaI. The above process was repeated five times to separate the microplastics in the sediment as much as possible. The sample, subjected to the density separation process, was filtered using a 20 µm sieve, placed in a 500 mL beaker, and dried in a natural circulation dryer (LDO-080N, Daihan Labtech Co., Namyangju, Korea) at 90 °C for 24 h. Next, the procedure occurred as in Section 2.2.1 (Figure 2).

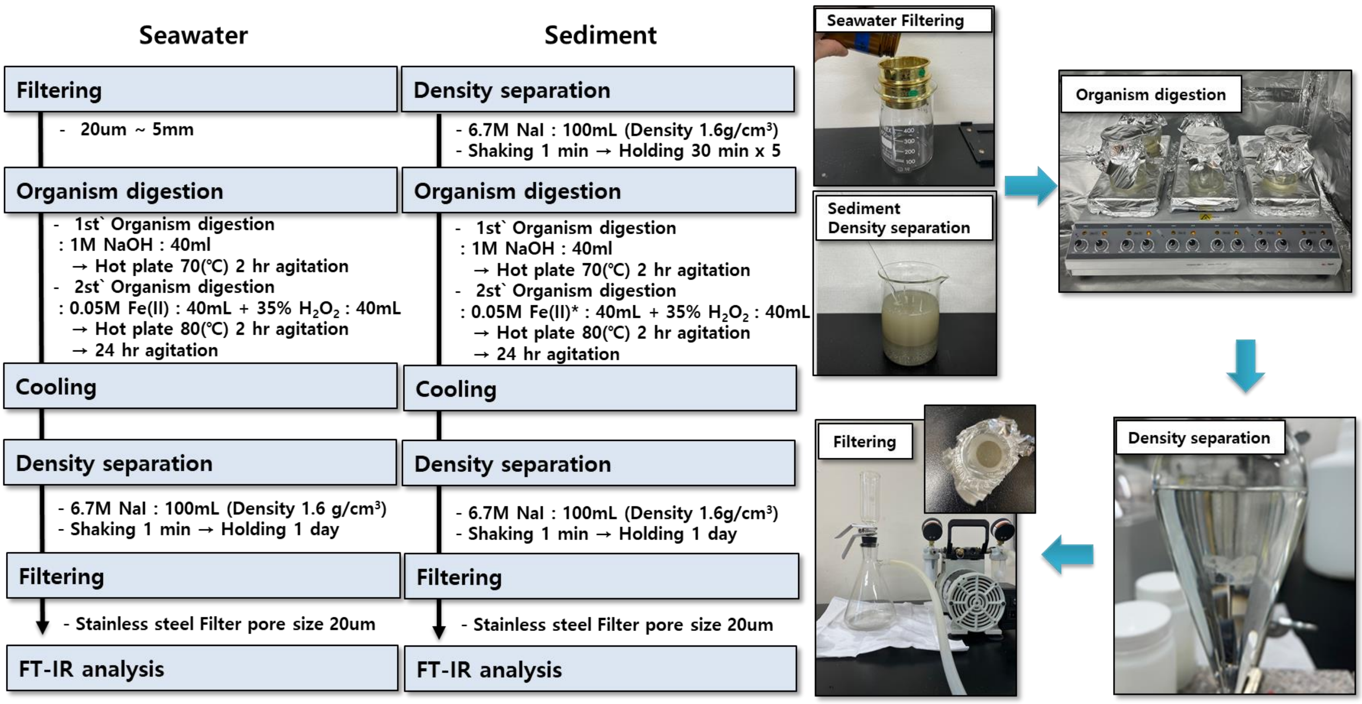

Figure 2. Flowchart of Microplastic Pretreatment Methods for Seawater and Sediments.

2.3. *Identifying the Polymer Types*

The device used in this study selected FT–IR (Nicolet iN10 MX, ThermoFisher, Waltham, MA, USA) equipped with array Detectors that can analyze up to 10 µm or more, extensively and in an automated manner that acquires data in a short time.

The FT–IR analysis condition was the imaging mode, and the detector type was set to imaging, using an array detector. In addition, the aperture, i.e., the IR interval, was set to 150 µm width 150 µm and height 150 µm, which are fixed values during the imaging mode. The background was analyzed by setting it as a collect background before the sample. The spectra with IR data were obtained with a measurement resolution of 4 cm-1 and in the wavelength range of 450–5000 cm^{-1}. The obtained spectra were compared to those of software databases (OMNIC, Thermo Fisher, Madison, WI, USA), and spectra with more than 80% similarity were considered plastic.

The target materials were selected as nine types in total: Polyethylene (PE), Polystyrene (PS), Polypropylene (PP), Polyester (PY), Acryl, Alkyd, Polyethylene terephthalate (PET), Polyvinyl chloride (PVC), and Nylon, as recommended by the National Institute of Fisheries Science's *Investigation Guidelines for Quantifying the Qualitative of Microplastics Residual* [28].

2.4. Microplastic Shape Classification and Size Measurement

The shapes of microplastics were classified using FT–IR microscopy, and the shapes were divided into five types: fragments, fibers, spheres, sheets, and pellets [28]. Feret's diameter was employed to measure the major axis of the particle using the ruler tool in OMNIC 9.13, the software of Nicolet iN10 MX FT–IR (Thermo Fisher, Madison, WI, USA) [28].

2.5. Quality Assurance/Quality Control (QA/QC)

One field blank per ten sampling points (i.e., a total of two field blanks) was collected to assess the contamination caused between collecting surface water in situ and the pretreatment process and FT–IR measurement [28]. Filtered distilled water (HPLC grade or higher-grade distilled water, Burdick and Jackson, Muskegon, MI, USA) was poured into a 1 L glass bottle from the sampling step in the ship. Notably, a new metal filter paper rinsed using distilled water was inspected to assess atmospheric contamination during FT–IR measurements before inspecting the samples [30]. As a result, only three microplastics were found in the two field blanks, and no microplastic was found in the metal filter blanks.

2.6. Statistical Analysis

The statistical analysis of detected microplastics was conducted using the SPSS (ver.25.0; SPSS Inc., Chicago, IL, USA) statistical program to verify the normal distribution of data.

2.7. Comparison between Other Literature and This Study

Previous studies on microplastic distribution primarily employed two methods depending on microplastic sample collection methods. The volume-reduced sampling method uses nets, such as the manta trawl net and bongo net [31–35]. Another method is the bulk water sampling method, in which a certain amount of seawater is collected using a bucket or an underwater pump [21,36]. The volume-reduced sampling method targets only those microplastics that are larger than the mesh size of the net (e.g., 300 µm mesh net, 0–1200 particles/m^3 [28,35]), while the bulk water sampling method uses relatively smaller mesh sizes (e.g., 20 µm mesh, 0–152,668 particles/m^3 [28,35]). For this reason, the two methods have a remarkable difference in measuring quantity [35]. Therefore, only the results of the bulk water sampling method, which is similar to the materials and methods of this study, were compared to distinguish and compare microplastic collection methods. In addition, for sediment microplastics, only the results when using grab samplers in areas other than beaches were compared.

