JEE Journal of Ecological Engineering

Journal of Ecological Engineering 2024, 25(9), 303–315
https://doi.org/10.12911/22998993/191506
ISSN 2299–8993, License CC-BY 4.0

Received: 2024.06.28
Accepted: 2024.07.23
Published: 2024.08.01

Microplastics Identification in Plastic Recycling Facility – Removal Efficiencies of the Treatment Plants and Its Potential Release to the Environment

Winda Islamiyah Umarie[1], Arseto Yekti Bagastyo[1*]

[1] Department of Environmental Engineering, Faculty of Civil, Environmental, and Geo Engineering, Institut Teknologi Sepuluh Nopember, Kampus ITS Sukolilo, Surabaya 60111, Indonesia

* Corresponding author's e-mail: bagastyo@enviro.its.ac.id

ABSTRACT

Plastic recycling facilities (PRF) are one of microplastic sources that may release into the environment. This study aims to identify the abundance and characteristics of microplastics potentially released from a PRF in Indonesia. Analysis of raw materials in the influent of the wastewater treatment plant (WWTP) showed a microplastic abundance of 485 particles/L. The dominant type of microplastic was the 2.5 µm – 5 mm size of fragmented high-density polyethylene (HDPE). In the effluent of floating clarifier 3, the microplastic abundance detected was 98 particles/L, with low-density polyethylene (LDPE) as the dominant fragmented plastic ranging from 1.2–2.5 µm. Meanwhile, in the WWTP sludge, microplastics were identified with an abundance of 364.81 particles/kg. The microplastics found in the sludge were predominantly in the fragment form, composed of HDPE, with sizes ranging from 0.2–2.5 µm. This information is crucial for understanding the extent to which PRFs contribute to microplastic pollution in the environment. These findings emphasize the importance of implementing more effective wastewater management technologies in PRFs to reduce the release of microplastics into the environment.

Keywords: microplastics, plastic recycling facilities, polyethylene, wastewater treatment plant.

INTRODUCTION

Microplastics are the result of degradation and weathering of larger plastic objects (GESAMP, 2015). In Indonesia, the Surabaya River, which serves as the raw water source for the city of Surabaya, has been contaminated by microplastics. The abundance of microplastics in the Surabaya River ranges from 1.47 to 43.11 particles/m^3. The size of the microplastics varies between 1001 to 5000 µm. The most common form of microplastic is a film, accounting for a dominant percentage of 45.8 to 92.9%, with the main polymer type being low-density polyethylene (LDPE), comprising 44–68% of the total microplastics (Lestari et al., 2020). Due to the abundance of microplastics in the Surabaya River, the water supply in Surabaya is contaminated with microplastics, with an abundance of about 26.8 to 35 particles/L (Radityaningrum et al., 2021).

Plastic recycling facilities (PRF) aim to reduce the amount of plastic in the environment by recycling it. However, during the recycling process, PRFs generate wastewater containing microplastics in addition to physical, chemical, and biological contaminants. Typically, the recycled used plastic bottles are thoroughly cleaned for decontamination in the initial step. The cleaned bottles then undergo further processing, such as primary separation, shredding, washing with cold or hot water, drying, and thermal drying. The resulting flakes are then washed again, dried with heat, and packaged for sale as raw materials for other products. Some of the water used for cleaning is renewed during the treatment process, although most is recycled in the washing line. The discharged wastewater is treated through a series of physical and biological processes (Guo et al., 2022).

According to a study by Brown et al. (2023), wastewater effluent from washing activity in PRF

contains microplastics ranging from 5.97×10^6 to 1.12×10^8 particle/m³. Microplastics smaller than 5 µm can be released into the environment, while those larger than 5 µm are usually retained in wastewater treatment installations (Brown et al., 2023). Another study found that microplastics in wastewater from recycling facilities ranged in size from 0.1 to 5 mm, with concentrations of 23.43 ± 1.04 mg/L to 1836.37 ± 31.73 mg/L. The concentration of microplastics in wastewater effluent decreased to 8.13 ± 0.42 to 83.83 ± 0.93 mg/L, while in sludge it ranged from 52.166 ± 2858 to 68.866 ± 2500 µg/g. The size of microplastics detected in the effluent ranged from < 1 mm to 2 mm (Guo et al., 2022). Research by Suzuki et al. (2022) indicated that microplastics in recycling facilities are predominantly in fragment form, with percentages ranging from 81 to 99%, although small amounts of aggregates, fibers, foam, and film were also found. The average size of microplastics ranged from 108 to 4613 µm. Generally, the colors of the microplastics found were white, gray, black, transparent, and green.

Microplastic removal technologies in wastewater treatment plants involve physical, chemical, thermal, and biological processes. Physical and chemical processes enhance the removal of microplastics, while biological and thermal processes help to degrade them (Reddy and Nair, 2022). The pre-treatment stage of WWTP is designed to remove large solids and sand to protect the treatment plant. Most microplastics in wastewater can be efficiently removed during the pre-treatment and primary treatment stages. In conventional primary treatment, about 65% of microplastics can be removed (Burns and Boxall, 2018). Studies have shown that initial treatments (sedimentation, filtration, flotation) can remove between 35% and 59% of microplastics, while primary treatments (adsorption, coagulation, ozonation) can remove between 50% and 98% of microplastics (Sun et al., 2019). In the secondary stage, biological processes in WWTP can remove 98.3% and 99.4% of microplastics (Lares et al., 2018). Biological treatment in WWTPs typically targets organic pollutants in wastewater and is not specifically designed to remove microplastics.

This study aims to assess the potential release of microplastics from PRF activities. The study focuses on the abundance, types, shapes, and sizes of microplastics that could potentially be released into the environment. In addition, a polymer identification was conducted to confirm the treatment plant efficiencies in removing microplastics.

METHODOLOGY

The sampling was conducted at the WWTP of a PRF located in Banten, Indonesia. This PRF processes plastic waste, specifically plastic bottles from mineral water bottles, with a wastewater discharge of 24 m³ per day. The incoming plastic waste primarily consists of mineral water bottle components, including bottle caps, bottle necks, and labels. Samples were taken from several critical points to evaluate the efficiency of microplastic removal across various treatment units. The sampling points are illustrated in Figure 1. The first wastewater sampling point was at the Pump

