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Elimination of Microplastics at Different Stages in Wastewater Treatment Plants

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Abstract: Microplastic pollution has been widely studied as a global issue due to increased plastic usage and its effect on human and aquatic life. Microplastics originate from domestic and industrial activities. Wastewater treatment plants (WWTPs) play an important role in removing a significant amount of microplastics; otherwise, they end up in bioaccumulation. This study provides knowledge about the characteristics of microplastics, removal efficiency, and the correlation between wastewater quality and microplastic concentrations from three different WWTPs that differ in the type of biological and advanced wastewater treatment techniques that are believed to play an important role in microplastic removal. Microplastics of different types, such as fragments, fibers, and beads, are identified by using an optical microscope before and after the treatment process at each stage to assess the effect of different treatment techniques. In the screening unit and primary clarifier unit, WWTP-B shows the highest removal efficiency with 74.76% due to a distribution flow system installed before the primary clarifier to ensure a constant flow of wastewater. WWTP-B uses a bioreactor consisting of a filter plate coated with activated carbon (BSTS II) that can enhance the adaptability and adhesion of microorganisms and showed that 91.04% of the microplastic was removed. Furthermore, only WWTP-A and WWTP-B were applied coagulation, followed by the disc filter; they showed significant results in microplastic removal, compared to WWTP-C, which only used a disc filter. In conclusion, from all WWTP, WWTP-B shows good treatment series for removing microplastic in wastewater; however, WWTP-B showed a high rate of microplastic removal; unfortunately, large amounts of microplastics are still released into rivers.

Keywords: wastewater; microplastics; removal; treatment plant; coagulation; disc filter

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Citation: Kwon, H.J.; Hidayaturrahman, H.; Peera, S.G.; Lee, T.G. Elimination of Microplastics at Different Stages in Wastewater Treatment Plants. *Water* **2022**, *14*, 2404. https://doi.org/10.3390/w14152404

Academic Editors: Jiangchi Fei, Qian Zhou, Zhenxing Wang and Lizhi Xiong

Received: 21 June 2022
Accepted: 29 July 2022
Published: 3 August 2022

1. Introduction

Plastics are widely used in various sectors of life, especially as packaging materials. Plastic has many advantages ranging from being cheap, durable, lightweight, and easy to obtain; however, after being used, all plastics end up as waste that accumulates in nature, especially in the aquatic environment [1]. The presence of plastic in water bodies is one of */the main factors affecting water pollution because it is difficult to control. In general, plastic waste is classified depending on size, such as megaplastics (more than 500 mm), macroplastics (50–500 mm), mesoplastics (5–50 mm), microplastics (<5 mm), and nanoplastics (less than 0.3 mm) [2,3]. Microplastics can be grouped into primary and secondary microplastics [4,5]. Primary microplastics are intentionally produced in micro sizes for skincare products, textile fibers, and other industrial uses. Secondary microplastics are generated from the degradation or breakdown process of large plastic particles [6,7].

Microplastics derived from various point sources and non-point sources are eventually carried by rivers into water bodies, including lakes, seas, and oceans [5,8]. In the last few years, the accumulation of microplastics in marine ecosystems has gradually increased, and microplastics found in aquatic environments have long-lasting detrimental effects on marine organisms [9,10]. For example, Albano et al. [11] studied microplastic pollution in *Pelagia noctiluca* in the Straits of Messina from forty-nine specimens. There were 55 fibrous shapes of microplastics with a size between 0.09 and 9.4 mm; the authors hypothesized that microplastics originated from direct Mauve stinger accidental ingestion and biomagnification. They also proposed the *Pelagia noctiluca* as a water column bioindicator for microplastic pollution in the Mediterranean Sea. The effect of polystyrene microspheres on the *Artemia salina* showed that microplastics were found in this organism after 6 h, increased exposure to concentrations resulted in decreased growth of *Artemia salina* due to lack of food sources, and mortality occurred after 96 h and 120 h after microplastic exposure [12]. Another study showed the presence of microplastics in marine fish (*Sparus aurata*) and freshwater fish species (*Cyprinus carpio*) at different life stages from two fish farms located in Italy and Croatia. The result shows that microplastic concentrations in *Cyprinus carpio* were lower than *Sparus aurata,* with microfiber abundance of 0.25 and 1.3 items/individual, respectively. Low microplastic concentration in these fish species depends on the concentration of microplastics in water and the presence of microplastics in feed aquaculture [13]. Microplastics are ingested by aquatic organisms and transported through the water bodies via the food chain, eventually causing irreparable damage to human health [14,15].

A WWTP receives wastewater containing high amounts of microplastics every day. Microplastics entering WWTP come from domestic, industrial, and agricultural wastewater [1]. Various studies explored the behavior of microplastics during the treatment process, and the results obtained depend on the type of wastewater and technology used. Microplastics of large size and low density are easily removed by physical treatment through screening, grit removal, and sedimentation processes in the primary clarifier, whereas large-shaped microplastics, such as fibers and fragments, are eliminated by floatation. On the other hand, high-density microbeads generally sink to the bottom of the sedimentation tank due to gravitational force. The physical treatment process aims to remove not only large debris particles but also large microplastics in order to maintain the performance of the facility for the subsequent treatment process [4,16,17]. Furthermore, smaller microplastic particles are bound with microorganisms in the form of flocs and deposited in the biological treatment and secondary clarifier [18,19]. Previous research showed that 90–98% of microplastics in wastewater could be removed after going through several processing stages and using advanced technology such as disc filter, rapid sand filtration, dissolved air floatation, or ozonation [16,19–21]. Most prior studies were focused on the concentration of microplastics in the effluent and then concluded WWTP as a source in the aquatic environment [22,23]. Another potential threat of microplastics is recycled activated sludge. Sludge is widely used as fertilizer in agriculture or as a raw material for concrete and finally becomes a part of pollutants in terrestrial [24].

