MICROPLASTIC POLLUTION AND REMOVAL IN WASTEWATER TREATMENT PLANTS IN THAILAND

BY

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ABSTRACT

Microplastic (MP) is an emerging pollutant that requires urgent attention. Wastewater treatment plants (WWTPs) are considered an important pathway to transport terrestrial MPs to water bodies. This study aims to investigate the level of MP pollution and removal at each treatment step to find the most efficient MP removal stage in four different WWTPs in Thailand. Removal efficiencies and the characteristics of MPs in the inlet of four WWTPs were compared. MP samples in three WWTPs were collected by filtration of wastewater through a set of sieves: 5 mm, 1 mm, 500 µm, and 53 µm. Grab sampling was performed in a WWTP built as a closed underground system. Sludge samples were collected from each WWTP by grab sampling. MPs found in the influent of four different WWTPs ranged from 3.50 – 77 particles per L, and the effluent ranged from 2.33 – 30.33 particles per L. The highest MP concentration in the inlet was found in Bang Sue WWTP. Despite the high MP loads in the influent, Bang Sue WWTP achieved the highest removal efficiency of 86.14% and 96.97% from the conventional treatment system and UF as a final polishing step, respectively. One of the reasons is that it was constructed as an underground treatment system to prevent atmospheric contamination. On the other hand, Sing Buri WWTP, with a waste stabilization pond, attained very low MP removal efficiency from both grab sampling and on-site filtration. There was a high concentration of MPs from atmospheric deposition in Sing Buri WWTP. The number of MPs detected in the influent of the largest WWTP in Bangkok (population equivalent to 1 million) is lower than Nong Khaem WWTP, which has a population equivalent to 520,000. Most of the MPs found in four WWTPs were fibers identified as polyethylene terephthalate (PET) or polyester. Fibers were derived from clothes shedding. Fragments identified as PE and films were less abundant. The results of this study show a variety of MP abundances in different WWTPs with different treatment technologies in Thailand. However, all of the studied conventional WWTPs, except the pond system, lack primary sedimentation which is reported to improve the removal of MPs. The addition of a primary sedimentation tank and advanced filtration as a final step in WWTP should be considered as it could decrease the number of MP released to the freshwater environment.

Keywords: Microplastic, Wastewater treatment plant, Sludge, Tertiary treatment, Waste stabilization pond

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Katekanya Tadsuwan

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Symbols/Abbreviations	Terms
A^2O	Anaerobic-Anoxic-Oxic
ATR	Attenuated total reflectance
BPA	Bisphenol A
CSO	Combined sewer overflow
DAPI	4’, 6-diamidino-2-phenylindole
DDS	Department of Drainage and Sewerage
DI	Deionized
EPDM	Ethylene propylene diene monomer
EECC	Environmental Education and Conservation Center
ESEM-EDS	Environmental scanning microscopy-energy dispersive X-ray spectroscopy
EU	European Union
FTIR	Fourier Transform Infrared
HDPE	High-density polyethylene
IR	Infrared radiation
LDPE	Low-density polyethylene
MBR	Membrane bioreactor
MP	Microplastic
NAFTA	North American Free Trade Agreement
PA	Polyamide
PAM	Polyacrylamide
PCD	Pollution Control Department
PE	Polyethylene

PES	Polyethersulfone
PET	Polyethylene terephthalate
POP	Persistent organic pollutant
PP	Polypropylene
PS	Polystyrene
PU	Polyurethane
PVC	Polyvinyl chloride
PVP	Polyvinylpyrrolidone
Pyr-GC/MS	Pyrolysis-gas chromatography coupled to mass spectrometry
SEM	Scanning electron microscopy
SEM-EDS	Scanning electron microscopy-energy dispersive X-ray spectroscopy
SS	Sewage sludge
SPI	Society of Plastic Industry
RO	Reverse osmosis
TDRI	Thailand Development Research Institute
UF	Ultrafiltration
UK	United Kingdom
US	United States
UV	Ultraviolet
VLR	Vertical loop reactor
WPO	Wet peroxide oxidation
WWTP	Wastewater treatment plant

CHAPTER 1
INTRODUCTION

1.1 Problem statement

Plastic is a synthetic polymer well-known as an inexpensive, lightweight, strong, durable, and corrosion-resistant material with thermal and electrical insulation properties (Thompson et al., 2009a). It has been widely used in many applications, including consumer products, construction, food packaging, and medical equipment (Agamuthu, 2018). The global production of plastic increased to 335 million tons in 2016, surpassing other man-made materials (Geyer et al., 2017). China was the most prominent plastic producer in 2020, accounting for 32% of worldwide production, followed by the North American Free Trade Agreement (NAFTA) countries and the rest of Asia (PlasticsEurope, 2021). The most significant proportion of plastic demand is packaging which accounts for 40.5% of the total demand (PlasticsEurope, 2021).

Plastic waste has become a global environmental concern due to a large amount of consumption. Plastic materials are produced from fossil-derived monomers which are non-biodegradable, such as ethylene and propylene (Geyer et al., 2017). Thus, due to its high stability and durability, plastic litter is highly persistent and tends to accumulate in many natural habitats and ends up in the ocean. The period that discarded plastics stay in the environment depends on the material's chemical nature, the environment's characteristics, and the degradation rate (Andrady & Neal, 2009). Environmental degradation can break down plastic debris into smaller size ranges, so-called microplastics (MPs), which are easily spread and ubiquitous in the marine environment (Cole et al., 2011). Thus, many studies have crucially investigated MP pollution to understand the abundance, pathway, and environmental impacts in freshwater and marine ecosystems.

MP is commonly defined as a plastic particle with a diameter ranging from 5 mm to 1 nanometer (Kershaw, 2015). It can be categorized into primary and secondary MPs. Primary MPs are manufactured in small sizes for industrial and domestic applications, such as virgin resin pellets, microbeads in personal care products, industrial scrubbers, and plastic powders. In contrast, secondary microplastics result from the fragmentation of bigger-sized plastics (Talvitie et al., 2017a). MPs are ubiquitous in various environments, including freshwater, marine, and terrestrial ecosystems (Eerkes-Medrano et al., 2015). They were present in the guts of a wide range of marine organisms, such as zooplankton (Rashid et al., 2021), mussels (Avio et al., 2015), and fish (Piyawardhana et al., 2022). Moreover, MPs were detected in tap and drinking water (Kirstein et al., 2021).

Concerns have been raised regarding the potential impacts of MPs on the environment, wildlife, and human health. MPs can be ingested by many organisms, resulting in physical effects such as internal abrasions and blockage (Wright et al., 2013). Toxic effects from plastic materials, chemical additives, and toxins adsorbed on the surface can cause chronic toxicity to humans and other living organisms (Li et al., 2018a). Persistent organic pollutants (POPs) from the surrounding environment tend to adsorb on the MP surface (Wang et al., 2021a). These chemicals can be transferred to organisms at higher trophic levels leading to biomagnification and bioaccumulation (Agamuthu, 2018).

Recent studies have focused on the source of MP in the aquatic and marine environment. The origins of MPs can be both aquatic and land-based sources. One of the land-based sources of MPs besides surface runoff is wastewater effluent. As MPs have been detected in wastewater effluent, many studies suggested that wastewater treatment plant (WWTP) is a pathway for MPs (Conley et al., 2019; Murphy et al., 2016; Ziajahromi et al., 2017). MPs such as synthetic fibers and microbeads from personal care products in the effluent of treatment plants have been found in the ocean (Talvitie et al., 2015). In addition, more than one-third of MPs in the ocean are synthetic fibers released from washing machines (Boucher & Friot, 2017; Browne et al., 2011). A schematic illustration of WWTPs as a pathway for MPs to water bodies is shown in Figure 1.1. However, there is a gap in knowledge of the fate of MPs and their transport behavior in wastewater treatment plants (Talvitie et al., 2015). Due to the small size of MP, the efficiency of the wastewater treatment process in every step until the discharge of final effluent needs to be focused on.

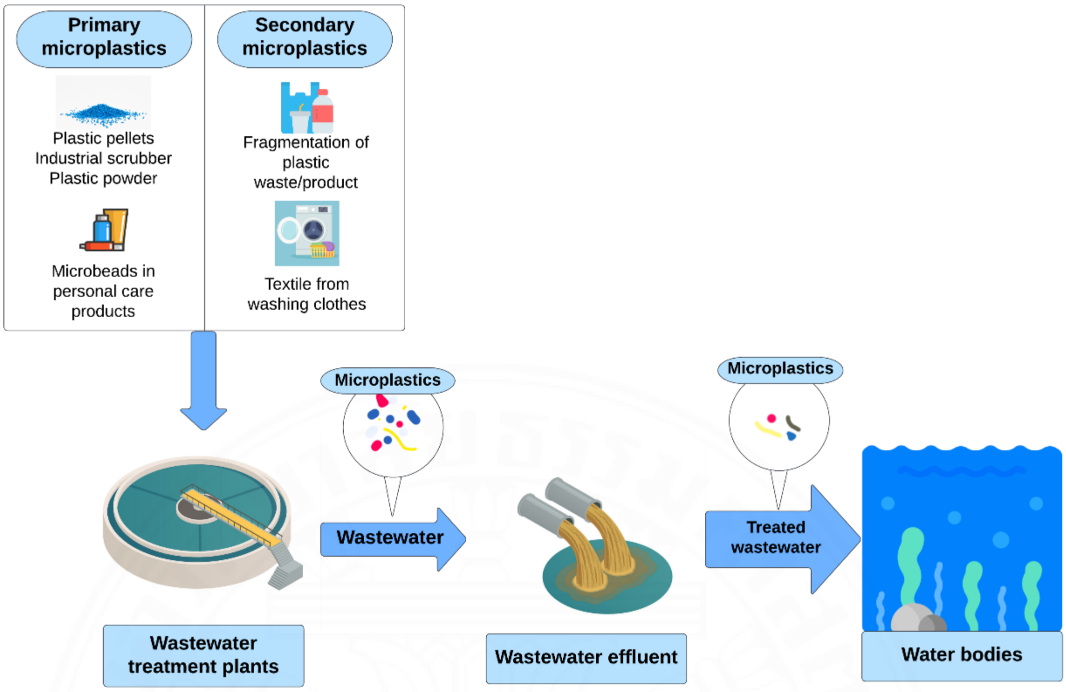


Figure 1.1 Wastewater treatment plants as a pathway for microplastics

Thailand is a top-ten contributor to marine debris (TDRI, 2021). About 0.15 – 0.41 million tons of marine debris were annually created from mismanaged plastic waste in Thailand (Jambeck et al., 2015). There is evidence that MPs are present near the coastline and marine biota. A study conducted along the eastern coast of Thailand detected a large number of MPs derived from anthropogenic activities along 21 beaches (Bissen & Chawchai, 2020). Another study investigated the number of MPs in sessile invertebrates (Thushari et al., 2017). A significant accumulation was found in bivalves, gastropods, and barnacles, and it is used as an indicator of MP contamination in the area (Thushari et al., 2017). In addition, there was an occurrence of MPs in the gastrointestinal tract in demersal and pelagic fish from the marine environment in Thailand (Klangnurak & Chunniyom, 2020). MPs investigated in densely-populated beaches in Phuket were likely derived from fishing activities or wastewater effluent (Akkajit et al., 2019). Thus, WWTP is a final barrier before MPs enter the water bodies.

Bangkok is a highly populated city with more than 2 million households. There are five major wastewater treatment plants and more than 15 plants under the supervision and regulation of the Department of Drainage and Sewerage (Sewerage).

Treated wastewater is released into canals around Bangkok connected to the Chao Phraya River. The river is the major water source for Thailand, and it ends up in the Gulf of Thailand, which is a habitat for many marine lives. However, most of the WWTPs in Bangkok are conventional WWTPs. They are not designed to specially remove their minute-sized pollutants (Iyare et al., 2020).

There is a lack of studies on MPs in a wastewater treatment plant in Thailand that acts as a pathway for MP transport. Therefore, this study investigates the abundance of MPs present in each treatment step in the wastewater system to find the most crucial unit process for MPs removal. The overall MP removal efficiencies of each WWTP were calculated. It is essential to assess the morphologies and types of MPs in the wastewater treatment system to find their potential sources.

1.2 Research objectives

1. To investigate the level of MP pollution in wastewater treatment plants in Thailand.
2. To estimate the removal efficiencies of wastewater treatment plants in Thailand and find the most effective unit process for removing MPs.
3. To characterize morphologies and types of MPs.
4. To propose technological and management strategies related to the findings.

1.3 Scope of the study

Figure 1.1 depicts the overview of this study. MP samples were collected from four WWTPs in Thailand: three WWTPs with different treatment technologies in Bangkok and one WWTP with a pond system in less-populated areas. Three WWTPs in Bangkok employ a conventional activated sludge system to treat domestic wastewater but have different configurations. One of the selected WWTPs in Bangkok was equipped with pilot-scale ultrafiltration (UF) unit, the only tertiary treatment in all WWTPs in the study. MP samples were collected mainly from influent, during treatment processes, sewage sludge, and final effluent from each WWTP. MP samples from two WWTPs were collected by in situ filtration from the influent, after the grit removal, and the effluent due to the accessibility of the pipelines. Another WWTP with a UF unit was built as a closed underground system. MPs were collected by grab sampling from the influent, after fine screen and grit removal, after the aeration tanks, the effluent from secondary treatment, and the effluent from UF. In addition, MPs from waste stabilization ponds were collected from the influent, the effluent from an anaerobic pond, a facultative pond, and a maturation pond. In this study site, both in situ filtration and grab sampling was performed to find the influence of sampling methods on the number of MPs. Moreover, MPs from atmospheric deposition were analyzed in this WWTP due to the long retention time.

The smallest size of MPs analyzed in this study was 53 µm in both wastewater and sludge samples. MPs were grouped into size ranges based on the sieve size. Samples were pre-treated to remove other organic and inorganic impurities. The abundances of 0.5 – 5 mm MPs were counted under an optical microscope. The 0.05 – 0.5 mm group was stained with Nile Red to perform fluorescence tagging for MP quantification. Fourier transform infrared (FTIR) spectroscopy in attenuated total reflectance mode (ATR) was employed to analyze polymer types of MPs larger than 0.5 mm. In contrast, smaller-sized MPs were analyzed by FTIR connected with a microscope (micro-FTIR) for more precise results. MP samples are prone to airborne contamination, affecting the false results; thus, procedural blanks were performed parallel to an actual sample, and contamination mitigation protocols were strictly followed.

MP removal at each treatment step was evaluated to find the most effective removal step. The overall MP removal efficiency of a WWTP was calculated based on the number of MPs in the influent and effluent. All studied WWTPs were compared to find the most efficient treatment technology for MP removal and improvement to prevent further release to the environment.

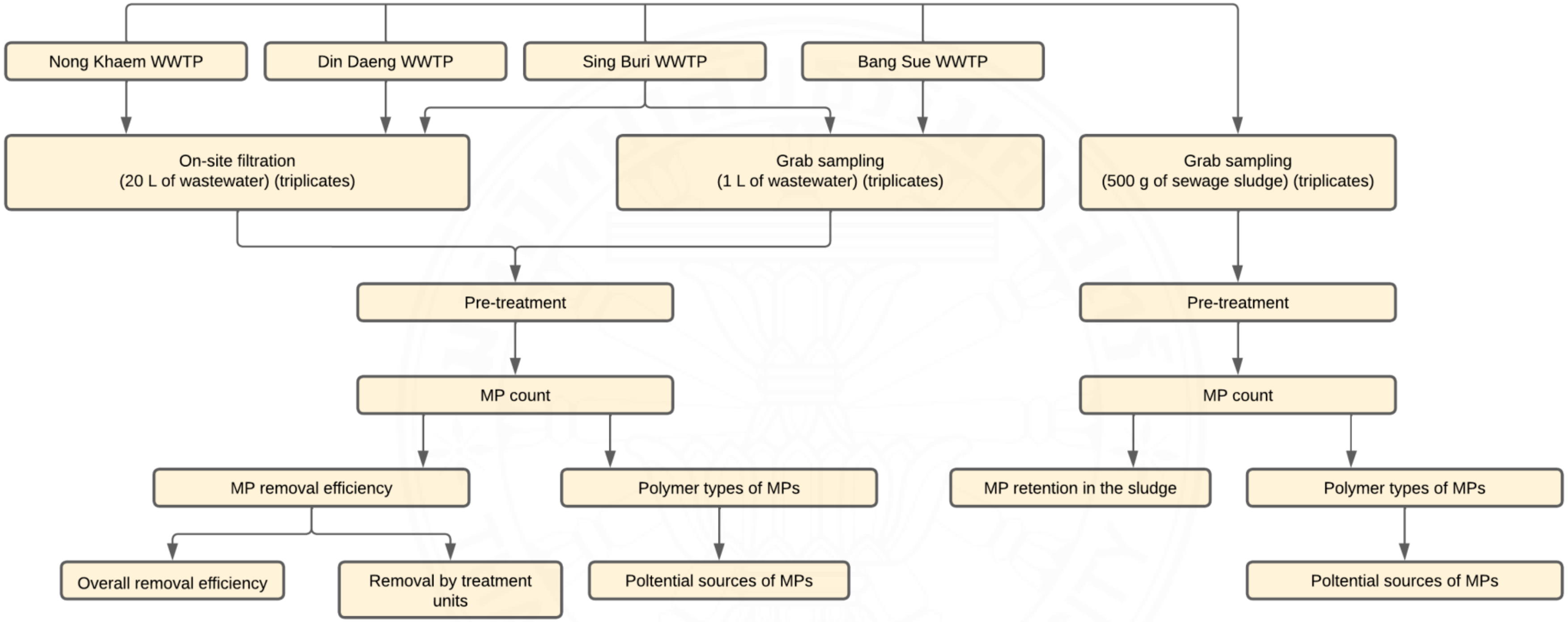


Figure 1.2 Flowchart of the overview of this study

CHAPTER 2
LITERATURE REVIEW

2.1 Plastic materials

Polymers are high molecular-weight organic molecules composed of repeated units called monomers (Valavanidis, 2016). Plastics are synthetic polymers prepared by the polymerization of monomers derived from fossil fuels together with some chemical additives to improve the properties of plastic (Gewert et al., 2015; Thompson et al., 2009b). The history of plastic began in the early 20th century when Bakelite, the first synthetic polymer, was invented (Geyer et al., 2017). The properties of plastic are strong, inexpensive, lightweight, durable, corrosion-resistant materials, and high thermal and electrical insulation, which make them suitable for a wide range of applications (Thompson et al., 2009a). The large scale of plastic production has continued to grow since then. It surpasses most other man-made materials such as metals, glass, papers, and other traditional materials due to its properties and low cost (Andrady & Neal, 2009; Geyer et al., 2017; Valavanidis, 2016). The total global production of plastic was 367 million tons in 2020, where half of the total production is in Asia (PlasticsEurope, 2021). In addition, the global trend has shifted from reusable plastic products to single-use plastic, which increases plastic packaging (Geyer et al., 2017). These disposable plastic products are designed to have a three-year service life; after that, they will be discarded (Gewert et al., 2015).

There are two main categories of plastics classified based on macromolecular structure and temperature-dependent physical properties; thermoplastics and thermosets (Klein, 2012). Thermoplastic consists of linear polymeric molecules without crosslinks (Shackelford, 2016). Thus, this type of plastic resin structure is resistant to chemicals and environmental effects (Klein, 2012). The application of heat can soften and melt thermoplastic, and it can be re-solidified by cooling (Klein, 2012; Shackelford, 2016). As thermoplastic is inexpensive, lightweight, and durable, it is used in many applications (Grigore, 2017). Another group of plastic is called thermosets, a hard and brittle material. The structure of thermosets is a narrow-crosslinked molecule, making its mechanical properties not temperature-dependent like thermoplastics (Klein, 2012). Moreover, heat cannot melt thermosets, and chemical decomposition occurs when the temperature exceeds decomposing temperature (Klein, 2012).

Even though there are more than hundreds of types of plastics, only eight groups of plastic are qualified as commodity plastic due to their high volume and relatively low price (Andrady & Neal, 2009). These groups include low-density polyethylene (LDPE), high-density polyethylene (HDPE), polypropylene (PP), polyvinyl chloride (PVC), polystyrene (PS), polyurethane (PU), polyethylene terephthalate (PET) and other. The percentage of global demand for each plastic type in 2016 is shown in Figure 2.1. In addition, the packaging is the largest plastic market, accounting for 39.9% of the total demand in 2016 (Plastics Europe, 2017).

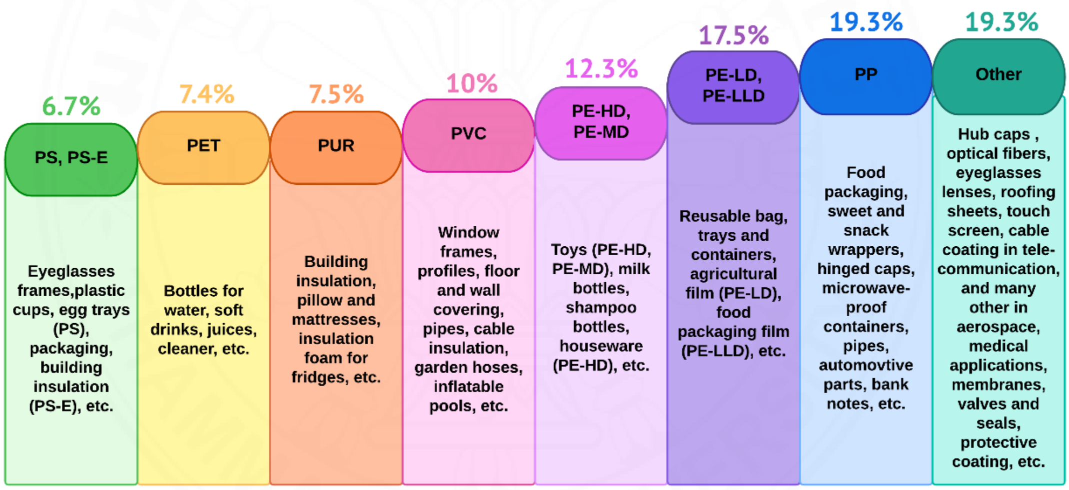


Figure 2.1 Fractional demand of polymer type based on a global basis in 2016 (adapted from Plastics Europe (2017)

Society of Plastic Industry (SPI) established a plastic coding system in 1988 for plastic packaging and containers for better waste sorting and recycling (The Office of Consumer Affairs, 2019). The majority of resin types in plastic products are divided into six groups: low-density polyethylene (LDPE), high-density polyethylene (HDPE), polypropylene (PP), polyvinyl chloride (PVC), polystyrene (PS), and polyethylene terephthalate (PET) (The Office of Consumer Affairs, 2019). The number is assigned to each resin type from 1 to 7 and printed on plastic products (Figure 2.2). PP is the most demanding polymer type in European Union (EU) countries followed by LDPE, and HDPE (PlasticsEurope, 2021). PP is a strong plastic with chemical resistance and is suitable for filling hot liquids because of its high melting point (American Chemistry Council, 2007). The applications of PP are mainly packaging, such as containers, bottle caps, and medicine bottles (American Chemistry Council, 2007). LDPE is used to manufacture reusable bags, containers, and packaging film, whereas HDPE is used for toys, bottles, pipes, and housewares (PlasticsEurope, 2021).