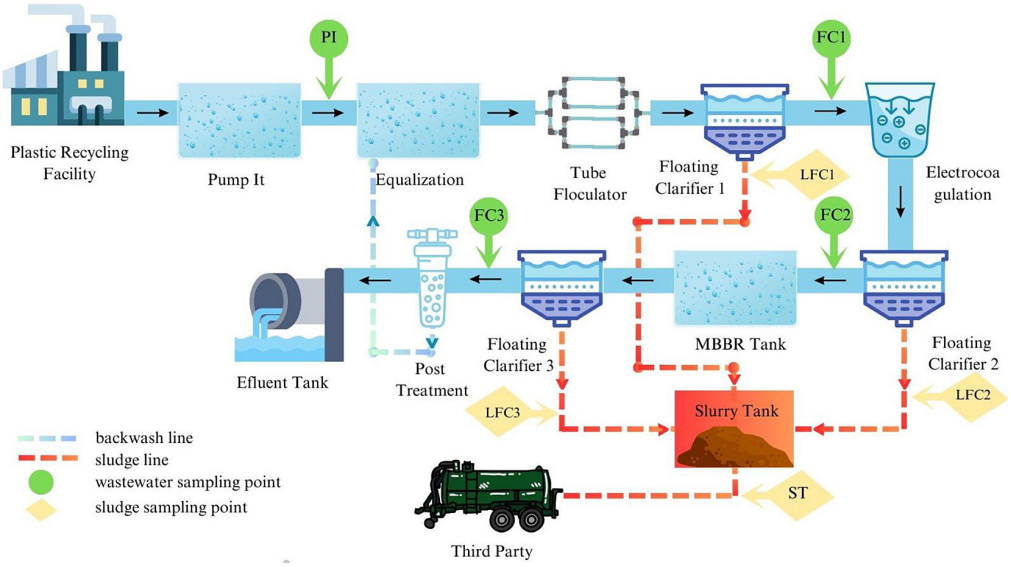

Figure 1. Sampling point at wastewater treatment in PRF

unit (Pump It/PI), marking the beginning of the treatment process. The first sludge sampling point was at the effluent of Floating Clarifier 1 (LFC1). In the Electrocoagulation unit, wastewater samples were taken from the effluents of Floating Clarifier 1 (FC1) and Floating Clarifier 2 (FC2). Electrocoagulation sludge samples were collected at the effluent of Floating Clarifier 2 (LFC2). In the Moving Bed Biofilm Reactor (MBBR) unit, wastewater sampling points were at the effluents of FC2 and Floating Clarifier 3 (FC3). The final sludge sample was collected from the effluent of the slurry tank (ST), where sludge from various treatment units is collected before further processing by a third party. A total of 4 wastewater samples and 4 sludge samples were analyzed.

Sampling was conducted in January-March 2024 using composite sampling methods. Samples were collected once during the operational condition of the WWTP and once during the non-operational condition. The composite sampling method involved collecting samples at three different times (morning, afternoon, and evening) for each condition. A 5 L sample was taken at each sampling time, both for wastewater and sludge, resulting in a total collected sample volume of 15 L for each sampling point in each condition. These composite samples were then homogenized to mix all the samples thoroughly. From the 15 L of homogenized collected samples, 5 L were taken as the final sample for further analysis. Operational conditions refer to an active PRF condition in processing raw materials for recycling, generating abundant wastewater influent in the treatment system. Non-operational conditions refer to when the inactive PRF condition at the stage of no process of raw materials occurred, thus no wastewater influent coming into the treatment system. In this case, only the existing wastewater within the system is processed.

Microplastic extraction

This study involved the extraction of microplastics from water and sludge samples, followed by microscopic observation to characterize the microplastics. The extraction method was modified from the National Oceanic and Atmospheric Administration (NOAA) method (Masura, 2015). A 1 L of water sample was subjected to wet peroxide oxidation (WPO) by adding 30% H_2O_2 and $FeSO_4$ and heated using a magnetic stirrer. After heating, density separation was performed by adding 5 M NaCl to isolate the floating microplastics. The samples were then filtered using polytetrafluoroethylene (PTFE) membranes in stages (2.5 µm, 1.2 µm, and 0.2 µm) and analyzed using a microscope. For sludge samples, the initial step involved preparation, including determining the dry weight and disaggregation with potassium metaphosphate. The sludge samples then underwent WPO and density separation, followed by filtration and microscopic analysis like the water samples.

Analytical methods

Microscopic observations were conducted using an XSZ 107BN Yazumi Binocular microscope with 40 to 100 times magnification to identify microplastics based on color, shape, and size. Following microscopic observation, microplastics were analyzed using a scanning electron microscope (SEM) with a Thermo Scientific Phenom ProX instrument to identify the morphology of microplastics at high resolution. The type of microplastics was determined using fourier-transform infrared (FTIR) spectroscopy, Agilent Cary 630, which provides information on the various chemical functional groups present in the material, including plastics. The FTIR results allow for the identification of specific types of plastics based on their chemical functional group patterns by comparing sample spectra with standard databases.

RESULT AND DISCUSSION

Abundance of microplastics

The abundance of microplastics was assessed at eight sampling points, encompassing both water and sludge samples. Table 1 presents the quantitative results of microplastic abundance across various measurement points in the wastewater treatment process. In the wastewater samples under operational conditions, the highest microplastic abundance was observed at the Pump It unit (485 particles/L), followed by a progressive decrease in effluents FC1 (221 particles/L) and FC2 (64 particles/L). However, a slight increase was noted at effluent FC3 (98 particles/L). During non-operational conditions, a similar trend was observed, albeit with generally lower abundances (65, 31, 45, and 92 particles/L for PI, FC1, FC2,

Table 1. Abundance of microplastic at WWTP in PRF

Condition	Wastewater			
	PI particles/L	FC1 particles/L	FC2 particles/L	FC3 particles/L
Operational	485	221	64	98
Non-operational	65	31	45	92
Condition	Sludge			
	FC1 particles/kg	FC2 particles/kg	FC3 particles/kg	ST particles/kg
Operational	826.09	3055	150.000	364.81
Non-operational	136.99	15.714	45.000	4000

Note: FC = floating clarifier

and FC3, respectively). The observed fluctuations in microplastic abundance throughout the treatment stages in our study, particularly the increase at FC3, suggest that the current wastewater treatment processes are not optimized for consistent microplastic removal.

These findings are consistent with recent literature on microplastic prevalence in various water treatment contexts. A study by Guo et al. (2022) examined three PET bottle recycling facilities, revealing microplastic abundances in effluent sewage treatment ranging from 96±11 to 201±17 particles/L. Furthermore, their analysis of sludge samples showed microplastic abundances between 773±21 and 1450±66 particles/kg. These results highlight the significant presence of microplastics in both water and sludge from facilities associated with plastic recycling processes. The variability in microplastic abundance across different facilities can be attributed to several factors, including the specific treatment methods used, the characteristics of the influent water, and the properties of the microplastics themselves. These parameters collectively influence the retention and removal efficiency of microplastics within the treatment processes.

The abundance of microplastics in sludge samples varied significantly across different sampling points, reflecting the complex dynamics of microplastic distribution in wastewater treatment processes. Under operational conditions, microplastic abundances in sludge were observed to be 826.09 particles/kg at LFC1, 3.055 particles/kg at LFC2, and 150,000 particles/kg at LFC3. The slurry tank, which serves as the collection point for sludge from various units, showed a notably lower concentration of 364.81 particles/kg.

