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# National Reconnaissance Survey of Microplastics in Municipal Wastewater Treatment Plants in Korea

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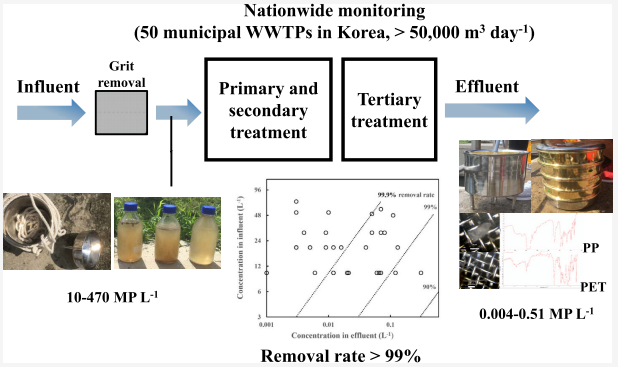
**ABSTRACT:** Large quantities of microplastics are thought to be emitted to freshwater environments via wastewater treatment plants (WWTPs). To evaluate the occurrence of microplastics in Korean WWTPs, a nationwide study was conducted for the first time in 50 representative WWTPs with large treatment capacities. Grab sampling and laboratory filtration were used for influents, whereas in situ filtration using a custom-made sampling device was used for effluents. The filtrates were pretreated using wet peroxidation and density separation prior to the identification of microplastics with a dissection microscope and Fourier-transform infrared spectroscopy. Pooled analyses of the microplastics revealed that they were predominantly fragment-shaped, and thermoplastics and synthetic fibers were the dominant microplastic materials in WWTPs. The concentration ranged from 10 to 470 $L^{-1}$ in influents and 0.004 to 0.51 $L^{-1}$ in effluents. The removal efficiency of microplastics during wastewater treatment was calculated to be 98.7−99.99% in 31 WWTPs. Additionally, WWTPs using advanced phosphorus removal processes exhibited higher removal efficiency than those not implementing such processes. Power-law distribution was successful in describing microplastic particle sizes down to 100 $\mu$m, although it was not applicable for smaller particles. This comprehensive monitoring study provides information on the current level and characteristics of microplastics in WWTPs in Korea.






## INTRODUCTION

Environmental contamination of microplastics, small plastic particles less than 5 mm in length,[1,2] occurs worldwide in various environmental media such as seawater,[3,4] beach soil,[5−7] freshwater,[8,9] and terrestrial soil.[10,11] Although this topic is under debate, microplastics are suspected to cause potential adverse effects on individual organisms,[12,13] ecosystems,[14] and human health.[15] Among the many anthropogenic sources of microplastics in the environment, municipal wastewater treatment plants (WWTPs) are regarded as important point sources to the aquatic environment,[16,17] although microplastics are effectively removed during particulate matter removal in WWTPs.[18−21]

With increasing concerns on microplastics in freshwater systems,[9,22] many studies have been conducted to determine the level of microplastics in WWTPs.[16−19,23−37] Given that no standard experimental protocols are in place to isolate and detect microplastics in WWTP influents and effluents, researchers have employed various methods for such procedures. Grab sampling methods were used to sample microplastics in WWTP influents, whereas in situ filtration using custom-made filtration devices were used for WWTP effluents[16,24,32] owing to the quality of sampled water such as suspended solids and differences in the order of magnitude in the concentration of microplastics in water. Large variations in sampling volumes were found in the literature, ranging from 0.1−30 to 2−232 000 L for WWTP influents and effluents, respectively.[19,25−27] Although most studies have adopted oxidative digestion and identification using Fourier-transform infrared (FT-IR) spectroscopy, differences in sampling methods result in variations in factors such as sample volume and pore size of the filtration system (0.7−330 $\mu$m), making it difficult to compare different study outputs. Owing to these reasons, and also to differences in treatment, population density, and product use, large variations occur in the reported concentrations in WWTP influents and effluents from different regions of the world. The reported range for influents is 0.28−640 and n.d.−65 $L^{-1}$ for effluents with removal efficiencies between 64.4[28] and 99.9%.[17]

Although the reported variations in the concentration and material type of microplastics in WWTP influents and effluents could be caused by differences in analytical methods, the

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Table 1. Basic Information of the Selected Wastewater Treatment Plants in Korea[a]