Figure 2.2 SPI Plastic Coding System

For the application of plastics, polymer resins are mixed with additives to improve the performance of the material, for example, the reinforcement from carbon and silica, pliability from plasticizers, thermal and ultraviolet stabilizers, flame retardants, and colorings (Thompson et al., 2009a). Moreover, some chemical additives used in the plastic industry can be released into the environment and are considered toxic (Thompson et al., 2009a). Additives raising public concerns include phthalate plasticizers, bisphenol A (BPA), brominated flame retardants, and anti-microbial agents (Thompson et al., 2009a). Therefore, plastic additives are strictly controlled in many countries, especially the application related to human health (Andrady & Neal, 2009). Plastic is not only persistent in the environment but also poses a risk to living organisms from chemical additives. Plastic becomes plastic waste if it is not disposed of or managed properly.

2.2 Plastic waste

The increase in the use of plastic has become problematic in waste management. Plastic waste accounts for approximately 10% of the total waste generation (Thompson et al., 2009b). Large amounts of plastic waste are discarded daily, accounting for 10%

of the gasoline consumed and imported by the US in the past few decades (Valavanidis, 2016). Most single-use plastics, such as packaging, are disposed of in landfills, while only a small proportion is recycled (Barnes et al., 2009). However, the plastic waste problem is not as local as it was a few decades ago since the increasing amount of plastic pollution is found on land and water bodies (Valavanidis, 2016).

Plastic waste contributes to a large proportion of marine litter (Barboza et al., 2018). Routes of plastic litter from the continents to the oceans are stormwater runoff, dumping along shorelines, and direct dumping at sea from ships (Walker et al., 2019). The number of plastics entering the oceans was estimated to be between 4.8 to 12.7 million metric tons in 2010 (Jambeck et al., 2015). About 1.7-4.6% of plastic waste generated in the oceans is mismanaged solid waste in many coastal countries (Galgani et al., 2019; Hoornweg & Bhada-Tata, 2012; Jambeck et al., 2015). In Thailand, the amount of marine debris generated from mismanaged plastic waste was 0.15-0.41 metric tons per year (Jambeck et al., 2015). The country has been listed as a top ten contributor to marine debris (TDRI, 2021). In 2020, Thailand generated approximately 6,300 tons of plastic waste per day (PCD, 2021). Plastic waste management requires a lot of attention due to the high amount of plastic waste produced daily.

The mobility of plastic waste is much higher than other man-made materials due to its lightweight. Moreover, none of the commercial plastics are biodegradable (Geyer et al., 2017). Due to its stability and durability, plastic is resistant to degradation and tends to accumulate in the environment (Gewert et al., 2015). A lifetime of discarded plastic depends on the chemical composition of materials, the characteristics of the environment, and the degree of degradation (Andrady & Neal, 2009). Plastic decomposition may take several to a hundred years (Avio et al., 2017). Moreover, plastic waste can be slowly degraded into smaller plastic particles, called 'microplastic', which spread easily and become pollution in the marine environment (Valavanidis, 2016).

2.3 Microplastics

Microplastic (MP) is usually defined as particles with sizes smaller than 5 mm to as small as 10 nm (Agamuthu, 2018; Avio et al., 2017). Owing to their minute sizes, they are challenging to monitor. Sources and the formation put MPs into two big categories: primary and secondary MP (Talvitie et al., 2017a). The key sources, types, and how MPs are formed are shown in Figure 2.3.

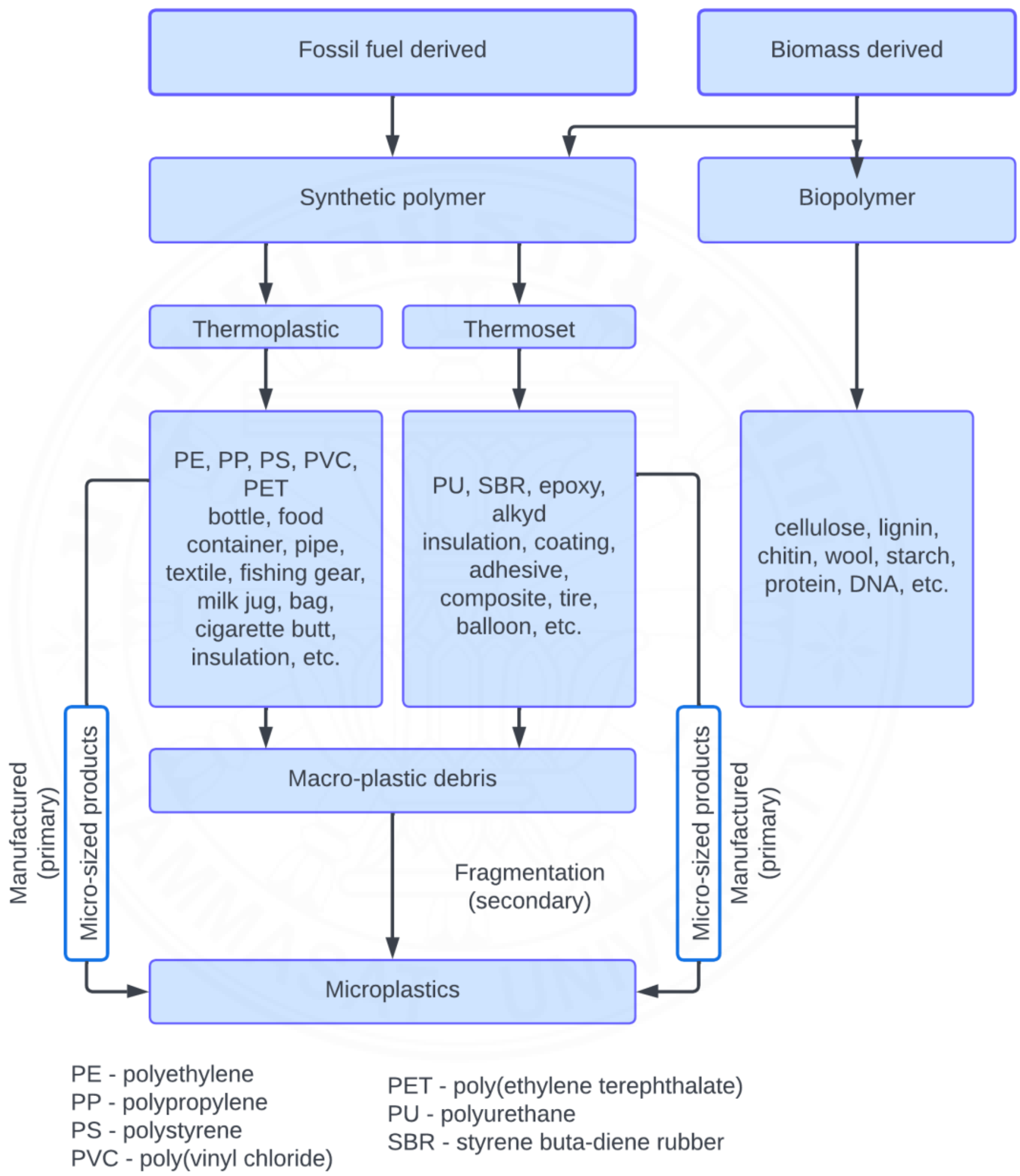


Figure 2.3 Key sources and types of MP (adapted from Kershaw, 2015)

2.3.1 Primary MP

Primary MPs, called 'plastic pellets', are manufactured directly in microscopic size for industrial and domestic products (Cole et al., 2011). They are mainly used in air blasting technology in industrial scrubbers and abrasive agents in personal care products in the form of microbeads (Cole et al., 2011; Conkle et al., 2018). The use of MP scrubbers in cosmetic products has significantly increased since 1988, and they come in various sizes, shapes, and compositions, according to the companies (Li et al., 2016). Moreover, primary MP can be produced as virgin resin pellets, with a size of 5 mm, to be used as feedstock for plastic manufacturing (Eerkes-Medrano et al., 2015). Improper handling at the processing facilities might cause the contamination of MP particles in surrounding water bodies (Rocha-Santos & Duarte, 2017). Therefore, legislation in some countries, such as the US, Canada, and the UK, has already banned primary MP in some consumer products, especially rinse-off cosmetics (Conkle et al., 2018). Microbeads have been officially banned in Thailand since January 2020 (FDA, 2019).

2.3.2 Secondary MP

Secondary MPs result from plastic weathering or fragmentation during their presence in the environment or the use phase of plastic (Rochman et al., 2016). This type of plastic constitutes the majority of MPs found in the ocean, where 75-90% of MPs originated from land-based activities (Duis & Coors, 2016; Hidalgo-Ruz et al., 2012). Littering, dumping plastic waste, and loss during landfilling and waste collection are important routes for MP to enter the environment (Duis & Coors, 2016). Typically, once the waste is deposited in the landfill, it is later covered with soil or synthetic material. A fence surrounds the landfill site to prevent wind loss (Duis & Coors, 2016). In addition, natural disasters, such as hurricanes, tsunamis, and storms, are contributed to the distribution of MPs in the ocean (Duis & Coors, 2016). Other secondary MPs are from plastic mulching which LDPE film is used in agriculture, abrasion of plastic materials, paints, plastic coating, the release of fibers from synthetic textiles and hygiene products, and material lost from fishing vessels and aquaculture facilities (Duis & Coors, 2016).

The study by Browne et al. (2011) found that the significant source of MP pollution in the marine environment is synthetic fiber from clothes (Browne et al., 2011). Most of the fibers detected were polyester, followed by acrylic, discharged mainly from the washing machines rather than the fragmentation of plastic cleaning tools (Li et al., 2016). Even though the efficiency of wastewater treatment facilities is nearly 99%, the amount of MPs released from the effluent is still significant (Li et al., 2016).

2.3.3 Degradation of plastics

Exposure to physical, chemical, and biological processes over a long period of time might lead to the reduction of structural integration followed by fragmentation of larger plastic (Browne et al., 2007a; Rocha-Santos & Duarte, 2017). These are the processes that break down break down macro-plastic (size > 5 mm) into MP (size < 5 mm). Physical degradation is an essential pathway to the formation of MP. It can be weathering elements, such as wetting/drying cycle, heating/cooling, and thawing/freezing, or abrasive forces made by animals or other abiotic factors in the environment, such as wind/water turbulence (Da Costa et al., 2016). This type of degradation does not alter the polymeric structure but decreases the size of particles and leads to morphological changes (Da Costa et al., 2018).

The degradation by ultraviolet (UV) radiation or photodegradation is the most efficient abiotic degradation occurred in the environment at the ambient condition. Plastic containing unsaturated chromophoric groups can absorb both visible light and high-energy UV radiation leading to photoinitiated chemical oxidation at the surface (Gewert et al., 2015). However, some types of plastic, such as PE and PP, do not contain chromophores, but the degradation is initiated by external impurities or structural abnormalities in macromolecule structure (Gewert et al., 2015). The molecular weight of photo-oxidized polymers becomes lower from bond scission and reduces mechanical strength. Plastic particles will become brittle and susceptible to fragmentation. When plastic fragments into smaller pieces, a higher surface area is ready for further biodegradation. Moreover, the photodegradation of plastic can lead to the leaching of chemical additives into the surrounding environment (Cole et al., 2011).

Thermal degradation is another factor that alters the properties of plastic because of the bond scissions of the main polymeric chain. Thermo-degradation can induce oxidation, leading to changes in tensile strength, molecular weight, crystallinity, and color of the materials (Da Costa et al., 2016). Plastic material with thermal pre-treatment, such as PP, is more subject to biodegradation (Da Costa et al., 2016). However, this mechanism does not generally occur since the melting temperature is higher than the environmental condition.

Moreover, plastic can undergo chemical degradation when polymers are in contact with chemicals, such as acid, base, solvent, and reactive gas (Klein et al., 2018; Wilczura-Wachnik). Chemical reactions involved in the deterioration of plastic are typical oxidation and hydrolysis (Matsumura, 2005). Oxidation strips out the side groups of polymers and produces acidic degradation products (Gewert et al., 2015). The oxidized polymers are prone to further degradation, and the acid catalyze can cause crazing to the material. Oxidation reactions can also be UV-induced or hydrolytically. Some polymers containing ester and ether groups are prone to hydrolytic degradation when submerged in water because these functional groups are covalently bonded (Engineer et al., 2011). In the hydrolytic reaction of polymers, the cleavage of the ester bond is controlled by the rate constant, the amount of absorbed water, the diffusion coefficient of chain fragments, and the solubility of the degraded product (Engineer et al., 2011). It results in a decrease in molecular weight and mechanical strength.

After physical and structural degradation, polymers are more subject to biodegradation because of the larger surface area for microorganisms to colonize. Extracellular enzyme-forming monomers or oligomers depolymerize plastic in aerobic and anaerobic conditions (Fotopoulou & Karapanagioti, 2019; Klein et al., 2018). After polymers are broken down into smaller molecules which can be assimilated into microbial cells, polymers can be depolymerized by intercellular enzymes and mineralized by microorganisms (Shah et al., 2008). The process of mineralization is the degradation of polymer into the end products such as CO_2, H_2O, and CH_4, which bacteria will later utilize (Shah et al., 2008). Moreover, different groups of bacteria can degrade different types of plastics. For example, *Brevibacillus borstelensis* is able to degrade PE (Hadad et al., 2005).

Plastic debris on beaches is directly exposed to UV radiation and high oxygen availability (Cole et al., 2011). The degradation rate of such an environment is higher than plastic floating in the ocean (Browne et al., 2007b). Plastic particles on the beach degrade rapidly, turning brittle and yellow, and due to loss of structural integrity, they are more susceptible to abrasion from winds and waves (Barnes et al., 2009; Cole et al., 2011). However, adding chemical additives such as UV, heat stabilizers, and anti-oxidants can retard light-induced degradation (Rochman et al., 2016).

Plastic waste disposed of in the environment can be degraded or broken down into MPs by various factors i.e., physical, chemical, and biological processes. Due to the large amount of plastic waste generated, the presence of MPs is ubiquitous in every environmental matrix.

2.3.4 Environmental impacts

Due to its widespread ubiquity of MP in the marine environment has raised scientific concern about the adverse effects of MP particles. The size is small, so they become bioavailable to a wide range of organisms through ingestion (Cole et al., 2011). Animals like birds and turtles mistake plastic particles for food, and there is a trace of plastic ingestion in more than 44% of marine bird species (Hohenblum et al., 2015). In addition, more than 35% of the fish sample from the North Pacific central gyre had MPs in the guts(Johansson & Ericsson, 2018). The effects of plastic ingestion include blockage of the intestinal tract, inhibition of gastric enzyme secretion, reduced feeding stimuli, decreased steroid hormone levels, delays in ovulation, and failure to reproduce (Li et al., 2016).

Moreover, some toxic effects might be associated with ingested MP particles from the leaching of chemical additives and the adhered pollutant on the MP surface (Cole et al., 2011). Persistent organic pollutants (POPs) are one of the toxic pollutants which can coat MP surfaces (Johansson & Ericsson, 2018). Since MPs can travel long distances, POPs are transported with them and pollute far-away ecosystems (Johansson & Ericsson, 2018). Moreover, POPs can transfer to humans through seafood consumption since MPs are found in fish, mollusks, and crustaceans (Johansson &

Ericsson, 2018). For example, a significant amount of MP was found in 3 species of sessile invertebrates collected from the eastern coast of Thailand (Thushari et al., 2017).

2.4 MP contamination in WWTPs

There is evidence that MPs are found in WWTPs. Potential sources of MPs in WWTPs, the removal of MPs by WWTPs, and the retention of MPs in the sludge are discussed in this section.

2.4.1 Sources of MPs in WWTPs

It has been reported that there is a widespread distribution of MPs in freshwater ecosystems. MPs can be from marine and land-based origins. One of the land-based sources of MPs is wastewater effluents. Many studies suggested that WWTPs are pathways for MPs to enter water bodies as MPs were found in wastewater effluent (Conley et al., 2019; Murphy et al., 2016; Ziajahromi et al., 2017). The study by Browne et al. (2011) found that MP samples from shoreline sediment and wastewater effluent disposal sites along the shoreline are of similar types, mostly polyester and acrylic fiber (Browne et al., 2011). Moreover, Talvitie et al. detected similar types of fibers and synthetic particles in both tertiary effluent from the wastewater treatment plant and seawater from the Gulf of Finland (Talvitie et al., 2015).

WWTPs receive terrestrial MPs before releasing to the aquatic environment. Thus, MPs in the wastewater treatment system come from various sources. Primary MPs in the form of microbeads in WWTPs mainly originate from cosmetics and personal care products such as toothpaste, soap, and scrub (Carr et al., 2016). Microbeads are mostly made of PE (Napper et al., 2015), and they are directly rinsed down household drains and enter the wastewater treatment system (Carr et al., 2016; Ngo et al., 2019). Up to 94,500 microbeads can be released from a single use of an exfoliant (Napper et al., 2015). Many countries have started to ban the use of microbeads in consumer products (Conkle et al., 2018). Industrial scrubbers are another form of primary MPs present in WWTPs. They are used in blasting clean surfaces, molding, and other processes, and are discharged directly into the wastewater collection system (Ngo et al., 2019).

Secondary MPs from the fragmentation of larger plastics are also found in WWTPs. MP fibers and filaments in wastewater treatment systems originate from the breakdown of synthetic textiles during washing (Hernandez et al., 2017). The major types of MP fibers in WWTPs are polyester, acrylic, and polyamide (Browne et al., 2011; Hernandez et al., 2017). A single garment wash can shed more than 1,900 fibers (Browne et al., 2011). In addition, a study by Napper and Thompson (2016) showed that an estimated 700,000 fibers are released by washing 6 kg of textiles from acrylic fibers (Napper & Thompson, 2016). Clothing can shed more fibers with higher washing temperatures and using detergent (Yang et al., 2019b). Fibers become problematic because of their higher volume-to-area ratio than other types of MPs (Astrom, 2016). MP fibers are likely to adsorb more chemicals and transfer them to living organisms.

Some secondary MPs are created by the breakdown of packaging, textile, and tires in concrete and highway construction (Kole et al., 2017). These MPs are present in the atmosphere and enter WWTPs through the wet sedimentation process (Ngo et al., 2019). Tire and road particles are considered important sources of MPs in the environment (Lassen et al., 2012). These particles are dispersed in the environment via several pathways: air emission, transportation by rainwater runoff into the soil, sewer systems, and surface water (Kole et al., 2017). Another source of MPs in sewer systems is landfill leachate. Plastic waste is buried in landfills under severe environmental conditions causing fragmentation into MPs (He et al., 2019). These MPs enter WWTPs through the discharge of leachate.

2.4.2 Function of wastewater treatment plants

WWTPs receive wastewater from households, businesses, industries, and sometimes urban runoff through combined sewer systems. WWTPs are designed to remove wastewater's solid debris, nutrients, and other organic pollutants. A conventional wastewater treatment system consists of preliminary, primary, and secondary treatment steps. Tertiary treatment is sometimes employed to improve the quality of treated wastewater.

The objective of preliminary treatment is to remove large solids and other materials in raw wastewater (Sonune & Ghate, 2004). Coarse and fine screens are responsible for diminishing large debris (Duis & Coors, 2016). Removed solids at this stage consist of wood, cloth, paper, plastics, garbage, and sometimes fecal matter (Sonune & Ghate, 2004). Sizes of coarse screen range from 6-150 mm, and fine screens are smaller than 6 mm in different WWTPs (Iyare et al., 2020). Grit chambers in preliminary treatment remove sand and other heavy particles (Tang & Hadibarata, 2021). Primary treatment is designed to remove organic and inorganic solids by sedimentation and floatation (Sonune & Ghate, 2004). Settleable solids are removed in a sedimentation tank in primary treatment. As a result, the total suspended solid of wastewater is reduced in this step (Westphalen & Abdelrasoul, 2018). Air floatation utilizes air bubbles to enhance the float of contaminants, such as solids and fibers, and allows them to be captured with oil and grease during mechanical skimming (Ngo et al., 2019).

The secondary treatment utilizes biological processes to remove further suspended solids and nutrients (Mason et al., 2016). These processes can efficiently eliminate biodegradable organic matter (Crini & Lichtfouse, 2019). Biological processes commonly used in secondary treatment in WWTPs include the activated sludge process, trickling filter, and rotating biological contractors (Westphalen & Abdelrasoul, 2018). Flocculation is a wastewater treatment process for suspended solids removal (Picos-Corrales et al., 2020). The formation of flocs is induced by a variety of flocculation agents such as polyacrylamide (PAM), aluminum salt polymers ($Al_2(SO_4)_3$), and iron salt polymers ($FeSO_4$; $FeCl_3$) (Li et al., 2020; Picos-Corrales et al., 2020; Zhang, 2014). Flocs settle to the bottom of the tank during the sedimentation process and are removed. Sewage sludge is a residue from the settling tank of wastewater treatment processes. There are several types of sludge treatment, e.g., lime stabilization, anaerobic digestion, composting, and thermal drying, before the sludge is applied to agricultural lands (Mahon et al., 2017).

Subsequently, tertiary treatment is introduced in some wastewater treatment plants. This step is specially designed to remove specific inorganic and organic pollutants to improve the discharge quality of treated wastewater (Iyare et al., 2020). Pollutants further removed by tertiary treatment are nutrients, pathogens, non-biodegradable compounds, heavy metals, dissolved solids and suspended solids, and micropollutants (Bassin et al., 2021). Thus, the final effluent meets a higher standard and can be reused for specific purposes such as irrigation, recreation, and drinking water (Gerba & Pepper, 2019). Types of tertiary treatment processes involve physicochemical processes such as coagulation, filtration, carbon adsorption, reverse osmosis, and disinfection (Gerba & Pepper, 2019).

2.4.3 Removal of MPs by WWTPs

Each treatment unit has different functions for removing specific contaminants. The effect of each wastewater treatment step on the removal of MPs is presented in this section.

2.4.3.1 Preliminary and primary treatment

Preliminary and primary treatments remove MPs based on physical mechanisms. A study by Murphy et al. showed that the grit and grease removal stage can efficiently eliminate MPs from raw wastewater (Murphy et al., 2016). Air bubbles from air floatation technology allow low-density MPs to float (Ngo et al., 2019). Lightweight MPs are captured along with the floating grease during skimming (Sun et al., 2019). Microbeads made of polyethylene PE are positively buoyant, thus, they can be easily skimmed off at the surface layer (Murphy et al., 2016). Ziajahromi et al. (2021) showed 69-79% MP removal by screening and grit removal stage (Ziajahromi et al., 2021). Primary treatment consecutively removes MPs by sedimentation process. Fibers and large-sized MPs can be easily separated by trapping in solid flocs and settling heavy particles during primary treatment (Liu et al., 2021). Primary sedimentation significantly removed fibers rather than synthetic particles (Talvitie et al., 2015). These findings correspond to a study by Carr et al. that fibers and MP fragments derived from personal care products were largely removed during the skimming and settling stages (Carr et al., 2016). On the contrary, Liu et al. (2021) found that grit and grease removal is not an effective step for MP removal. In contrast, the primary settling stage exhibited excellent removal efficiency (Liu et al., 2021). However, the removal efficiency at this stage also depends on the density and shapes of MPs in raw wastewater (Ngo et al., 2019).

The combination of grit and grease removal with the primary sedimentation could improve the removal rate of MPs (Liu et al., 2021). The efficiency of sedimentation together with an aerated grit chamber in removing MPs was 40.7% in Wuhan, China (Liu et al., 2019). The sedimentation process reduced MPs by up to 71.67% in Beijing, China, and 91.7% in Vancouver, Canada (Gies et al., 2018). Two studies achieved higher efficiencies from the sedimentation process due to denser polymers of MPs, such as PET and polyester, which can be easily eliminated by physical sedimentation (Ngo et al., 2019). On the other hand, low-density MPs (PE and PP) and moderate-density MPs (PS and polyamide: PA) can float at the surface of the sedimentation tank by air floatation technology (Ngo et al., 2019). Therefore, the density of MP particles significantly influences the removal by sedimentation and skimming processes.