During non-operational conditions, a different pattern emerged. Microplastic abundances were lower at LFC1 (136.99 particles/kg) and LFC3 (45.000 particles/kg) compared to operational conditions, while higher concentrations were observed at LFC2 (15.714 particles/kg) and in the slurry tank (4.000 particles/kg). This variability between operational and non-operational states suggests that factors such as sludge retention time, treatment processes, and sampling timing may significantly influence microplastic abundance.

The lower microplastic abundance in the slurry tank, particularly during operational conditions, may be attributed to periodic sludge removal, which could potentially mask the actual microplastic levels at the time of sampling. It is noteworthy that the wastewater treatment plant under study does not incorporate specific sludge treatment processes. Instead, the collected sludge is transferred to third-party entities for incineration. This practice, while preventing the direct release of microplastics into the environment through land application, raises new considerations regarding the fate of microplastics during the incineration process.

Shapes of microplastics

Analysis of microplastic composition in wastewater and sludge samples shown in Figure 2 indicates a predominance of fragments as the primary identified form. In wastewater samples, fragments constituted over 54.84% of the total microplastics detected. During operational conditions, the percentage of fragments varied across sampling points, i.e., 89.07%, 95.93%, 64.06% and 94.90% in the effluent of PI, FC1, FC2, and FC3, respectively. Under non-operational conditions, fragment percentages were 81.54%, 54.84%, 55.56% and 84.78% in PI, FC1, FC2, and FC3, respectively.

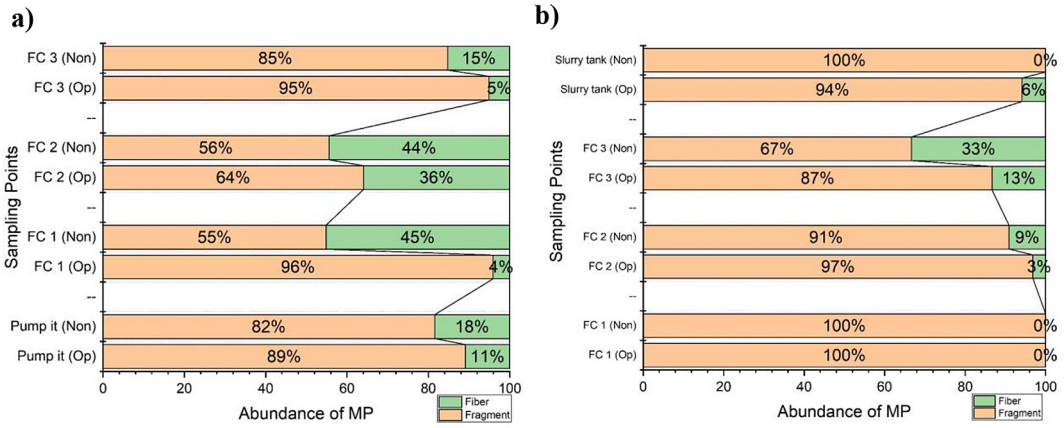

Figure 2. Microplastic shapes identified in: a) wastewater, b) sludge

Sludge samples also exhibited a dominance of fragments, with percentages exceeding 87% during operational conditions. In FC1 and FC2 effluent sludge, fragments reached 100%, while in FC3 effluent sludge and the slurry tank, percentages were 87% and 94.12%, respectively. Under non-operational conditions, fragments in FC1 and slurry tank effluents attained 100%, while in FC2 and FC3 effluents, they constituted 90.91% and 66.67%, respectively.

These findings align with recent studies, such as those conducted by Çolakoğlu & Uyanık (2024) and Suzuki et al. (2022) where fragment microplastics were identified as the most prevalent form, with percentages ranging from 76% to 99%. Besides fragments, other microplastic shapes like fibers and films were also detected in smaller percentages. The predominance of fragments in these samples indicates that plastic bottle recycling activities are a primary source of the identified microplastics. Fragment microplastics typically originate from bottlenecks and labels, while fibers are characterized by their elongated shape with smaller diameters. The consistency of these findings across various studies strengthens the hypothesis that plastic bottle recycling processes contribute significantly to microplastic production, particularly in fragment form.

Size of microplastics

Analysis of microplastic distribution and abundance in water treatment facilities in Figure 3 shows significant variations based on particle size, sampling location, and operational conditions. This study focuses on three microplastic size ranges: 2.5–5 mm, 1.2–2.5 µm, and 0.2–1.2 µm, which were analyzed in water and sludge samples under both operational and non-operational conditions.

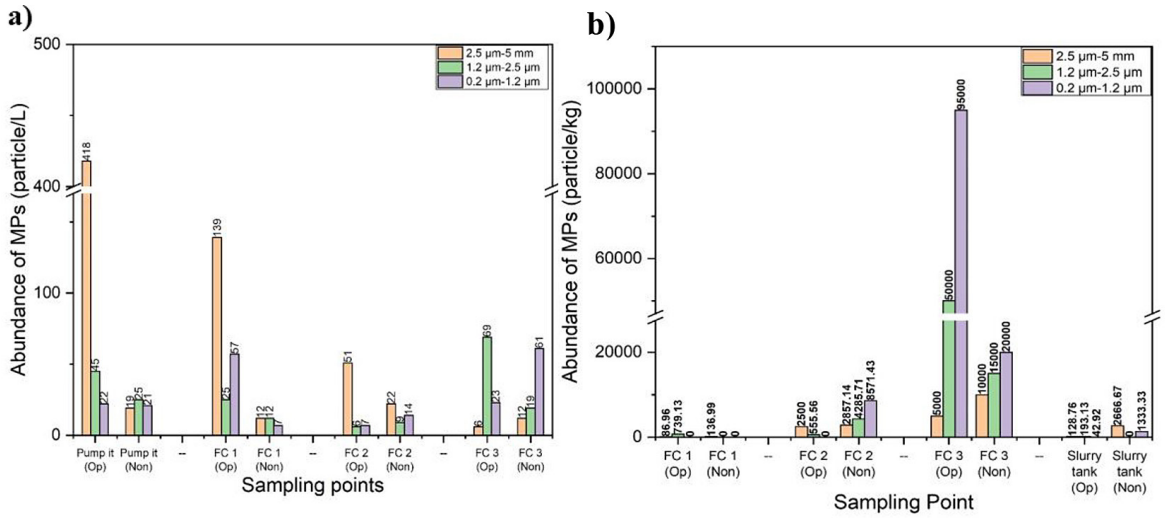

Figure 3. Microplastic sizes identified in: a) wastewater, b) sludge

Under operational conditions, the abundance of microplastics in water for the 2.5–5 mm size range peaked at PI with 418 particles/L, then gradually decreased in the effluents of FC1, FC2, and FC3 to 139, 51, and 6 particles/L, respectively. This declining pattern demonstrates the system's effectiveness in removing larger microplastics. For the 1.2–2.5 µm range, microplastic abundance varied from 45 particles/L at PI, then decreased at FC1 and FC2, however, increased to 69 particles/L at FC3. This increase at FC3 may be due to the fragmentation of larger particles or limitations in the treatment system to remove particles in this size range. Meanwhile, for the smallest range of 0.2–1.2 µm, the abundance fluctuated from 22 particles/L at PI, increasing at FC1, decreasing at FC2, and rising again at FC3, reflecting the complexity in handling very small microplastics. Likewise, non-operational conditions showed similar patterns with lower abundances compared to operational conditions across all size ranges. This may be due to reduced water flow and microplastic input during non-operational periods.