3. Results

3.1. Spatial Distribution of Water Temperature and Salinity in the Surface Water

Table 1 and Figure 3 show the water temperature and salt concentration results in Gwangyang Bay and southern coastal waters.

Looking at the surface water temperature results in Gwangyang Bay, the water temperature range was 24.30–24.90 (average 24.60 ± 0.20) °C. In the case of horizontal distribution, the water temperature was somewhat low at the site of GY01, closest to Jungbang Stream, located in the Yeosu area, and the site of GY03, located in the Seomjin River basin. The range for salt was 26.00–28.94 (average 27.78 ± 1.02) psu, and in the case of horizontal distribution, meager salt results were found at the site of GY01, which showed low water temperature results. It is judged that the influence of river water flowing from land is significant.

Table 1. Results of seawater microplastic abundance, composition(polymer types), size, shape, water temperature and salinity in the South Sea Coast and Gwangyang Bay of Korea.

Site	MPs Abundance (Particles/L) [(a)]									MPs Size (%)				MPs Shape (%) [(b)]				Tem. (°C)	Sal. (psu)
	PP	PE	PY	PS	PET	PVC	Alkyd	Acryl	Total	0.02–0.3 mm	0.3–0.6 mm	0.6~1.0 mm	1.0–5.0 mm	Fragment	Fiber	Sphere	Sheet		
YS01	3.60	0.46	0.06	0.02	0.04	N.D.	0.16	0.02	4.36	92	5	1	2	87	11	2	N.D.	24.50	26.00
YS02	1.36	0.32	N.D.	N.D.	N.D.	N.D.	0.02	N.D.	1.70	96	4	N.D.	N.D.	89	6	2	2	24.90	27.36
YS03	2.06	0.46	0.10	0.02	0.04	0.02	0.08	N.D.	2.78	88	7	4	N.D.	78	17	3	3	24.30	28.24
YS04	2.94	1.44	0.16	N.D.	0.12	N.D.	0.18	0.02	4.86	97	2	1	N.D.	88	7	5	1	24.60	28.35
YS05	1.48	0.50	0.04	0.04	0.10	N.D.	N.D.	N.D.	2.16	84	10	4	2	81	2	17	N.D.	24.70	28.94
Min	1.36	0.32	0.04	0.02	0.04	0.02	0.02	0.02	1.7	-	-	-	-	-	-	-	-	24.30	26.00
Max	3.60	1.44	0.16	0.04	0.12	0.02	0.18	0.02	4.86	-	-	-	-	-	-	-	-	24.90	28.94
Mean	2.29	0.64	0.09	0.03	0.08	0.02	0.11	0.02	3.17	92	6	2	1	85	8	6	1	24.60	27.78
SD	0.86	0.41	0.05	0.01	0.04	-	0.06	0.00	1.23	-	-	-	-	-	-	-	-	0.20	1.02
SS01	0.05	N.D.	0.01	N.D.	N.D.	N.D.	N.D.	N.D.	0.06	83	N.D.	N.D.	17	83	17	N.D.	N.D.	26.67	30.36
SS02	0.03	N.D.	0.01	N.D.	N.D.	N.D.	N.D.	N.D.	0.04	50	25	25	N.D.	75	25	N.D.	N.D.	25.84	30.95
SS03	0.06	N.D.	0.01	N.D.	N.D.	N.D.	N.D.	N.D.	0.07	57	29	14	N.D.	76	14	N.D.	N.D.	25.71	31.18
SS04	0.01	0.01	N.D	N.D.	N.D.	N.D.	N.D.	N.D.	0.02	50	N.D.	50	N.D.	100	0	N.D.	N.D.	25.72	31.14
SS05	0.06	0.05	N.D	N.D.	N.D.	N.D.	N.D.	N.D.	0.07	100	N.D.	N.D.	N.D.	100	0	N.D.	N.D.	27.39	31.95
SS06	0.03	0.01	0.05	N.D.	N.D.	N.D.	N.D.	N.D.	0.13	23	23	8	46	8	62	N.D.	N.D.	27.88	33.12
SS07	0.04	N.D.	0.06	N.D.	N.D.	N.D.	N.D.	N.D.	0.11	9	45	27	18	45	55	N.D.	N.D.	27.62	32.33
SS08	0.08	N.D.	N.D	N.D.	N.D.	N.D.	N.D.	N.D.	0.08	75	25	N.D.	N.D.	100	0	N.D.	N.D.	25.26	31.15
SS09	0.08	0.02	0.01	N.D.	N.D.	N.D.	N.D.	N.D.	0.09	78	11	11	N.D.	89	11	N.D.	N.D.	25.58	31.30
SS10	0.35	N.D.	0.01	N.D.	N.D.	N.D.	N.D.	N.D.	0.38	84	11	3	3	98	3	N.D.	N.D.	26.87	30.97
SS11	0.01	0.03	N.D	N.D.	N.D.	N.D.	N.D.	N.D.	0.01	100	N.D.	N.D.	N.D.	100	0	N.D.	N.D.	26.55	31.47
SS12	0.10	0.03	0.02	N.D.	N.D.	N.D.	N.D.	N.D.	0.15	73	7	7	13	80	20	N.D.	N.D.	25.57	31.56
SS13	0.07	0.05	0.07	N.D.	N.D.	N.D.	N.D.	N.D.	0.19	63	21	5	11	63	37	N.D.	N.D.	25.53	31.60
SS14	0.02	N.D.	0.03	N.D.	N.D.	N.D.	N.D.	N.D.	0.05	80	N.D.	N.D.	20	60	40	N.D.	N.D.	25.08	31.50
SS15	0.02	0.03	0.04	N.D.	N.D.	N.D.	N.D.	N.D.	0.09	33	22	33	11	67	33	N.D.	N.D.	25.07	31.46
Min	0.01	-	-	-	-	-	-	-	0.01	-	-	-	-	-	-	-	-	24.67	30.36
Max	0.35	0.05	0.05	-	-	-	-	-	0.38	-	-	-	-	-	-	-	-	27.88	33.12
Mean	0.07	0.01	0.01	-	-	-	-	-	0.10	64	15	12	9	70	21	-	-	26.02	31.47
SD	0.08	0.02	0.02	-	-	-	-	-	0.09	-	-	-	-	-	-	-	-	0.97	0.62

[(a)] Only the detected polymer types (PE, PS, PP, PY, Acryl, Alkyd, PET, PVC, and nylon) are shown. [(b)] Only the observed shapes (fragment, fiber, sphere, sheet, and pellet) are shown. N.D.: not detected.