In this study, determination of the type of microplastic polymer will not be conducted; information regarding the removal of microplastics during the wastewater treatment process was focused on investigating the abundance and shape of microplastics in three full-scale WWTP and evaluating the removal rate of microplastics at different treatment stages. Finally, the number of microplastics in treated water was calculated as the emission of microplastic pollution from WWTP to the nearby river. This study provides insight into which stage of treatment is most useful for removing microplastics significantly.

2. Materials and Methods

2.1. Sampling Sites

This study was conducted at three full-scale WWTPs in Gyeongsangbuk-do Province, Republic of Korea. All WWTPs have a similar process from beginning to end, with some differences in the technology used (Table 1). Firstly, the physical process will treat wastewater entering the WWTP in bar screening, grit removal, and primary clarifier for separating solid organic matter from wastewater. Secondly, the biological process will continue to treat water from the previous process by using microorganisms that play a role in removing organic pollutants and small particles by binding or absorbing these pollutants into flocs. Next, flocs of microorganisms and any remaining organic sediment are settled in the secondary clarifier. Furthermore, the effluent of the secondary clarifier then flows into the coagulation process to remove total phosphorus, followed by a disc filter to improve water quality in the wastewater treatment process before discharge into the river.

Table 1. Characteristics of WWTPs.

WWTP	Source of Wastewater	Capacity (m^3/day)	Physical Treatment	Biological Treatment	Advanced Treatment
A	Industrial Domestic	80,000	Grit removal and rectangular sedimentation tank	TEC-BNR [1] and circular clarification	Coagulation and disc filter
B	Domestic	26,000	Grit removal and rectangular sedimentation tank	BSTS II [2] and circular clarification	Coagulation and disc filter
C	Domestic	13,000	Grit removal and rectangular sedimentation tank	IFAS [3] and rectangular sedimentation tank	Disc filter

[1] TEC-BNR: Taeyoung External Carbon for Biological Nutrient Removal. [2] BSTS-II: BioMecca Sewage & Wastewater Treatment System-II. [3] IFAS: Integrated Fixed film Activated Sludge.

2.2. Sample Collection

Three days were randomly selected during the Autumn (October to November 2019) to collect samples and reduce the effect of algae (Table S1) [25]. In order to study the removal efficiency of each treatment process in the WWTPs, the microplastics in raw wastewater and the effluent water of each technological step were sampled. As shown in Figure 1, water samples were collected at raw wastewater influent (W1), effluent water of primary clarifier (W2), effluent water of secondary clarifier (W3), effluent water of coagulation (W4), and disc filter outlet or final effluent water (W5). Two liters of water samples were collected at each treatment sampling point [16]. The grab sampling method was selected for sample collection, utilizing a custom-made sampler. The sampler was constructed of a stainless-steel container with rope as a tool to take a water sample from approximately 30 cm depth of wastewater and sludge stream. All samples were transferred to the laboratory and stored at 4 °C until further processing.

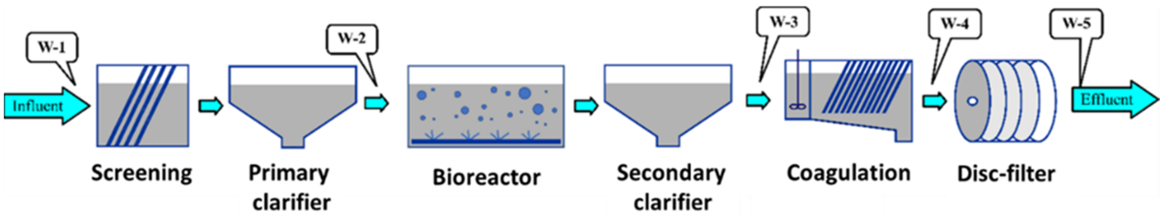

Figure 1. Sampling points and typical wastewater treatment process.

2.3. Extraction of Microplastic

Several steps were taken to reduce the incidence of microplastic contamination. Natural fabric cloth was worn underneath the clean white lab coat. All laboratory equipment was cleaned three times with distilled H_2O before use, and the work surface was wiped down with 70% ethanol (Duksan, Pure Chemical, Kyungkido, Korea). Wet digestion was applied to remove organic matter from wastewater (W1–W3) [18,26] using 30% hydrogen peroxide (H_2O_2) (Santoku Chemical Industry, Sendai, Japan) until the sample became clear. For the other samples (W4 and W5), no pre-treatment is needed because the water sample is relatively clear. After pre-treatment, all wastewater samples were filtered using a glass microfiber filter with a 1.2 µm pore size (Whatman GF/C) (GE Healthcare, Buckinghamshire, UK) and 30% hydrogen peroxide (H_2O_2) added to the remaining samples the filter paper to make sure no organic material in the sample water. Adding H_2O_2 in post-treatment will also change the physical properties of the natural fiber surface. Finally, filter papers were transferred to glass Petri-dishes for visual analysis using a digital microscope.