Analysis of sludge samples revealed different dynamics. In the 2.5–5 mm range, there was an increase in microplastic abundance from the effluent of FC1 to FC3, with higher values during non-operational conditions. Similar patterns were observed for the 1.2–2.5 µm range, with increased abundance in the effluents of FC2 and FC3 under both operational and non-operational conditions. Most notably, the abundance of microplastics in the 0.2–1.2 range was very high in the FC3 sludge effluent, indicating a significant accumulation of very small particles in the sludge. These findings suggest that sludge can act as a sink for microplastics, especially for smaller particles that may escape the water treatment process.

Comparing these results with studies conducted by Guo et al. (2020), Suzuki et al. (2022), and Çolakoğlu and Uyanık, (2024) at plastic recycling facilities, which identified microplastics with a minimum size of 75–100 µm, this research demonstrates the ability to detect and analyze microplastics in a much smaller size range. This difference highlights the importance of considering very small microplastics in assessing contamination and treatment effectiveness, which may be overlooked in studies focusing solely on larger particles. It is important to note that, according to Guo (2022), the crushing process in plastic recycling facilities affects the size distribution of particles and the proportion of differently-sized particles entering the wastewater.

Types of microplastics

This study identified several types of microplastics in water and sludge samples from the wastewater treatment system, including HDPE, LDPE, and a category of "others". The "others" category comprised EVA (Ethylene Vinyl Acetate) and commercial PCL (Polycaprolactone), which were grouped as they are classified within the seventh category of plastic resins, often referred to as "other" or "miscellaneous plastics". This classification system includes the six main plastic resin categories (PETE, HDPE, PVC, LDPE, PP, and PS) and a seventh category for other types of plastics. The analysis was conducted on nine microplastic samples extracted from wastewater, with particles larger than 500 µm subjected to FTIR analysis for identification.This identification was conducted using FTIR spectroscopy, comparing the FTIR spectra of samples with standard spectra for verification. FTIR analysis (Fig. 4) revealed characteristic peaks for each type of microplastic. LDPE exThis identification was conducted using FTIR spectroscopy, comparing the FTIR spectra of samples with standard spectra for verification. FTIR analysis revealed characteristic peaks for each type of microplastic. HDPE showed characteristic peaks at 2914 cm^{-1} and 2846 cm^{-1} (C-H stretching vibrations), 1462 cm^{-1} (C-H bending vibrations), and 718 cm^{-1} (CH_2 rocking vibrations). LDPE exhibited similar peaks at 2914 cm^{-1} and 2846 cm^{-1} (C-H stretching), 1467 cm^{-1} (C-H bending), and 715 cm^{-1} (CH_2 rocking). PCL was characterized by peaks around 2940 cm^{-1} and 2860 cm^{-1} (C-H stretching), 1722 cm^{-1} (C = O stretching of ester carbonyl group), and 1160 cm^{-1} (C-O-C symmetric stretching). EVA was identified through peaks at 2917 cm^{-1} and 2848 cm^{-1} (C-H stretching), 1740 cm^{-1} (C = O stretching), 1469 cm^{-1} (CH_2 and CH_3 bending), and 1020 cm^{-1} (C-O stretching). The FTIR results for the samples closely matched these standard spectra, confirming the presence of these specific microplastic types in the wastewater and sludge samples.

The sources of microplastics were also successfully identified, with HDPE originating from bottle caps and bottlenecks in the recycled materials. Then, LDPE was traced to plastic bottle labels, while EVA and PCL were detected from other items that entered the recycling process, such as sacks, insulation, or adhesives that were processed within the system. This detailed characterization of microplastic types and their