| location | name | treatment capacity in 2017 ($m^3$ $day^{-1}$) | treatment processes | advanced phosphorus removal treatment |
|---|---|---|---|---|
| Seoul | STP-1 | 1 337 000 | modified A2O, A2O, MLE, AS | N |
| Seoul | STP-2 | 1 584 000 | MLE, modified A2O | N |
| Busan | STP-3 | 330 000 | AS, MLE, MBR | N |
| Busan | STP-4 | 404 000 | A2O | N |
| Daegu | STP-5 | 491 000 | A2O | Y (precipitation) |
| Daegu | STP-6 | 401 000 | A2O | Y (disk filter) |
| Incheon | STP-7 | 231 000 | AS, MLE | N |
| Incheon | STP-8 | 232 000 | modified A2O, MLE | N |
| Gwangju | STP-9 | 552 000 | modified A2O | Y (precipitation) |
| Gwangju | STP-10 | 102 000 | MLE, A2O | Y (filtration) |
| Daejeon | STP-11 | 619 000 | modified A2O, MLE | Y (precipitation) |
| Ulsan | STP-12 | 217 000 | modified A2O | N |
| Ulsan | STP-13 | 107 000 | modified A2O | N |
| Sejong | STP-14 | 36 000 | modified A2O | Y (filtration) |
| Gyeonggi | STP-15 | 522 000 | modified A2O | N |
| Gyeonggi | STP-16 | 336 000 | modified A2O, A2O | Y (disk filter) |
| Gyeonggi | STP-17 | 146 000 | MLE | Y (filtration) |
| Gyeonggi | STP-18 | 191 000 | MLE | Y (precipitation) |
| Gyeonggi | STP-19 | 701 000 | modified A2O | Y (filtration) |
| Gyeonggi | STP-20 | 386 000 | modified A2O, modified AS | N |
| Gyeonggi | STP-21 | 190 000 | MLE | N |
| Gyeonggi | STP-22 | 139 000 | MBR, MLE | Y (precipitation) |
| Gyeonggi | STP-23 | 93 000 | modified A2O | Y (precipitation) |
| Gyeonggi | STP-24 | 99 000 | modified AS | Y (filtration) |
| Gyeonggi | STP-25 | 165 000 | modified A2O | N |
| Gyeonggi | STP-26 | 21 000 | modified A2O | N |
| Gangwon | STP-27 | 130 000 | modified A2O | Y (floatation) |
| Gangwon | STP-28 | 139 000 | modified A2O | Y (disk filter) |
| Gangwon | STP-29 | 68 000 | modified A2O | N |
| Chungbuk | STP-30 | 269 000 | modified A2O | Y (precipitation) |
| Chungbuk | STP-31 | 62 000 | modified AS, modified A2O | Y (floatation) |
| Chungnam | STP-32 | 175 000 | modified A2O, MBR | N |
| Chungnam | STP-33 | 26 000 | modified AS | Y (disk filter) |
| Chungnam | STP-34 | 63 000 | modified A2O | N |
| Chungnam | STP-35 | 40 000 | modified A2O | N |
| Jeonbuk | STP-36 | 334 000 | modified A2O | Y (precipitation) |
| Jeonbuk | STP-37 | 147 000 | modified A2O | N |
| Jeonbuk | STP-38 | 88 000 | MLE | Y (filtration) |
| Jeonnam | STP-39 | 59 000 | modified A2O | N |
| Jeonnam | STP-40 | 81 000 | modified A2O | N |
| Jeonnam | STP-41 | 105 000 | modified A2O | N |
| Jeonnam | STP-42 | 23 000 | AS | N |
| Gyeongbuk | STP-43 | 214 000 | modified A2O | N |
| Gyeongbuk | STP-44 | 92 000 | MLE | N |
| Gyeongbuk | STP-45 | 64 000 | modified A2O | Y (floatation) |
| Gyeongbuk | STP-46 | 261 000 | modified A2O | Y (filtration) |
| Gyeongnam | STP-47 | 306 000 | modified A2O | N |
| Gyeongnam | STP-48 | 135 000 | modified A2O | Y (floatation) |
| Gyeongnam | STP-49 | 108 000 | modified A2O | Y (floatation) |
| Gyeongnam | STP-50 | 75 000 | modified A2O | Y (precipitation) |

[a]Abbreviations: A2O, anaerobic−anoxic−aerobic treatment process; MLE, modified Ludzack−Ettinger process; AS, activated sludge treatment process; and MBR, membrane bioreactor process.

variation in the occurrence of microplastics in WWTPs also likely results from regional characteristics such as the local consumption of plastic products as well as regional industrial activities, hydrologic and (micro)climate factors, and unit processes in wastewater treatment. For example, poly(ethylene terephthalate) (PET) was found to be the dominant material type of microplastics in Finnish WWTPs with the concentration of microplastics in WWTP influent at 500−6000 $L^{-1}$,[29] whereas polypropylene (PP) was the dominant material type in seven Chinese WWTPs with an influent concentration of 1.0−14 $L^{-1}$.[30]

In Korea, almost all sewers are connected to approximately 600 small and large municipal WWTPs;[38] thus, microplastics that originate from sewers likely pass through these facilities. However, only a few studies have reported microplastics in a few of these WWTPs[31] or water bodies receiving WWTP effluents.[39] Therefore, a nationwide monitoring study was planned to estimate WWTP microplastic loads, as well as the amount of microplastics discharged into public waterways from WWTPs. We report the occurrence of microplastics in influents and effluents of 50 WWTPs as preliminary results of this monitoring program in Korea. Treatment facilities were chosen to cover a wide range of treatment processes, discharge volume, and locations. Approximately 1700 microplastic particles were recovered and analyzed. Based on the screening analysis of 50 WWTPs, the characteristics of microplastics in


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WWTPs were evaluated, including dominant material type, morphology, size distribution, removal efficiency, per capita WWTP microplastic loads, and the amount of discharge from the WWTPs.

## MATERIALS AND METHODS

**Municipal Wastewater Treatment Plants.** To monitor microplastics in the influents and effluents of municipal WWTPs in Korea, 50 facilities capable of treating more than 50 000 $m^3$ $day^{-1}$ were chosen. This covers approximately 8% of the 625 WWTPs in Korea and provides a representative sample size for nationwide microplastic monitoring. Basic information of the selected WWTPs is summarized in Table 1. Fourteen facilities discharge their effluent to coastal waters and others to rivers and lakes. Thirty-nine facilities (78%) apply anaerobic−anoxic−aerobic (A2O) or modified A2O treatment processes. The population size serviced per WWTP ranged from 51 00 to 3 339 000, and the treatment capacity ranged from 21 000 to 1 584 000 $m^3$ $day^{-1}$ in 2017.[38] The locations of all WWTPs are shown in Figure S1 (Supporting Information); these cover the entire nation and four major river systems receiving WWTP effluents in Korea.