2.4. Inspection and Identification of Microplastics

Microplastics in filter papers were visually inspected with a digital microscope (Leica DM 750 combined with camera), 100× magnification, and light projected from above. The IMT iSolution Lite Version 22.1. is used to take photographs and measure microplastic dimensions. All suspected microplastics were counted and categorized by shape (fragment, fiber, and microbead). The following details should be noted when using an optical microscope for microplastics identification: (1) clear and uniform color distribution, (2) the object has no metallic luster, (3) no visible tissue or natural organic structures attached to it, (4) same dimensions throughout the entire length and thickness of the synthetic fiber, (5) there are twists (convolutions), striations, zigzags, or jagged and irregular widths along the natural fibers, and (6) by adjusting the angle of incidence of light and the brightness, the edges of the transparent microplastics will be seen [18,27,28].

2.5. Statistical Analysis

In situ wastewater quality data are collected from each process at the same point and day. Statistical analysis was used to find a correlation between water quality and the number of microplastic. Furthermore, this correlation was analyzed by multiple comparisons with a significance level set at 0.05. IBM SPSS Statistics version 24.0 was used for statistical analysis and for generating some graphs.

3. Results and Discussion

3.1. Overall Microplastic Removal

WWTP treats wastewater containing significant amounts of microplastics from various sources (domestic, industrial activities) before being discharged into nearby rivers. WWTP can remove a significant amount of microplastics, and it can be seen in Figure S1 that microplastic concentration decreased drastically from influent to effluent; however, microplastics are still released into aquatic ecosystems in a definite amount. All WWTP show a good result of microplastic removal (Figure 2A). Microplastics are always found in varying amounts, and this is related to the performance of each treatment process applied to all WWTPs. As shown in Figure S1, the range concentrations of microplastic in the effluent from three WWTPs were 91–175 MP/L, higher than the amount of microplastic in the Nakdong River (0.29–4.76 MP/L) as reported by [29].

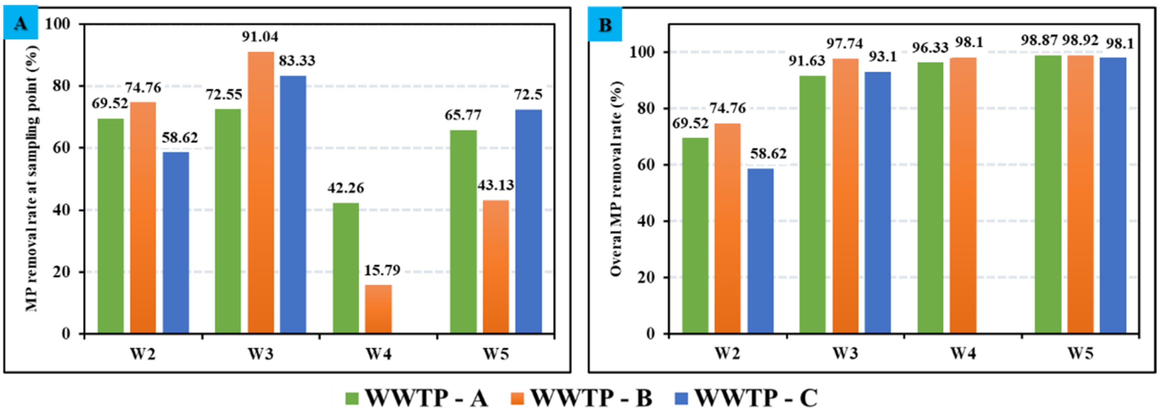

Figure 2. Removal of microplastic in different treatment processes. (**A**) Percentage of microplastics removed at samping point; (**B**) Overall percentage of microplastic removal.

Among related studies, the microplastic concentration varies from influent to effluent. Some studies showed high microplastics concentration in influent (2223–10,044 MP/L) and effluent (29–447 MP/L), meanwhile low microplastic concentration were also found in influent (1 MP/L) and effluent (0.00088 MP/L) [22,30]. These results are caused by different sampling techniques, identification methods, and technology applied in every WWTP [23]. For example, the filter paper pore size used to capture the microplastic from the sample is different and often varying [30–33]. Currently, no standard method for microplastic analysis exists from the beginning to the end; therefore, it is necessary to standardize the procedures of research on microplastic. Several factors such as differences in population density, local/regional development area, and catchment area also affect the microplastic emissions in an area. Although in general, there is a positive correlation between microplastic emission and those factors.

In terms of shape, microplastic samples were observed under the microscope and grouped into microbeads, fibers, and fragments (Figure 3). Based on Figure 4, the proportion of fragments was the highest in all WWTPs, with the lowest proportion of 53.63% only in the influent of WWTP-A. The proportion of fragments decreased gradually, with only 21.18% in the effluent of WWTP-A. The proportion of microbeads increased gradually in the subsequent treatment sections and reached 55.29% in the effluent of WWTP-A. During the entire processing sequence, fragments and fibers were removed in large quantities. As a result, the proportion of these two types of microplastics was decreased in the effluent portion while the proportion of microbeads was increased.