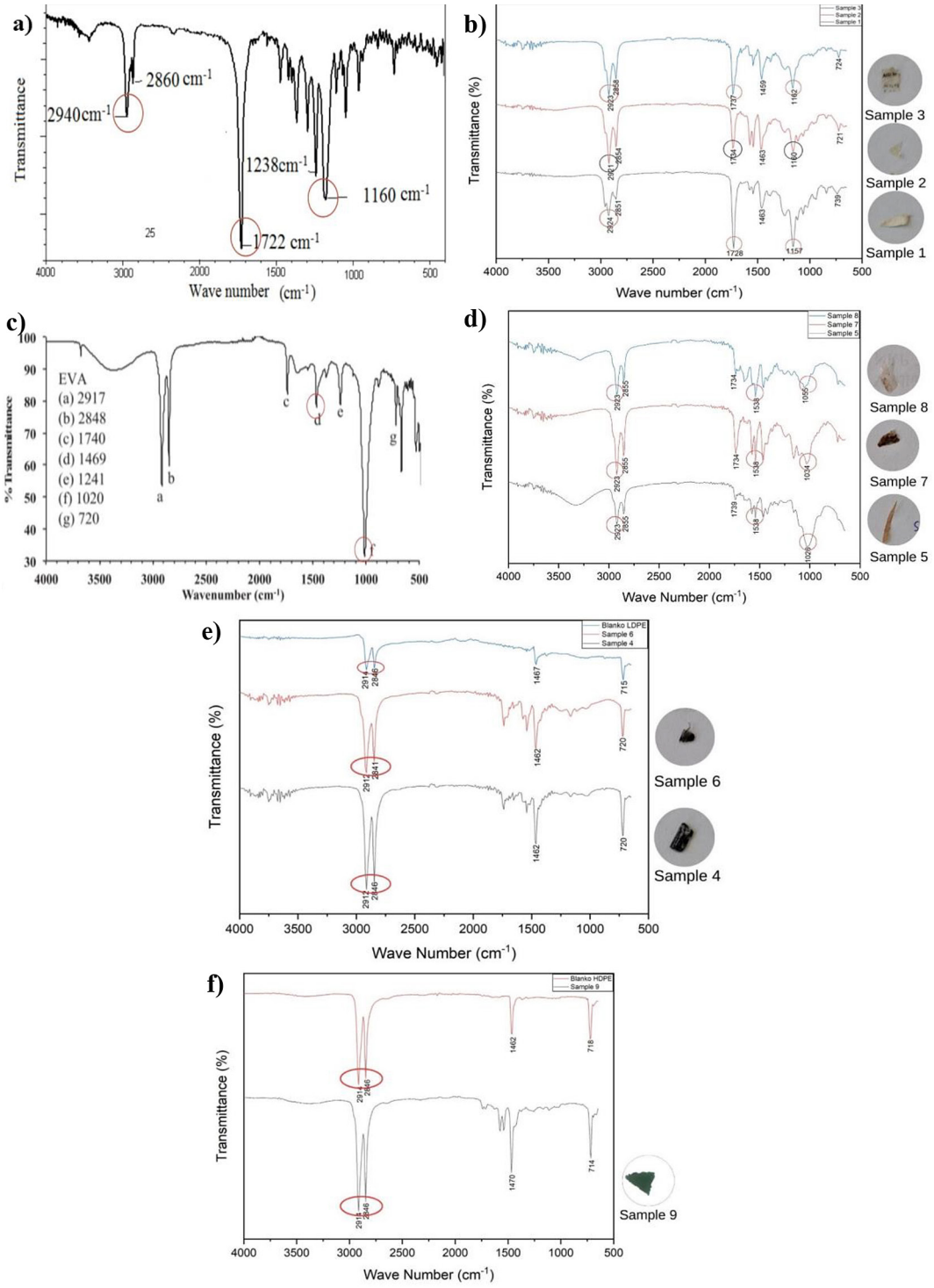

Figure 4. FTIR spectra comparison of various microplastic types identified in wastewater and sludge samples: a) FTIR spectrum of PCL from literature (Aqil et al., 2015), b) FTIR spectra of Samples 1, 2, and 3 identified as PCL, c) FTIR spectrum of EVA from literature (Jung et al., 2018), d) FTIR spectra of Samples 5, 7, and 8 identified as EVA, e) Comparison of FTIR spectra between pure LDPE and Samples 4 and 6, f) Comparison of FTIR spectra between pure HDPE and Sample 9

sources provides valuable insights into the origin and fate of various plastic materials in the recycling and wastewater treatment processes. The presence of these diverse microplastic types, including those classified as "others" (EVA and PCL). FTIR results were then further corroborated and extended by SEM-EDS (Scanning Electron Microscopy with Energy Dispersive X-ray

Spectroscopy) analysis as shown in Figure 5. SEM-EDS provided crucial information on the surface morphology and elemental composition of the microplastics, reinforcing the FTIR findings and offering additional insights. HDPE particles exhibited a smooth, shiny surface with a solid form. LDPE particles appeared rougher and more irregular in shape. EVA showed a rough, wavy surface. PCL displayed long, rough fibers with fragments. Various microplastic types identified in the plastic recycling facility wastewater samples are shown in Figure 6. EDS analysis confirms the presence of carbon (C) and oxygen (O) in the samples, supporting the FTIR results

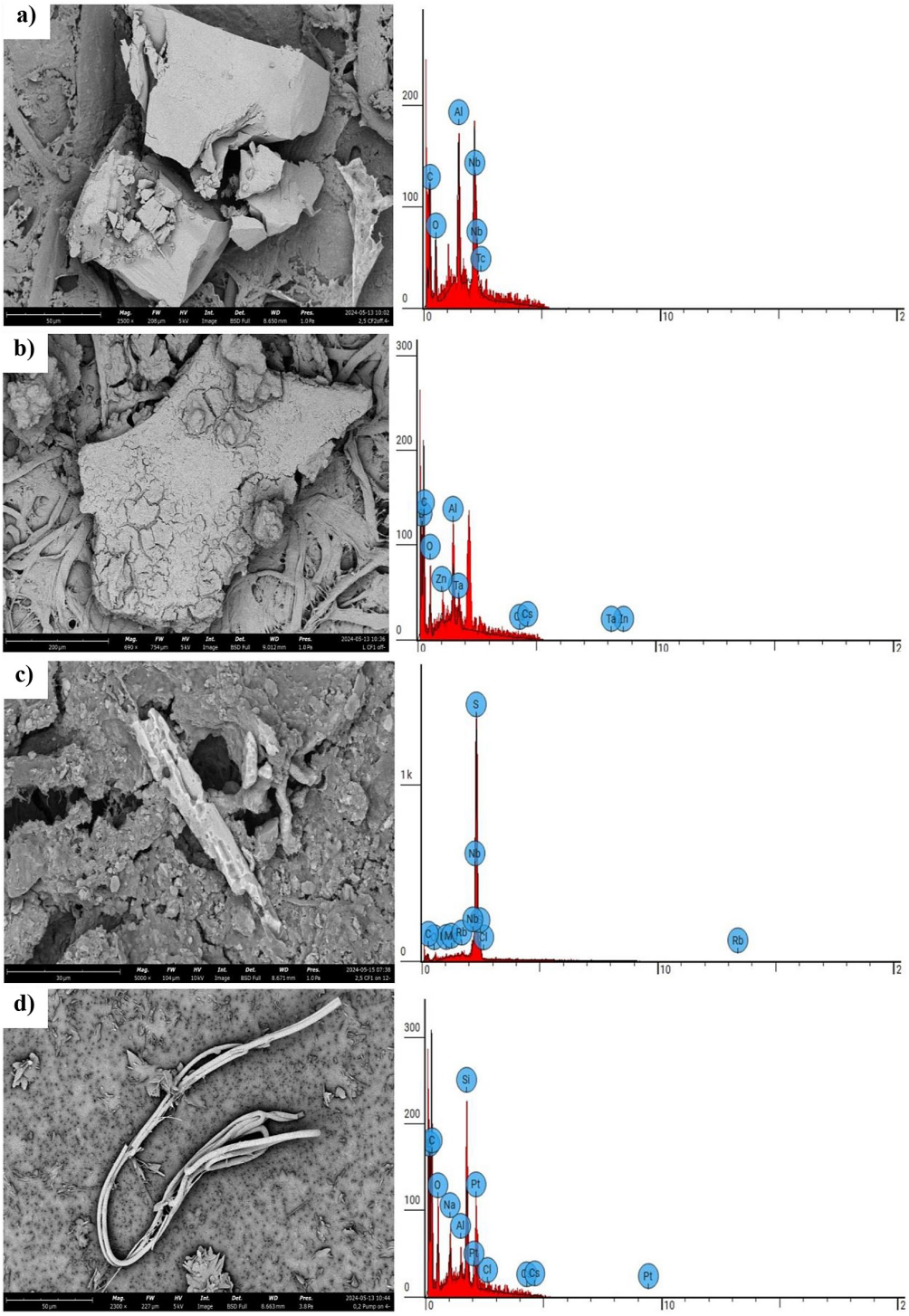

Figure 5. SEM-EDS results of microplastics identified at various sampling points: a) HDPE identified in FC2 wastewater effluent, b) LDPE identified in FC1 sludge effluent, c) EVA identified in PI station, d) PCL identified in FC1 effluent

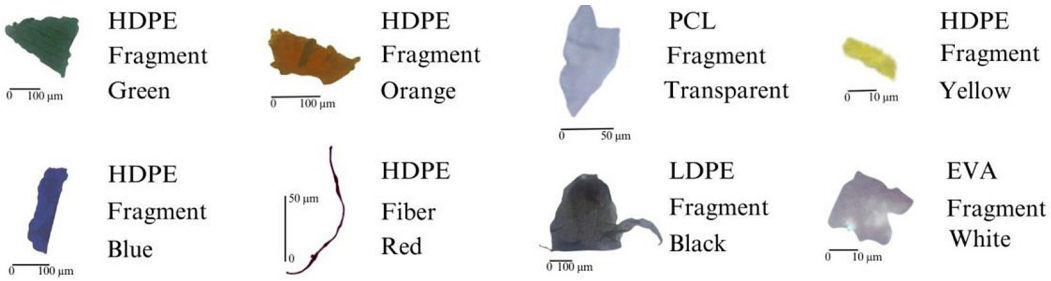

Figure 6. Various microplastic types identified in PRF wastewater

in identifying the different types of microplastics. Additionally, the analysis revealed the presence of other elements such as silicon (Si), aluminum (Al), boron (B), chlorine (Cl), and platinum (Pt). These elements, likely originating from environmental contamination or sample preparation processes, highlight the potential for microplastics to adsorb and transport various elements. This finding is particularly concerning as it suggests that when released into the environment, microplastics may act as vectors for potentially harmful substances, posing additional risks to ecosystems and organisms.