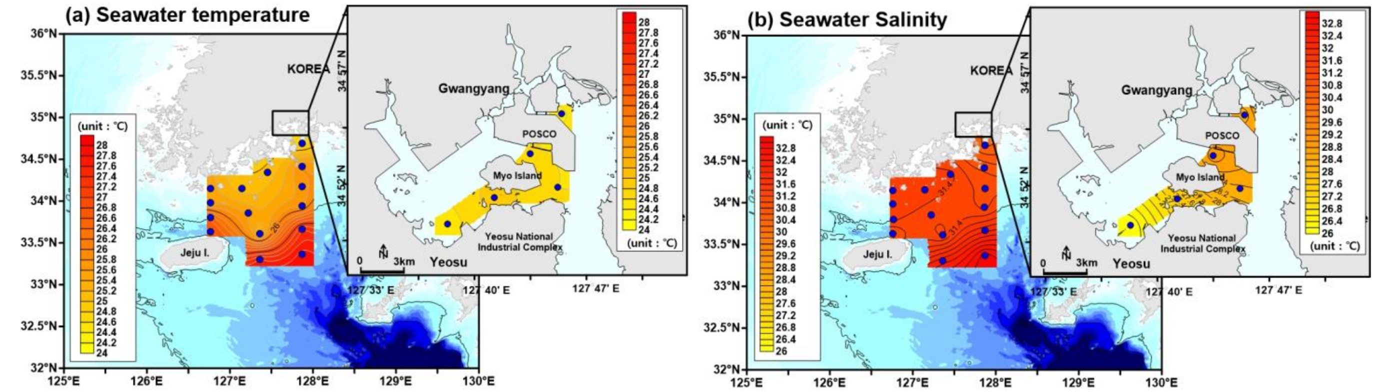

Figure 3. Seawater temperature and Salinity Distribution Chart for the South Sea Coast and Gwangyang Bay of Korea (**a**): Seawater temperature, (**b**): Sea-water Salinity. (Blue circle: sampling sites in the study area).

The results of the surface water temperature on the southern coast showed that the water temperature range was 24.67–27.88 (average 26.02 ± 0.97) °C. In the case of horizontal distribution, the water temperature tended to increase as it headed toward the SS07 site and SS06 site, located in the outer sea, centering on the study area. The range of salt was 30.36–33.12 (average 31.47 ± 0.62) psu. In the case of horizontal distribution, similar to the water temperature results, the salt tended to increase as it headed outward around the study area.

3.2. The Distribution on Numerical Microplastics Abundance

Tables 1 and 2 and Figure 4 show the results of microplastics abundance in surface seawater and surface sediments at the site of Gwangyang Bay and southern coastal waters.

Looking at the results of surface seawater in Gwangyang Bay, all microplastics were detected in all site surface seawater samples. The abundance range was investigated as 1.70–4.86 (average 3.17 ± 1.23) particles/L. In the case of horizontal distribution, the water temperature and salt results were somewhat low, and the sites of GY01 and GY04, located in the sea where ship inflows were most frequent, showed very high abundance. Looking at the results of surface sediments, the abundance range was investigated as 240.0–656.0 (average 462.4 ± 143.9) particles/kg, and in the case of horizontal distribution, contrary to the results for surface seawater, the abundance was somewhat higher at the sites not adjacent to the river.

The results for surface seawater on the southern coast showed that all microplastics were detected in all site surface seawater samples. The abundance range was considered as 0.01–0.38 (average 0.10 ± 0.09) particles/L. In the case of horizontal distribution, the abundance was somewhat high at the sites of SS10 close to the coast of Jeju Island and the sites of SS06 and SS07 located in the outer sea, and somewhat low at the sites located in the eastern sea centered on the study area. The results of surface sediments showed that the abundance range was 28.0–136.0 (average 50.6 ± 29.7) particles/kg. In the case of horizontal distribution, contrary to the results for surface seawater, the site located in the southern coastal sea showed a somewhat higher abundance.

Table 2. Results of surface sediment microplastic abundance, composition (polymer types), size, and shape in the South Sea Coast and Gwangyang Bay of Korea.

Site	MPs Abundance (Particles/kg) [(a)]								MPs Size (%)				MPs Shape (%) [(b)]			
	PP	**PE**	**PY**	**PS**	**PET**	**PVC**	**Alkyd**	**Total**	**0.02–0.3 mm**	**0.3–0.6 mm**	**0.6~1.0 mm**	**1.0–5.0 mm**	**Fragment**	**Fiber**	**Sphere**	**Sheet**
YS01	16.0	200.0	12.0	N.D.	4.0	N.D.	8.0	240.0	97	3	N.D.	N.D.	98	2	N.D.	N.D.
YS02	84.0	244.0	8.0	8.0	8.0	4.0	12.0	368.0	96	1	2	1	100	N.D.	N.D.	N.D.
YS03	124.0	336.0	20.0	16.0	16.0	N.D.	8.0	520.0	95	4	1	N.D.	95	2	3	N.D.
YS04	284.0	356.0	8.0	N.D.	8.0	N.D.	N.D.	656.0	100	N.D.	N.D.	N.D.	100	N.D.	N.D.	N.D.
YS05	84.0	388.0	20.0	N.D.	12.0	4.0	20.0	528.0	96	2	2	N.D.	96	4	N.D.	N.D.
Min	16.0	200.0	8.0	8.0	4.0	4.0	8.0	240.0	-	-	-	-	-	-	-	-
Max	284.0	388.0	20.0	16.0	16.0	4.0	20.0	656.0	-	-	-	-	-	-	-	-
Mean	118.4	304.8	13.6	12.0	9.6	4.0	12.0	462.4	97	2	1	0	98	2	1	-
SD	89.8	71.0	5.4	4.0	4.1	-	4.9	143.9	-	-	-	-	-	-	-	-
SS01	24.0	8.0	N.D	N.D.	N.D.	N.D.	N.D.	32.0	88	13	N.D.	N.D.	100	N.D.	N.D.	N.D.
SS03	32.0	N.D	N.D	N.D.	N.D.	N.D.	N.D.	32.0	88	13	N.D.	N.D.	100	N.D.	N.D.	N.D.
SS04	32.0	16.0	8.0	N.D.	N.D.	N.D.	N.D.	56.0	79	21	N.D.	N.D.	100	N.D.	N.D.	N.D.
SS05	16.0	8.0	12.0	N.D.	N.D.	N.D.	N.D.	36.0	67	11	N.D.	22	67	33	N.D.	N.D.
SS07	20.0	12.0	4.0	N.D.	N.D.	N.D.	N.D.	36.0	78	N.D.	11	11	78	22	N.D.	N.D.
SS08	28.0	N.D	N.D	N.D.	N.D.	N.D.	N.D.	28.0	100	N.D.	N.D.	N.D.	100	0	N.D.	N.D.
SS09	16.0	24.0	8.0	N.D.	N.D.	N.D.	N.D.	48.0	83	8	8	N.D.	83	17	N.D.	N.D.
SS10	36.0	N.D	N.D	N.D.	N.D.	N.D.	N.D.	36.0	89	11	N.D.	N.D.	100	N.D.	N.D.	N.D.
SS11	56.0	12.0	4.0	N.D.	N.D.	N.D.	N.D.	72.0	78	11	6	6	94	6	N.D.	N.D.
SS13	36.0	8.0	N.D	N.D.	N.D.	N.D.	N.D.	44.0	91	9	N.D.	N.D.	100	N.D.	N.D.	N.D.
SS15	84.0	40.0	12.0	N.D.	N.D.	N.D.	N.D.	136.0	94	3	N.D.	3	91	9	N.D.	N.D.
Min	16.0	8.0	4.0	-	-	-	-	28.0	-	-	-	-	-	-	-	-
Max	84.0	40.0	12.0	-	-	-	-	136.0	-	-	-	-	-	-	-	-
Mean	34.5	16.0	8.0	-	-	-	-	50.5	85	9	2	4	92	8	-	-
SD	6.4	3.32	3.27	-	-	-	-	29.7	-	-	-	-	-	-	-	-