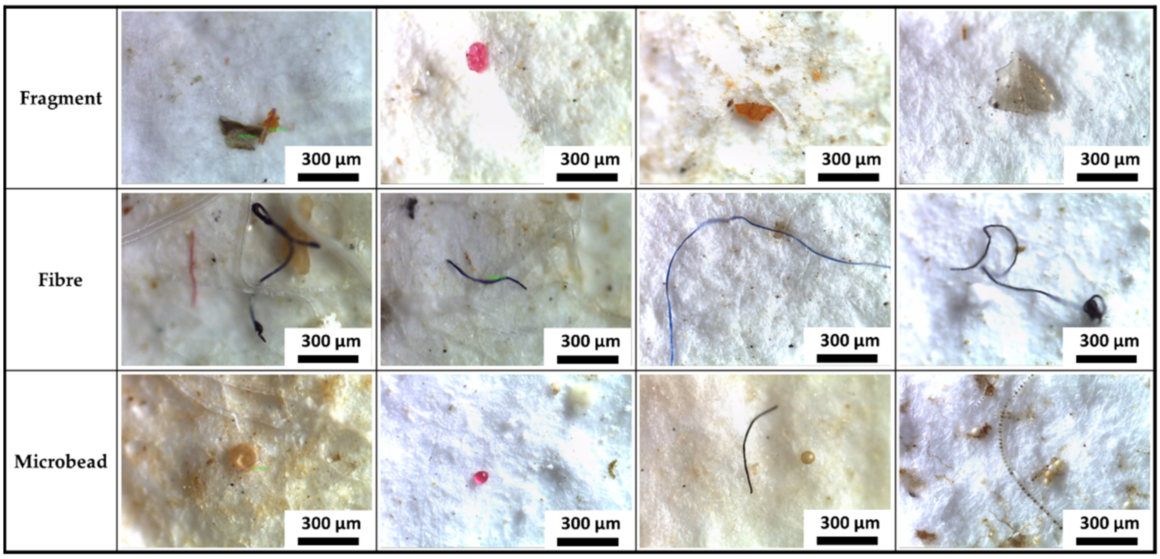

Figure 3. Result of microscope observation for microplastic particles.

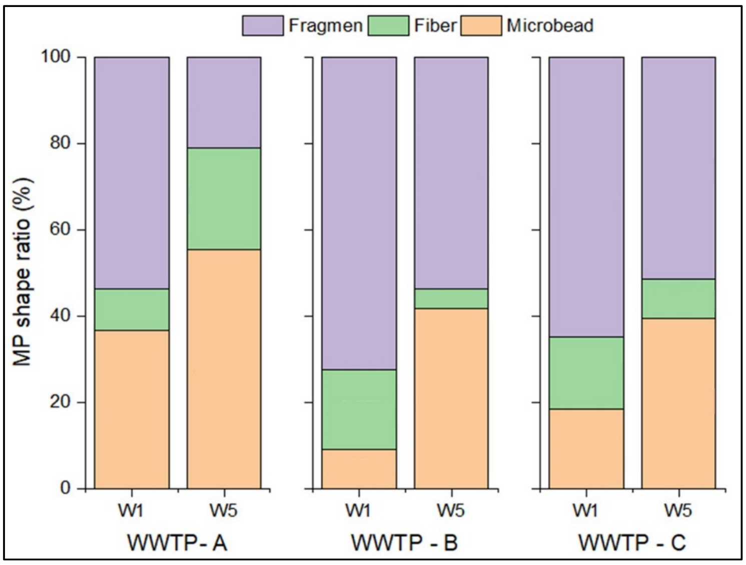

Figure 4. Comparison portion of microplastic shape between influent (W1) and effluent (W5).

Irregular fragments are mainly secondary microplastics, which usually come from the fragmentation of larger plastic objects, such as tires, bottles, and plastic bags [34,35]. Many types of facial cleansers, facial scrubs, or exfoliants often contain plastic particles, and they are considered to be one of the sources of microbeads [36]. Possible sources of fibers in wastewater came from the laundering of synthetic fabrics and shedding of textiles during the aging process for cloth, linen, carpets, etc. [37]. The washing of synthetic materials could release a large number of plastic fibers, so their presence in surface water may be due to the inflow of sewage, according to the reported studies [37]. Airborne contamination of open wastewater treatment plant systems must also be considered when assessing microplastic emission [38].

3.2. Microplastic Removal with Different Treatment Processes

3.2.1. Screening and Primary Clarifier

The main purpose of the screening process is to separate solid particles such as organic and inorganic materials from the wastewater and allow the remaining solids particle to sink to the bottom of the primary clarifier. In general, the design for this process is rectangular and has a hydraulic retention time of ~2 h. It is expected that the removal rate of the total suspended solids will reach 50–70% [39]. In addition, by skimming and sedimentation processes, microplastics are also expected to be removed from wastewater [22]. The flow of wastewater also affects the effectiveness of the primary clarifier. If the flow is too fast, it will be difficult for the solid particles to sink to the bottom of the system and vice versa [40].