Figure 7 shows the results of microplastic identification by type in both wastewater and sludge. Under operational conditions, the Pump It unit showed the highest abundance of HDPE (419 particles/L), followed by LDPE (43 particles/L) and others (23 particles/L). The abundance of HDPE generally decreased along the treatment process, while LDPE and other plastics showed fluctuating patterns. In sludge samples, FC3 effluent contained significantly higher abundances of all plastic types, with HDPE reaching 55.000 particles/kg, LDPE 35.000 particles/kg, and others 40.000 particles/kg.

Non-operational conditions generally showed lower abundances of microplastics across all sampling points for both water and sludge samples. For instance, in water samples, the Pump It unit contained HDPE (36 particles/L), LDPE (11 particles/L), and others (18 particles/L). Sludge samples under non-operational conditions also showed lower concentrations, with FC2 and FC3 effluent sludge having higher concentrations than FC1. Çolakoğlu & Uyanık (2024) reported a broader range of polymer types in plastic recycling facilities' effluents, with PE (56%) being dominant, followed by PP (16%), PUR (7%), PS (6%), and others (15%). While our study also found PE (HDPE and LDPE) to be dominant, we did not identify significant proportions of PP, PUR, or PS. Suzuki (2022) observed varying dominant polymer types across different facilities, including PS, PET, and PP as major components, which contrast with our findings of HDPE and LDPE dominance. These differences highlight the variability in microplastic pollution across

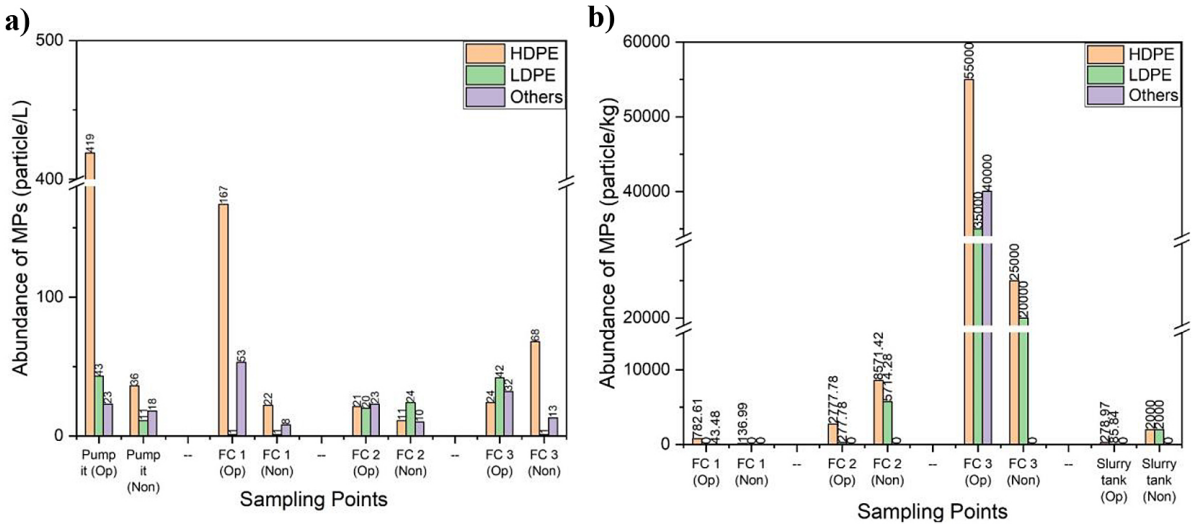

Figure 7. Microplastic types identified in: a) wastewater, b) sludge

different plastic recycling facilities, likely due to factors such as local processing methods, types of plastics recycled, and analytical techniques used.

Potential release of microplastics at different treatment units

Tube flocculator

The initial stage of treatment revealed a microplastic abundance of 485 particles/L in the influent. Following passage through the tube flocculator, the microplastic abundance in the FC1 effluent decreased to 221 particles/L, demonstrating removal efficiencies of 54.43% and 52.31% under operational and non-operational conditions, respectively.

The tube flocculator, designed to remove suspended particles through the addition of coagulants, promotes coagulation and flocculation processes. Microplastic removal via coagulation is influenced by the physical and chemical properties of microplastics, wastewater characteristics, and coagulant type and dosage (Reddy and Nair, 2022). In this study, aluminum chlorohydrate (ACH) was employed as the coagulant, with a dosage of 5.2 L/h, complemented by 9 L/h of polymer.

Under operational conditions, the removal efficiency for HDPE and LDPE microplastics was 60.14% and 97.67%, respectively. In non-operational conditions, LDPE removal reached 90.91%, while HDPE removal was 38.89%. Overall microplastic removal ranged from 44.44% to 66.76% across both operational and non-operational conditions.

Size-dependent removal efficiencies were observed, with the highest removal occurring in the 2.5 µm–5 mm range under operational conditions and in the 0.2 µm – 1.2 µm range under non-operational conditions. Regarding microplastic shapes, fibers were more efficiently removed in the tube flocculator under operational conditions, with a removal rate of 83.02%. Conversely, fragments showed higher removal efficiency (67.92%) under non-operational conditions.

Comparing these results with other relevant studies provides valuable insights. Of relevance is the laboratory-scale research conducted by Lapointe et al. (2020), which also used ACH as a coagulant. Their study, employing ACH at dosages ranging from 0.91 to 3.64 mg Al/L, revealed that pure polyethylene (PE) microplastics were more resistant to coagulation, with a removal rate of 82%, while weathered PE exhibited a higher removal rate of 99%. This difference highlights the significant impact of microplastic surface properties on removal efficiency in treatment processes.

This study's removal efficiencies (54.43% under operational conditions and 52.31% under non-operational conditions) are lower than those reported by Lapointe et al. (2020). However, it's important to note that our study was conducted in a full-scale wastewater treatment plant, dealing with a complex mixture of microplastics and other contaminants, whereas Lapointe et al.'s research was performed under controlled laboratory conditions.