[(a)] Only the detected polymer types (PE, PS, PP, PY, Acryl, Alkyd, PET, PVC, and nylon) are shown. [(b)] Only the observed shapes (fragment, fiber, sphere, sheet, and pellet) are shown. N.D.: not detected.

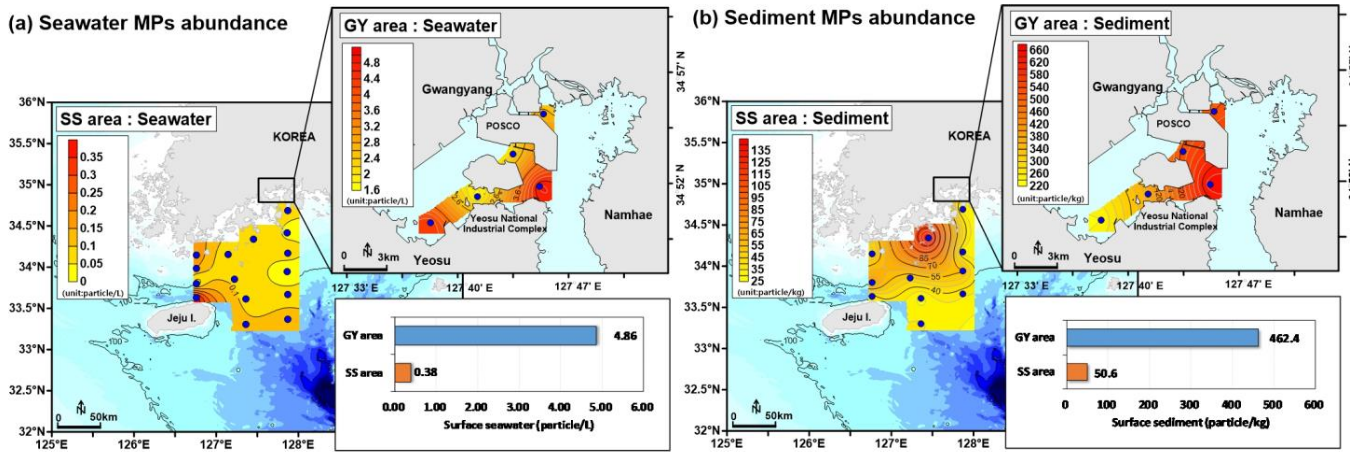

Figure 4. Horizontal distribution chart of the abundance of seawater and sediment microplastics in the South Sea Coast and Gwangyang Bay of Korea. (**a**): seawater MPs abundance; (**b**): sediment MPs abundance. (Blue circle: sampling sites in the study area).

3.3. The Distribution of Microplastic Polymer Types

Tables 1 and 2 and Figure 5 show the results of microplastic materials on surface seawater and surface sediments in Gwangyang Bay and southern coastal waters.

The seawater results for Gwangyang Bay's surface showed PE, PS, PP, PY, PET, and PVC, excluding Alkyd and nylon materials, among nine microplastics. Looking at the seven types of plastic compositions detected, PP materials averaged 73%, PE materials 20%, PY materials 2%, PS materials 1%, PET materials 2%, PVC materials 0.1%, and Acryl materials 2%. It was shown that PP materials were the most distributed in surface seawater. In the case of the most commonly detected PP material, the percentage was very high at 83% at the site of GY01 adjacent to Jungheung Stream in Yeosu. In addition, the GY02 site, which is close to this site, decreased somewhat as it moved away from the land. The surface sediment results showed that PE, PS, PP, PY, PET, and PVC were detected, excluding Alkyd and nylon materials, among the nine types of microplastics. Looking at the seven types of plastic compositions detected, i.e., PP material 23%, PE material 68%, PY material 3%, PS material 1%, PET material 2%, PVC material 0.4%, and Acryl material 2%, showed that PE material was most commonly distributed in surface sediment. The most commonly detected PE material showed a very high percentage of 83% at the site of GY01 adjacent to Jungheung Stream in Yeosu.

Looking at the results of surface seawater on the southern coast, only three of the nine types of microplastics were detected: PE, PP, and PY. Looking at the three types of plastic compositions detected, the average PP material was 66%, the PE material was 13%, and the PY material was 21%, indicating that PP material was most commonly distributed in surface seawater. The most widely detected PP material showed a very high percentage in the coastal and outer waters centered on the study area. Looking at the results of the surface sediments, only three, PE, PP, and PY, were detected out of the nine types of microplastics. Looking at the three types of plastic composition detected, the average PP material was 71.5%, the PE material was 20.3%, and the PY material was 7.9%, indicating that PP material was most commonly distributed in surface sediment. The most widely detected PP material showed a very high percentage at the site in the sea near Jeju Island.