During the screening and primary clarifier, the microplastic removal rate in WWTP-A was 69.52%, followed by WWTP-B was 74.76%, and WWTP-C was 58.62 % (Figure 2B). In terms of shape, this series of processes had a significant removal rate for fiber in the range of 64.60–79.59% and 21.88–68.42% of fragments and microbeads, respectively (Figure 5A). This study proved that most of the microplastic was removed during this process. WWTP-B shows good results for eliminating microplastics due to a distribution flow system installed before the primary clarifier and ensuring that the wastewater flow rate is the same for each primary clarifier unit. On the other hand, WWTP-A and WWTP-C did not use this flow system, and wastewater flow depends on the actual discharge.

The retention time is the time required for a certain amount of wastewater to pass through a sedimentation tank at a specific flow rate. Inside the tank, the microplastic particles in the wastewater take time to cross the sediment tank and settle at the bottom of the tank as it flows slowly through the tank. WWTP-B has the lowest retention time, followed by WWTP-A and C with 0.104 days, 0.107 days, and 0.256 days, respectively. According to our data, retention time has correlated with microplastic removal; the lowest retention time shows the highest microplastic removal. It means that when the retention time is low, wastewater will take a long time to pass through the tank so that microplastic attached with another particle has the opportunity to settle more at the bottom of the tank. This parameter also controls the performance of the primary clarifier.

Microplastics in wastewater are generally suspended individually or attached to larger particles such as paper, wood branches, or larger plastic particles. Most of the microplastics adsorbed to these larger particles will be easily removed during the screening process [41]. The microplastics that settle during this process are microplastics attached to the sand particles so that they settle very easily [18]. Some microplastics float on the surface of the wastewater because they have a lower density, which can then be easily removed by skimming.

A similar study showed that the concentration of microplastics in wastewater is reduced significantly through the screening, skimming, and settling process [16]. The results show a consistent trend of microplastic shape removal rate with previous studies, in which fiber was removed due to being easily entrapped in solid floc particles during screening and settling on a primary clarifier [18]. Hongprasith et al. [42] stated that microplastics were suspended together with other fine particles to form suspended solids or were mutually adsorbed between the two.

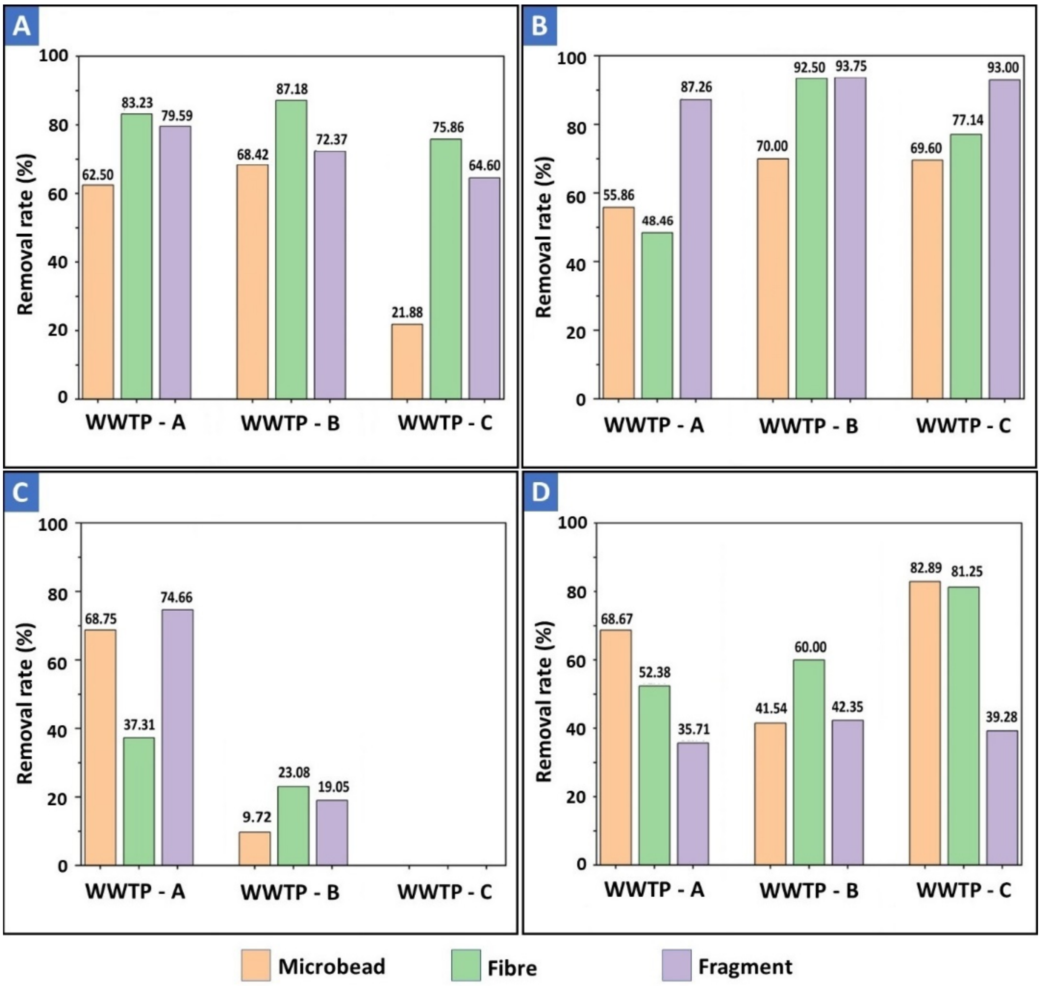

Figure 5. Removal efficiency of microplastic shapes at each treatment process. (**A**) Microplastic removal rate of screening and primary clarifier; (**B**) Microplastic removal rate of bioreactor and secondary clarifier; (**C**) Microplastic removal rate of coagulation; (**D**) Microplastic removal rate of disc-filter.