**Sample Collection.** Sampling was conducted, avoiding rainfall events between September 5 and November 1, 2018. Prior to sampling, the hydraulic retention time of each facility was obtained by interviewing operators. The effluent sample was collected after the influent sample was obtained, considering the retention time of the facility. Table S1 (Supporting Information) details the sampling dates and times.

Owing to the water quality and differences in the concentration of microplastics in the WWTP influents and effluents, different sampling methods were used. For sampling microplastics in WWTP influents, the grab sampling method was used. In this case, a stainless steel bucket was submerged into the WWTP influent reservoir at depths of 0.4−0.5 m and the collected water was poured into a 1 L amber glass bottle. Influent samples were collected after grit removal; influent reservoir depth was typically <5 m. Although larger debris with densities greater than that of water [e.g., poly(tetrafluoroethylene), ca. 2.2 g $cm^{-3}$] tend to settle, the retention time in the reservoir was not long enough to settle microplastics with moderate density (e.g., PET, 1.38 g $cm^{-3}$). This sampling process was repeated three times to collect triplicate influent samples. For sampling microplastics in WWTP effluents, the in situ filtration method was used. For this process, a custom-made filtration device (Figure S2, Supporting Information) was constructed as described by Ziajahromi et al.[32] The device was prepared using commercial stainless steel sieves with 5 mm, 1 mm, 300 $\mu$m, and 100 $\mu$m pore sizes. The pumping rate was approximately 21 L $min^{-1}$, and a total volume of 1000 L of WWTP effluent was filtered through the custom-made filtration device at depths of 0.5−1 m from the surface layer of the effluent reservoir. All materials in contact with water were composed of stainless steel, except for the polybutylene tubing. As discussed subsequently, no polybutylene microplastics were identified in the results. The amber glass bottles and the sieve assembly were wrapped using aluminum foil and secured immediately after sampling and were transported to the laboratory for isolation of microplastics.

**Isolation of Microplastics from Wastewater Influents and Effluents.** To remove particles other than microplastics, the wet peroxidation method using a 30% $H_2O_2$ solution (Daejung Inc., Siheung, Korea) was employed.[23,32,33,40] To remove high-density particles such as soil minerals, density separation using 3.66 M $ZnCl_2$ (Daejung Inc.) solution was carried out at $d$ = 1.36 g $cm^{-3}$. Distilled water and $ZnCl_2$ solution were filtered through a Whatman grade GF/C glass microfiber filter with a pore size of 1.2 $\mu$m (Merck KGaA, Darmstadt, Germany) to filter out any residual particles from the water and reagents. The analyzed volume of influent samples varied. Initially, three 20 mL aliquots from each influent sample were combined, digested, and filtered to count the microplastics. If the total number of isolated microplastics from 60 mL was less than 10, microplastics were additionally isolated from 3 × 50 mL of influent samples to give a total volume of 210 mL. The influent sample was mixed with the same volume of 30% $H_2O_2$ solution in a glass tube, and the resulting solution was heated in a water bath at 60 °C for 3 h. If 3 h was not sufficient for removing organic matter and the solution was not clarified, an additional volume of $H_2O_2$ solution was added, and the mixture was heated until the solution became clear enough to proceed with the isolation of the microplastics. After wet peroxidation, the glass tube was placed in a drying oven at approximately 105 °C to evaporate the solution completely. Because the sample was removed immediately after the solution had completely evaporated, the actual temperature of the solution should have been below 100 °C. For density separation, 20 mL of $ZnCl_2$ solution was added to the glass tube and microplastic particles were allowed to float near the surface for 3 h. Particles on the surface of the solution were carefully collected and filtered through a 25 mm diameter and 45 $\mu$m pore size stainless steel filter. All of the filtrates were secured in a clean Petri dish to minimize potential contamination and further dried in a vacuum desiccator.

For effluent samples, the particles on each sieve were detached from the sieve using a distilled water squeeze bottle and then transferred to a glass tube. All particles from the same WWTP effluent were combined. Wet peroxidation, density separation, and filtration were then conducted as described previously for the pretreatment of WWTP influents.

**Prevention of Potential Contamination.** Given the potential for microplastic sample contamination during the sampling and isolation steps, the experiments were conducted with extreme care. Amber glass bottles used for WWTP influent sampling were washed thoroughly and dried at 105 °C overnight. The bottles were then rinsed with distilled water before sampling. The influent samples were stored in a refrigerator at 4 °C prior to the wet peroxidation procedure to minimize potential microbial degradation. Custom-made sieves (Figure S2, Supporting Information) were thoroughly washed with distilled water and dried before sampling. The sieves were kept wrapped in aluminum foil until the laboratory analysis. The particles on sieves were collected in a glass tube as early as possible using the method described in the previous section to minimize the potential loss of microplastics. Although no field control samples were employed in this study, it is unlikely that influent and effluent samples were affected during transportation because they were completely sealed and wrapped immediately after sampling.