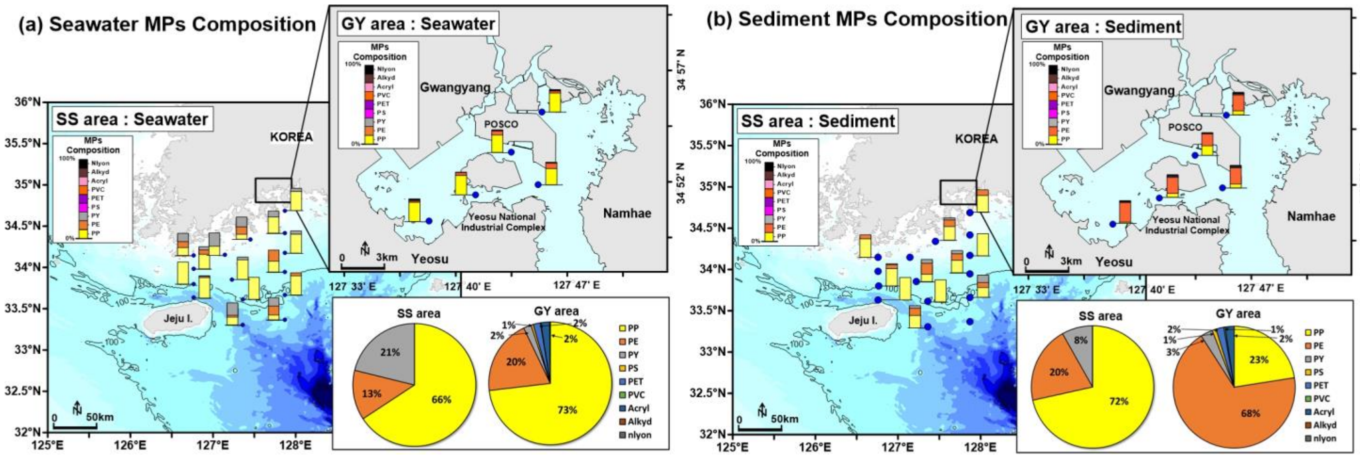

Figure 5. Horizontal distribution chart of the composition of seawater and sediment microplastics in the South Sea Coast and Gwangyang Bay of Korea. (**a**): seawater MPs' composition; (**b**): sediment MPs' composition. (Blue circle: sampling sites in the study area).

3.4. The Distribution on Microplastics' Shape

Tables 1 and 2 and Figure 6 show the results for microplastics for surface seawater and surface sediments in Gwangyang Bay and southern coastal waters.

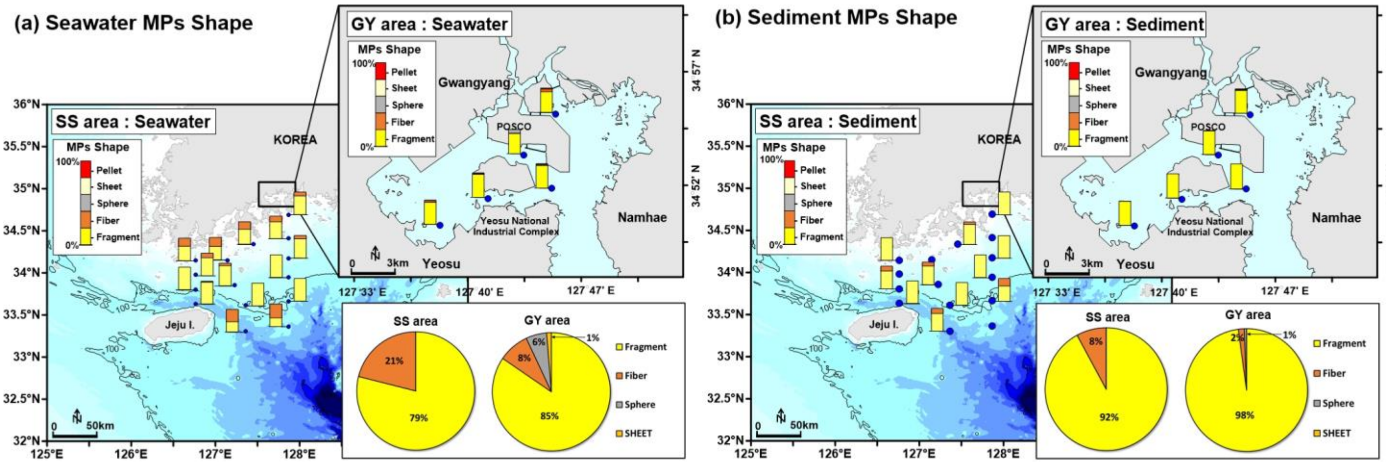

Figure 6. Horizontal distribution chart of the shape of seawater and sediment microplastics in the South Sea Coast and Gwangyang Bay of Korea. (**a**): seawater MPs shape; (**b**): sediment MPs shape. (Blue circle: sampling sites in the study area).

The results for surface seawater in Gwangyang Bay were classified into fragment shape, fiber shape, sphere shape, sheet shape, and pellet shape through the FT–IR microscope and were observed in four of these forms, except for pellet shape. Among the observed microplastic forms, the fragment shape showed a very high percentage of 85% on average. The percentages were shown in the order of fiber shape (average 8%), sphere shape (average 6%), and sheet shape (average 1%). The surface sediment results were observed in three forms, except for the sheet and pellet shapes. Among the observed microplastic forms, the fragment shape showed a high percentage of 98%. The percentages were shown in the order of fiber shape (average 2%) and sheet shape (average 1%).

The results for surface seawater on the southern coast showed that only the fragment and fiber shapes were observed when classifying them into fragment, fiber, sphere, sheet, and pellet shapes through the FT–IR microscope. Among the observed microplastic forms, the fragment showed a very high percentage, averaging 79%. The average fiber shape was 21%, slightly higher than the results of surface seawater in Gwangyang Bay. Looking at the results of the surface sediments, only the fragment and fiber shapes were observed, in a similar trend to that of the results for surface seawater. The fragment shape showed a very high percentage, averaging 92%. The fragment shape was shown to be 8% on average, showing a similar trend to the results for surface sediment in Gwangyang Bay.

3.5. The Distribution of Microplastics' Sizes

Tables 1 and 2 and Figure 7 show the results for microplastic size for surface seawater and surface sediments in Gwangyang Bay and southern coastal waters. Microplastics were categorized into sizes 0.02 to 0.30 mm, 0.30 to 0.60 mm, 0.60 to 1.00 mm, and 1.00 to 5.0 mm.

The results for seawater on the surface of Gwangyang Bay found all four size classifications. The size of the four microplastics was an average of 92% (average 2.86 paricels/L) at 0.02 to 0.30 mm, an average of 6% (average of 0.16 particles/L) at 0.30 to 0.60 mm, an average of 2% (average of 0.06 particles/L) at 0.60 to 1.0 mm, and an average of 1% (average of 0.02 particles/L) at 1.0 to 5.0 mm. 0.02 to 0.30 mm, which corresponds to the small size among microplastics most often observed in the size classification group, which is likely to be bio-concentrated, as most of the microplastics detected in shellfish were reported to be less than 0.2 mm [36]. The surface sediment results show that all four sizes were found. Looking at the four sizes of microplastics, an average of 97% (average 447.0 particles/kg) was found at 0.02 to 0.30 mm, an average of 2% (average 10.0 particles/kg) at 0.30 to 0.60 mm, an average of 1% (average 8.0 particles/kg) at 0.60 to 1.0 mm, and an average of 0.2% (average 4 particles/kg) at 1.0 to 5.0 mm, as shown in the results for surface seawater.