3.2.2. Bioreactor and Secondary Clarifier

Bioreactor treatment aims to destroy the organic material contained in the wastewater; the resulting suspended particles were deposited in the secondary clarifier. The activated sludge used in this process can indirectly reduce the number of microplastics in wastewater. The number of microplastics further declined during biological treatment and secondary sedimentation. Microplastic removal reached 72.55–91.04% after bioreactor treatment (Figure 2A,B). According to the shape distribution shown in Figure 5B, the fragment was the most dominant fraction removed in samples from all WWTPs, contributing to about 87.26–93.75%. Sheets were also a dominant fraction, ranging from 55.86 to 70.00% in WWTP-B and WWTP-C.

Same as the previous process, WWTP-B showed a more dominant microplastic removal rate compared to WWTP A and WWTP C. Based on Table 1, each WWTP uses different technologies. BSTS II is a biological treatment technology that applies the microbial control tank in the last part of the bioreactor. This tank will enhance the adaptability of microorganisms to the wastewater and promote activated sludge activity in the bioreactor. There is a filter plate in the tank coated with activated carbon, which is useful for increasing the adhesion of microorganisms so that the bioreactor works efficiently. This technology can remove organic matter, nutrients, and microplastic from wastewater. Coupled with the clarifier, increasing microplastic removal can be achieved. The CNR technology applied in WWTP-C has relatively the same process as WWTP-B by using a filter medium in the aerobic tank to stabilize microorganisms. The low microplastic removal rate is due to the design of the secondary sedimentation system, which is made in a rectangular shape without any skimming process on the surface of the tank/pond. Secondary processing at WWTP-A has the lowest microplastic removal rate compared to other WWTPs. The main factor is the TEC-BNR technology used. The technology modifies conventional bioreactor technology by combining activated sludge with fermentation solutions from food waste. By adding the fermentation solution, biological degradation will increase the performance of the bioreactor to remove organic matter and nutrients; however, during the fermentation process, the microplastic from food packages still exists even in a small portion. It will increase the number of microplastics in the wastewater and decrease microplastic removal efficiency in the bioreactor. The trend of microplastic removal rate in the secondary treatment process has the same trend as BOD, COD, SS, and T-P removal at each WWTP (Figure S2).

Microplastics were removed together with dissolved organic matter through the activity of microorganisms and sedimentation. Hongprasith et al. [42] showed that activated sludge greatly contributes to the microplastic removal process. The hydrophobic characteristics of microplastics also help accelerate the binding process of microplastics with organisms or sludge in biological reactors. Fragments might be trapped into sludge flocs by the ingestion process of microorganisms as activated sludge [43].

3.2.3. Coagulation

Coagulation was designed in all WWTPs to treat total phosphorus that cannot be completely removed from previous treatment processes; however, this process can also remove microplastics in wastewater efficiently. The performance of microplastic removal was investigated with different dosages of Poly Aluminum Chloride (PAC) as a coagulant at WWTP-A and WWTP-B for ± 72 mg/L and ± 36 mg/L, respectively. Only WWTP-C does not use coagulation to remove total phosphorus. As shown in Figure 2B, the removal efficiency of the coagulation process in WWTP-A was 42.26% compared to WWTP-B, with microplastic removal efficiency being 15.79%. According to this study, the low removal rate in WWTP-B is related to the lack of interaction between the coagulant and microplastic to generate flocs. Although the dose of coagulant used in WWTP-A shows more effectiveness in removing microplastics, WWTP-B cannot apply this dose directly to their plant, which requires further research.

Figure 5C shows the microplastic removal rate during coagulation. The result indicates that WWTP-A can remove 68.75% and 74.66% of microbeads and fragments, respectively. In WWTP-B, the highest removal rate was fiber at 23.08%. These results indicate that all shapes of microplastic will be agglomerated into floc particles and settled down in a sedimentation tank. Another factor regarded as important for microplastic removal in coagulation is the surface of the microplastic. The efficiency of coagulation increased when the plastic surface was weathered, especially fragments [44].

Similar studies of coagulation experiments using microbeads/microsphere showed high removal efficiency >90% [45,46]. Ma et al. [47] reported that the microplastic removal efficiency was 36.89% for 15 mmol/L (calculated as 405 mg/L) with Al-based as a coagulant. In contradiction, Rajala et al. [46] reported 98.2% microplastic removal with Polyaluminum Chloride at a metal dosage of 1.4 mmol/L. Wang et al. [48] also reported that during coagulation combined with sedimentation, the microplastic removal rate was 40.5–54.5% with high Al-based salt concentration, and 50.7–60.6% fibers were removed through this process. The different results are mainly caused by the different dosages of coagulant, microplastic type, and wastewater characterize used during the experiment.