**Spectroscopic Determination of Microplastics.** The suspected particles isolated on the stainless steel filter were analyzed with an FT-IR spectroscope equipped with a dissection microscope (Nicolet iN10; ThermoFisher Scientific, Waltham, MA) for confirmation and identification of the



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**Table 2. Occurrence of Microplastics in 50 WWTPs**

| | influent samples | | | effluent samples | |
|---|---|---|---|---|---|
| name | volume analyzed (mL) | number counted | concentration ($L^{-1}$) | number counted | concentration ($L^{-1}$) |
| STP-1 | 210 | 14 | 67 | 20 (1/10)[a] | 0.20 |
| STP-2 | 210 | 11 | 52 | 30 (1/10)[a] | 0.30 |
| STP-3 | 60 | 13 | 220 | 20 | 0.020 |
| STP-4 | 60 | 19 | 320 | 59 | 0.059 |
| STP-5 | 210 | 7 | 33 | 6 | 0.006 |
| STP-6 | 210 | 12 | 57 | 6 | 0.006 |
| STP-7 | 210 | 13 | 62 | 15 (1/10)[a] | 0.15 |
| STP-8 | 210 | 25 | 120 | 21 (1/10)[a] | 0.21 |
| STP-9 | 210 | 17 | 81 | 29 (1/2)[d] | 0.058 |
| STP-10 | 150 | 7 | 47 | 11 (1/2)[d] | 0.022 |
| STP-11 | 210 | 14 | 67 | 12 (1/10)[a] | 0.12 |
| STP-12 | 60 | 10 | 170 | 49 (1/5)[b] | 0.245 |
| STP-13 | 60 | 11 | 180 | 25 (1/10)[a] | 0.25 |
| STP-14 | 210 | 14 | 67 | 20 | 0.02 |
| STP-15 | 60 | 28 | 470 | 8 | 0.008 |
| STP-16 | 150 | 5 | 33 | 6 | 0.006 |
| STP-17 | 210 | 11 | 52 | 19 | 0.019 |
| STP-18 | 210 | 20 | 95 | 20 | 0.02 |
| STP-19 | 60 | 12 | 200 | 17 (1/5)[b] | 0.085 |
| STP-20 | 210 | 6 | 29 | 55 | 0.055 |
| STP-21 | 210 | 35 | 170 | 26 | 0.052 |
| STP-22 | 210 | 20 | 95 | 8 | 0.008 |
| STP-23 | 210 | 16 | 76 | 22 | 0.022 |
| STP-24 | 60 | 9 | 150 | 18 (1/5)[b] | 0.09 |
| STP-25 | 210 | 25 | 120 | 14 (1/2)[d] | 0.028 |
| STP-26 | 210 | 20 | 95 | 12 (1/10)[a] | 0.12 |
| STP-27 | 150 | 4 | 27 | 17 (1/5)[b] | 0.085 |
| STP-28 | 210 | 18 | 86 | 13 (1/10)[a] | 0.13 |
| STP-29 | 210 | 2 | 10 | 41 (1/10)[a] | 0.41 |
| STP-30 | 210 | 9 | 43 | 29 (1/10)[a] | 0.29 |
| STP-31 | 210 | 13 | 62 | 26 (1/3)[c] | 0.078 |
| STP-32 | 210 | 30 | 140 | 9 | 0.009 |
| STP-33 | 210 | 17 | 81 | 10 (1/10)[a] | 0.10 |
| STP-34 | 150 | 14 | 67 | 43 (1/10)[a] | 0.43 |
| STP-35 | 210 | 20 | 95 | 21 (1/2)[d] | 0.042 |
| STP-36 | 210 | 23 | 110 | 4 | 0.004 |
| STP-37 | 210 | 22 | 100 | 18 (1/10)[a] | 0.18 |
| STP-38 | 210 | 6 | 29 | 11 (1/2)[d] | 0.022 |
| STP-39 | 210 | 6 | 29 | 15 (1/10)[a] | 0.15 |
| STP-40 | 210 | 9 | 43 | 28 (1/3)[c] | 0.084 |
| STP-41 | 210 | 8 | 38 | 51 (1/10)[a] | 0.51 |
| STP-42 | 210 | 3 | 14 | 13 | 0.013 |
| STP-43 | 210 | 11 | 52 | 22 (1/10)[a] | 0.22 |
| STP-44 | 60 | 12 | 180 | 15 (1/10)[a] | 0.15 |
| STP-45 | 60 | 16 | 270 | 17 (1/3)[c] | 0.051 |
| STP-46 | 210 | 12 | 57 | 27 | 0.027 |
| STP-47 | 210 | 13 | 62 | 14 | 0.14 |
| STP-48 | 210 | 7 | 33 | 4 | 0.004 |
| STP-49 | 210 | 8 | 38 | 13 | 0.013 |
| STP-50 | 210 | 26 | 120 | 11 | 0.11 |

[a]One of every 10 particles analyzed. [b]One of every five particles analyzed. [c]One of every three particles analyzed. [d]One of every two particles analyzed.

material type. All visible particles from the WWTP influent samples were analyzed using FT-IR. However, due to the extremely high number of suspected particles in the WWTP effluent samples, every 2, 3, 5, or 10 particles were analyzed depending on the total number of suspected particles to minimize processing time. A minimum of 10 microplastic particles were ultimately counted in each WWTP effluent sample.