Looking at the results for seawater on the surface of the southern coast, all four size classifications were found. Looking at the size of the four microplastics, it was shown that they were high in the 0.02 to 0.30 mm size range with an average 64% (average 0.07 particles/L), in 0.030 to 0.60 mm with an average 15% (average 0.03 particles/L), in 0.60 to 1.0 mm with an average 12% (average 0.01 particles/L), and in 1.0 to 5.0 mm with an average 9% (average 0.02 particles/L). As described above, it was observed to be high in the 0.02 to 0.30 mm classification group, which is likely to be bio-concentrated but at a lower level than the Gwangyang Bay coast. Looking at the surface sediment results, all four size classifications were found. Looking at the four size classifications for microplastics, it showed an average of 85% (average of 43.27 particles/kg) at 0.02 to 0.30 mm, an average of 9% (average of 5.33 particles/kg) at 0.30 to 0.60 mm, an average of 2% (average of 4.0 particles/kg) at 0.60 to 1.0 mm, and an average of 4% (average of 5 particles/kg) at 1.0 to 5.0 mm. As with the results for surface seawater, it was very high at 0.02 to 0.30 mm, which is a small classification.

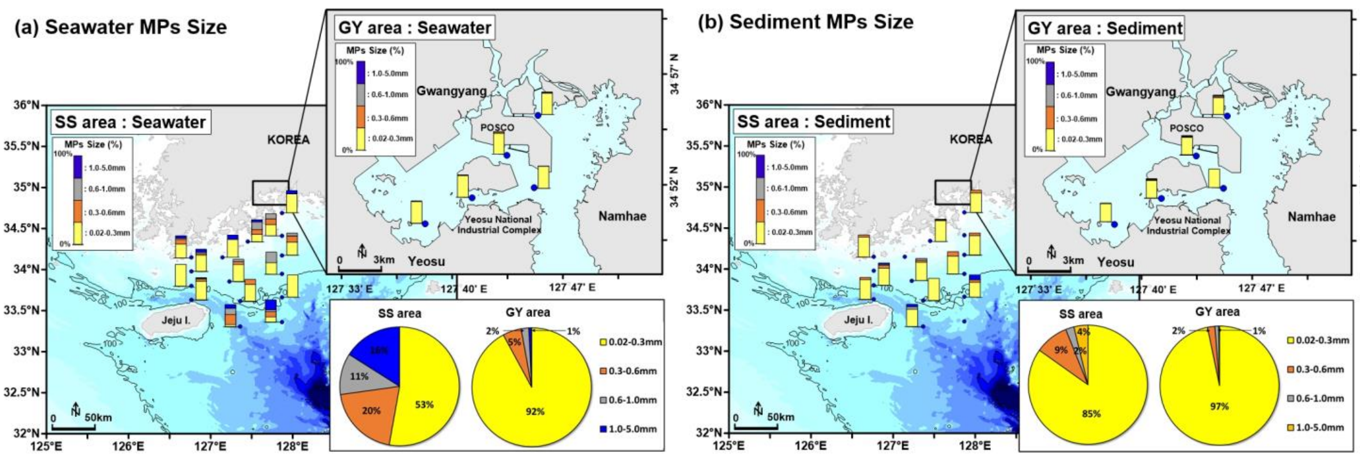

Figure 7. Horizontal distribution chart of the size of seawater and sediment microplastics in the South Sea Coast and Gwangyang Bay of Korea. (**a**): seawater MPs size; (**b**): sediment MPs size. (Blue circle: sampling sites in the study area).

4. Discussion

In Gwangyang Bay, there is a national industrial complex, where several plastic manufacturing factories are located. Additionally, it is adjacent to five regions (Yeoseo, Suncheon, Gwangyang in Jeollanam-do, and Hadong and Namhae in Gyeongsangnam-do), making it a densely populated area. Therefore, due to the environmental characteristics surrounding Gwangyang Bay, a significant number of microplastics are expected to enter Gwangyang Bay and then flow into the southern coast of Korea following ocean currents. Consequently, a study was conducted on microplastics distribution patterns and characteristics in Gwangyang Bay and the southern coast of Korea.

Gwangyang Bay is more than just a curve of the coast, and the sea moves toward land. The southern coastal waters have a topography at the border between land and outer seas. Therefore, the results of other literature studied in waters with this topography were compared with the results of this study (Tables 3 and 4). Seawater results were examined. In the Bay type case, this study showed a higher level than the results conducted on Cheonsu Bay, Hampyeong Bay, Deukryang Bay, Gwangyang Bay, and Yeongil Bay in Korea [21]. However, in the case of Gwangyang Bay, the results of this study were 2.362 ± 1.022 particles/L, which were higher than those of other literature. However, there was no significant difference. In addition, the results of this study were higher than those of the results surveyed in Jiaozhou Bay, China [37]. We looked at the survey results on the coast, similar to the southern coastal waters. This showed a much lower level than the results for Korea's Ulsan, Incheon, and Busan coasts [21]. However, the results of the Ulsan and Busan coasts, excluding the Incheon coast, showed a lower level than the results of Gwangyang Bay in this study. The coast of Incheon is the downstream point of the Han River, where Seoul (population: 9,86,034; [38]) and Gyeonggi-do (population: 13,638,821; [38]), the capital of Korea, are adjacent. Therefore, the result is thought to have been very high on the coast of Incheon in relation to the population in this area. In addition, the results conducted on the coast of China [39] were higher than those of Gwangyang Bay and the southern coastal waters of this study. In addition, the results in the Yellow Sea, South Sea, and East Sea in Korea [26,27], the North Yellow Sea in China [40], and Malaysian beaches [41] showed higher levels than in the southern coastal waters. However, these were shown to be much lower than the results of Gwangyang Bay. The offshore results of this study were higher than those conducted in the central basin of the Arctic [42], and the Atlantic Ocean [43]. In addition, the results conducted in China's South China Sea [44] and Greenland Sea [39] showed the same level between the southern coastal waters and the Gwangyang Bay as in this study.

The results for surface sediments were examined. In the form of a bay with a shape similar to that of Gwangyang Bay, the results of this study were much lower than those conducted in China's Qinzhou Bay (outside the Mangrove) [45] and Sanggou [46]. Results conducted in Laizhou Bay [47] and Belgian Bay [48] in China were higher than those of this study. However, only Gwangyang was shown to be at a lower level than these results. In addition, this study's results were lower in China's Qinzhou Bay (in Mangrove) [45] and Singapore's Lim Chu Kang [49]. Looking at the survey results on the coast similar to the southern coastal waters, the results conducted in Tidal on the west coast of Korea [50], China's North Yellow Sea [51] and Maowei Sea [52] seemed to be higher than the results of this study. The results conducted in Venice [53] in Italy and North African Coasts [54] in the Mediterranean Sea were similar to the results of Gwangyang Bay in this study. In addition, the results for the Belgian Coast [48], the Atlantic Coasts of France [55], and the Spanish coast of the Mediterranean Sea [56] appeared to be similar to those of the southern coastal waters of this study.

Table 3. Comparison of results of other studies on microplastic abundance in surface seawater in the South Sea Coast and Gwangyang Bay of Korea.