The morphology of MPs is another factor that influences the removal rate. Fibers are considered the most shape of MPs to remove from wastewater (Long et al., 2019). On the contrary, fragments and granules are the most straightforward shape to eliminate from the wastewater stream (Long et al., 2019). The surface of fibers and pellets are smoother than other shapes, which makes them less resistant to wastewater and more difficult to be captured by treatment technologies (Anderson et al., 2018), whereas the angular, bifurcate. The twisted shape of fragments and granules increases the ability to be captured in solid flocs and the chance of microbial colonization for a higher degree of sedimentation and degradation (Ngo et al., 2019).

Fibers are considered the most challenging type of MPs to retain in WWTPs. Even though they are trapped during flocculation and settling, due to their longitudinal shape, fibers can easily escape treatment processes (Liu et al., 2021). The neutral buoyant property of fibers also hinders the removal by the skimming process (Ngo et al., 2019). Studies found that fibrous MPs are the dominant shape in wastewater effluent (Zhang et al., 2021; Ziajahromi et al., 2017; Zou et al., 2021). Since WWTPs cannot completely remove this type of MPs regarding its nature, it is crucial to focus on reducing fibers at sources.

2.4.3.2 Secondary treatment

The secondary treatment step employs biological processes to reduce further suspended and dissolved solids remaining in wastewater from primary treatment (Iyare et al., 2020). MPs are captured by the accumulation of MPs in the sludge flocs (Ngo et al., 2019). Flocs containing MPs are eliminated after settling in the clarification tanks. Flocculation agents added in secondary treatment enhance the aggregation of suspended solids and capture MPs in the sludge flocs (Murphy et al., 2016; Sun et al., 2019). However, MPs trapped in unstable flocs might result in a redistribution of particles in the tank and subsequent escape during clarification (Carr et al., 2016). Factors influencing MP removal in activated sludge processes are retention time and nutrient levels (Carr et al., 2016; Rummel et al., 2017). Smaller MPs (106-300 µm) have a higher removal rate by secondary treatment than larger MPs (>300 µm) because small particles are easily adsorbed to sticky media such as biofilm or floc (Lee & Kim, 2018).

However, some configurations of secondary treatment processes do not remove MPs efficiently. For example, Anaerobic-anoxic-oxic (A^2O) process is the most widely used system in WWTPs. A^2O showed a relatively low removal rate of MPs due to the sludge being returned, and about 20% of MPs attached to the sludge would return to the aqueous phase (Liu et al., 2021). The biodegradation rate of MPs in A^2O is low due to the short hydraulic retention time, which is ineffective for microbial degradation (Liu et al., 2021). Therefore, this study concluded that a conventional activated sludge process is not ideal for MP removal.

2.4.3.3 Tertiary treatment

Tertiary treatments, including variations of filtration processes, further removed MPs by 5-20% after the secondary treatment (Iyare et al., 2020). The application of tertiary treatment technologies can increase the overall MP removal efficiency of WWTPs by 10-97% (Sun et al., 2019). MP removal efficiency of a WWTP with membrane bioreactor (MBR) technology was reported as 99.9% (Talvitie et al., 2017a). Another study with a pilot scale MBR also achieved a 99.4% MP removal rate with a pore size of 0.4 µm (Lares et al., 2018). However, a study by Lv et al. (2019) reported

99.5% removal based on MP mass, but only 82.1% of the number of MPs were removed (Lv et al., 2019). Bayo et al. (2020a) demonstrated a 79.01% MP removal rate of a WWTP with MBR. It was concluded that advanced technologies did not perform better than conventional systems (Bayo et al., 2020a). Adsorption is essential in MP removal within the MBR system (Liu et al., 2021).

Membrane filtration, such as ultrafiltration (UF), is another technology in tertiary treatment that helps reduce the concentration of MPs by intercepting larger MPs than the pore size of membranes (Liu et al., 2021). The sizes of MPs are generally larger than the pore size of the membrane; thus, the UF completely rejects MP particles (Ma et al., 2019). Hydrophobic interactions and surface repulsion forces control the adsorption rate of MPs on membrane surfaces (Enfrin et al., 2020). Several studies reported that WWTPs equipped with UF units attained more than 95% removal efficiencies (Mintenig et al., 2017; Yang et al., 2019a; Ziajahromi et al., 2017). UF, together with reverse osmosis (RO), completely eliminated particles >190 µm, while smaller particles were retained in the tertiary effluent (Ziajahromi et al., 2017). However, membrane fouling was induced by pore blockage from MPs (Enfrin et al., 2020). Therefore, membrane fouling must be strictly controlled to ensure the long-term operation of membrane filtration (Kumar & Ismail, 2015).

Biofilters are another tertiary technology designed to degrade specific dissolved pollutants such as pharmaceuticals (Zhang et al., 2019). A study by Liu et al. (2020) reported that biofilter as a polishing step completely removed MPs larger than 100 µm (Liu et al., 2020). Liu et al. (2021) showed that biofilters had the highest removal performance of MPs among other treatment technologies (Liu et al., 2021). MPs in a biofilter process are removed by the mechanisms of biofilm filtration and adsorption (Liu et al., 2021). MPs and excess microbes were easily eliminated from the biofilter by backwashing in the ascendant water flow (Rocher et al., 2012).

Sand filtration is filtration-based technology used in tertiary treatment. Sand filtration was found to reduce MP concentration from the secondary effluent by 50% (Magni et al., 2019). A WWTP with rapid sand filtration had a removal rate of 97% (Talvitie et al., 2017a). It is also considered a simple and cost-effective method compared to membrane filtration (Iyare et al., 2020). However, a study by Carr et al.

(2016) found that the effect of the gravity sand filter on MP removal was minimal (Carr et al., 2016). This study concluded that MPs were mainly removed in primary treatment during the skimming and settling.

Advanced oxidation processes, including chlorination and UV oxidation, are commonly used in WWTPs. The chlorination process increased MP abundances in WWTPs by cracking MPs from the attack of chlorine (Kelkar et al., 2019; Lv et al., 2019; Ruan et al., 2019). Chlorination altered MPs' physical and chemical properties as chlorine is a potent oxidizing agent (El-Shahawi et al., 2010). This change in the carbon-chlorine bond of MPs results in a higher adsorption and accumulation rate of other harmful contaminants (Wang et al., 2018). UV oxidation changed MP topography and chemical characteristics (Cooper & Corcoran, 2010). UV-oxidation at the surface of MPs produced cracks and flakes, which were easy to break into more minor MPs and nano-plastics (Cai et al., 2018). However, the toxicity of UV-oxidized MPs is still unknown. Ozone is another oxidation process for removing inorganic ions and refractory organic pollutants in wastewater (Hidayaturrahman & Lee, 2019). Ozone shows a good influence on polymer degradation. A degradation rate of more than 90% was reached after a 30-min exposure to ozone (Chen et al., 2018). It has been suggested that even with the high cost of ozone as a strong oxidant, treated water has a lower quantity of residue (Chen et al., 2018). Hidayaturrahman and Lee (2019) found that a WWTP with ozone technology had the highest percentage removal of MPs among other WWTPs with disc-filter and rapid sand filtration at the tertiary treatment step. The ozonation reduced 89.9% of MPs after the coagulation process (Hidayaturrahman & Lee, 2019). Fitri et al. (2021) detected weight loss and structural changes in PE treated with ozone and suggested ozonation as a pre-treatment step prior to the biodegradation of PE MPs (Fitri et al., 2021).

On the contrary, not all types of advanced and tertiary treatments are suitable for MP removal. A study found that MP concentration in the effluent from tertiary granular filtration was not lower than the secondary WWTPs (Sutton et al., 2016). Talvitie et al. (2017b) found no significant impact of active biological filters on MP reduction (Talvitie et al., 2017b). Small-sized MPs (20 µm to 100 µm) escaped all treatment stages and were released into recipient water (Cesa et al., 2017).

2.4.4.4 Major treatment units for MPs removal

Several studies suggested that skimming and settling processes are the most effective unit for MP removal (Carr et al., 2016; Murphy et al., 2016; Zhang et al., 2021). Large-sized fragments tend to settle during primary sedimentation (Liu et al., 2021). Due to the positive buoyancy, PE fragments derived from personal care products can be easily skimmed during grease removal (Murphy et al., 2016). On the contrary, some studies reported that biological processes in secondary treatment play an important role in MPs removal (Jiang et al., 2022; Yang et al., 2021a). The removal rate of MPs during this stage can be as high as 95.2% (Jiang et al., 2022). However, the most effective step for MP retention is still controversial. The secondary treatment removed < 20% of MPs in primary effluent (Okoffo et al., 2019). Anaerobic-anoxic-oxic (A^2O) process attained a lower removal rate than the activated sludge process (Liu et al., 2019; Yang et al., 2019a; Ziajahromi et al., 2017). In the A^2O process, MPs are likely to return to the system when sludge is recycled (Liu et al., 2021). On the contrary, a study showed no significant difference in MP removal from three configurations of the activated sludge process (Lee & Kim, 2018). Therefore, the results of removal efficiency by secondary treatment are still uncertain.

The tertiary treatment acts as a final polishing step which can increase the overall percentage removal of WWTPs (Talvitie et al., 2017a). In this stage, MPs with specific properties and very small particle sizes are efficiently removed (Liu et al., 2021). No MP was found in the effluent of a WWTP with tertiary treatment in a study by Carr et al. (Carr et al., 2016). MBR has become the most popular technology for removing contaminants regarding its high removal capability, and it is the most efficient MP removal method among other advanced technologies (Hamidian et al., 2021; Ngo et al., 2019). MBR technology has been suggested as the most cost-effective method for MP removal (Vuori & Ollikainen, 2022). Despite an effective removal by MBR, the WWTP still releases 800,000 fibers daily (Michielssen et al., 2016). Fibers can pass through a filter with a pore size as small as 0.08 µm (Leslie et al., 2017). Small-sized MPs (20-100 µm) bypassed every treatment step, including tertiary treatments (Salvador Cesa et al., 2017).

2.4.4 MP abundances and removal efficiencies of WWTPs

The abundance of MPs in WWTPs is influenced by various factors such as population served, type of wastewater (municipal or industrial), economy, and lifestyle of surrounding communities (Liu et al., 2021). The population is positively correlated to the abundance of MPs in WWTPs (Zou et al., 2021). A higher number of MPs were detected in municipal and industrial WWTPs than in municipal WWTPs (Liu et al., 2021). Human activities in the served catchment, such as wearing synthetic clothes and using plastic products, affect MP discharge from households to WWTPs (Sun et al., 2019). In addition, factors such as the combined sewer system, flow rate of the WWTP, and tertiary filtration also affect the number of MPs in the effluent (Mason et al., 2016). MPs from other sources, such as combined sewer overflows and runoff, possibly enter the WWTPs during the peak flow rate (Hamidian et al., 2021). The variability of MP concentration also depends on sampling, isolation, and detection method, as well as the smallest mesh size (Iyare et al., 2020). A study using a 10 μm filter reported a significantly higher number of MPs (Vollertsen & Hansen, 2017). Thus, comparing different studies is still challenging due to a lack of standardized methods.

Several studies have reported negligible MP concentrations (lower than 0.5 MPs/L) in the WWTP effluents (Lv et al., 2019; Razeghi et al., 2021). However, a large volume of treated wastewater is discharged daily. Murphy et al. estimated the amount of MP entering the aquatic environment to be 6.5×10^{7} per day (Murphy et al., 2016). In China, approximately 6.5×10^{8} MPs are discharged daily from seven WWTPs into Xiamen Bay (Long et al., 2019). Municipal WWTPs in the US were estimated to discharge approximately 4 million MPs/facility/day (Mason et al., 2016). Higher MP concentrations were detected downstream from WWTP effluent outfall sites in nine rivers in the US (McCormick et al., 2016).

Table 2.1 summarizes studies on MPs in the WWTPs and their removal efficiencies by different treatment technologies. Removal efficiencies range from 40 to 99.9% with different lowest mesh sizes. A study by Carr et al. (2016) found that the removal efficiency of WWTP was 99.9% from secondary treatment effluent, and no MP particle was found in the effluent of tertiary treatment (Carr et al., 2016). It is also further suggested that secondary and tertiary treatment processes significantly contribute to MP removal. In addition, the investigation of Mintenig et al. found that a WWTP where there is additionally installed post-filtration can remove up to 97% of MP pollution.

On the other hand, WWTPs without this technology showed a considerable number of MPs (Mintenig et al., 2017). Murphy et al. (2016) examined a WWTP with only primary and secondary treatment and demonstrated a 98.41% removal by skimming and settling stage (Murphy et al., 2016). On the contrary, a WWTP with an oxidation ditch showed relatively low removal based on MP number (53.6%) but 97% based on mass (Lv et al., 2019). An MBR system from a study by (Lv et al., 2019) reduced 82.1% of MPs based on the number and 99.5% based on mass. Moreover, there is a variation in the removal percentage of a WWTP with a disc filter (40-98.5%) (Talvitie et al., 2017a). Due to the different lowest ends of the mesh and sampling methods, there might be variability in the results.

Table 2.1 Comparison of studies on MPs removal from WWTPs in different countries

Location	Type of facility	Mesh size	Influent (MPs/L)	Effluent (MPs/L)	Removal efficiency	References
US	Seven tertiary treatment plants and one secondary treatment plant	0.4 to 0.02 mm	N/A	0.0088 to not detected	99.9% in tertiary plants	Carr et al. (2016)
Scotland	Primary and secondary treatments	0.065 mm	15.70±5.23	0.25±0.04	98.41%	Murphy et al. (2016)
Finland	WWTPs with different tertiary treatments (A) Membrane bioreactor (MBR) (B) Rapid sand filter (C) Dissolved air floatation (D) Disc-filter	0.3, 0.1, and 0.02 mm	(A) 6.9±1.0 (B) 0.7±0.1 (C) 2.0±0.07 (D) 0.5-2.0	(A) 0.005 (B) 0.02 (C) 0.1 (D) 0.03-0.3	(A) 99.9% (B) 97% (C) 95% (D) 40-98.5%	(Talvitie et al., 2017a)
Germany	12 WWTPs 6 with primary skimming tanks, 4 with tertiary treatment	0.01 mm	N/A	0-0.05 (MPs>0.5 mm) 0.01-9 (MPs<0.5 mm)	97% with post-filtration	Mintenig et al. (2017)
Australia	3 WWTPs (A) Only primary treatment (B) Primary and secondary processes with UV disinfection (C) Tertiary treatment with disinfection, ultrafiltration, reverse osmosis (RO), and decarbonation	0.5, 0.19, 0.1, 0.025 mm	N/A	(A) 1.5 (B) 0.48 (C) 0.28 after UF and 0.21 after RO	90% after tertiary treatment [a]	Ziajahromi et al. (2017)
China	Two parallel treatment systems 1. Oxidation ditch	0.5, 0.25, 0.125, 0.625, and 0.025 mm	0.28±0.02	1. 0.13±0.01 2. 0.05±0.01	1. 53.6% (97%)[b] 2. 82.1% (99.5%)[b]	Lv et al. (2019)

	2. Membrane bioreactor (MBR)					
China	A conventional WWTP with an activated sludge process	0.047 mm	79.9	28.4	64.4%	Liu et al. (2019)
Italy	Pre-treatment Primary treatment Secondary treatment Tertiary treatment (sand filter)	5, 2, and 0.063 mm	2.5±0.3	0.4±0.1	84%	Magni et al. (2019)
Netherlands	7 WWTPs	0.3 to 0.01 mm	68-910	51-81	72%	Leslie et al. (2017)
South Korea	3 WWTPs with tertiary treatments (A) coagulation and ozone (B) coagulation membrane disc-filter (C) coagulation and rapid sand filtration	0.0012 mm	(A) 4,200 (B) 31,400 (C) 5,840	(A) 33 (B) 297 (C) 66	(A) 99.2% (B) 99.1% (C) 98.9%	Hidayaturrahman and Lee (2019)
Turkey	2 WWTPs with primary and secondary treatments	0.055 mm	(A) 26.55±3.17 (B) 23.44±4.10	(A) 6.99±0.76 (B) 4.11±0.32	(A)73% (B)79%	Gündoğdu et al. (2018)
Turkey	3 WWTPs (A) Anaerobic-anoxic-oxic (B) primary and secondary treatments (C) primary and secondary treatments	0.026 mm	(A) 3.1 (B) 2.6 (C) 1.5	(A) 1.6 (B) 0.7 (C) 0.6	(A) 48% (B) 73% (C) 60%	Akarsu et al. (2020)
Spain	A WWTP with an activated sludge process	0.00045	14.23±2.70	1.23±0.15	90.1%	Bayo et al. (2020b)
Spain	A WWTP with an A^2O process	0.375 to 0.025 mm	171±43 [c]	10.7±5.2	93.7%	Edo et al. (2020)

[a] the number of MPs from primary effluent was used for calculation.

[b] based on plastic mass

[c] primary effluent

2.4.5 Retention of MPs in sewage sludge

MPs removed from wastewater are retained in the sludge through both primary and secondary treatment sedimentation. MP concentration in sewage sludge has been reported from 1,000 to 240,300 items/kg (dry weight) (Okoffo et al., 2019). Sewage sludge treatments, such as anaerobic digestion, thermal drying, and lime stabilization, are performed before land application or disposal. A study found a lower MP concentration in the sludge after the anaerobic digestion, whereas lime stabilization increased the number of MPs in the sludge due to the shearing effect (Mahon et al., 2016). Moreover, MPs were detected in bottom ash, a by-product from an incinerator (Yang et al., 2021b).

Sewage sludge is considered one of the important sources of MPs in the terrestrial environment (Bläsing & Amelung, 2018). Studies show evidence of MP spread in agricultural soil after sludge application. Microbeads and fibers, similar to those found in WWTPs, were present in sludge-amended soil (Chen et al., 2020). More than 30,000 tons of MPs enter the agricultural soil of Europe and North America every year (Nizzetto et al., 2016). MPs in soil negatively impact flora and fauna in the soil environment (Kumar et al., 2020). Soil nutrient cycling is also altered by this type of pollutant (Huang et al., 2022). Moreover, MPs tend to accumulate at the roots of some edible plants and transport them to leaves (Li et al., 2021). Chemicals in MPs pose a threat to human health when transferred through the food chain (Kumar et al., 2020).

2.5 Methods for MP analysis

Currently, there is no standardized method for sampling and analysis of MP from wastewater and sludge. Different researchers used a variety of sampling equipment/methods. The procedures for sample preparation also differed. Lastly, various techniques for MP characterization are discussed.

2.5.1 Sampling methods

Various sampling methods have been employed in MP collection from wastewater samples (Table 2.2). The smallest mesh sizes are also varied in different studies. Different containers, such as steel buckets, glass jars, and Ruttner samplers, were used for grab sampling to collect wastewater samples (Gies et al., 2018; Lares et al., 2018; Magni et al., 2019; Magnusson & Norén, 2014; Murphy et al., 2016). Container collection is suitable for wastewater influents due to high organic and solid loads (Talvitie et al., 2017b). Pumping coupled with filtration has been widely used to obtain a large volume of wastewater effluent (Hu et al., 2019). Several studies utilized a stacked stainless-steel mesh to collect MP samples based on their size distribution (Carr et al., 2016; Dyachenko et al., 2016; Ziajahromi et al., 2017). A stack of different sieve sizes assembled from coarse to fine is used for on-site filtration or in a laboratory to isolate MP particles. A study by Talvitie et al. (2015) developed a filter tube with 200 and 100 µm connected to a centrifugal pump (Talvitie et al., 2015). An automatic sampler has been used in some studies to yield representative samples (Dris et al., 2015; Simon et al., 2018). In addition, surface filtration was adopted for MP sampling by skimming the water surface at the effluent discharge (Carr et al., 2016). The range of mesh sizes used in collecting MPs from wastewater matrices was from 1 to 500 µm (Hamidian et al., 2021). A filter with a 300 µm mesh size has been commonly used for microliter sampling regarding the size of plankton nets (Magnusson & Norén, 2014). Mesh or pore sizes of sieves, filters, and sampling devices influence the number of collected MPs (Magnusson & Norén, 2014). The selection of mesh size for MP sampling should be decided carefully.

Table 2.2 A summary of sampling methods for collecting MPs from wastewater treatment plants

Method	Device	Smallest mesh size	References
Grab sampling	Glass jar	63 µm	Gies et al. (2018)
	Steel bucket	63 µm	Gies et al. (2018)
		0.25 mm	Lares et al. (2018)
		63 µm	Magni et al. (2019)
	Ruttner sampler	300 µm	Magnusson and Norén (2014)
Filtration	Pumping coupled with filtration	0.125 mm	Mason et al. (2016)

	Filter tube connected to a pump	100 µm	Talvitie et al. (2015)
	Stacked units of stainless-steel mesh screens	25 µm	Ziajahromi et al. (2017)
		20 µm	Carr et al. (2016)
		0.125 mm	Dyachenko et al. (2016)
Automatic sampler	Teledyne ISCO Glacier Portable Water Sampler	63 µm	Gies et al. (2018)
	ISCO 3700	20 µm	Talvitie et al. (2017b)
Surface filtration	Skimming surface water at effluent discharge by a filtering assembly	125 µm	Carr et al. (2016)

MPs are present in sewage sludge because they are transferred from wastewater to the solid phase. Thus, the collection of sludge samples is conducted to investigate MP retention. Filtration of sewage sludge cannot be conducted directly due to its viscous matrix containing organic and inorganic materials and microorganisms (Zhang & Chen, 2020). Thus, sludge samples are commonly collected in small amounts from glass jars or beakers from sludge treatment units (Koyuncuoğlu & Erden, 2021).

2.5.2 Sample preparation

Wastewater samples are organic-rich matrices that contain MPs. Pre-treatment is required to isolate MPs from wastewater samples. The following steps prepare MP samples prior to spectroscopic analysis.

2.5.2.1 Purification

MP particles in the environment are attached to natural organic and inorganic materials. Removing these particles before spectroscopic analysis (Löder & Gerdts, 2015a). Using a strong oxidizing agent to remove biological material can degrade or damage plastic samples (Lusher et al., 2017). The purification processes of MP samples can be divided into chemical degradation and enzymatic degradation (Li et al., 2018a). The most widely used technique for MP purification is wet peroxide oxidation (WPO) by hydrogen peroxide (H_2O_2) (Li et al., 2018a; Magni et al., 2019; Tagg et al., 2015; Ziajahromi et al., 2017). However, exposure to H_2O_2 may not suffice for removing organic residue in large volumes of wastewater (Tagg et al., 2017). The combination with an iron catalyst, the so-called Fenton's reaction, was effectively used to extract MPs from organic-rich wastewater and sludge without altering MP shapes and chemistry (Hurley et al., 2018; Tagg et al., 2017). Fenton’s reagent also showed a higher MP recovery rate than other alkaline-based digestion, such as NaOH and KOH (Hurley et al., 2018).

An alternative for purifying MP samples is enzymatic digestion. Specific enzymes, including protease, cellulase, and chitinase, are used to eliminate lipids, protein, and carbohydrates in the environmental samples (Löder et al., 2017). It showed a high recovery rate without damaging plastic material.