The shapes of identified microplastics were divided simply into fibers and fragments. The fragment particle lengths were estimated assuming an oval shape. The lengths of the major and minor axes were measured using photographs of the



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Table 3. Comparison of Concentration of Microplastics in WWTP Influents and Effluents

| location | concentration[a] influent ($L^{-1}$) | effluent ($L^{-1}$) | method used (mesh size) | reference |
|---|---|---|---|---|
| Canada | 31.1 ± 6.7 | 0.5 ± 0.2 | oil extraction followed by $H_2O_2$ digestion and filtration (63 $\mu$m) | 37 |
| China | 79.9 ± 9.3 | 28.4 ± 7.0 | sieved, digested using $H_2O_2$ and filtrated (47 $\mu$m) | 28 |
| China | 5.43 ± 4.53 | 0.59 ± 0.49 | pumped through stacked sieves followed by $H_2O_2$ digestion (43 $\mu$m) | 30 |
| China | 0.28 ± 0.02 | 0.13 ± 0.01 | filtered followed by $H_2O_2$ digestion (20 $\mu$m) | 36 |
| | | 0.05 ± 0.01 | | |
| Finland | 610 | 13.5 ± 2.9 | pumped through stacked sieves (20 $\mu$m) | 16 |
| Finland | 568 ± 165 | 2.5 ± 1.5 | pumped through stacked sieves (20 $\mu$m) | 29 |
| | 640 ± 255 | 0.6 ± 0.2 | | |
| Finland | 57.6 ± 32.8 | 1.0 ± 1.1 | pumped through stacked sieves followed by $H_2O_2$ digestion (250 $\mu$m) | 33 |
| France | 293 | 35 | filtration through glass fiber membrane (1.6 $\mu$m) | 34 |
| Korea | 10−470 | 0.004−0.51 | influent: digested using $H_2O_2$ and filtered (45 $\mu$m) | this study |
| | | | effluent: pumped through stacked sieves followed by $H_2O_2$ digestion (100 $\mu$m) | |
| Netherlands | 73 ± 13 | 65 ± 67 | filtration through glass fiber membrane (0.7 $\mu$m) | 26 |
| Scotland | 15.7 ± 5.23 | 0.25 ± 0.04 | sieved and vacuum filtrated (65 $\mu$m) | 25 |
| Sweden | 15.1 ± 1.54 | 0.00825 ± 0.0017 | filtered through plankton net (300 $\mu$m) | 17 |
| United States | 136.73 ± 48.8 | 13.65 ± 8.80 | filtration followed by $H_2O_2$ digestion (43 $\mu$m) | 35 |

[a]Mean value ± standard deviation.

particles. For each background and sample spectrum, an absorption spectrum was obtained by scanning 16 times at a resolution of 4 $cm^{-1}$ in a wavelength range of 4000−675 $cm^{-1}$. The spectra were compared with the internal infrared (IR) database, which is a commercial polymer database, including a spectrum library, and were confirmed using the OMNIC Picta expert software (ThermoFisher Scientific) with a similarity of 70% and above. Particles identified as rayon were not counted because the spectra of anthropogenic rayon and natural cellulose overlap.[41]

**Extraction Recovery.** To verify the recovery rate of the microplastic isolation process used, an extraction test was performed using three material types. Standard microplastics of polyethylene (PE) and PP were fabricated in forms of fibers, 100−150 $\mu$m in diameter with a plastic extruder (LME-230, Dynisco, Franklin, MA), and poly(ethylene terephthalate) fiber was purchased from Goodfellow Corp. (Coraopolis, PA). The fibers were cut into 100−150 $\mu$m fragments; an example of the prepared standard microplastics are illustrated in Figure S3 (Supporting Information). Standard microplastics in quantities of 10, 20, 30, 40, and 50 were counted and spiked into a 20 mL influent solution. After the same pretreatment procedure as that described above was completed, the number of standard microplastics filtered was counted using a stereomicroscope (Luxeo 4Z; Labo America Inc., Fremont, CA) to obtain pretreatment recovery for the fragment-type microplastics.

The treatment temperatures during wet peroxidation and solvent evaporation were rather high in this study to shorten the pretreatment time required to process a large number of samples. Therefore, a comparison of treatment methods was also conducted using standard PE microplastics using wet peroxidation temperatures of 40 and 60 °C and evaporation temperatures of 60 and 105 °C in a drying oven.

To account for potential contamination during pretreatment in the laboratory, five blank samples (i.e., distilled water) were treated following the same procedure as that used for the influent samples. For five effluent blanks, debris was detached from the sieves using distilled water following the same procedure used for WWTP effluents and was then collected and stored. The final filtrates on the stainless steel filter with a 45 $\mu$m pore size were then analyzed by visual identification and FT-IR analysis.

**Statistical Analyses.** Correlation analysis and principal component analysis on the relationship of the population served by WWTPs, wastewater volumetric flow rate to WWTPs, and incoming load of microplastic particles to the WWTPs were conducted using the R software.[42] A paired *t*-test was conducted for the removal efficiency of microplastics for two groups of WWTPs with and without tertiary treatment processes using Microsoft Excel 2016.