Despite the tube flocculator's effectiveness in removing a substantial portion of microplastics from wastewater, some microplastics, particularly in the 2.5 µm – 5 mm range, still escaped the process. These escaped microplastics were predominantly fragments, with HDPE being the most prevalent type. This observation is consistent with findings from Talvitie et al. (2017), who reported that smaller microplastics (< 300 µm) were more likely to pass through conventional wastewater treatment processes.

Electrocoagulation

The results indicate significant removal efficiency, with values of 71.04% under operational conditions. However, under non-operational conditions, an accumulation of microplastics was observed, particularly in the size range of 0.2–1.2 µm.

The effectiveness of the EC process is influenced by several factors, including anode material selection, current density, pH, and the characteristics of microplastics (type, shape, and abundance) (Shen et al., 2022). In this study, an aluminum (Al) anode was employed with a current density of 304–310 A/m² at pH 7.5. Under these conditions, microplastics in the size range of 0.2–1.2 µm exhibited the highest removal efficiency of 87.71%, with fragments being the most readily removed form.

Comparative analysis with related studies indicates that EC has significant potential for microplastic removal, with some researchers reporting removal efficiencies up to 99%. Perren et al. (2018) demonstrated that the EC process could remove 99.24% of microplastics at a wastewater pH of 7.5. Shen et al. (2022) reported peak removal efficiencies of 93.2% for PE, 91.7% for PMMA, and 98.4% for PP using an aluminum anode at pH 7.2. Elkhatib et al. (2021) achieved 99% removal efficiency for microplastics sized 350 and 850 µm, and 100% for those sized 1500

μm, at a current density of 2.88 mA/cm² and pH 4. Furthermore, a study in Bangkok, Thailand, by Gabisa and Ratanatamskul (2024) utilizing plastic industry waste treated with EC method, demonstrated 96% removal of microplastics, particularly PET and PS types, using a voltage of 10V and an aluminum anode. These findings indicate that EC has significant potential in removing various types of microplastics.

Despite the high removal efficiencies observed, some microplastics still escape the EC process. These are generally characterized by relatively small sizes, particularly in the range of less than 2.5–5 mm and 0.2–1.2 μm. The most prevalent form of these escaped microplastics is fragments, which are small pieces of larger plastic materials. HDPE and LDPE are among the types of microplastics frequently found post-treatment.

Moving bed biofilm reactor

The moving bed biofilm reactor (MBBR) is a biological wastewater treatment process where microorganisms grow as biofilm on suspended carrier media. The plastic media used in MBBR is designed to provide a large surface area for microbial growth, primarily aimed at removing organic pollutants from wastewater. In this study, sampling points for the MBBR unit were taken from FC2 and FC3 effluents.

Contrary to expectations, the observed phenomenon in the MBBR system was the accumulation of microplastics rather than their removal. The results showed an increase in microplastic abundance in the size range of 0.2–2.5 μm. Microplastic abundance escaping the MBBR stage exhibited relatively stable values in the FC3 effluent, with 98 particles/L under operational conditions and 92 particles/L under non-operational conditions. The size distribution varied between conditions, with microplastics predominantly ranging from 1.2–2.5 μm under operational conditions, while under non-operational conditions, the most common size was 0.2–1.2 μm.

Fragments were the most prevalent form of microplastics in the FC3 effluent, with LDPE dominating under operational conditions and HDPE prevailing under non-operational conditions. In sludge samples from FC3 effluent, microplastic abundance reached 150,000 particles/kg under operational conditions and 45.000 particles/kg under non-operational conditions. The majority of microplastics in sludge were 0.2–1.2 μm in size and predominantly fragmented. HDPE was the most common type of microplastic in FC3 sludge, with significant amounts under both operational and non-operational conditions. Interestingly, a study conducted by Lee and Kim (2018) presented contrasting results. Their research on biological treatment using a microbe carrier process demonstrated significant microplastic removal of 99%. For microplastics sized 106–300 μm, abundance decreased from 10.165 particle/L to 0.1 particle/L, while for sizes > 300 μm, it reduced from 3.7 particle/L to 0.18 particle/L. Microplastic abundance in sludge for the 106–300 μm range was 10.615 particle/g and 2.585 particle/g for sizes > 300 μm. In their study, microplastics were removed through adsorption to the biofilm.

However, this current study observed accumulation rather than removal of microplastics. This accumulation occurred under both operational and non-operational conditions, highlighting the complexity of processes within the reactor. Several factors may contribute to this accumulation, including process instability due to influent fluctuations and variations in wastewater retention time. This phenomenon may be attributed to the fact that the received wastewater is not always directly from the influent but sometimes stored beforehand, leading to extended retention times. Additionally, the possibility that the biofilm in the MBBR had not fully matured could be a factor affecting the system's ability to remove microplastics significantly. The difficulty in capturing small-sized microplastics by the biofilm media, resulting in their escape to the effluent, further complicates the situation. The observed accumulation phenomenon in this study potentially indicates specific factors that require further investigation to comprehensively understand microplastic dynamics in MBBR systems. Future research should focus on elucidating these factors and optimizing MBBR performance for effective microplastic removal in wastewater treatment processes.

CONCLUSIONS

This study provides critical insights into the behavior of microplastics within a plastic recycling facility (PRF) in Indonesia, focusing on their abundance, characteristics, and the efficiency of various wastewater treatment processes in their removal. The research confirms significant microplastic contamination in PRF wastewater, with initial concentrations of 485 particles/L in the influent.

Fragments were identified as the predominant form of microplastics, constituting over 54.84% of the total detected, with HDPE and LDPE being the most common types. The study also identifies a wide range of microplastic sizes, from 0.2–5 mm, highlighting the complexity of the contamination.

The effectiveness of different treatment stages varied considerably. The tube flocculator demonstrated moderate removal efficiency (54.43% under operational conditions), with performance varying based on microplastic size and polymer type. Electrocoagulation shows promising results with up to 71.04% removal efficiency under operational conditions, although its performance was highly dependent on operational parameters. Meanwhile, the Moving Bed Biofilm Reactor (MBBR) exhibited an accumulation of microplastics rather than removal, particularly for smaller sizes (0.2–2.5 μm), so further investigation of this unexpected phenomenon is required.

Despite the treatment efforts, a significant amount of microplastics still escaped into the effluent, with concentrations of 98 particles/L under operational conditions. Moreover, high microplastic accumulation was observed in sludge samples, particularly in the MBBR stage, raising concerns about potential environmental transfer through sludge disposal. These findings emphasize the persistent challenge of managing microplastic pollution from PRFs and the need for more effective mitigation strategies. Optimizing existing treatment processes, with a particular focus on improving the removal of smaller microplastics (<2.5 μm). This could involve exploring the integration of advanced treatment technologies specifically designed for microplastic removal, such as membrane filtration. Furthermore, strategies to reduce the generation of microplastics at source in PRF operations should be implemented, such as improved pre-cleaning processes for recycled materials.