2.5.2.2 Density separation

Density separation is an approach aiming to separate MP from non-plastic materials. It is the most widely used method to separate MPs from inorganic substances, such as sediment and sand (Li et al., 2018a). The initial separation can be done by adding a saturated salt solution. Densities of most commercial plastics are approximately 0.8-1.4 g/cm^3, and salt solution is slightly denser than plastic which makes them float due to supernatant, while high-density particles are likely to sink to the bottom (Li et al., 2018a; Ribeiro Claro et al., 2016). MPs can be separated by collecting supernatant particles.

Sodium Chloride (NaCl: 1.2 kg/L) is the suggested salt solution by Thompson et al. (2004) for density separation of MPs, and it is the most commonly used brine solution among all due to low-cost, availability, and environmental-friendliness

(Hamidian et al., 2021). However, the NaCl solution performed better in extracting low-density MPs than high-density particles (Claessens et al., 2013). Zinc Bromide ($ZnBr_2$: 1.7 kg/L) showed the highest MP recovery rate in the segregation of denser MPs among tested brine solutions (Quinn et al., 2017). However, $ZnBr_2$ is costly and hazardous to the environment (Quinn et al., 2017). Sodium Iodide (NaI: 1.6-1.8 kg/L) exhibited a similar recovery rate to $ZnBr_2$, but it is cheaper and more environmentally friendly (Quinn et al., 2017). Moreover, NaI solution is recyclable without any changes in density by filtration (Kedzierski et al., 2017). The use of NaI can significantly reduce the cost of MP isolation. Zinc chloride ($ZnCl_2$: 1.5-1.8 kg/L) is also suggested for density separation due to the recyclability by pressure filtration (Löder & Gerdts, 2015b). $ZnCl_2$ is an alternative brine solution for environmental and economic reasons.

Density separation is generally performed after purification for wastewater samples, whereas density-based separation is completed before the step of purification for sludge samples. A summary of sample preparation methods for wastewater and sludge samples is shown in Table 2.3.

Table 2.3 Purification and separation methods for MP samples from wastewater and sludge

Sample type	**Purification**	**Density separation**	**References**
Wastewater and sludge	30% H_2O_2	Canola oil	Gies et al. (2018)
Sludge	30% H_2O_2	NaCl	Li et al. (2018b)
Wastewater and sludge	15% H_2O_2	NaCl	Magni et al. (2019)
Wastewater	30% H_2O_2	-	Tagg et al. (2015)
Wastewater	30% H_2O_2	NaI	Ziajahromi et al. (2017)
Wastewater	30% H_2O_2 with 0.05 M $FeSO_4$	-	Dyachenko et al. (2016)

Wastewater	30% H_2O_2 with 0.05 M $FeSO_4$	NaI	Gündoğdu et al. (2018)
Wastewater and sludge	30% H_2O_2 with 0.05 M $FeSO_4$ and cellulase enzyme	-	Lares et al. (2018)
Wastewater	30% H_2O_2 with 0.05 M $FeSO_4$	$ZnCl_2$	Lee and Kim (2018)

2.5.2.3 Nile Red staining

Large-sized MPs are visually sorted under an optical microscope, but it requires expert knowledge and is time-consuming (Maes et al., 2017). The staining approach allows a quick and inexpensive quantification of MPs in various environmental matrices (Tamminga et al., 2017). MP staining has been proposed as a complementary method for enumeration to avoid the risk of over- or underestimation (Kershaw et al., 2019). Nile Red is a lipophilic dye that was first used in microbiology (Tamminga et al., 2017). Due to the hydrophobic properties of plastics, they are fluorescent when they are excited with certain wavelengths (Nalbone et al., 2021). Chloroform has been tested as the most suitable solvent for Nile Red as it achieved the highest recovery rate of Europe's most demanded polymer types (Tamminga et al., 2017). Stained filter papers are identified under a fluorescence microscope to enumerate MP abundances at each sampling location.

2.5.3 MP characterization

After sampling and pre-treatment prior to spectroscopic analysis, MPs will be characterized and identified based on morphology, such as size, shape, color, chemical composition, and concentration (Rocha-Santos & Duarte, 2017). Various methods can be used for identification.

2.5.3.1 Visual Identification

Visual identification or visual sorting can be made by the naked eye or under optical microscopy in order to sort out MPs from non-plastic materials (Hidalgo-Ruz et al., 2012). It can be used to characterize the shape of MPs and classify them into primary or secondary MPs, which infer the origin of that particle (Rocha-Santos & Duarte, 2017). Optical microscopy can also be used to measure the length of MP samples if they are not spherical (Rocha-Santos & Duarte, 2017). In addition, Norén (2007) suggested criteria for visual sorting as follows: no structure of organic origin, homogenous thickness for fibers, and clear or homogenous color (Norén, 2007). However, this method may lead to misidentification in a smaller size (< 500 µm) MPs if being used solely (Löder & Gerdts, 2015a). Moreover, identification accuracy highly depends on the skills of the sorting person to discriminate MPs from other substances and the quality and magnification of microscopy (Lusher et al., 2017; Rocha-Santos & Duarte, 2017). Plastic samples should be pre-treated with enzymes or oxidizing agents to avoid misidentification of similar size and shape natural debris (Li et al., 2018a). Therefore, another complementary method, such as spectroscopic analysis, must be applied together to verify the visual identification and provide more accuracy.

2.5.3.2 Raman spectroscopy

Raman spectroscopy is a high-reliability method that can be used to identify the chemical composition of MP samples from various media backgrounds with the ability to assess very small size particles (< 20 µm) (Araujo et al., 2018). It indicates the sample's chemical structure, such as the type of atom, bonding, and intermolecular interaction from the interaction of laser light with the molecules (Ribeiro Claro et al., 2016). Results of Raman spectroscopy are from the differences in the frequency of backscattered light and the background laser frequency (Löder & Gerdts, 2015a). This analysis can be used as a complementary method with visual inspection to increase the success rate in MP identification. The advantages of using Raman spectroscopy are minimal sample preparation, high selectivity, and reproducibility (Löder & Gerdts, 2015a). In addition, compared with IR spectroscopy, Raman spectroscopy covers a wider spectral range and provides better resolution while having less interference from moisture (Rocha-Santos & Duarte, 2015). However, the limitations of Raman spectroscopy are the long measurement time and spectral distortion from sample impurities (Araujo et al., 2018).

Due to the limited sample size of the conventional Raman spectroscopic method, some studies adopted Raman micro-spectroscopy to identify smaller particles down to 1 µm including MP in drinking water which is usually overlooked (Ivleva et al., 2017; Schymanski et al., 2018). Moreover, Raman spectroscopic analysis is time-consuming and not automated. Therefore, the novel Raman spectroscopy has been developed to reduce measurement. Frère et al. (2016) proposed semi-automated Raman spectroscopy for large-scale identification, resulting in a 75% identification rate (Frère et al., 2016). Moreover, the study by Araújo et al. (2018) came up with automated Raman spectroscopy with real-time detection using stimulated Raman scattering coupled with flow cytometry (Araujo et al., 2018). The operation time is reduced, and fluorescence interference from plastic is overcome (Araujo et al., 2018).

2.5.3.3 Fourier-transform Infrared (FT-IR) spectroscopy

Fourier-transform Infrared (FT-IR) spectroscopy is another widely used identification technique with high accuracy. FT-IR and Raman spectroscopy are complementary techniques for visual identification to confirm the types of polymers (Löder & Gerdts, 2015a). It is suitable for visible MPs having a size over 100 µm (Ribeiro Claro et al., 2016). FT-IR differs from Raman spectroscopy in that infrared radiation (IR) is used rather than laser light (Ribeiro Claro et al., 2016). Some molecular vibration, which is inactive in Raman analysis, can be detected by IR (Ribeiro Claro et al., 2016). The results of IR spectra can be used to compare with reference IR spectra to identify types of polymers (Ivleva et al., 2017). It also shows the sample's degree of oxidation when weathering is exposed (Ribeiro Claro et al., 2016).

There are three operating modes in FT-IR; transmission, reflection, and attenuated total reflectance (ATR) (Ivleva et al., 2017). ATR mode is usually used to analyze MP particles larger than 500 µm, which can be handled by tweezers (Ivleva et al., 2017). While FT-IR combined with optical microscopy, so-called micro-FTIR, is used in the analysis of smaller-size samples (Ivleva et al., 2017). Recently, the use of micro-FT-IR is increased due to the ability to simultaneous visualization, mapping of samples, and collection of spectra while being able to analyze irregular-shaped MPs

(Rocha-Santos & Duarte, 2015). This technique is being developed because it is still time-consuming, requires a trained operator, and is high cost.

FT-IR was used to verify visual sorting in the study of Leslie et al. (2017). In comparison, the study of Zhang et al. (2017) applied the FT-IR technique to confirm the type of polymers of MP samples collected from three Gorges Reservoirs (Zhang et al., 2017). It was found that most of the MP particles are PE, PP, and PS (Zhang et al., 2017). In addition, the technique of FT-IR has proved to be a promising method for identifying the type of polymers in many studies (Dyachenko et al., 2016; Mani et al., 2015; Mintenig et al., 2017; Murphy et al., 2016; Wang et al., 2017).

2.5.3.4 Pyrolysis-gas chromatography coupled to mass spectrometry (Pyr-GC/MS)

Pyrolysis-gas chromatography coupled to mass spectrometry or Pyr-GC/MS is another frequently used technique to identify polymer types and other chemicals associated with MPs. Samples are heated up to 500-1400 ºC into individual fragmented substances, which are later separated by a fused silica capillary column and interpreted by mass spectrometry (Kusch et al., 2016). Since the pyrolysis method directly assesses the solid polymer and copolymer, pre-treatment is unnecessary (Rocha-Santos & Duarte, 2015). This technique analyzes the thermal degradation product of MPs in one run without any solvent required (Li et al., 2018a). Thus, it prevents background contamination. However, the method is destructive, so the sample cannot be analyzed which other techniques to compare the results (Rocha-Santos & Duarte, 2015). Moreover, it is time-consuming because it requires a manual placement on the instrument, and only one particle can be analyzed at once (Lusher et al., 2017).

Pyr-GC/MS was applied to identify polymer type and organic plastic additives in sediment samples (Fries et al., 2013). However, this technique has some limitations: mass and size limitations. It can identify only low-mass samples lower than 350 µg and diameter less than 1.5 mm, which is the size of a thermal desorption tube (Fries et al., 2013). Therefore, sample preparation is obligatory.

2.5.3.5 Scanning electron microscopy (SEM)

Scanning electron microscopy (SEM), scanning electron microscopy-energy dispersive X-ray spectroscopy (SEM-EDS), and environmental scanning microscopy-energy dispersive X-ray spectroscopy (ESEM-EDS) can produce the image of surface morphology from MP samples (Rocha-Santos & Duarte, 2015). It employs the interaction of electron beam and diffraction/reflection of emitted radiation from MP surfaces (Rocha-Santos & Duarte, 2015). Only SEM requires the coating of samples because of the high vacuum (Li et al., 2018a). This method is also destructive, and the analysis has charge effects. On the contrary, SEM-EDS and ESEM-EDS can determine the elemental composition of MPs which can be inorganic additives in MP samples (Rocha-Santos & Duarte, 2015). These two techniques do not require sample preparation or coating because the testing condition is the low vacuum.

The study by Erikson et al. (2013) employed SEM analysis to verify the results from visual identification. It was found that 20% of particles less than 1 mm initially identified as MP were aluminum silicate from fly ash (Eriksen et al., 2013). Thus, SEM is an alternative method to confirm whether particles are MPs or not and to provide a high-resolution image of the morphological surface of the samples.

Various methods can analyze MP samples to obtain the quantity and chemical composition. The advantages and limitations of identification methods for MPs are presented in Table 2.4.

Table 2.4 Advantages and limitations of identification methods for MPs

Identification methods	Advantages	Limitations
Microscopy	Simple, fast, and easy	• depends on the skills of the sorting person • high possibility of misidentification • high possibility of underestimation of smaller particles • No data on the chemical composition
Raman spectroscopy	• Non-destructive	• Time-consuming

	• Minimal sample preparation, high selectivity, and reproducibility • More comprehensive spectral range and better resolution than FTIR • Suitable for smaller-sized MPs (down to 1 µm)	• Spectral distortion from sample impurities • High cost
FTIR spectroscopy	• Non-destructive • Able to detect molecular vibration that is inactive in Raman • Suitable for samples >100 µm	• Time-consuming • Requires trained operator • High cost
Pyr-GC/MS	• Simultaneous analysis of polymer types and chemical additives • It does not require pre-treatment or any solution	• Destructive method • Mass and size limited • Time-consuming
SEM	• Provide high-resolution images of MP morphologies	• Requires sample coating • Destructive method

CHAPTER 3
METHODOLOGY

3.1 Study sites

There were four WWTPs selected for this study. Three of the WWTPs are in Bangkok, Thailand, with different treatment technologies, and another WWTP is in a suburban area with less population equivalent. The summary of each WWTP is shown in Table 1. Din Daeng WWTP is the biggest WWTP in Bangkok with the highest treatment capacity and the largest population served. Nong Khaem WWTP was selected because of its different configurations of the activated sludge process. Nong Khaem WWTP is in the sub-urban area of Bangkok, surrounded by the textile industry.

On the other hand, Bang Sue WWTP is currently the only WWTP with advanced technology on a pilot scale. The treatment system of Bang Sue WWTP is constructed underground unlike other WWTPs. Lastly, Sing Buri WWTP employs waste stabilization ponds to treat domestic wastewater in Sing Buri province, 150 km from Bangkok. The area is less populated than Bangkok, and the lifestyles of citizens are also different.

Table 3.1 Summary of four WWTPs in the study with population equivalent, treatment technologies, and treatment capacity per day

WWTP	Population equivalent	Treatment technology	Treatment capacity (m^3/day)	Service area (km^2)
Nong Khaem WWTP	520,000	Vertical loop reactor	157,000	44
Din Daeng WWTP	1,080,000	Biological activated sludge	350,000	37

Bang Sue WWTP	227,660	Biological-activated sludge coupled with ultrafiltration (UF)	120,000 (dry season) 300,000 (rainy season)	20.7
Sing Buri WWTP	17,000	Waste stabilization pond	4,500	4

3.1.1 Nong Khaem WWTP

Nong Khaem Wastewater Treatment Plant is located in Nong Khaem district, Bangkok, Thailand. The plant serves an area of 44 square kilometers in Nong Khaem, Pasi Charoen, and Bang Kae districts with 520,000 population. The total length of the pipe from Nong Khaem WWTP is 46 kilometers, connected to 8 pumping stations. Vertical Loop Reactor (VLR) is the treatment technology used in this plant. The biological processes of this WWTP include anoxic and aerobic tanks. Ferric Chloride is a flocculation agent added during the clarification process. The treatment capacity of Nong Khaem WWTP is 157,000 m^3/day. Moreover, this location includes a sludge treatment plant that receives sewage sludge from WWTP all over Bangkok. Sewage sludge is treated by anaerobic digestion before being used as a soil amendment. The capacity of sludge treatment is 500 m^3/day. The flow diagram and sampling location for MPs were shown in Figure 3.2.

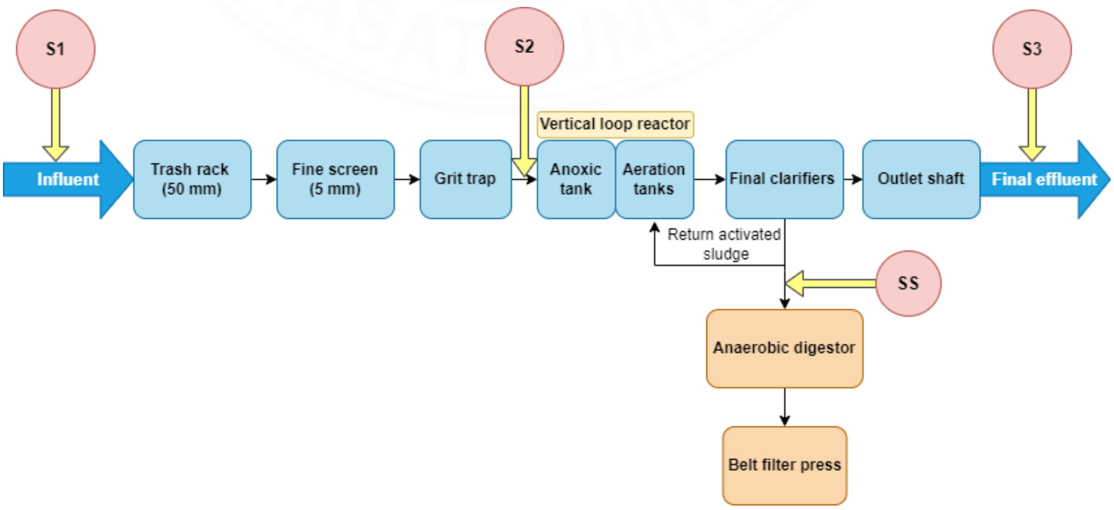


Figure 3.1 Flow diagram of Nong Khaem WWTP with sampling locations

S1 = influent, S2 = after grit trap, S3 = final effluent, and SS = sewage sludge

3.1.2 Din Daeng WWTP

Din Daeng wastewater treatment plant is the biggest WWTP in Thailand which serve more than 1 million population in Din Daeng, Pathum Wan, and partly Phaya Thai, Dusit, Ratchathewi, Pom Prap Sattru Phai, Samphanthawong, and Phra Nakorn district of Bangkok which covers the area of 37 square kilometers. The plant is operated under the Water Quality Management Office, Department of Drainage and Sewerage. It is in an area of 27,200 square meters. This WWTP uses a combined sewer system connected to 0.15-3.2 m diameter pipe and extended to 63 kilometers in distance. The treatment technology employed in this WWTP is a biological activated sludge process with nutrient removal with a capacity of 350,000 cubic meters per day. Moreover, the final effluent of Din Daeng WWTP is released to Sam Sen and Makkasan canal. It is also responsible for improving water quality in Ong Ang, Bang Lam Pu, Maha Nark, Saen Saeb, Prem Prachakorn, Samsen, and Padung Krung Kasem canals.

Din Daeng wastewater treatment plant comprises seven treatment units; coarse screens, inlet pumping station, rake screens, aerated grit channels, activated sludge tanks, clarifier tank, and belt filter press. Sampling locations are shown in Figure 3.1.

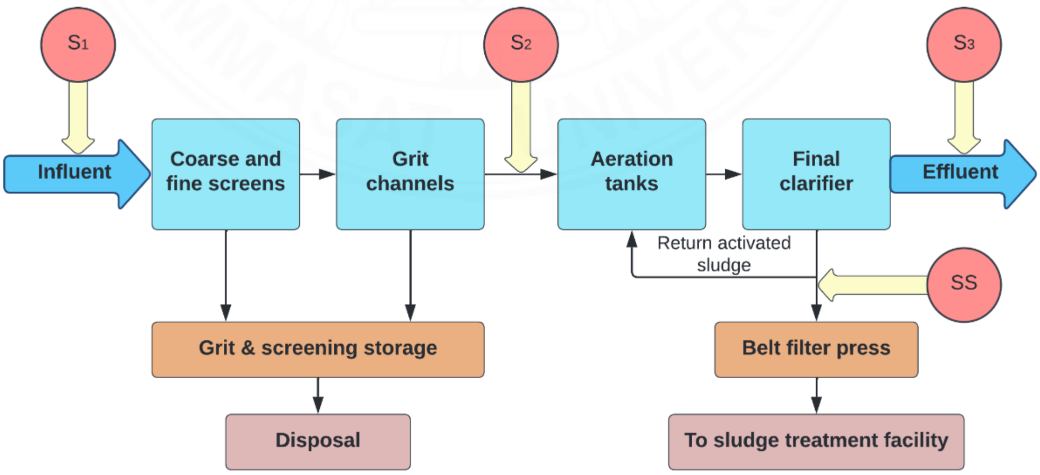


Figure 3.2 Flow diagram of Din Daeng WWTP with sampling locations

S1 = influent, S2 = after grit channels, S3 = final effluent, and SS = sewage sludge

3.1.3 Bang Sue WWTP

Bang Sue WWTP is the underground WWTP of Bangkok Bang Sue Environmental Education and Conservation Center (Bang Sue EECC). The WWTP serves an area of 20.7 km^2 in 4 districts of Bangkok. It serves a population of 227,660. The collection system pipeline is 49.4 km in length. The wastewater collection system is a combined sewer system. The treatment capacity of this WWTP is 120,000 m^3/d and 300,000 m^3/d in dry and rainy seasons, respectively. The treatment plant is a closed underground system to prevent air and noise pollution. This wastewater treatment plant includes primary, secondary, and tertiary treatment. This WWTP employs anoxic-aerobic tanks in secondary treatment, and treatment technology in tertiary treatment is ultrafiltration (polyether sulfone (PES)/polyvinylpyrrolidone (PVP) blend membrane) with a pore size of 0.1 microns. There are two sets of ultrafiltration in the WWTP. Each set can treat up to 1,000 m^3 of wastewater daily. Treated water by ultrafiltration in Bang Sue EECC for cleaning and decorative purposes and the effluent from the final clarifier is discharged into Prem Prachakorn canal. Samples were collected at treatment units shown in Figure 3.3.

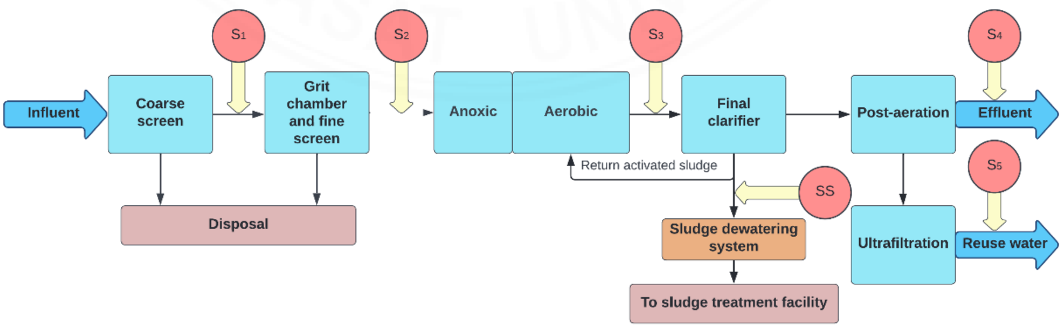


Figure 3.3 Flow diagram of Bang Sue WWTP with sampling locations

S1 = influent, S2 = after grit chamber, S3 = after aeration tanks, S4 = effluent after secondary treatment, S5 = effluent after ultrafiltration (UF), and SS = sewage sludge

3.1.4 Sing Buri WWTP

Sing Buri WWTP serves a population of approximately 17,000. WWTP mainly receive domestic wastewater from restaurants, hotels, housing estate, and hospitals. The treatment technology employed is a waste stabilization pond which consists of an anaerobic pond, a facultative pond, and a maturation pond. The storage capacity of anaerobic, facultative, and maturation ponds are 56,000, 16,000, and 17,900 m^3, respectively. The treatment capacity of this WWTP is 4,500 m^3/day to support the population growth of this area. However, the average flow of wastewater is only 1,500 m^3/day. Treated wastewater is either discharged to the Lop Buri River or recycled as irrigation water. When there is a high demand for water in the growing season, treated wastewater will be sent directly from the outlet of the facultative pond to agricultural fields without passing through the maturation pond. MP samples were collected from the effluent of each pond as shown in Figure 3.4.