## RESULTS AND DISCUSSION

**Recovery of Spiked Plastic Particles and Laboratory Controls.** As presented in Table S2 (Supporting Information), the pretreatment recoveries of standard PE, PP, and PET microplastics were 86.8 ± 8.2, 89.4 ± 8.5, and 78.7 ± 9.8%, respectively. Upon comparison, no significant differences were observed between digestion temperatures at 40 and 60 °C and evaporation temperature at 60 and 120 °C using standard PE microplastics (Table S3, Supporting Information). Although the average recovery did not exceed 90%, we believe that the recovery range was acceptable for at least PE and PP fragments, as well as PET fibers, 100 $\mu$m or longer. Image analysis of standard PE microplastics also confirmed that the melting of PE surfaces during pretreatment was not serious because sharp edges were maintained (Figure S4, Supporting Information). The slightly lower recovery of PET fibers compared to that of PE and PP might be due to their longer shape and higher density. However, the denser $ZnCl_2$ solution (1.58 g $cm^{-3}$) did not significantly improve the recoveries for PE and PET microplastics (Table S3, Supporting Information). The major causes of loss would be physical adherence to glass and stainless steel surfaces. It should be noted that the employed method might not be applicable for high-density plastic materials such as poly(vinyl chloride) and poly(tetrafluoroethylene) because the density of the $ZnCl_2$ solution used in the density separation step was 1.36 kg $L^{-1}$, even though an acceptable recovery rate was observed for PE, PP, and PET.

One PE fragment was isolated from five laboratory blank samples for WWTP influents, while one PE fragment and one


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PET fiber were isolated from five laboratory blank samples for WWTP effluents (Figure S5, Supporting Information). This supports that potential contamination during the laboratory procedure was not significant compared with the total number of microplastics isolated per stainless steel filter. Atmospheric fallout or fabric fibers in the laboratory might account for the slight contamination observed in negative controls.

**Occurrence of Microplastic Particles in WWTP Influents and Effluents.** All visible particles on the filter were analyzed using FT-IR and photographed to record the shapes and sizes of all identified microplastics. In total, 1720 microplastic particles were identified in 50 WWTPs, with 700 and 1020 particles found in the influents and effluents, respectively. Representative example photos of fragment- and fiber-type microplastics are shown in Figure S6, Supporting Information. Table 2 lists the number of counted microplastics and the concentration in each influent and effluent sample. The number of microplastics counted in the influents were between 2 and 35, resulting in a concentration of 10 (STP-29) to 470 $L^{-1}$ (STP-15). For the WWTP effluents, the number of counted microplastics were 4−59 on each filter, and the resulting concentration was 0.004−0.59 $L^{-1}$. Table 3 compares the concentration of microplastics in this study with those reported in the literature for both influent and effluent concentrations.[16,17,25,26,28−30,34−37] Considering that there were no standardized protocols (i.e., different studies employed different methods), the results could not be directly compared. However, more number of microplastics tended to be counted when a smaller mesh size was used. Two studies using larger mesh sizes[17,33] reported lower concentrations. However, concentrations of microplastics in both influent and effluent samples were higher by approximately 1 order of magnitude when filtered through a 20 $\mu$m pore size[16,29] compared to a 250 $\mu$m size.[33] This could potentially explain the lower microplastic concentrations observed in this study compared to earlier studies that employed smaller mesh sizes.[16,29,33−35] Understanding size distribution of microplastics would provide a quantitative comparison tool for studies with varying filtration cutoffs.[20]

Fragment-type microplastics were found to be dominant in both influents and effluents. In the influents, 68.2% were classified as fragments and 31.8% were fibers, whereas 82.3% were fragments and 17.7% were fibers in the effluents. The lower fraction of fibers in WWTP effluents was consistent with Talvitie et al.[19] These percentages show that the ratio of the fragment-type in the effluent was higher than that in the influent, which might be attributed to differences in sampling methods between the influents and effluents. During long sampling times, fibers can be washed away more easily than fragments from the sieve. Moreover, most of the isolated fibers exhibited diameters below the 45 $\mu$m pore size of the stainless filter. Thus, a higher fraction of fibers might have been isolated if a smaller pore size (e.g., 20 $\mu$m) was used.[19,29] Another explanation is that, unlike with microplastic fragments, the elongated shape of fibers may make them prone to entanglement during wastewater treatment and gravitational settling.[43]

PP was found to be the most dominant material type in the WWTP influents and effluents. In influent samples, the percentages of PP, PE, and PET were 39.6, 25.6, and 21.3%, respectively. Other identified materials include polystyrene, acrylics, polyamide, polyurethane, and polyether. In the effluent samples, PP was the most dominant, at 63.3%, followed by PE and PET at 13.8 and 13.3%, respectively. Although the same three material types were dominant in the influents, the amount of PP was much greater in the effluents. In addition, the number of fibers of polymer PET composition was decreased in the effluents owing to the higher removal rates or the differences in sampling methods described above.

The patterns of isolated material types identified in this study differ somewhat from those reported in earlier studies in other regions. For example, PET was the major material type in Finnish WWTP effluents, at 28.38−100%,[29] and PE was dominant in German WWTP effluents, at 33.6−77.7%.[24] The predominance of PP in the present study agrees with the research results reported for Chinese WWTPs.[30] Although a review by Kang et al. reported large variations among WWTPs, polyethylene was found to be the most dominant material type, at 15−78%.[20] This might be related to the production, export, and usage of plastic products in Korea. Although the amounts of low-density PE (LDPE) and PP produced globally were 17.5 and 19.3% in 2017, respectively,[44] those in Korea were 18 and 26%, respectively.[45] The PP used for many consumer products might be the source of the PP microplastics found in WWTPs in Korea. The predominance of PP in the WWTPs is also consistent with the microplastics found in the Nak-Dong River.[39] However, further investigation using smaller mesh sizes is required to determine whether PET and other fiber-type microplastics were lost during sampling due to the relatively larger mesh size used in this study.