Effective sludge management is essential, given the high concentration of microplastics found in sludge. Developing and implementing advanced sludge treatment methods before disposal or reuse is essential to prevent the transfer of microplastics to the environment. In addition, establishing a comprehensive monitoring program to track microplastic levels during the treatment process and in the receiving environment will provide valuable data for ongoing optimization efforts. Although recycling is crucial for sustainable plastic management, it is equally important to address the potential environmental risks associated with microplastic release. Future research and technological innovations should focus on developing more effective to mitigate microplastic pollution from these facilities, ensuring that plastic recycling contributes positively to environmental sustainability without compromising ecosystem health.

Acknowledgements

The authors gratefully acknowledge the Institut Teknologi Sepuluh Nopember for the internal support under the Scientific Research Scheme No. 1151/PKS/ITS/2024. The first author (Winda I.U.) acknowledges the Indonesian Endowment Fund for Education/Lembaga Pengelola Dana Pendidikan (LPDP), Ministry of Finance Indonesia for providing the financial support (scholarship grant).

REFERENCES

1. Aqil, M., Abderrahim, B., Abderrahman, E., Mohamed, A., Fatima, T., Abdesselam, T., Krim, O. 2015. Kinetic thermal degradation of cellulose, polybutylene succinate and a green composite: Comparative study. World Journal of Environmental Engineering, 3(4), 95–110. https://doi.org/10.12691/wjee-3-4-1
2. Brown, E., MacDonald, A., Allen, S., Allen, D. 2023. The potential for a plastic recycling facility to release microplastic pollution and possible filtration remediation effectiveness. Journal of Hazardous Materials Advances, 10, 100309. https://doi.org/10.1016/j.hazadv.2023.100309
3. Burns, E.E., Boxall, A.B.A. 2018. Microplastics in the aquatic environment: Evidence for or against adverse impacts and major knowledge gaps. In Environmental Toxicology and Chemistry 37(11), 2776–2796. Wiley Blackwell. https://doi.org/10.1002/etc.4268
4. Çolakoğlu, E.B., Uyanık, İ. 2024. Plastic waste management in recycling facilities: Intentionally generated MPs as an emerging contaminant. Waste Management, 181, 79–88. https://doi.org/10.1016/j.wasman.2024.04.005
5. Elkhatib, D., Oyanedel-Craver, V., Carissimi, E. 2021. Electrocoagulation applied for the removal of microplastics from wastewater treatment facilities. Separation and Purification Technology, 276. https://doi.org/10.1016/j.seppur.2021.118877
6. Gabisa, E.W., Ratanatamskul, C. 2024. Effects of operating conditions on removal of microplastics (PET, PP, PS) from wastewater by electrocoagulation systems and kinetics of chromium removal

in the presence of microplastics. Journal of Water Process Engineering, 61. https://doi.org/10.1016/j.jwpe.2024.105313

7. GESAMP. 2015. Science for Sustainable Oceans. www.imo.org
8. Guo, J.J., Huang, X.P., Xiang, L., Wang, Y.Z., Li, Y.W., Li, H., Cai, Q.Y., Mo, C.H., Wong, M.H. 2020. Source, migration and toxicology of microplastics in soil. In Environment International, 137. Elsevier Ltd. https://doi.org/10.1016/j.envint.2019.105263
9. Guo, Y., Xia, X., Ruan, J., Wang, Y., Zhang, J., LeBlanc, G.A., An, L. 2022. Ignored microplastic sources from plastic bottle recycling. Science of the Total Environment, 838. https://doi.org/10.1016/j.scitotenv.2022.156038
10. Jung, M.R., Horgen, F.D., Orski, S.V., Rodriguez C.,V., Beers, K.L., Balazs, G.H., Jones, T.T., Work, T.M., Brignac, K.C., Royer, S.J., Hyrenbach, K.D., Jensen, B.A., Lynch, J.M. 2018. Validation of ATR FT-IR to identify polymers of plastic marine debris, including those ingested by marine organisms. Marine Pollution Bulletin, 127, 704–716. https://doi.org/10.1016/j.marpolbul.2017.12.061
11. Lapointe, M., Farner, J.M., Hernandez, L.M., Tufenkji, N. 2020. Understanding and improving microplastic removal during water treatment: Impact of coagulation and flocculation. Environmental Science and Technology, 54(14), 8719–8727. https://doi.org/10.1021/acs.est.0c00712
12. Lares, M., Ncibi, M.C., Sillanpää, M., Sillanpää, M. 2018. Occurrence, identification and removal of microplastic particles and fibers in conventional activated sludge process and advanced MBR technology. Water Research, 133, 236–246. https://doi.org/10.1016/j.watres.2018.01.049
13. Lee, H., Kim, Y. 2018. Treatment characteristics of microplastics at biological sewage treatment facilities in Korea. Marine Pollution Bulletin, 137, 1–8. https://doi.org/10.1016/j.marpolbul.2018.09.050
14. Lestari, P., Trihadiningrum, Y., Wijaya, B.A., Yunus, K.A., Firdaus, M. 2020. Distribution of microplastics in Surabaya River, Indonesia. Science of the Total Environment, 726. https://doi.org/10.1016/j.scitotenv.2020.138560
15. Masura, J. 2015. Laboratory Methods for the Analysis of Microplastics in the Marine Environment: Recommendations for quantifying synthetic particles in waters and sediments.
16. Perren, W., Wojtasik, A., Cai, Q. 2018. Removal of microbeads from wastewater using electrocoagulation. ACS Omega, 3(3), 3357–3364. https://doi.org/10.1021/acsomega.7b02037
17. Radityaningrum, A.D., Trihadiningrum, Y., Mar'atusholihah, Soedjono, E.S., Herumurti, W. 2021. Microplastic contamination in water supply and the removal efficiencies of the treatment plants: A case of Surabaya City, Indonesia. Journal of Water Process Engineering, 43. https://doi.org/10.1016/j.jwpe.2021.102195
18. Reddy, A.S., Nair, A.T. 2022. The fate of microplastics in wastewater treatment plants: An overview of source and remediation technologies. In Environmental Technology and Innovation, 28. Elsevier B.V. https://doi.org/10.1016/j.eti.2022.102815
19. Shen, M., Zhang, Y., Almatrafi, E., Hu, T., Zhou, C., Song, B., Zeng, Z., Zeng, G. 2022. Efficient removal of microplastics from wastewater by an electrocoagulation process. Chemical Engineering Journal, 428. https://doi.org/10.1016/j.cej.2021.131161
20. Sun, J., Dai, X., Wang, Q., van Loosdrecht, M.C.M., Ni, B.J. 2019. Microplastics in wastewater treatment plants: Detection, occurrence and removal. In Water Research, 152, 21–37. Elsevier Ltd. https://doi.org/10.1016/j.watres.2018.12.050
21. Suzuki, G., Uchida, N., Tuyen, L.H., Tanaka, K., Matsukami, H., Kunisue, T., Takahashi, S., Viet, P.H., Kuramochi, H., Osako, M. 2022. Mechanical recycling of plastic waste as a point source of microplastic pollution. Environmental Pollution, 303. https://doi.org/10.1016/j.envpol.2022.119114