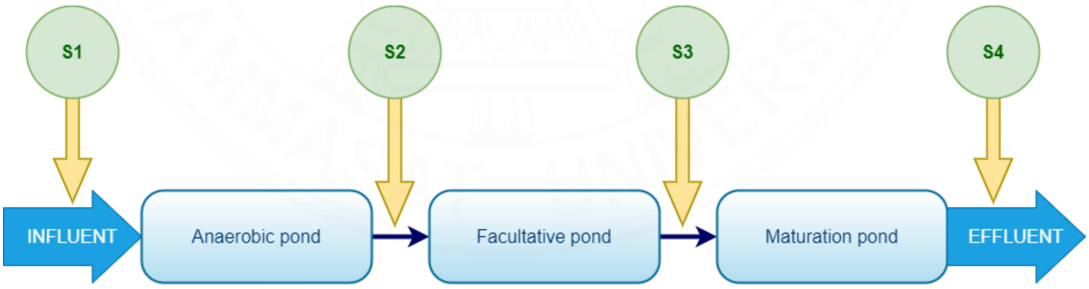


Figure 3.4 Flow diagram of Sing Buri WWTP with sampling locations

S1 = influent, S2 = effluent from anaerobic pond, S3= effluent from facultative pond, and S4 = final effluent

3.2 Sampling methods

In-situ filtration was conducted in Nong Khaem and Din Daeng WWTP. The volume of wastewater filtered through a set of Tyler sieves was 20 L for both WWTPs (n=3). Stacked sieves comprised 5 mm, 1 mm, 500 µm, and 53 µm stainless steel sieves. Samples were collected mainly from the influent, after grit removal, and the final effluent of Nong Khaem and Din Daeng WWTP. Samples retained on each sieve size were rinsed with deionized (DI) water and kept separately in glass bottles. Plastic pieces on 5-mm-sieve were separated and excluded from the total count since they were classified as macro-plastic. Sludge samples were collected from excess sludge from final clarifiers. They were taken from randomized points and homogenized in a glass bottle (~0.5 kg).

Bang Sue WWTP is a closed underground treatment system. Hence, 1L grab samples were collected in triplicates. Stainless steel bucket was used to collect samples primarily from the influent after the coarse screen, after the grit chamber, after aeration tanks, the effluent from secondary treatment, and the effluent from an ultrafiltration unit. Sludge samples were taken from excess sludge after the final clarification to find MP retention in the sludge.

A paired sampling method was used in sample collection from Sing Buri WWTP to find the influence of different sampling methods on the number of MPs. A set of sieves was used to filter 20 L of wastewater from this WWTP's influent and effluent. Grab samples of 1 L wastewater were also collected. Sludge was collected from the bottom of the facultative pond by grab sampling to find the settlement of MPs in a pond system. Due to a long retention time of a pond-based wastewater treatment system, the approach of atmospheric deposition was conducted using a passive sampler. A stainless-steel funnel with a diameter of 12 cm connected with a glass bottle was placed near the ponds and left for 10 hours to find the rate of MP dry deposition in this area. The deposition rate was expressed in the unit of MP/m^2/day.

3.3 Sample processing

Before quantifying and characterizing MPs, samples require pre-treatment and extraction from other inorganic materials. A summary of procedures for sample processing is illustrated in Figure 3.5.

3.3.1 Wet peroxide oxidation

Samples are kept in the oven at 60ºC until dry. Wet peroxide oxidation adopted by Masura et al. was employed for removing organic residue contaminated in MP samples (Masura et al., 2015). The use of H_2O_2 alone may not be sufficient for treating organic-rich wastewater. Thus, Fenton's reagent was effectively used to analyze the large volume of wastewater while enabling MPs for more accurate spectroscopic analysis (Tagg et al., 2017). Fenton's reagent was prepared by mixing 20 mL of 30% hydrogen peroxide (H_2O_2) with 20 mL of iron catalyst ($FeSO_4.7H_2O$). The solution was added to the samples and heated up to 60ºC to speed up the reaction while preventing mass losses of MPs following Munno et al. (2018). An additional 20 mL of H_2O_2 was added to the samples until there was no visible organic matter (Dyachenko et al., 2016). Samples were left to cool down for 24 hours and filtered through a set of sieves. The residue from Fenton's reagent was washed away with DI water. Subsequently, samples were transferred to beakers and dried at 60ºC.

3.3.2 Density separation

Density-based separation was employed to separate MPs from inorganic materials such as sediment and sand. NaI solution (~ 1.5 g/cm^3) was used in this study due to its high recovery rate, cost-effectiveness, and recyclability (Kedzierski et al., 2017; Quinn et al., 2017). NaI solution was mixed with dried samples, stirred continuously for 15 minutes, and left to settle for at least 24 hours. Floating particles were isolated and transferred to a set of sieves to wash away salt residue and categorized into size fractions: 1-5 mm, 0.5-1 mm, and 0.5-0.05 mm. Samples in each size fraction were vacuum filtered on a cellulose nitrate membrane filter with a pore size of 0.45 µm.

3.3.3 Nile Red staining

To avoid underestimation and overestimation of smaller-sized MPs, a staining technique was performed to discriminate synthetic polymers from other inorganic particles (Shim et al., 2016). Filter papers with a size fraction of 0.05-0.5 mm were stained with Nile Red in chloroform (1 mg/mL). Filter papers were placed on a petri dish, covered with aluminum foil, and left in the fume hood for 24 hours. Stained filter papers were later inspected under a fluorescence microscope.

3.3.4 Sludge samples

Sludge samples were collected from the excess sludge, which was wet sludge. Samples were dried at 105ºC until they had a constant weight. The total solid content of sewage sludge was calculated from the weight before and after drying. Sewage sludge samples were processed similarly with liquid fraction. However, density separation preceded wet peroxide oxidation to eliminate a large volume of sediment in the sludge. Saturated NaI solution was added to dried sludge samples in the beakers and stirred for 15 min. The top water layer with floating particles was extracted and washed with DI water on a sieve. Samples were dried at 60ºC before pre-treatment. Fenton's reagent was added to sludge samples and heated up to 60ºC to accelerate the reaction. Samples were left to cool down for 24 hours. Wet sieving was performed to wash away the chemical residue and categorize sludge samples as per the size of the sieves they retained.

3.4 MP quantification and characterization

MPs were enumerated by visual sorting under a microscope and identified using the spectroscopic method. Figure 3.6 depicts the quantification and characterization approach for different size ranges.

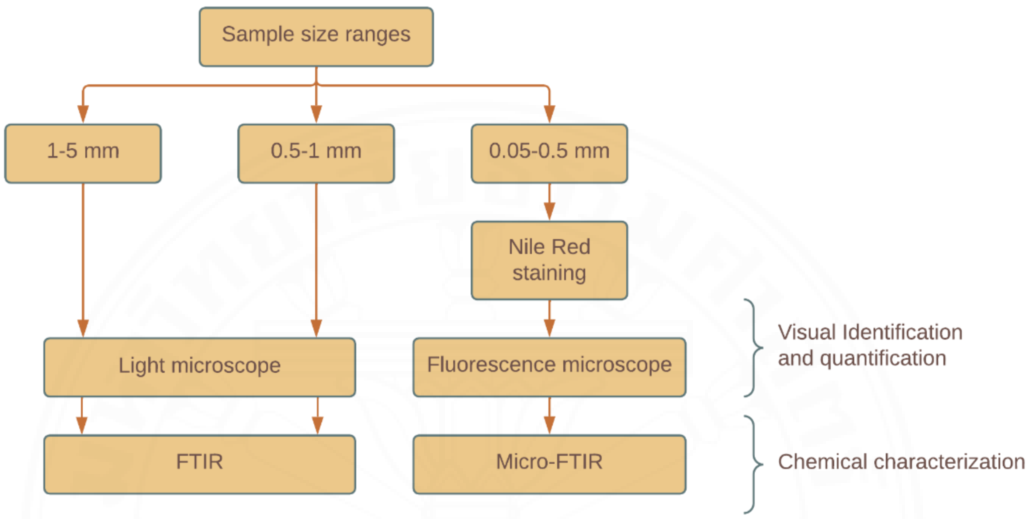


Figure 3.6 Quantification and characterization approach for different sample sizes

3.4.1 Visual identification

Filter papers containing samples with size fractions of 1-5 mm and 0.5-1 mm were inspected under a light microscope (Olympus CX41) with ×4 and ×10 magnification. The number of MPs on each filter paper was counted and grouped into shapes and colors. Shape categories for MPs were fiber, fragment, film, and bead. Organic material, such as plant parts and insect parts, was ignored from the total count during the inspection. The Nile Red staining method was employed for subsequent fluorescent tagging for 0.05-0.5 mm MPs to quantify the number of MPs in this size range. Stained particles with Nile Red were observed through a fluorescence microscope (Delta Vision™ Elite cell Imaging System) under DAPI (4', 6-diamidino-2-phenylindole) filter (blue fluorescence, excitation: 390/18 nm, emission: 435/48 nm) with ×4 magnification. Images of the whole filters were captured from the camera connected to the Delta Vision microscope, and the number of MPs was assessed by counting visible fluorescent spots.

3.4.2 Polymer type identification

The polymer types of MPs were obtained by spectroscopic method. MP sample sizes, 1-5 mm and 0.5-1 mm, were picked individually by tweezers and analyzed by FT-IR (Nicolet iS50, Thermo Scientific) in the ATR mode with a diamond microtip. The resolution of the analysis was 4 cm^{-1} with 32 scans. Filter papers containing particles smaller than 0.5 mm were analyzed using FT-IR connected with a microscope (micro-FTIR: Nicolet iN10, Thermo Scientific) with a resolution of 8 cm^{-1} and 64 scans. Ten spots on each filter paper were randomly selected to identify by micro-FTIR. The obtained spectra were compared with an OMNIC software reference library. The match factor of ≥ 0.70 was used to confirm the polymer types of MPs.

3.5 MP abundance and removal efficiency in WWTPs

The average number of MPs at each sampling location in the liquid fraction was expressed in the unit of MP particles per L of wastewater. The abundance of MPs in sewage sludge was expressed in MP particles per kg of dried sludge. The removal efficiency of each WWTP was calculated following equation 3.1.

$$\%\ \text{removal efficiency} = \frac{\text{influent (MPs/L)}-\text{effluent (MPs/L)}}{\text{influent (MPs/L)}} \times 100\ \% \qquad (3.1)$$

To obtain MP loads discharged daily to water bodies, the treatment capacity of each WWTP is considered and calculated following equation 2.

$$\text{MPs/day} = \text{Final effluent (MPs/L)} \times \text{Treatment capacity (L/day)} \qquad (3.2)$$

3.6 Statistical analysis

Statistical analysis was performed in Minitab 20 software and Microsoft excel (Home & Student). Significant differences between two sampling points and size fractions were determined from a two-sample t-test. When the p-value is less than 0.05, it is considered significantly different, with $T_{stat} > T_{critical}$.

3.7 Contamination mitigation

To prevent MP contamination, mitigation protocol was strictly followed at every step. A cotton lab coat was worn all the time during the experiment. All glassware was rinsed with DI water before use, and the lab bench was wiped with 70% ethanol. To avoid plastic contamination, every container used in the sampling and analysis, including sampling devices, was not plastic. Glassware and metal equipment were used in the analysis. Most of the experimental procedures were performed under a fume hood. Beakers and Petri dishes were covered with aluminum foil to prevent airborne contamination. Blank samples (DI water) were analyzed parallel to the actual samples. Filter papers of blank samples were observed under a microscope, and the number of MPs was quantified and considered background contamination. MPs from blank samples were deducted from the total count in actual samples.

CHAPTER 4
RESULTS AND DISCUSSION

This chapter presents the results of MP abundances and characteristics in 4 WWTPs in Thailand. The MP removal rate for each treatment unit and the overall removal efficiency of each WWTP has been reported. The comparison of 4 WWTPs and the effective unit for MP removal have also been discussed in this chapter.

4.1 MP abundance and removal in the WWTPs

The number of MPs at each sampling location, the removal by each treatment unit, the overall removal efficiency of WWTPs, and the MP characterization of all studied WWTPs will be demonstrated in this section.

4.1.1 Nong Khaem WWTP

4.1.1.1 MP abundance

MP samples were categorized into three size fractions based on their retained sieve size: 1–5 mm, 0.5–1 mm, and 0.05–0.5 mm. Plastic pieces larger than 5 mm were not found in this WWTP. The group of 0.05-0.5 mm MPs was the dominant group found in this WWTP, accounting for more than 70% at every sampling point and about 69.40% in sewage sludge (SS) (Figure 4.1). Figure 4.2 shows the abundance of MPs per liter of wastewater. The abundance of MPs was in ascending order with smaller size ranges. The average number of MPs contaminated in the influent of Nong Khaem WWTP was 26.6 MPs/L. A total of 14.75 MPs/L was discharged with the final effluent.

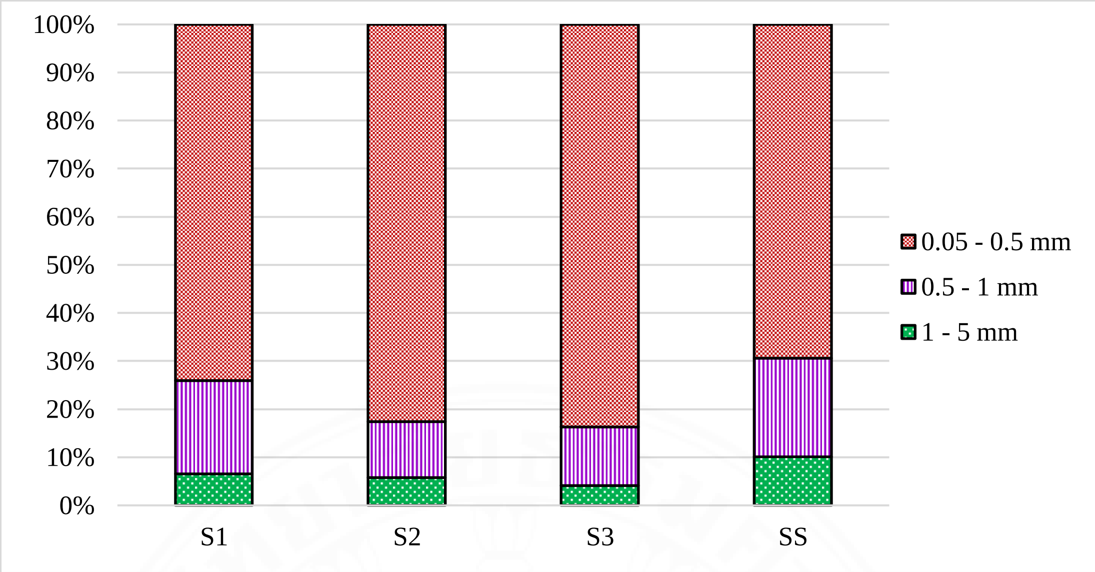


Figure 4.1 Percentage distribution of the different size classes of MPs from various locations in Nong Khaem WWTP

S1 = influent, S2 = after the grit trap, S3 = final effluent, and SS = sewage sludge

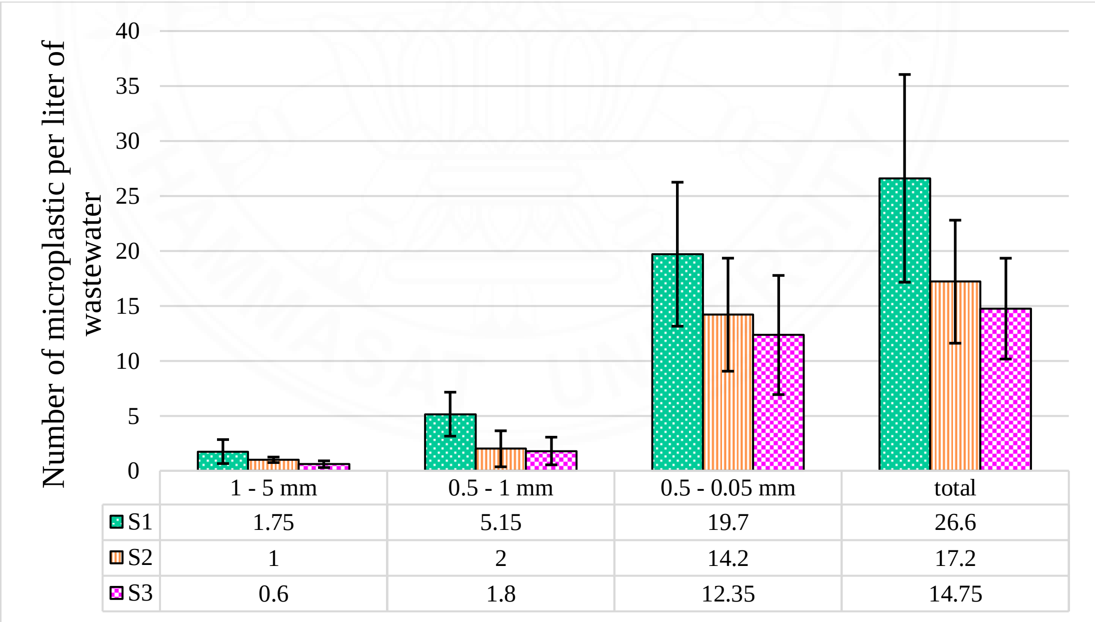


Figure 4.2 Number of MPs per liter of wastewater from treatment steps of Nong Khaem WWTP

S1 = influent, S2 = after the grit trap, S3 = final effluent

4.1.1.2 MP removability

In this WWTP, larger size groups of MPs were more efficiently removed than the size 0.05-0.5 mm (Table 4.1). Screening and grit removal contributed to a higher removal rate of MPs in all size ranges. Secondary treatment achieved a higher removal efficiency (40%) in 1-5 mm MPs than in the other two groups (Table 4.1). Only 14.24% of the total number of MPs were removed during secondary treatment. The overall MP removal efficiency of Nong Khaem WWTP was 44.55%. When this WWTP's treatment capacity is considered, MP loads discharged daily are equal to 2.32 billion MPs/day (Table 4.2).

Table 4.1 Removal efficiencies of different size fractions at each treatment unit based on the average number in Nong Khaem WWTP

Size fraction	Removal efficiency (%)		
	Screening and grit removal	Secondary treatment	Overall
1-5 mm	42.86	40.00	65.71
0.5-1 mm	61.17	10.00	65.05
0.05-0.5 mm	27.92	13.03	37.31
Total	35.24	14.24	44.55

Table 4.2 Number of MPs released per day based on the treatment capacity from each sampling location in Nong Khaem WWTP

Sampling site	MPs/L	million MPs/day
S1	26.6 (±9.45)	4176 (±1483)
S2	17.2 (±5.59)	2700 (±878)
S3	14.75 (±4.58)	2316 (±720)

4.1.1.3 MP morphologies and characterization

In the influent (S1), fibers accounted for 87% of the total MPs, and the proportion of fibers was reduced to about 65% in the effluent (S3) (Figure 4.3). Some of the fibers were removed by secondary treatment because a large proportion of fibers (77.09%) was detected in sewage sludge (Figure 4.3). However, the proportion of fragments tends to increase during treatment processes. It might be a result of MP breakdowns during the operation.

Only FT-IR analyzed MPs from this study site. Suspected particles were picked individually with tweezers to identify their polymer types. The result spectra were matched with reference spectra from polymer libraries in OMNIC software. Polyester fibers contributed to the most significant proportion of MPs (49%), followed by polypropylene (PP) (20%) and polyethylene (PE) (13%) (Figure 4.4). Even though polyester fibers were the most abundant type obtained from FT-IR, some of the fibers failed to be identified due to the small contact area of fiber with the microtip of FT-IR. Thus, micro-FTIR was employed to identify smaller-sized MPs in the following study sites for more precise results.

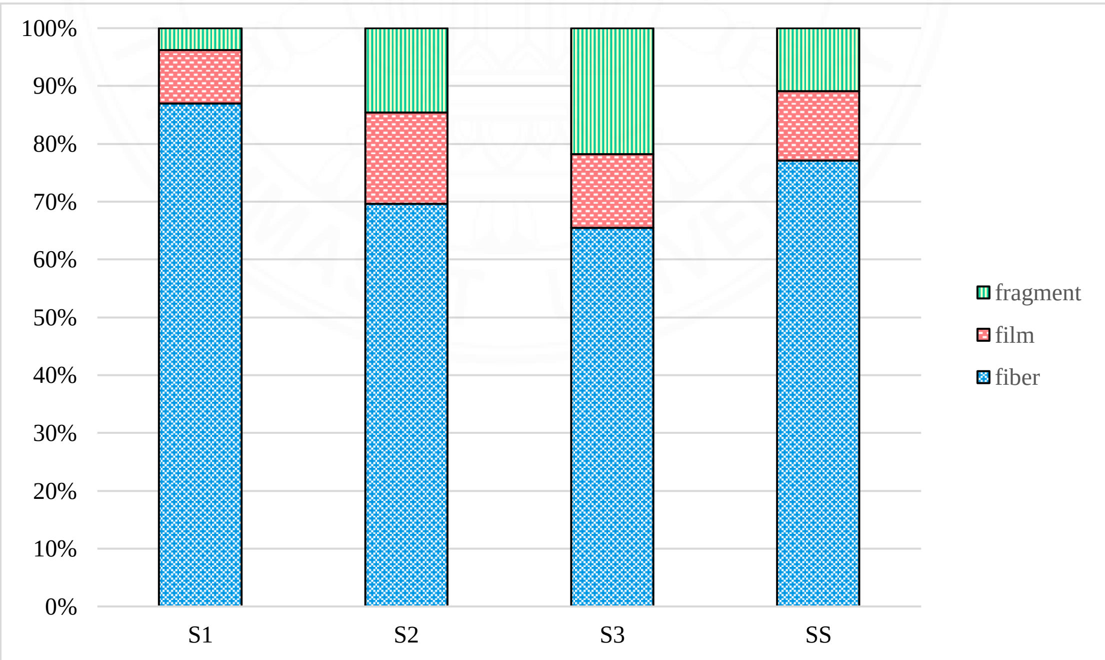


Figure 4.3 Percentage distribution of shape categories in every treatment step of Nong Khaem WWTP

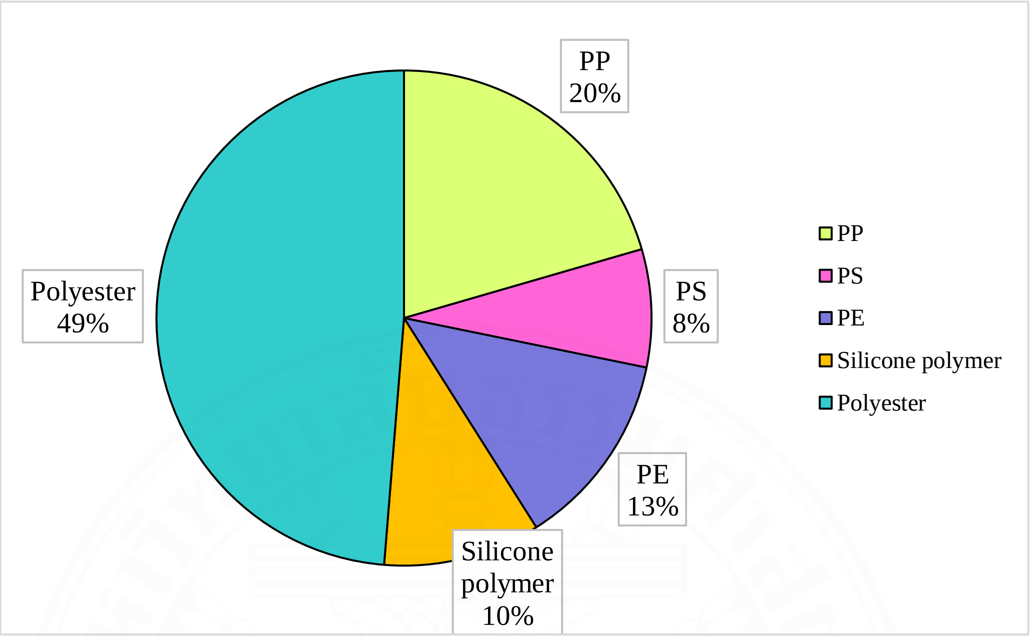


Figure 4.4 Polymer-type distribution of MPs from all sampling locations of Nong Khaem WWTP identified by FT-IR

4.1.1.4 MP retention

MP abundances in sewage sludge were quantified and expressed in the unit of MP particles/kg of sludge (dry weight). The total number of MPs retained in the sludge from Nong Khaem WWTP was 81167±284 per kg of dried sludge (Figure 4.5). The 0.05-0.5 mm size range contributed to the most significant proportion of MPs retained in the sludge (Figure 4.5). It corresponds to wastewater samples' most significant proportion of 0.05-0.5 mm MPs. Sludge from this WWTP plant was subsequently sent to a sludge treatment facility for anaerobic digestion. Sewage sludge was air-dried and applied for soil amendment. The number of MPs contaminated in sewage sludge indicates the potential risk of MP contamination in agricultural soil.