Nation	Area		Investigation	n	Collected Vol. (Mesh Size)	Abundance (Particles/L)	References
China	Jiaozhou	Bay	2017	14	50 L (20 μm)	mean 1.602 ± 1.274	[37]
Korea	Cheonsu	Bay	2016/2017	5	100 L (20 μm)	mean 0.784 ± 0.272	[21]
Korea	Hampyeong	Bay	2016/2017	5	100 L (20 μm)	mean 1.548 ± 0.211	[21]
Korea	Deukryang	Bay	2016/2017	5	100 L (20 μm)	mean 1.146 ± 0.423	[21]
Korea	Youngil	Bay	2016/2017	5	100 L (20 μm)	mean 1.688 ± 0.496	[21]
Korea	Gwangyang	Bay	2016/2017	5	100 L (20 μm)	mean 2.362 ± 1.022	[21]
Korea	Gwangyang	Bay	2020	5	100 L (20 μm)	mean 3.17 ± 1.1.23	This study
Korea	Ulsan	Coastal	2016/2017	5	100 L (20 μm)	mean 1.764 ± 1.006	[21]
Korea	Incheon	Coastal	2016/2017	5	100 L (20 μm)	mean 4.064 ± 1.075	[21]
Korea	Busan	Coastal	2016/2017	6	100 L (20 μm)	mean 1.020 ± 0.279	[21]
Korea	Yellow Sea	Coastal	2018	9	200 L (20 μm)	mean 0.266 ± 0.459	[26]
Korea	South Sea	Coastal	2018	5			
Korea	East Sea	Coastal	2018	8	200 L (20 μm)	mean 0.289 ± 0.280	[26]
Korea	Southwest Sea	Coastal	2020	23	30 L (20 μm)	mean 0.46 ± 0.27	[27]
China	China	Coastal	-	16	100 L (50 μm)	mean 4.5	[39]
China	north Yellow Sea	Coastal	2016	50	25 L (30 μm)	mean 0.545 ± 0.282	[40]
Malaysia	Malaysia	Coastal	2018	-	2.041 L/s (20 μm)	mean 0.211 ± 0.104	[41]
the Ross Sea of the Arctic		Coastal	2010	15	600–2000 m^3	mean 0.002 ± 0.0003	[42]
Korea	South Sea	Coastal	2020	15	100 L (20 μm)	mean 0.10 ± 0.09	This Study

Table 4. Comparison of results of other studies on microplastic abundance in sediment in the South Sea Coast and Gwangyang Bay of Korea.

Nation	Area		Investigation	Sample Collection	Abundance (Particles/kg)	References
China	Qinzhou (mangrove Side)	Bay	spoon	7	mean 1298 ± 2207	[45]
China	Qinzhou (mangrove in)	Bay	spoon	7	mean 42.9 ± 26.8	[45]
China	Sanggou	Bay	Van Veen grab	8	mean 1674 ± 526	[46]
China	Laizhou	Bay	Van Veen grab	58	mean 461.6 ± 167	[47]
China	Lim Chu Kang	Bay	-	7	mean 36.8 ± 23.6	[49]
China	Belgian	Bay	Van Veen grab	11	mean 167	[48]
Korea	Gwangyang	Bay	Van Veen grab	5	mean 462.4 ± 143.9	This study
Korea	West coast tidal	Coastal	spoon	7	mean 2191	[50]
China	North Yellow Sea	Coastal	Box sampler	28	mean 499.76 ± 370.07	[51]
China	Maowei Sea,	Coastal	-	10	520 ± 8–2310 ± 29	[52]
Belgian	Belgian coast	Coastal	-	6	mean 97.2	[48]
French	Atlantic coastal	Coastal	box-core	3	mean 67 ± 76	[55]
The Mediterranean	North African coasts	Coastal	Corer/Visual	4	182.66–649.33	[54]
	The Spanish Mediterranean	Coastal	Box-corer	10	mean 113.2 ± 88.9	[56]
Korea	South coast	Coastal	grab	11	mean 50.6 ± 29.7	This study

As such, Gwangyang Bay showed similar or higher levels than the domestic and foreign microplastic abundance results. On the contrary, the southern coastal waters showed much lower levels than other coastal results, as did the topography in the form of a bay. Therefore, an independent sample *T*-test was conducted to confirm these two regions' differences [57]. However, the independent sample *T*-test should show normality with more than 30 samples, but the number of samples in this study was less than 30 (5 in Gwangyang Bay and 15 in the southern coastal waters), so it showed no normality. Therefore, the Mann–Whitney test, a nonparametric verification method, was conducted. In the results conducted, both microplastic seawater abundance and surface sediment abundance showed significant results ($p < 0.01$) in Gwangyang Bay and southern coastal waters (Figure 8). As a result, it is judged that the abundance of microplastics is affected by different factors in the Gwangyang Bay and southern coastal waters.

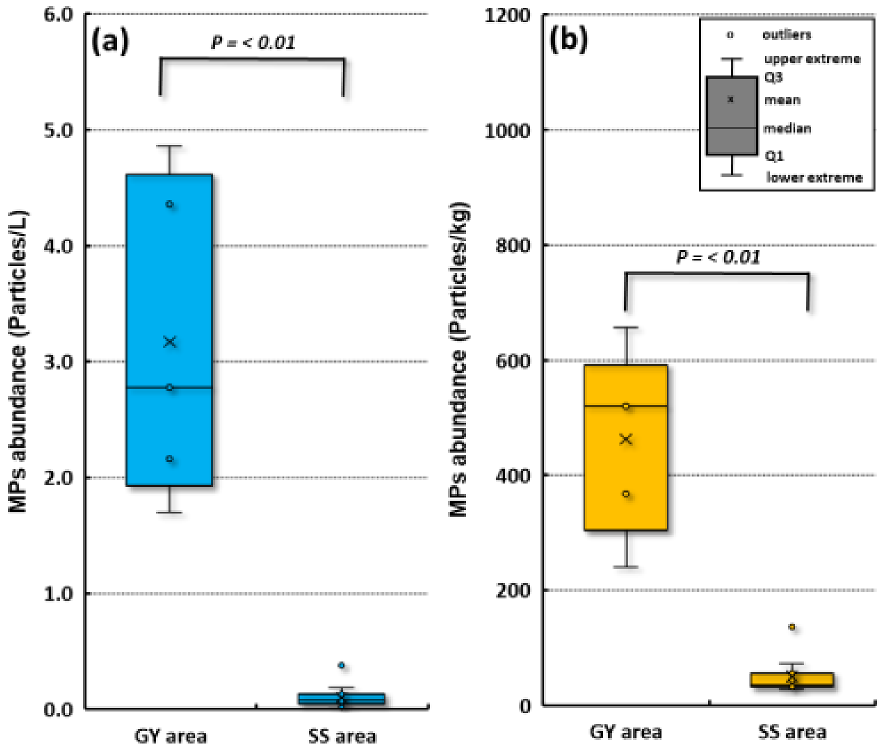

Figure 8. Comparison of seawater and deposited microplastic abundance in the South Sea Coast and Gwangyang Bay of Korea. (**a**): seawater, (**b**): sediment.