3.2.4. Disc Filter

The concentration of microplastics in the effluent of the disc filter represents the number of microplastics released into the river. The removal efficiency of the microplastics by the disc filter was 43.13–72.50% (Figure 2B), and total microplastic removal for WWTP-A, WWTP-B, and WWTP-C was 98.87%, 98.92%, and 98.10%, respectively. In all WWTP, fiber was the most efficient removal process of microplastics during the disc filter process (52.38–81.25%), followed by microbead (43.67–62.89%) and fragment (35.71–42.35%) (Figure 5D). Generally, as the size of microplastics determine whether they can pass through the filter, the disc filter should have retained microplastics whose size is more than the pore size of the filter mesh. In addition, the shape of the microplastics also needs to be considered. The result indicates that the fragment size is larger than the other shapes, so the disc filter process cannot remove the fragment efficiently compared to other shapes. As a result, the proportion of fiber and microbead shape was decreased in the effluent compared to the previous treatment.

Changing the flow rate and pressure during the disc filter process will influence the microplastic removal rate. In this case, the pressure in all WWTPs is the same, but WWTP-B uses a larger flow rate than WWTP-A and C. The results showed that the microplastic removal rate in WWTP-B was the lowest (Figure 2B) due to the larger flow rate and pressure that can damage the filter quicker than normal. The larger flow rate can reduce microplastic removal efficiency because microplastic sizes, which are relatively larger than the fiber's pores, will be forced out. Although the disc filter performance can remove microplastic in all WWTPs, microplastics are still present in the treated water in all WWTPs. This is probably due to the maintenance of the disc filter process by activating high-pressure backwash and some microplastics passing through the system.

Previous studies also proved that the microplastic removal rate after disc filter use was 40–98.5% [38] and advanced filtration in Germany WWTP was 93–95% [20]. Talvitie et al. [38] reported that fibers were removed efficiently during the disc filter process and contributed 20–100% after the treatment of total microplastics. Once this process is in progress, fiber and other shapes of microplastics can pass through the disc filter longitudinally; even when the pore of the membrane filter is 0.08 microns in size [49]. This condition shows that the movement of microplastics can occur at WWTPs applied to membrane technology in smaller pore sizes [38,50]. As an advanced technology, disc filters need to be developed to become a promising technology for removing microplastics in wastewater [38,51]. The microplastic removal rate will be affected by various technologies applied to tertiary treatments [38,52]. Our study proved that coagulation and disc filter treatments increase the microplastic removal rate for all WWTP.

3.3. Correlation with Wastewater Quality Data

Wastewater quality parameters are monitored regularly at all stages of treatment, such as biochemical oxygen demand (BOD), chemical oxygen demand (COD), suspended solids (SS), total nitrogen (T-N), and total phosphorus (T-P). Wastewater quality parameters decrease after passing through in all treatment processes with different efficiency removal from three WWTP (Table S2). From all WWTP, the microplastic concentration has a positive correlation with all water quality parameters (Figure 6). Suspended solid shows a high positive correlation with the microplastic removal at all treatment stages, followed by COD and BOD. Suspended solids in wastewater contain microplastics and other particulate material, and together, they can be removed through all series of treatment processes. COD and BOD are indicators of organic content that are indirectly related to the number of microorganisms and microplastics in water bodies. When the wastewater treatment unit removes organic elements, at the same time, microplastics are degraded along with microorganisms, for example, in bioreactors or deposited together in the filtration process. These results are in line with research conducted by Kataoka et al. [53], which showed that biochemical oxygen demand as wastewater quality has a positive relationship with microplastic removal and suspended solid particles in the wastewater. Peller et al. [54]

showed that even though microplastic is a part of total suspended solids (TSS), there is no obvious correlation between microplastic and TSS concentration. Additional data should be collected and analyzed from different weather conditions to justify the significant relationship between wastewater quality and microplastic concentration.