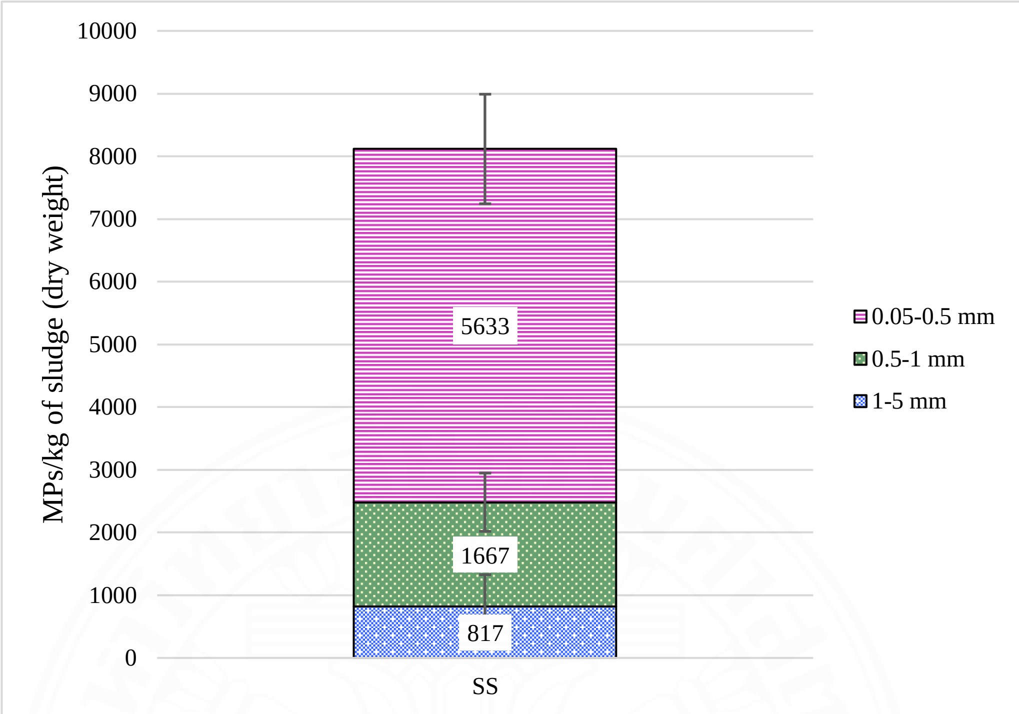


Figure 4.5 MP abundance based on size classes in sewage sludge samples from Nong Khaem WWTP

4.1.2 Din Daeng WWTP

4.1.2.1 MP abundance

No plastic particles bigger than 5 mm were found in the collected samples. The most abundant group of MPs in wastewater samples (S1, S2, and S3) and sludge samples (SS) was the size fraction of 0.05–0.5 mm (Figure 4.6). There was no significant difference between S1, S2, and S3 ($p > 0.05$). In sludge samples (SS), 0.05-0.5 mm MPs were found in the largest proportion (43.19%), but it was not significantly higher than the size of 0.5-1 mm MPs (31.47%) ($p > 0.05$).

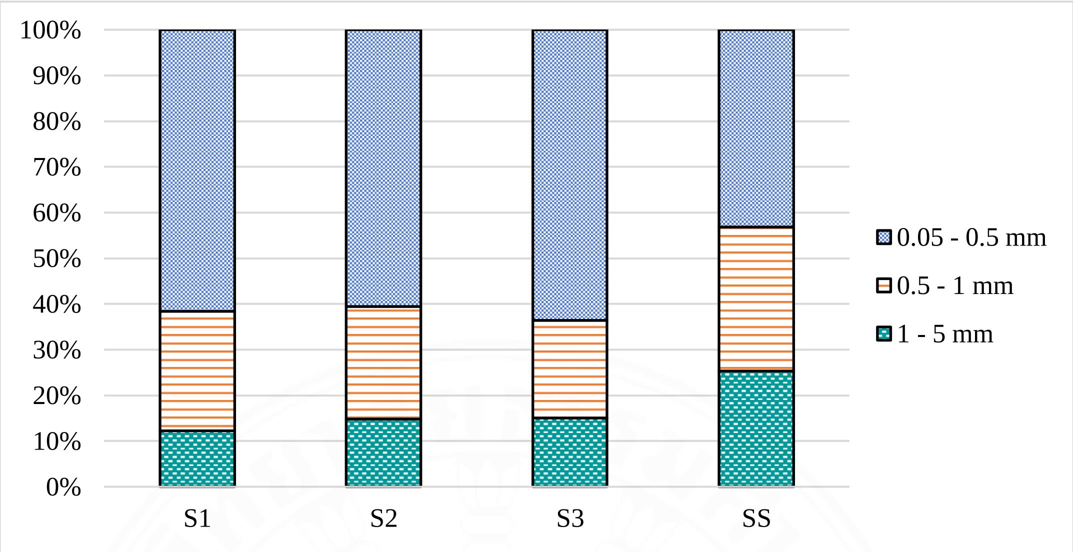
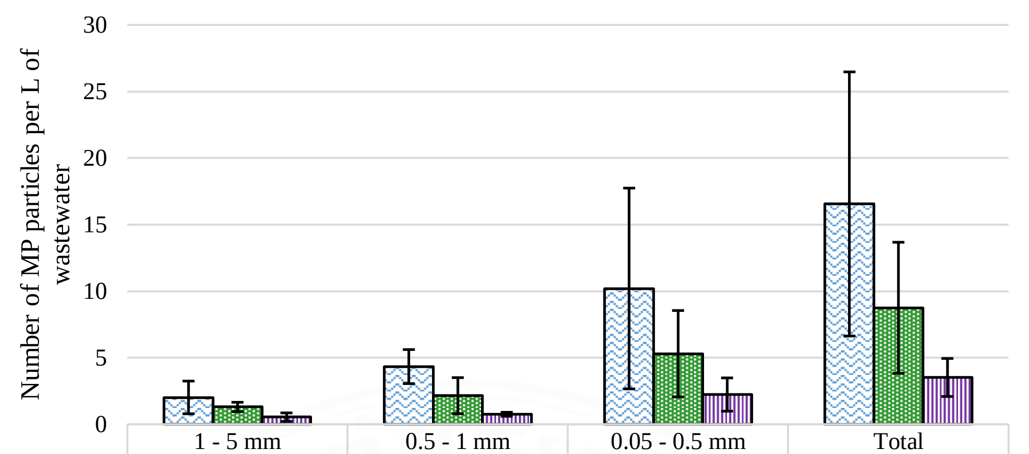


Figure 4.6 Percentage distribution of the different size classes of MPs during wastewater treatment steps from Din Daeng WWTP

S1 = influent, S2 = after the grit trap, S3 = final effluent, and SS = sewage sludge.

Figure 4.7 indicates the number of MPs per L of wastewater in different sampling locations based on different size classes. The average of MPs coming into the wastewater system of Din Daeng WWTP was 16.55±9.92 MPs/L, and the number of MPs discharged to the environment was 3.52±1.43 MPs/L. The largest proportion of MPs contaminated in the inlet of WWTP was the group of 0.05-0.5 mm MPs (10.2 MPs/L) followed by 0.5-1 mm (4.33 MPs/L) and 1-5 mm (2.02 MPs/L). The size fraction of 0.05-0.5 mm also contributed to the largest proportion in the final effluent (2.23 MPs/L) followed by 0.5-1 mm (0.75 MPs/L) and 1-5 mm (0.53 MPs/L).



	1 - 5 mm	0.5 - 1 mm	0.05 - 0.5 mm	Total
S1	2.02	4.33	10.2	16.55
S2	1.3	2.15	5.3	8.75
S3	0.53	0.75	2.23	3.52

Figure 4.7 Number of MP particles per liter of wastewater from treatment steps of Din Daeng WWTP

S1 = influent, S2 = after the grit trap, and S3 = final effluent

4.1.2.2 MP removability

Table 4.3 shows each treatment unit's removal efficiencies of different size classes. The overall removal efficiencies of 1-5 mm, 0.5-1 mm, and 0.05-0.5 mm were 73.76%, 82.68%, and 78.14%, respectively. It resulted in a 78.73% overall MP removal efficiency of Din Daeng WWTP. After secondary treatment, MPs were removed 47.13% by grit trap and 59.77%. Secondary treatment contributed to higher percentage removal in all size fractions than screening and grit trap. The removal of 1-5 mm after screening showed the least contribution among other size classes. Concerning the treatment capacity of this WWTP (350,000 m^3/day), a considerable amount of 1.23 billion MPs is discharged to the environment every day (Table 4.4).

Table 4.3 Removal efficiencies of different size fractions at each treatment unit based on the average number of MPs in Din Daeng WWTP.

Size fraction	Removal efficiency (%)		
	Screening and grit removal	Secondary treatment	Overall
1-5 mm	35.64	59.23	73.76
0.5-1 mm	50.35	65.12	82.68
0.05-0.5 mm	48.04	57.92	78.14
Total	47.13	59.77	78.73

Table 4.4 Number of MPs released per day based on the treatment capacity from each sampling location in Din Daeng WWTP

Sampling site	MPs/L	million MPs/day
S1	16.55(±9.92)	5793(±3472)
S2	8.75(±4.92)	3063(±1724)
S3	3.52(±1.43)	1231(±500)

4.1.2.3 MP morphologies and characterization

The results from visual sorting and identification found that the highest proportion of fiber was found in the liquid fraction. At the same time, fragments were the predominant group in the sewage sludge sample (Figure 4.8). However, the number of fibers and fragments in the influent (S1) was not significantly different. Only 1 microbead was found in the influent, and 2 microbeads were found after the grit removal. Therefore, the number of microbeads was mostly negligible.

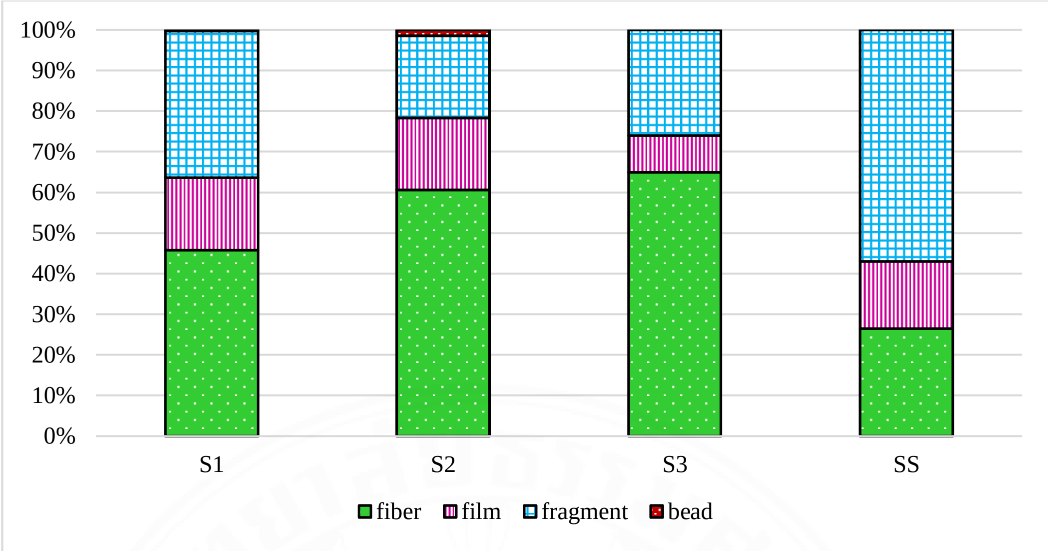


Figure 4.8 Percentage distribution of shape categories in every treatment step of Din Daeng WWTP

S1 = influent, S2 = after the grit trap, and S3 = final effluent

Fingerprint spectra from FT-IR and micro-FTIR were compared to reference libraries. The success rate of FTIR analysis of suspected particles was 46.05% in this WWTP. The results show that the most abundant group of polymers is polyethylene (PE), followed by polyethylene terephthalate (PET) and acrylate polymers (Figure 4.9). PE was observed in the form of transparent fragments and films because PE is used in a variety of packaging and containers. They were secondary MPs derived from the fragmentation and abrasion of plastic products.

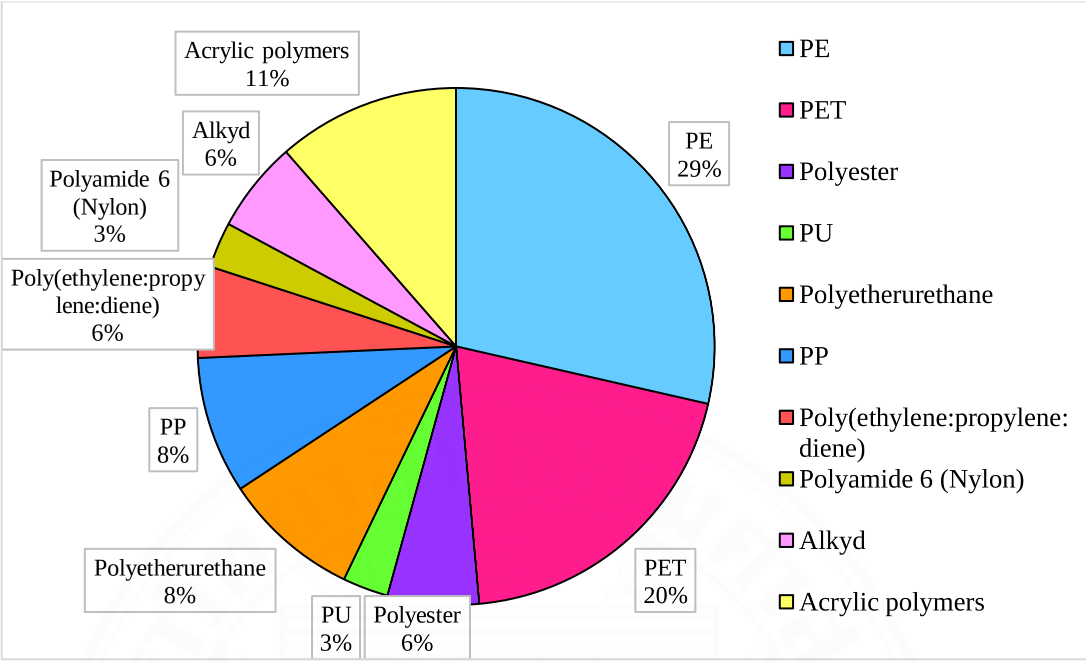


Figure 4.9 Polymer-type distribution of MPs from all sampling locations of Din Daeng WWTP identified by FT-IR and micro-FTIR

4.1.2.4 MP retention

Figure 4.10 shows the abundance of MPs in sewage sludge from Din Daeng WWTP. The size fraction of 0.05-0.5 mm was the most abundant group (20,450 MPs/kg) followed by 0.5-1 mm (14,910 MPs/kg) and 1-5 mm (12,000 MPs/kg). The total number of MPs retained in the sludge was 47,352±15,481 MPs/kg of dried sludge. Dried sewage sludge, after anaerobic digestion, was mostly used for soil application. A high number of MPs might pose a risk of soil contamination.

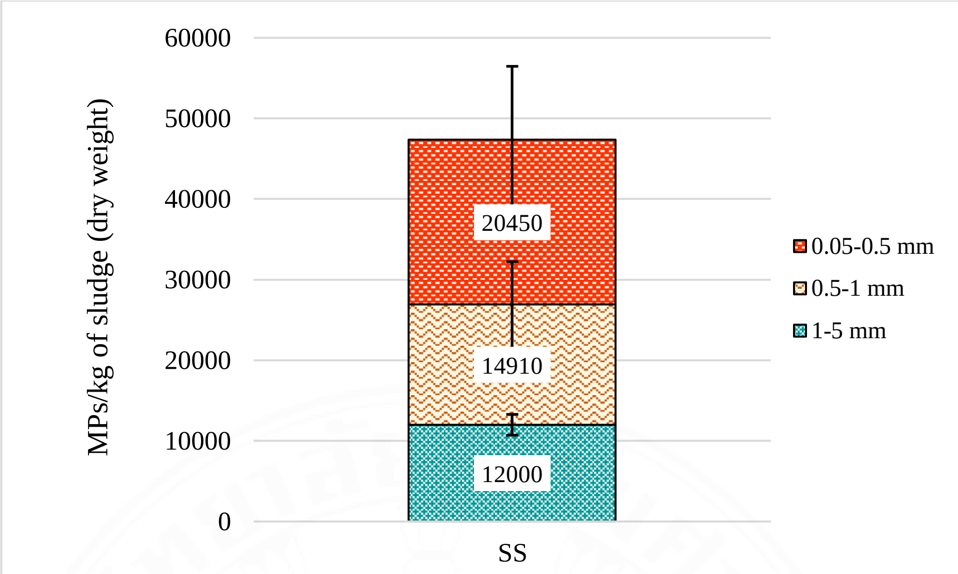


Figure 4.10 MP abundance based on size classes in sewage sludge samples from Din Daeng WWTP

4.1.3 Bang Sue WWTP

4.1.3.1 MP abundance

MPs samples were categorized into three size fractions according to the sieve size they were retained on: 1 - 5 mm, 0.5 - 1 mm, and 0.05 – 0.5 mm. Plastics larger than 5 mm were not found in this study. The number of MPs counted by an optical microscope (size fractions 1-5 mm and 0.5-1 mm) was combined with the number of 0.05-0.5 mm sized MPs from a fluorescence microscope. Percentage distributions of different size ranges of MPs are shown in Figure 4.11. It was found that most MPs in the treatment system were 0.05 – 0.5 mm (Figure 4.11). The size fraction of 0.05-0.5 mm MPs was the only group in the effluent from the UF unit (S5). No 1-5 mm MP was found in the effluent of secondary treatment (S4), which indicates that they were successfully removed after final clarification.

Figure 4.12 exhibits the number of MPs per L of wastewater at each treatment step. The average number of MPs entering the system was 77±7.21 particles/L (S1), and a total of 10.67±3.51 particles/L were discharged with the final effluent (S4) (Figure 4.12).

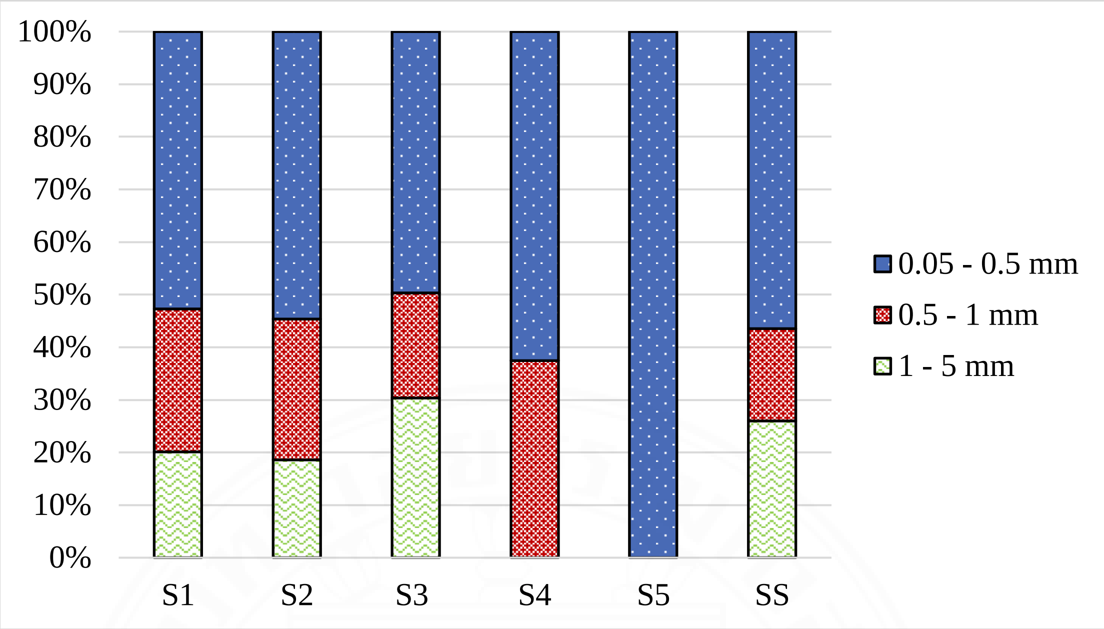
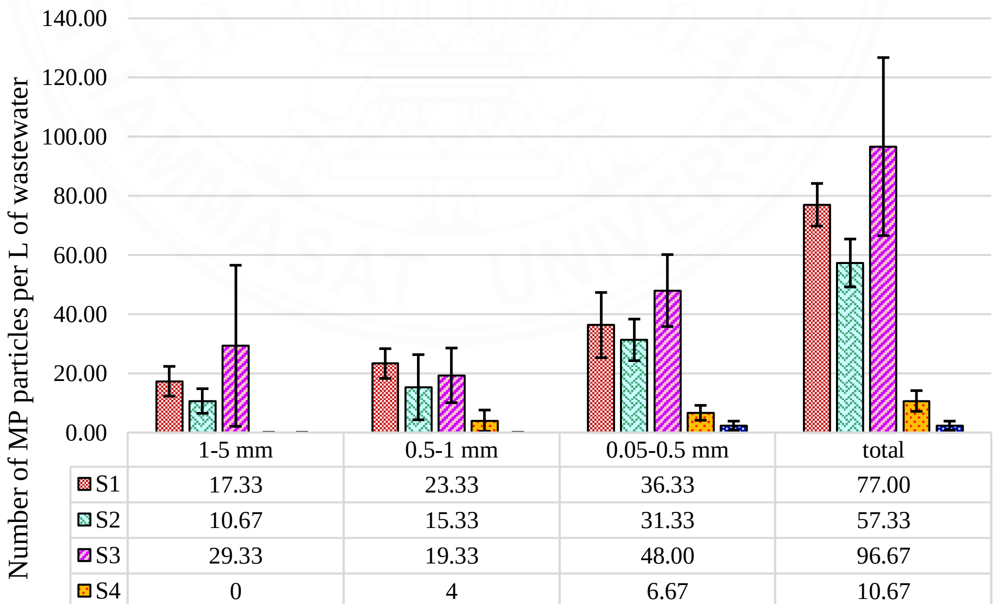


Figure 4.11 Percentage distribution of the different size classes of MPs during wastewater treatment steps of Bang Sue WWTP

S1 = after inlet pumping, S2 = after the grit trap, S3 = after the aeration tank, S4 = effluent from the final clarifier, S5 = effluent from ultrafiltration, and SS = sewage sludge



	1-5 mm	0.5-1 mm	0.05-0.5 mm	total
S1	17.33	23.33	36.33	77.00
S2	10.67	15.33	31.33	57.33
S3	29.33	19.33	48.00	96.67
S4	0	4	6.67	10.67
S5	0	0	2.33	2.33

Figure 4.12 Number of MP particles per liter of wastewater from treatment steps of Bang Sue WWTP

S1 = after inlet pumping, S2 = after the grit trap, S3 = after the aeration tank, S4 = effluent from the final clarifier, S5 = effluent from ultrafiltration, and SS = sewage sludge

4.1.3.2 MP removability

The size fraction of 1-5 mm was successfully removed after the secondary treatment (Table 4.5). The number of MPs removed after the grit chamber and fine screen (S2) was 25.55% (Table 4.5). The number of MPs increased in the aeration tank (S3) (Table 4.5) because MPs were retained in the sludge, returned to the tank, and resuspended, and there were no MPs removed during this process. Additionally, 81.91% of MPs were removed after the final clarifier (S4). Furthermore, there was a 78.16% reduction of MPs after the ultrafiltration in the tertiary treatment (S5). The removal efficiency was based on the number of MPs at the influent and effluent. The calculation of the overall removal efficiency resulted in 86.14% after the final clarifier and 96.97% after the tertiary treatment, respectively. The capacity of this WWTP is 120,000 m^3/day in the dry season during the sample collection. If the maximum flow rate is reached, up to 1.28±0.42 billion MP particles could be released daily into a freshwater ecosystem (Table 4.6). Adding a UF unit may reduce the MP loads released to the water bodies to 280±183 million MPs/day.