**Size Distribution.** Except for those engineered to a desired small size, microplastics in the environment are known to form via weathering processes of larger plastic particles.[46] If the plastic fragmentation is a scale-independent process within the 45 $\mu$m to 5 mm range of microplastics recovered in this study, the size distribution of the microplastics should follow the power-law.[47] Figure 1 describes the particle size distribution,

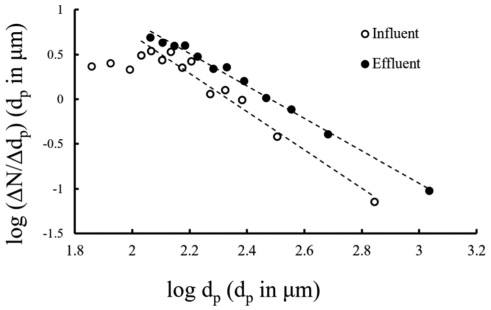


**Figure 1.** Particle size distribution of microplastics identified in WWTP influents and effluents. Groups of 30 microplastics were used to represent the size distribution. Particles with a minor axis length greater than 45 $\mu$m in influent samples and 100 $\mu$m in effluent samples were pooled and used for analysis, including 380 and 339 particles in the influent and effluent, respectively.

$\log(\Delta N/\Delta d_p)$ versus $\log d_p$, pooling all fragment-type microplastics, including 380 for influents and 339 for effluents, where $\Delta N$ is the number of microplastics in a size group and $\Delta d_p$ is the difference in the median length between the two adjacent size groups. All microplastics for which the minor axis is larger than 45 $\mu$m for microplastics in influents and larger than 100 $\mu$m in effluents were pooled and analyzed. The power-law distribution appears to be applicable for microplastics in the range of 100 $\mu$m to 5 mm. The slopes in Figure 1 are −2.14 and −1.81 for microplastics in influents and effluents,


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respectively. It is interesting to note that the slopes of the power-law distribution in this study are similar to those obtained by the same analysis by Kang et al.[20] using values available in the literature.[19,32]

In a laboratory batch fragmentation test conducted in previous research, the size distribution of microplastics formed via degradation of macro-sized plastic followed the power-law down to 20 $\mu$m in size.[48] Although smaller microplastics were not determined in this study, the fragmentation processes in an isolated system likely led to smaller microplastics of at least a few micrometers in size. However, the size distribution observed in field studies has been controversial. Recent monitoring studies of microplastics from Arctic Sea ice core samples with a cutoff size of 11 $\mu$m[49] and marine water samples in Denmark with a cutoff size of 10 $\mu$m[50] showed that smaller size microplastics were more abundant than larger plastic particles, which generally follows the power-law down to the cutoff size of the study. Conversely, the size distribution of floating microplastics sampled from the Mediterranean Sea showed that microplastics of about 2 mm in size were the most abundant; smaller microplastic particles were less abundant.[51] Similarly, the size distribution of microplastics collected from beach sand in Korea with a cutoff size of 20 $\mu$m showed that the abundance of microplastics decreased below 100 $\mu$m in size.[5] Unlike that in a closed batch system, microplastics in the environment may undergo several degradation processes such as settling and burial, fragmentation, and chemical decomposition.

The power-law successfully explains the size distribution of microplastics in the effluent samples with a cutoff size of 100 $\mu$m in this study. However, it appears that the tendency was not continued with smaller microplastics in the influent samples with a cutoff size of 45 $\mu$m. It is expected that the rate of chemical and biological reactions increases with the increase in the surface-to-volume ratio, leading to accelerated degradation of microplastics of smaller sizes. Faster natural weathering processes for smaller microplastics before entering WWTPs might explain the observed tendency. However, further investigation is needed to determine the method in which the accelerated decomposition of microplastics results in the size distribution of microplastics in various environmental media.

**Microplastics Load to WWTPs.** The load of microplastics to each municipal WWTP was obtained by multiplying the measured concentration and the volumetric flow rate with the population size served by each WWTP (Table S4, Supporting Information). As shown in Figure 2, the microplastics load to the WWTPs increased with an increase in both the volumetric flow rate and the population size. Multivariate and principal component analyses on these three variables showed that all are strongly correlated (Figure S7 and Table S5, Supporting Information). Assuming that microplastics flowing into WWTPs are derived mainly from human activities, the per capita production of microplastics to WWTPs was estimated to be 4000−480 000 day$^{-1}$. Conley et al.[35] estimated 35 654−115 832 microplastic particles per day per capita for three WWTPs. Considering the cutoff size and population size served by WWTPs, the range reported by Conley et al. overlaps with those in the present study. However, the contribution of other indirect sources to the overall microplastics load to municipal WWTPs remains unclear. Thus, further studies comparing extreme WWTPs (i.e., a relatively large volume flow rate with low population vs a relatively small volume flow rate with high population) would be helpful to determine the relationship between population size and microplastic input to municipal WWTPs.