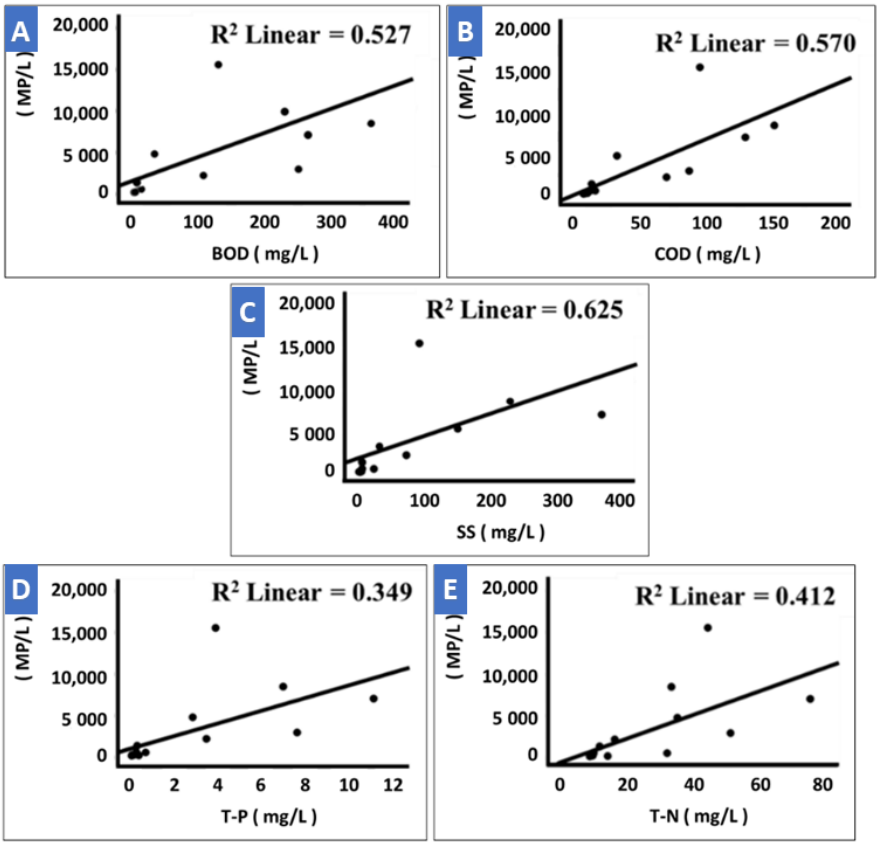

Figure 6. Correlation of microplastic concentrations with five wastewater quality parameters. 3.4. Microplastic pollutant load. (**A**) Microplastic concentration relationship with biological organic demand (BOD); (**B**) Microplastic concentration relationship with chemical oxygen demand (COD); (**C**) Microplastic concentration relationship with suspended solids (SS); (**D**) Microplastic concentration relationship with total phosphorus (T-P); (**E**) Microplastic concentration relationship with total nitrogen (T-N).

This study showed different total numbers of microplastics released from all WWTP. Although microplastic concentration in treated water is low, considering the large amount of wastewater discharged daily, we found that the number of microplastics also released along with treated water is very high. Based on this fact, this is in accordance with [4], who stated that WWTP could be considered a point source for releasing microplastics into the aquatic environment. Considering that the WWTP–C discharges at the lowest flow rate with an average flow of 8845 m^3/day, approximately 1.17 billion microplastics are emitted to the nearby river daily, reaching up to the Nakdong River (Table 2). The Nakdong River is one of the most important rivers in South Korea. It plays a significant role in providing a water source to metropolitan cities such as Daegu and Busan. It is also a habitat for native and migratory fish.

Table 2. Average microplastic loading amount.

WWTP	Average Number of Microplastic in the Treated Water (MP/L)	Average Flow Rate (m^3/day)	Average Microplastic Released to the Nearby River (billion/day)
A	172.5	52,000	8.97
B	90	22,925	2.09
C	32	8845	1.17

A high number of microplastics in treated water released may threaten the aquatic ecosystem. El Hadri et al., for instance, found that fibers were more toxic to the *Ceriodaphnia dubia* compared to other shapes [8]. Microplastics can be dangerous if toxic materials are adsorbed onto the microplastic surface and are eaten by aquatic organisms [55]. Furthermore, aquatic organisms can enter the food chain [56]. Further research is needed to assess the potential risk of the microplastic released by WWTP to the aquatic ecosystems of the Nakdong River.

4. Conclusions

The characteristics and removal of microplastics were studied from three full-scale WWTPs in Gyeonsangbuk-do, South Korea. The results showed the efficiency of the WWTPs in removing microplastics was high, with a removal rate of >98% from influent to final effluent. The main proportion of microplastic in all WWTPs were microbeads and fragments. Microplastic removal mainly occurs in screening, biological treatment, and sedimentation. Coagulation followed by disc filter showed a better microplastic removal (WWTP-A and WWTP-B) than only applied disc filter (WWTP-C). Despite the high efficiency of microplastic removal at each WWTP, many particles escape through the discharge of treated wastewater. This research proves that microplastics are still found in the WWTP effluent. Creating new technology and modifying the current WWTP system for removing microplastics from wastewater is needed in order to reduce the release of microplastics into the aquatic environment. In addition, it can be used as information input for environmental authorities in South Korea to improve regulations on plastic waste and plastic pollution.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/w14152404/s1, Figure S1: Average number of microplastics in different treatment unit; Figure S2. Comparisons removal rate of microplastic and water quality in Bioreactor and second-ary clarifier. (A) Microplastic removal rate of bioreactor and secondary clarifier; (B) Removal rate of biological organic demand (BOD); (C) Removal rate of chemical oxygen demand (COD); (D) Removal rate of suspended solids (SS); (E) Removal rate of total nitrogen (T-N); (F) Removal rate of total phosphorus (T-P); Table S1: Sampling locations and date; Table S2: Percentages of removal efficiency at each treatment stage for organic material (BOD and COD), suspended solids (SS), total nitrogen (T-N), and total phosphorus (T-P); Table S3: Overall percentages removal rate for organic material (BOD and COD), suspended solids (SS), total nitrogen (T-N), and total phosphorus (T-P).

Author Contributions: Conceptualization, methodology and writing—original draft preparation: H.J.K.; writing and validation: H.H.; formal analysis: H.H.; review and editing, S.G.P.; supervision, project administration, T.G.L.; funding acquisition, T.G.L.; All authors have read and agreed to the published version of the manuscript.

Funding: This research was supported by the Bisa Research Grant of Keimyung University in 2019 (20190682).

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Conflicts of Interest: The authors declare no conflict of interest.

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