Table 4.5 Removal efficiencies of different size fractions at each treatment unit based on the average number in Bang Sue WWTP.

Size fraction	**Removal efficiency (%)**				
	Screening and grit removal	**Secondary treatment**	**Ultra-filtration (UF)**	**Overall removal by a conventional treatment system**	**Overall removal by UF**
1-5 mm	38.43	61.57	-	100	100
0.5-1 mm	34.29	73.91	100	82.85	100
0.05-0.5 mm	30.88	78.71	65.07	85.29	94.86
Total	25.55	81.91	78.16	86.14	96.97

Table 4.6 Number of MPs released per day based on the treatment capacity from each sampling location in Bang Sue WWTP

Sampling site	MPs/L	million MPs/day
S1	77(±7.21)	9240±865
S2	57.33(±8.08)	6880±970
S3	96.67(±30.09)	11600±3611
S4	10.67(±3.51)	1280±421
S5	2.33(±1.53)	280±183

4.1.3.3 MP morphologies and characterization

Fibers were found in both liquid fraction and sewage sludge (≥ 60 %) (Fig. 4.13). Even though large proportions of fibers were found in the effluent from secondary treatment (S4) and UF (S5), some of the fibers were removed by the final clarifier, and they were found in the sewage sludge (SS) (Figure 4.13). Although the mesh size of the UF is very fine, fibers can still escape due to their small size and morphology. A small proportion of film and fragments were also detected in the sludge (SS). Only 5.13 % (2 items) of microbeads were found in S2.

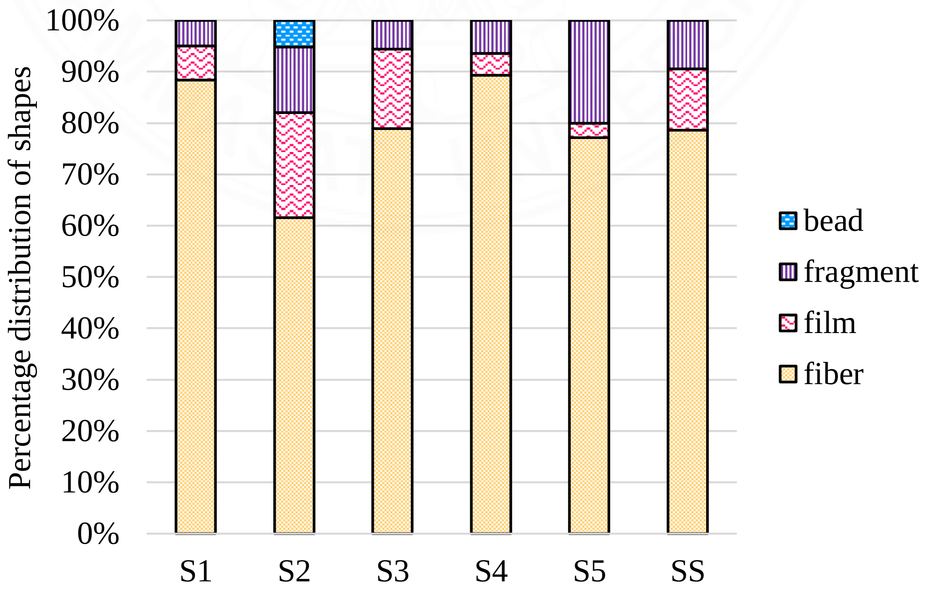


Figure 4.13 Percent distribution of different shapes of MP samples found at each sampling point in Bang Sue WWTP.

S1 = after inlet pumping, S2 = after the grit trap, S3 = after the aeration tank, S4 = effluent from the final clarifier, S5 = effluent from ultrafiltration, and SS = sewage sludge

MPs with a size fraction of 1 – 5 mm and 0.5 – 1 mm were analyzed with FTIR. Selected MP samples were large enough to be handled with tweezers. The 0.05 – 0.5 mm size range was tested by FT-IR connected with a microscope (micro-FTIR) and photographed. FT-IR and micro-FTIR successfully tested only 50% of all suspected particles. The results from both FT-IR and micro-FTIR showed that nine types of polymers were found in MP samples (Figure 4.14). The predominant polymer type was polyethylene terephthalate (PET) from suspected microfibers (Figure 4.14) (see Appendix B.1). Some fibers were identified as natural polymers, such as silk and cotton, and some particles were tested as organic debris, such as chipboard. The number of other natural materials was deducted from the total count.

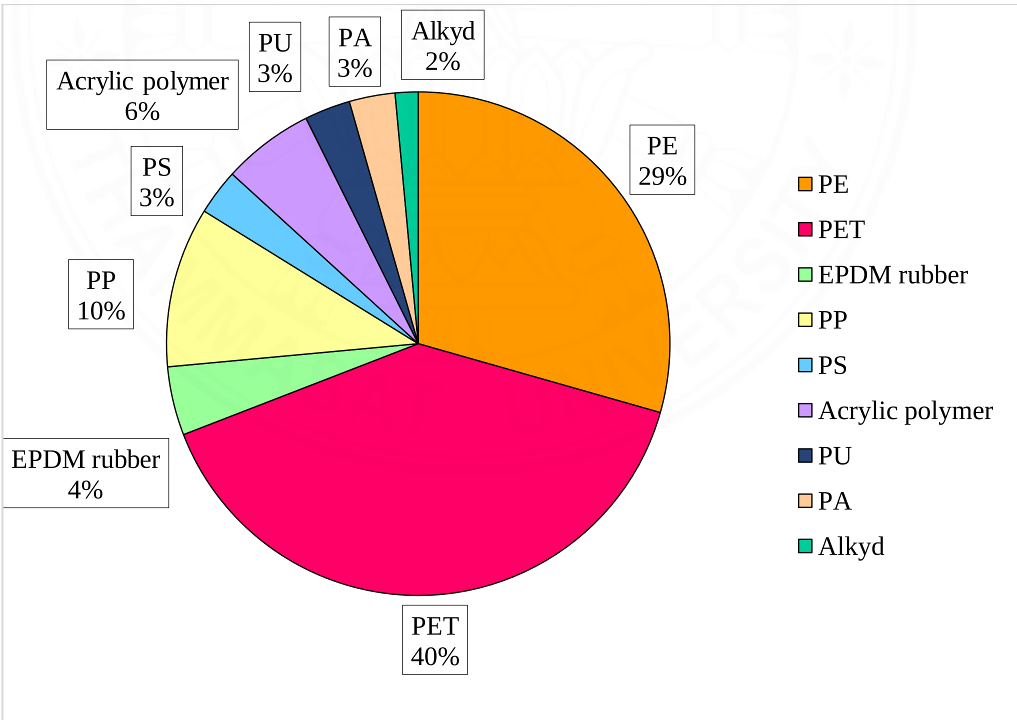


Figure 4.14 Polymer-type distribution of MP samples from all sampling locations of Bang Sue WWTP

4.1.3.4 MP retention

Figure 4.15 shows the number of MPs based on size groups in sewage sludge samples from Bang Sue WWTP. The group of 0.05-0.5 mm MPs shows the largest contribution in sewage sludge (10,990 MPs/kg). The three size fractions of MPs combined resulted in 26,325 ±15,482 MP particles/kg of sludge. However, the number of MPs in the sludge of this WWTP was only from excess sludge after final clarification. Sludge cake from UF was not included in the sampling and analysis.

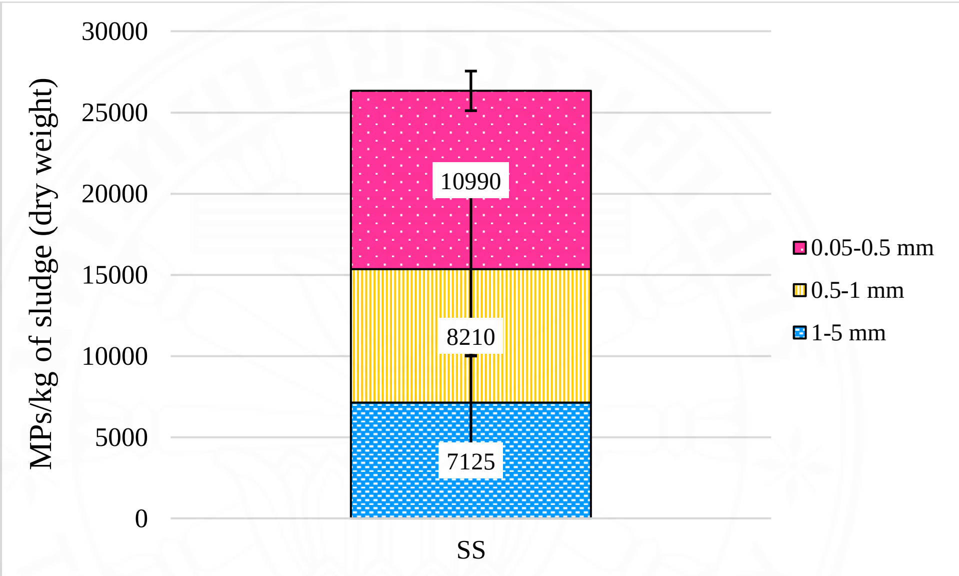


Figure 4.15 MP abundance based on size classes in sewage sludge samples from Bang Sue WWTP

4.1.4 Sing Buri WWTP

4.1.4.1 MP abundance

The abundance of MPs at each treatment step was assessed by the 1-L grab sampling method. The investigation showed that particles larger than 5 mm were not found. The 0.05 – 0.5 mm size range is dominant in S2 and S4 (Figure 4.16). However, there is no significant difference between size ranges ($p < 0.05$) (Figure 4.16). The number of MPs found in the influent is 23±7.81 MPs/L, while an average of 27.67±6.66 was detected at the effluent (Figure 4.17). There was no effective removal in any stage of the waste stabilization pond. Thus, the removal efficiency of MPs cannot be calculated from this pond-based wastewater system. The number of MP discharged to the river can be as high as 124.51 million particles per day if the plant reaches its maximum flow rate (Table 4.7). However, the average flow rate of this WWTP is only 1,500 m^3/day. Thus, about 41.5 million MPs are discharged daily (Table 4.7).

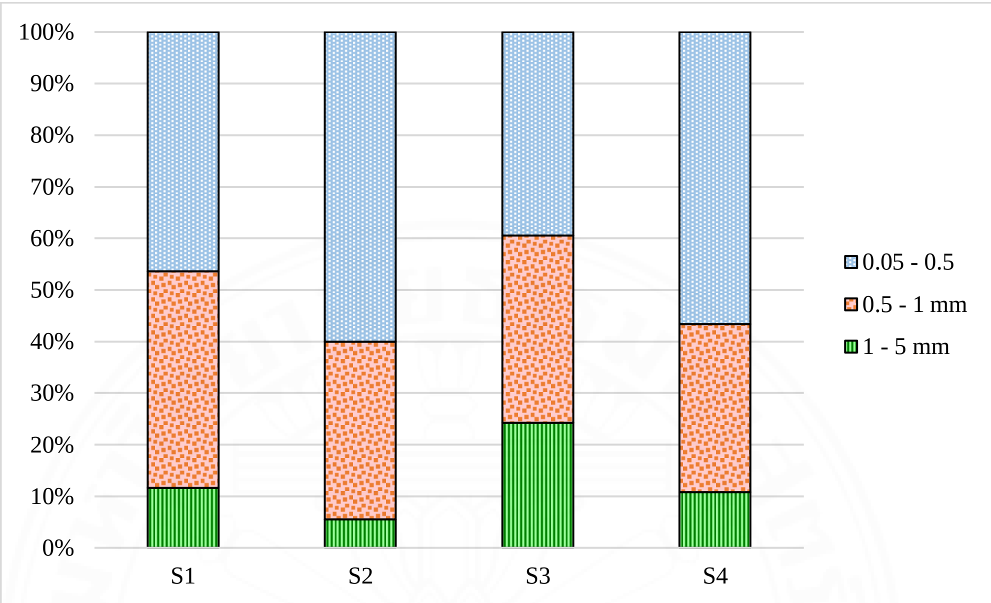


Figure 4.16 Percentage distribution of the different size classes of MPs during wastewater treatment steps from Sing Buri WWTP

S1 = influent; S2 = outlet of anaerobic pond; S3 = outlet of facultative pond; S4= outlet of maturation pond or the effluent

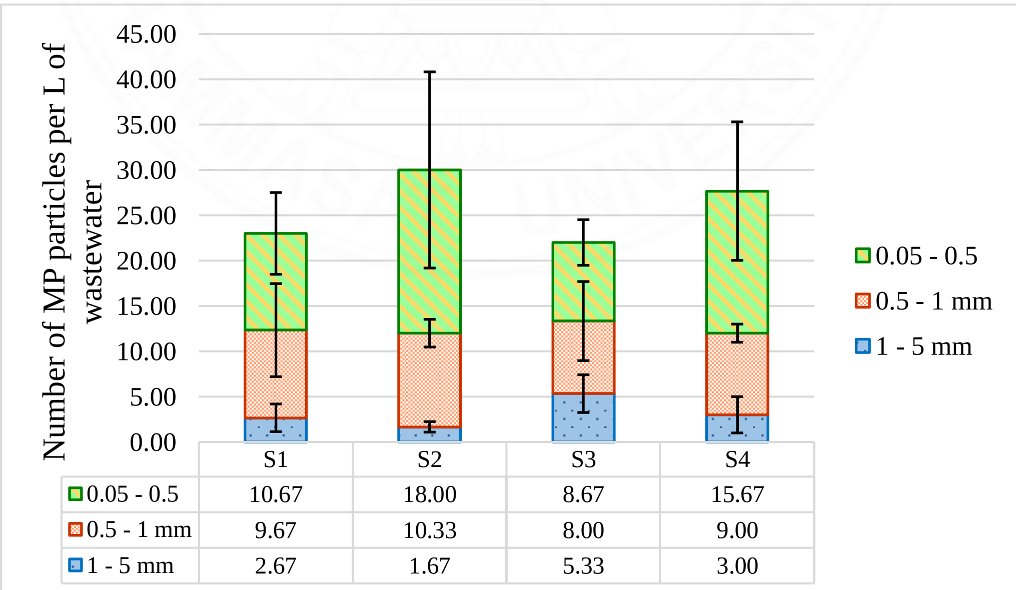


Figure 4.17 Number of MP particles per L of wastewater from treatment steps of Sing Buri WWTP

S1 = influent; S2 = outlet of anaerobic pond; S3 = outlet of facultative pond; S4= outlet of maturation pond or the effluent

Table 4.7 Number of MPs released per day from each sampling location based on the average number in Sing Buri WWTP

Sampling site	MPs/L	million MPs/day (average flow rate)	million MPs/day (maximum flow rate)
S1	23(±7.81)	34.5(±11.72)	103.5(±35.15)
S2	30(±10.15)	45(±8.76)	135(±26.28)
S3	22(±6.08)	33(±9.12)	99(±27.37)
S4	27.67(±6.66)	41.5(±9.99)	124.51(±29.96)

4.1.4.2 MP morphologies and characterization

The dominant shape found in S2 to S4 was fiber, while fragments and fibers were not significantly different in S1 ($p < 0.05$) (Figure 4.18). An example of MPs identified by micro-FTIR is shown in appendix B.2. No microbead was found in this WWTP. A smaller proportion of fragments in S2 to S4 can be explained by settling high-density fragments to the bottom of the ponds. However, fibers are predominantly found in effluent like other WWTPs. This shape of MPs was considered the most challenging to eliminate from WWTPs.

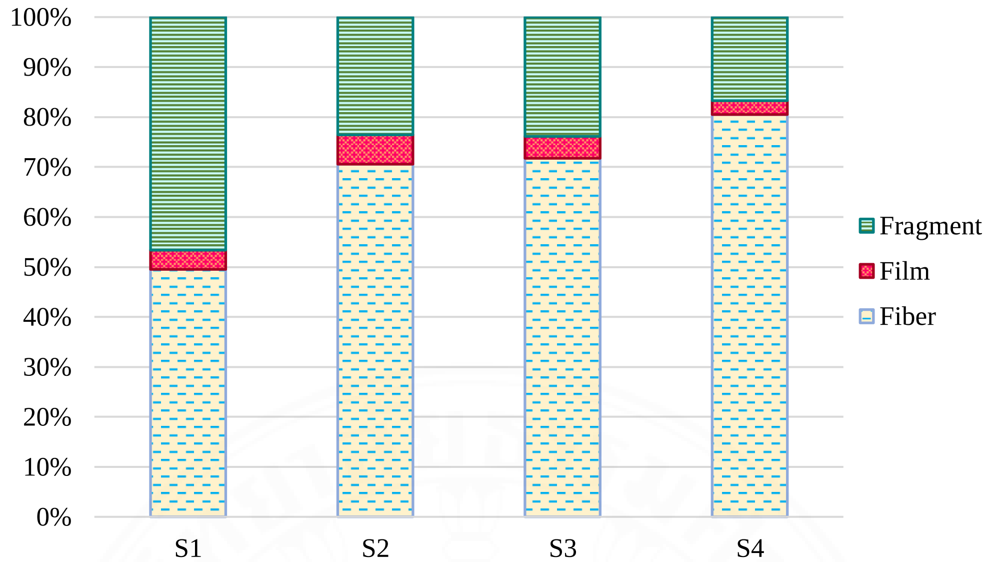


Figure 4.18 Percent distribution of different shapes of MP samples found at each sampling point in Sing Buri WWTP.

S1 = influent; S2 = outlet of anaerobic pond; S3 = outlet of facultative pond; S4= outlet of maturation pond or the effluent

The results from FTIR and micro-FTIR show that PE is the most significant proportion of polymers in Sing Buri WWTP, followed by PET (Figure 4.19). PE was mainly observed in the form of fragments. Some other non-synthetic particles, such as zein, cellulose, and linen, were also detected in WWTP. These natural polymers were subtracted from the total count of MPs. There are various synthetic fibers in this WWTP, e.g., PET, polyester, Nylon, Rayon, and linen, a natural fiber. It reflects the lifestyle of the surrounding communities that the WWTP serves.

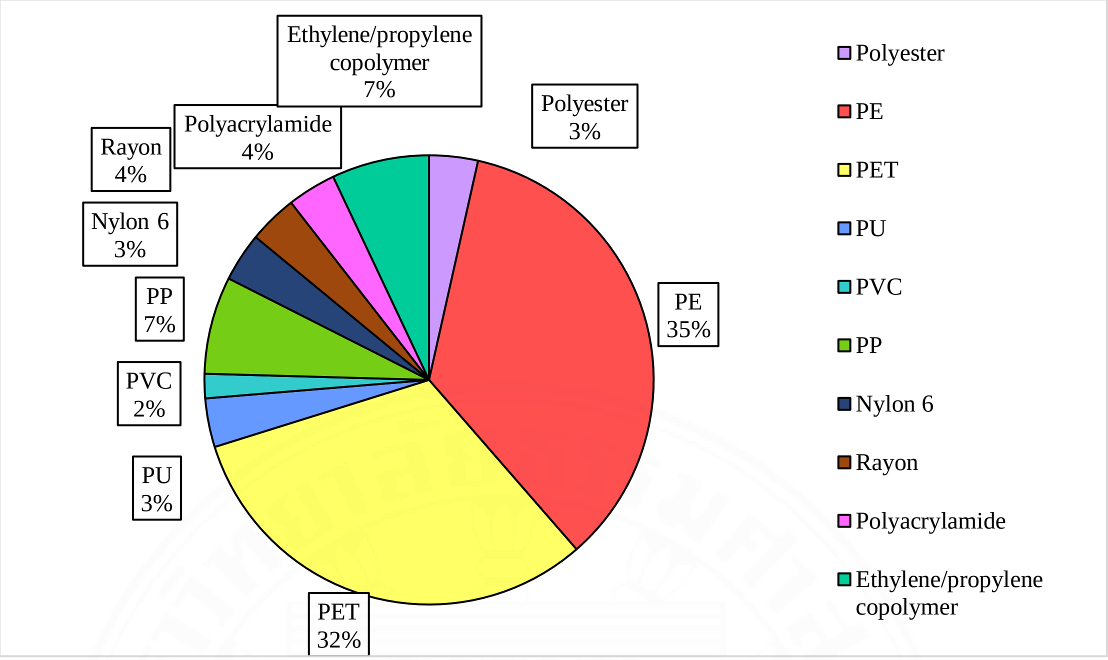


Figure 4.19 Polymer-type distribution of MP samples from all sampling locations of Sing Buri WWTP

4.1.4.3 In situ filtration vs. grab sampling

MP samples were collected by two different sampling methods to find the influence of sampling methods on the abundance of MPs. Samples were collected mainly from the influent and the effluent of the WWTP. In situ filtration was conducted by filtering 20 L of wastewater through a set of sieves, and 1 L of wastewater was collected in glass bottles with three replicates for grab sampling. The number of MPs collected by filtration and grab sampling is shown in Figure 4.20. The number of MPs captured by grab sampling was almost ten times higher than in situ filtrations (Figure 4.20). The magnitude of MPs captured by grab sampling was higher than by net-based sampling (Barrows et al., 2017; Green et al., 2018; Watkins et al., 2021). However, on-site filtration can filter a large volume of wastewater and yield a representative sample.