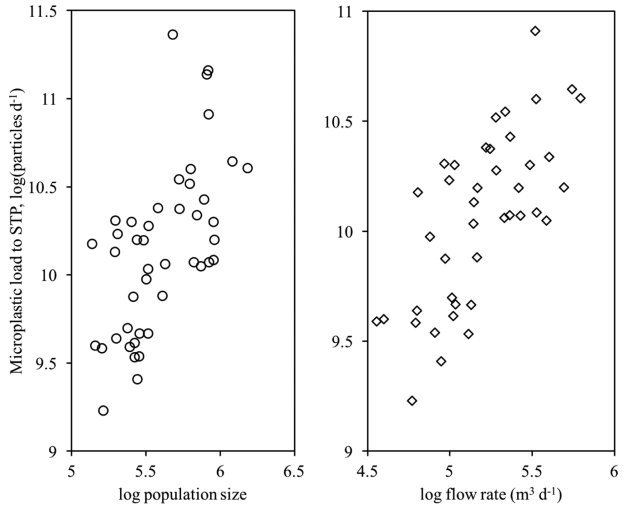


**Figure 2.** Microplastics load to wastewater treatment plants (WWTPs) with increasing populations and volumetric flow rates.

**Removal Efficiency of Fragment-Type Microplastic Particles in WWTP.** Differences in the cutoff size and sampling method employed lead to difficulties in deriving the removal efficiency simply using the influent and effluent microplastic concentrations. Thus, the removal rate was calculated for WWTPs ($n$ = 31) in which at least two particles with a minor axis length greater than 100 $\mu$m were found in both influent and effluent samples (Table S6, Supporting Information). The calculated values ranged from 98.7 to 99.99%, which suggests that incoming microplastics to WWTPs are easily removed. They were likely removed by gravitational settling to sewage sludge. Thus, the appropriate management of sewage sludge should be important to prevent the release of microplastics removed from WWTPs, because wastewater sludge is reused in agriculture in some countries.[21]

It is worth noting that the 16 WWTPs outfitted with advanced phosphorus removal units exhibited greater removal efficiency than the 15 facilities without such units according to Student's $t$-test ($p$ = 0.047). However, no statistically significant differences were identified among different phosphorus removal unit processes. Most phosphorus removal unit processes in Table 1 apply chemical coagulation followed by filtration, floatation, or sedimentation. Additional physical unit processes after secondary treatment are likely to be beneficial for microplastic removal. Given the nationwide scope of our study, only the influents after grit removal and the final effluents were investigated herein. Further research is needed to determine the removal efficiency of each unit process in municipal WWTPs, including tertiary phosphorus removal processes.

**Implications of the Study.** Providing the largest scale of sampling locations of WWTPs reported, this study offers collective information on the occurrence of microplastics in municipal WWTPs in Korea. Although sampling was conducted once for each WWTP, observations with such a large sample size could enhance our understanding of microplastics in WWTPs. Pooled analyses of 50 WWTPs revealed that thermoplastics and synthetic fibers were dominant microplastic materials with a typical concentration


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ranging from 10 to 470 $L^{-1}$ in influents and 0.004 to 0.51 $L^{-1}$ in effluents. However, it should be noted that microplastic concentrations in individual WWTPs are not likely representative of the site due to inter- and intraday variations in WWTP microplastic loads. Microplastics appeared to be removed effectively during wastewater treatment, especially when advanced phosphorus removal treatment processes were included. However, a few important questions remain. Seasonal patterns or variations in the microplastics load to WWTPs might occur. For example, the observation in this study might not reflect the possible effects of heavy rainfall because almost 60% of the rainfall in Korea is concentrated in the summer season.[52] The performance of each unit process used in WWTPs that were not investigated in this study also warrants further research. If coagulation followed by gravitational settling is responsible for the removal of microplastics in WWTPs, mechanistic experimental and modeling studies would provide a better understanding of the fate of microplastics in WWTPs. Although the removal efficiencies in 31 WWTPs were mostly greater than 99%, it is unclear whether microplastics smaller than 100 $\mu$m are also removed as efficiently as larger microplastics. Considering that aquatic organisms are more likely affected by smaller microplastics[12,13,53−55] and that the abundance of smaller microplastics is likely higher in aquatic environments, further studies are required to extend our understanding of the removal efficiency of smaller microplastics in WWTPs. As only large facilities were investigated in this study, comparing large and small WWTPs with different wastewater treatment levels may also provide useful insights. The controversy regarding the size distribution of environmental microplastics is also worthy of further investigation. Although the power-law distribution was successful in describing microplastic particle size down to 100 $\mu$m, it was not applicable for smaller microplastics in this study. Therefore, our understanding of the rate of fragmentation and degradation of microplastics is too limited to achieve a consensus.

## ■ ASSOCIATED CONTENT

### Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.9b04929.

Sampling date and time for influent and effluent with the hydraulic retention time of each facility; percent recovery of standard microplastics of three different material types; comparison of percent recoveries of standard PE and PET microplastics using different pretreatment conditions; log conversion of serving population, volumetric flow rate, microplastic load in each WWTP per day; correlation table from multivariate analysis; estimation of removal efficiencies of microplastics in 31 WWTPs; sampling locations of 50 municipal wastewater treatment plants; microplastics found in negative blank tests; example photos of isolated microplastics under microscope and their FT-IR spectra; biplot of principal component analysis on the microplastic load to WWTP, serving population size and volumetric flow rate (PDF)

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### Notes

The authors declare no competing financial interest.

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