Looking further at this, in the case of Gwangyang Bay, in the microplastic form of seawater (Figure 4a) and surface sediments (Figure 4b), the fragment form was shown to display a higher percentage than the fiber form [40] caused by aquaculture and fishing activities. This area is where Gwangyang Bay is designated and managed as a particular management sea area by Article 15 of the Marine Environment Management Act, a Korean law. Therefore, aquaculture activities and fishing activities are restricted. As a result, it is judged that the inflow of microplastic products (PE production capacity: 4018 thousand tons/year (LG Chem: 2076 thousand tons/year, Lotte Chemical: 1060 thousand tons/year, Hanwha Solution: 882 thousand tons/year), PP production capacity: 1480 thousand tons/year (LG Chem: 380 thousand tons/year, Lotte Chemical: 1100 thousand tons/year), PS production capacity: 552 thousand tons/year (LG Chem: 140 thousand tons/year, Lotte Chemical: 80 thousand tons/year, Kumho Petrochemical: 332 thousand tons/year), PCV production capacity: 1670 thousand tons/year (LG Chem: 890 thousand tons/year, Hanwha Solution: 780 thousand tons/year), [58]) or by-products in the form of fragments produced in industrial complexes will be significant due to industrial activities, along with the inflow from surrounding cities (Yeoseo, Suncheon, Gwangyang in Jeollanam-do, and Hadong and Namhae in Gyeongsangnam-do) rather than in the form of fibers [23]. In addition, in the results for microplastic abundance in seawater (Figure 4a), the abundance was somewhat high at the site inside Gwangyang Bay, where Suncheon, Gwangyang, and Yeosu regions of Jeollanam-do are bordered, and the site close to the Yeosu Wolnae water end treatment plant is considered to be evidence of this.

In addition, after high tide, the direction of the ocean current in Gwangyang Bay moves from the western sea to the eastern sea around Myo Island in Gwangyang Bay. After that, it passes through the Yeosu Strait by combining with the water mass flowing south from the Seomjin River [59,60]. Among the results of this study, the horizontal distribution of surface seawater microplastic abundance showed very high microplastic abundance on the west side of Gwangyang Bay due to the flow of ocean currents. In addition, the surface sediment microplastic abundance was higher at the site on the east side than on the west side of Gwangyang Bay. There is a similar trend between the flow of ocean currents and the microplastic deposition mechanism [61], and it is judged that the results of surface seawater and surface sediment are different. However, at the GY04 site located on the east side of Gwangyang Bay, the surface seawater microplastic abundance was somewhat high. It is thought that this was introduced from the Yeosu Wolnae water end treatment plant (processing capacity: 198 m^3/day, [62]) located near the GY04 site, and showed a somewhat high abundance.

Meanwhile, the Tsushima Current moving northward in summer turns clockwise around Jeju Island and flows quickly into the Jeju Strait at 50 cm/s in July. After passing through the Jeju Strait, the Jeju warm current water moves to the SS area, the study area [63]. After that, it can be confirmed that it flows strongly into the Korean Strait, combined with the Tsushima Current [64], moving northward from the southeastern sea of Jeju Island [26]. Therefore, microplastics introduced into Gwangyang Bay are expected to escape the Yeosu Strait and enter the southern coastal waters, while some will be deposited on the seafloor. The remaining microplastics have shown much lower results in the southern coastal waters than in Gwangyang Bay because they move toward the Korean Strait rather than flowing into the southern coastal waters, due to the Jeju warm current and Tsushima current that occur very strongly in summer. In addition, the surface sea microplastic abundance showed a higher abundance than the site adjacent to the land in the southern coastal waters (the waters where Tsushima Current moves north), which is the 93 area (Figure 4a). On the other hand, the surface sediment microplastic abundance was opposite to the surface seawater microplastic results (Figure 4b). Therefore, it is judged that entering one source of pollution does not affect the distribution of microplastics in Gwangyang Bay and southern coastal waters, but this is due to different forms.

However, this study could not obtain samples in the middle of Gwangyang Bay and southern coastal waters. Therefore, further research is underway to clarify the results of microplastic distribution characteristics in the southern coastal waters after inflow from Gwangyang Bay.

5. Conclusions

This study aims to understand the distribution characteristics of microplastics introduced from industrial facilities and densely populated areas and to provide primary data for other microplastic studies.

Gwangyang Bay surface seawater results averaged 3.17 $\pm$ 1.23 particles/L, and sediment results averaged 462.4 $\pm$ 143.9 particles/kg. Seawater results in southern coastal waters averaged 0.10 $\pm$ 0.09 particles/L, and sediment results averaged 50.6 $\pm$ 29.7 particles/kg. Gwangyang Bay, which is adjacent to land, showed more diverse and abundant microplastic materials than the southern coast. Comparing our microplastic results to those of similar studies, we found that Gwangyang Bay exhibited significantly higher levels than reported in other literature, while the southern coast showed lower levels. This comparison highlights the relevance and progress of our current research in understanding microplastic distribution in Korean bays and coasts.

Because microplastics from Gwangyang Bay flow into the southern coastal waters and move in the direction of the Korean Strait due to the Jeju warm current and Tsushima current, which are strongly generated in summer, the southern coastal waters showed much lower microplastic abundance than Gwangyang Bay. Microplastics have shown much lower results in the southern coastal waters than in Gwangyang Bay because they move toward the Korean Strait rather than flowing into the southern coastal waters due to the Jeju warm current and Tsushima current that occur enormously in summer. In addition, the surface sea microplastic abundance showed a higher abundance than the site adjacent to the land in the southern coastal waters (the waters where Tsushima Current moves north), which is the study area. On the other hand, the surface sediment microplastic abundance was opposite to the surface seawater microplastic results. Therefore, it is judged that entering one source of pollution does not affect the distribution of microplastics in Gwangyang Bay and southern coastal waters, but this is due to different forms.

Author Contributions: Conceptualization, B.-K.M., C.-R.C., H.-Y.S. and H.-S.C. (Hyeon-Seo Cho); methodology, B.-K.M., C.-R.C. and H.-S.C. (Hyeon-Seo Cho); software, B.-K.M.; validation, B.-K.M., C.-R.C., H.-Y.S. and H.-S.C. (Hwi-Su Cheon); formal analysis, B.-K.M. and C.-R.C.; investigation, B.-K.M. and H.-S.C. (Hwi-Su Cheon); resources, B.-K.M. and C.-R.C.; data curation, B.-K.M. and C.-R.C.; writing—original draft preparation, B.-K.M.; writing—review and editing, C.-R.C., H.-Y.S. and H.-S.C. (Hyeon-Seo Cho); visualization, B.-K.M.; supervision, H.-S.C. (Hyeon-Seo Cho); project ad-ministration, H.-Y.S. and H.-S.C. (Hyeon-Seo Cho); funding acquisition, H.-S.C. (Hyeon-Seo Cho). All authors have read and agreed to the published version of the manuscript.

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