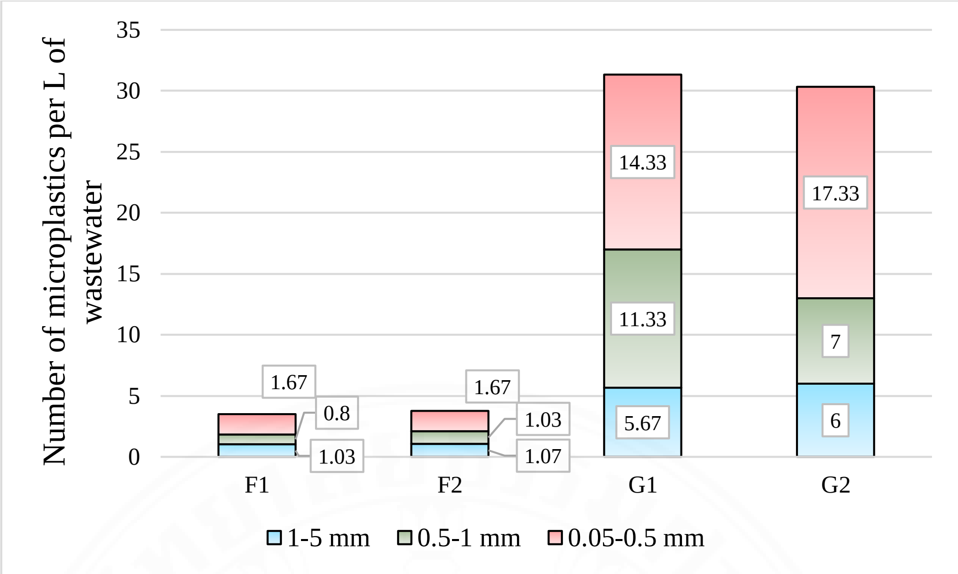


Figure 4.20 Number of MP particles per L of wastewater at the influent and effluent of Sing Buri WWTP from in situ filtration and grab sampling

F1 = influent by filtration; F2 = effluent by filtration; G1 = influent by grab sampling; G2 = effluent by grab sampling

4.1.4.4 MP retention

Sludge samples were collected from the bottom of the facultative pond by grab sampling. MPs were retained in the sludge but not as high as WWTPs in this study. Figure 4.21 shows the abundance of MPs in the sludge from Sing Buri WWTP. It indicates that some of the MPs settled down to the bottom of the pond with the sludge. The average number of MPs was 1613±500 MPs/kg of dried sludge. However, the number of 0.05-0.5 mm MPs was the lowest among the three size fractions in the sludge samples. It implies that this size fraction has a lower ability to settle down due to its size and lightweight.

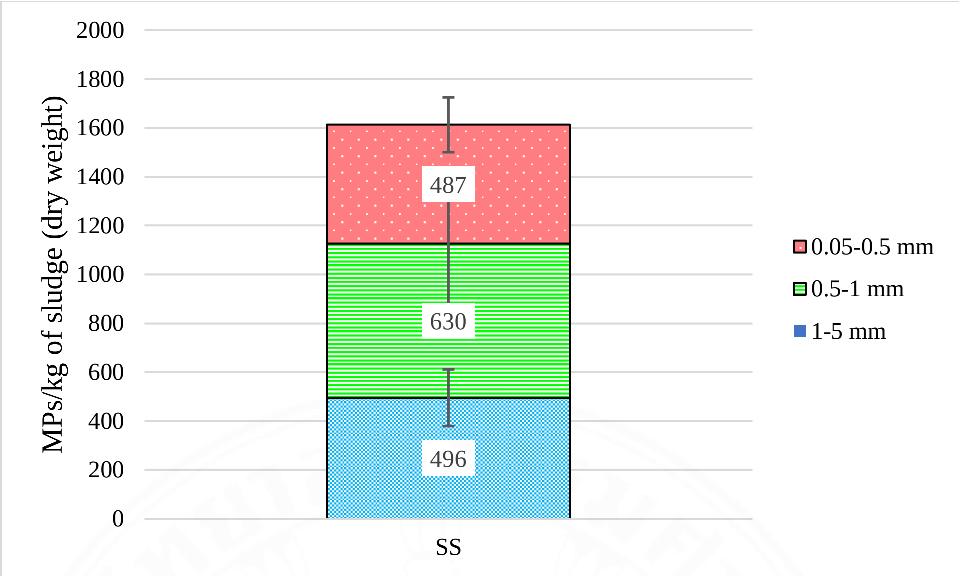


Figure 4.21 MP abundance based on size classes in bottom sludge from Sing Buri WWTP

4.1.4.5 Atmospheric deposition

Passive samplers were left at random locations near waste stabilization ponds for 10 hours. The area of a receiving funnel was taken into consideration. The dry deposition rate of MPs was expressed as MPs/m^2/day. Before counting and analysis, samples were processed in the same manner as MP samples in the liquid phase. The number of MPs was counted under a microscope. The results show that the atmospheric deposition rate of MPs in Sing Buri WWTP is 161.60±28.93 MP/m^2/day. The total area of three ponds is approximately 107,520 m^2. It results in the deposition of 17.37 million MPs/day. The most significant proportion of airborne MPs was 0.05-0.5 mm, which is predominantly 100 times higher than 1-5 mm MPs (Figure 4.22). Fiber accounted for 74.75% of the total MPs, and fragments were 25.25% of the total count (Figure 4.23). No other shapes of MPs were detected. Carbon soot produced from incomplete combustion of fossil fuel may be a source of contamination in atmospheric samples. Due to its inert property, carbon soot cannot be digested by wet peroxide oxidation. The atmospheric contamination may influence the higher number of MPs in the pond system.

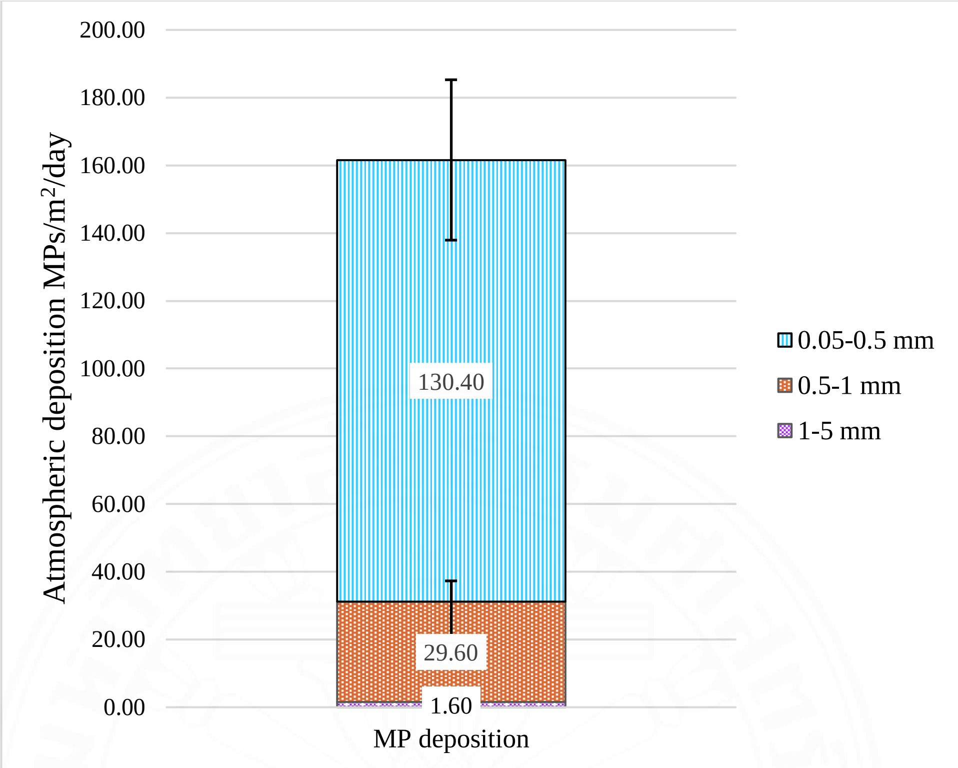


Figure 4.22 Size-based MP atmospheric deposition in Sing Buri WWTP

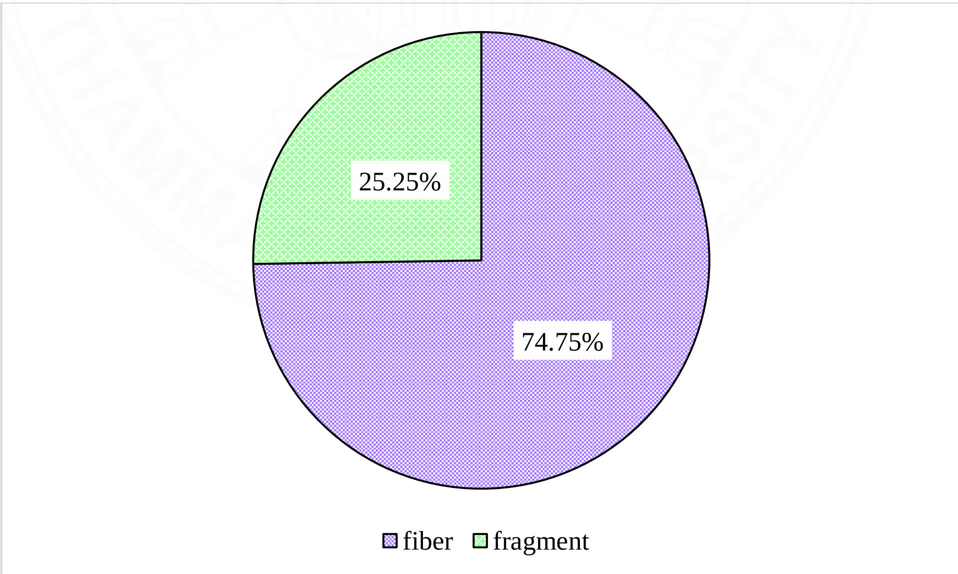


Figure 4.23 Shape percentage distribution of atmospheric MPs from Sing Buri WWTP

4.2 Comparison of MP abundances and removability in WWTPs

Table 4.8 indicates summarized data of the studied WWTPs, the level of MP contamination, and the overall MP removal efficiencies. Combined sewer overflow is a system that collects wastewater together with runoff and sends it to every WWTP in this study. The highest number of MPs in the influent was from Bang Sue WWTP, with a population of 227,660. Despite the larger population in the service area of Din Daeng and Nong Khaem WWTPs, the influent had fewer MPs. The number of different types of residents in the service area of the three WWTPs in Bangkok is shown in Appendix A. Three WWTPs employ activated sludge processes with different configurations for wastewater treatment, but Bang Sue WWTP was equipped with the UF unit. It resulted in the highest MP removal efficiency (96.97%) among all studied WWTPs. When only a conventional system of Bang Sue WWTP was considered, it also exhibited the highest overall removal percentage (86.14%) among all studied conventional systems. One of the reasons is that Bang Sue WWTP was constructed as a closed underground treatment system that can lower airborne MP contamination. There is also a scum skimmer at the clarification tank of Bang Sue WWTP, which can remove low-density and floating MPs (Figure 4.24). On the contrary, the clarifiers of other WWTPs (Nong Khaem and Bang Sue) are placed outdoors, prone to atmospheric contamination.

Table 4.8 Summary of MP abundances and overall removal efficiencies of four studied WWTPs.

WWTPs	Population equivalent	Treatment technology	Sampling method	Inlet (MPs/L)	Outlet (MPs/L)	Overall removal efficiency (%)
Nong Khaem WWTP	520,000	Vertical loop reactor	20-L in situ filtration	26.6±11.8	14.75±4.58	44.55
Din Daeng WWTP	1,080,000	Activated sludge	20-L in situ filtration	16.55±9.92	3.52±1.43	78.73

Bang Sue WWTP	227,660	Activated sludge (Anoxic-aerobic) coupled with Ultrafiltration (UF)	1-L grab sampling	77±7.21	10.67±3.51 (2.33±1.53) [a]	86.14 (96.97) [a]
Sing Buri WWTP	17,000	Waste stabilization pond	1-L grab sampling	31.33±6.66	30.33±3.21	3.19
			20-L in situ filtration	3.50±1.13	3.77±0.90	-

[a] the effluent of a UF unit

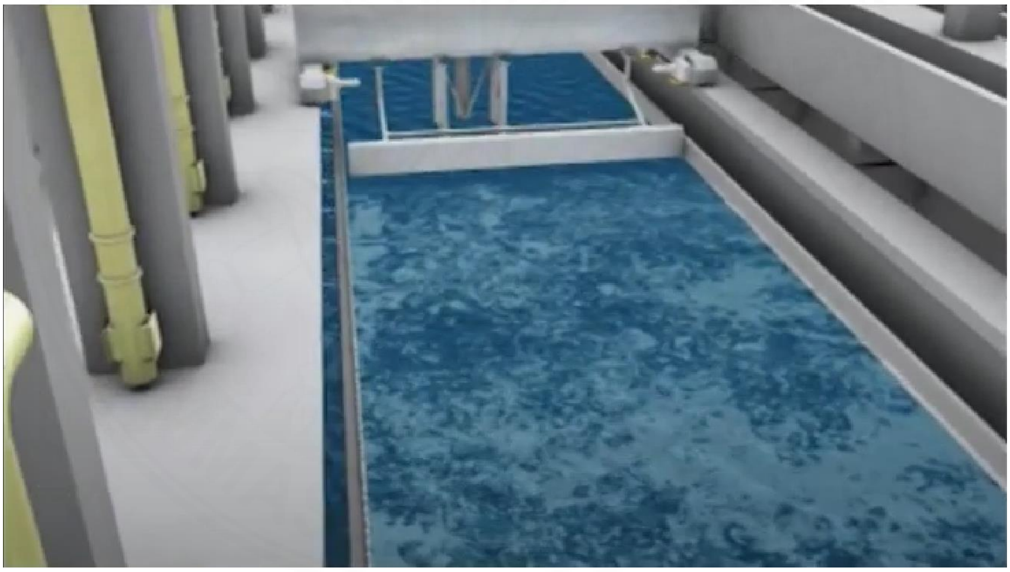


Figure 4.24 A scum skimmer at the clarifiers of Bang Sue WWTP

Nong Khaem WWTP is the only WWTP where the flocculation agent ($FeCl_3$) is added during the clarification process. Flocculation agents can enhance the aggregation of flocs and the removal of MPs with the settled sludge flocs (Murphy et al., 2016; Sun et al., 2019). However, the WWTP achieved the lowest removal efficiency among conventional WWTPs, and only 14.24% of MPs were removed by secondary treatment. The important factor affecting MP removal is retention time in the clarifiers. It might be due to a short retention time or the redistribution of MPs in the aeration tanks by recycled sludge (Carr et al., 2016). MPs in this WWTP comprised a large proportion of fibers (>80%) because the textile industry surrounded the WWTP. Fibers are considered the most challenging type of MPs to remove due to their smooth surface and morphology (Long et al., 2019). It might be a reason for the relatively low removal rate in Nong Khaem WWTP.

Sing Buri WWTP has the lowest population equivalent, but the number of MPs from 1 L grab sampling was higher than 2 WWTPs in Bangkok besides Bang Sue WWTP. On the other hand, MP abundances from on-site filtration are ten times lower than grab samples. Due to water pressure from a huge volume of water continuously filtered, small MP particles and fibers may escape even the smallest size of the sieve. It implies that different sampling methods influence the abundance of MPs in the same WWTP. The removal efficiency of Sing Buri WWTP was not significant and relatively low (3.19%) compared to other studied WWTPs. Despite the number of MPs found in the bottom sludge of Sing Buri WWTP, the abundance of MPs in the atmospheric deposition was considerably high. Results show from this study show that the waste stabilization pond is ineffective in reducing MPs. Gao et al. (2021) found a high concentration of MPs in duckweed (aquatic plant) in waste stabilization ponds, and as high as 789,000 MP particles were released daily from the pond effluent. However, this study's MP removal efficiency of waste stabilization ponds was not estimated (Gao et al., 2021).

A conventional system of Bang Sue WWTP removed MPs by 86.14%, which is higher than conventional WWTPs in other studies (Akarsu et al., 2020; Gündoğdu et al., 2018; Liu et al., 2019) but lower than conventional WWTPs in several studies (Bayo et al., 2020b; Edo et al., 2020; Murphy et al., 2016). The removal efficiency of Bang Sue WWTP by the UF unit following a study by Mintenig et al. (2017). A WWTP with post-filtration achieves a 97% removal rate (Mintenig et al., 2017). MP retention rate of Din Daeng WWTP (78.73%) also corresponds with conventional WWTPs in a study by Gündoğdu et al. (2018) (73 and 79%), and the similar smallest mesh sizes were used in the analysis. On the contrary, the percentage removal of Nong Khaem WWTP is the lowest compared to other conventional WWTPs, as shown in Table 2.1, because of a large proportion of fibers which is the most difficult to remove. In addition, the removal rate of Sing Buri WWTP is not comparable with the same type of WWTP because no study has been conducted on the MP removal efficiency of waste stabilization ponds until now. However, comparing the removal with other studies may not be reliable as the unit process design may differ in each treatment plant. It also depends on the MPs present in the influent to WWTPs.

4.3 Size- and shape-based removal of MPs

The 0.05-0.5 mm MPs group was the most significant proportion of MPs in the effluent from four WWTPs. Due to the lack of primary sedimentation tanks in Din Daeng, Nong Khaem, and Bang Sue WWTP, the removal of MPs depends on only final clarification after secondary treatment. Many studies suggested that the primary settling tank is the most crucial unit for MP removal (Gies et al., 2018; Liu et al., 2019; Murphy et al., 2016). Skimming and settling processes in primary treatment can remove a significant number of MPs (Carr et al., 2016; Lares et al., 2018). In addition, fibers can be easily trapped with grit or attached to larger particles and settled down in the primary settling tank (Talvitie et al., 2017b). The size range of 0.5-1 mm MPs was easily removed during primary treatment, while smaller particles (< 0.5 mm) were more likely to be trapped during final clarification (Liu et al., 2021).

Despite the utilization of UF with very fine pore sizes, MPs were found in the effluent from the UF unit. These particles might escape from larger-sized pores, membrane imperfections, or gaps between pipework (Ziajahromi et al., 2017). MPs with a size range of 20-100 µm could bypass every treatment stage, including tertiary treatment (Salvador Cesa et al., 2017). Advanced treatment technologies may increase the removal rate but not completely diminish these small-sized particles.

Fibers were detected as the most abundant shape in the effluent of all studied WWTPs. Three WWTPs with a conventional treatment system do not contain primary sedimentation as it is suggested to be an effective step for removing textile fibers (Talvitie et al., 2015). On the other hand, secondary treatment can significantly reduce synthetic particles more efficiently (Talvitie et al., 2015). MP fibers were mostly made of PET which is a high-density polymer. Thus, PET polymer is easily removed by physical sedimentation in primary treatment (Ngo et al., 2019). Adding a primary sedimentation tank may improve the removal efficiency of fibers, which is the most problematic MPs in wastewater treatment systems. However, some fibers are neutrally buoyant, which are not easily eliminated by a skimmer (Ngo et al., 2019). Due to their longitudinal shape, fibers can escape every treatment process (Liu et al., 2021). Fibers were found after membrane filtration due to their small size and shape (Talvitie et al., 2017a).

On the other hand, low-density MPs, such as fragments made of PE, float on the surface of the clarification tank. The skimming process can easily eliminate them. A scum skimmer may enhance the removal of low-density MPs.

4.4 Potential sources of MPs in WWTPs

WWTPs in this study receive wastewater from not only domestic wastewater but also combined sewer overflows. Fibers, as the most abundant group, were confirmed by FT-IR to be either PET fibers or polyester. PET is a sub-group of polyester fibers and is a predominant group in fiber production (Militky, 2009). These types of fibers are likely derived from clothes. Moreover, other types of fibers were also observed in the WWTPs, e.g., Nylon, Rayon, and linen. Even though linen is a natural polymer, natural polymers have been suggested as a carrier for harmful substances due to their treatment with chemical additives during manufacturing (Talvitie et al., 2017b). Laundering was suggested to be a significant source of MP fiber in WWTPs (Zambrano et al., 2019). A single garment can shed more than 1,900 fibers per wash (Browne et al., 2011), or approximately 700,000 fibers are discharged per average wash load (Napper & Thompson, 2016). Among synthetic textiles, clothes made from polyester lose the highest amount of fibers during laundry (Almroth et al., 2018). Washing using detergent could release more fibers than washing with only water, and the increasing washing temperature also affects fiber shedding (Yang et al., 2019b). Due to the high surface-to-area ratio of fibers compared to other shapes of MPs, fibers can adsorb more contaminants and pose a threat to human health and living organisms (Astrom, 2016).

However, not every fiber in WWTPs comes from washing clothes. Plastic fibers are spun in the form of strings, ropes, cables, and optical fibers. Moreover, some plastic products are fragmented into fibers depend on the fabrication method. For example, thermosets, cross-linked polymer chains, include vinyl ester, epoxy, phenolic, cyanate ester, polyurethane, polyimide (Erden & Ho, 2017). Fibers are commonly incorporated to enhance the properties of plastics leading to composite materials such as glass fibers which are used as reinforcement for plastics (Biron, 2020).

Personal care products were expected to be abundant in WWTPs. However, the number of microbeads found in this study was mostly negligible. One reason is the legislation of the microbead ban in Thailand since January 1, 2020 (FDA, 2019). On the other hand, microbeads might be fragmented during treatment processes resulting in irregular-shaped PE. Most of the PE fragments observed in this study are either opaque or transparent. They might be derived from the fragmentation of plastic containers. Unlike a study by Carr et al., irregular-shaped PE fragments were blue similar to those in toothpaste (Carr et al., 2016). It indicates that microbeads in personal care products can appear in the form of fragments due to the breakdown during the treatment processes.

PP is another large group of polymers in all studied WWTPs. PP is used in manufacturing food packaging, wrapper, hinged caps, containers, and pipes (Plastics Europe, 2017). PP found in the form of MPs can be derived from the fragmentation of sewage pipes or packaging, wrapper, and household containers.

Some of the MPs in Din Daeng and Bang Sue WWTPs were identified as a group of acrylic polymers. Acrylic polymers are used for lighting, electronic screen, automotive components, and outdoor glazing in architecture and construction (Chan et al., 2022). Acrylics have also been used in the coating industry due to their UV stability (Bierwagen et al., 2017).

Silicone polymer is another type of polymer detected in Nong Khaem WWTP. Silicone can be made in the form of fluids, greases, emulsions, elastomers (rubber), and resins (Greenwood & Earnshaw, 2012). Silicones have been used in various applications, e.g., insulation, sealant, and adhesive (Greenwood & Earnshaw, 2012).

PS is the last group of polymers found in Nong Khaem WWTP and a small percentage in Bang Sue WWTP. PS is used for plastic cups, egg trays, packaging, and building insulation (Plastics Europe, 2017).

Other types of polymers found in Din Daeng WWTP were polyether urethane (medical applications), poly(ethylene:propylene: diene) (EPDM: synthetic rubber), PU (furniture and insulation), and alkyd (polyester resins used for coating). Poly(ethylene:propylene: diene) was also found in Bang Sue and Sing Buri WWTPs in the form of EPDM rubber and ethylene-propylene co-polymers, respectively, based on the spectral library.

Despite the high demand for PU, PS, and PVC plastics, according to PlasticsEurope (2021), they were detected in a small proportion, and PVC was present in only Sing Buri WWTP. Fragmentation of this plastic product can create MPs, but they may not be directly discharged into the household sewer.

4.5 MP contamination in sludge

The number of MPs in sludge samples from this study ranged from 1,613–26,326 MP particles per kg of sludge (dry weight) (Figure 4.25). MPs in the sludge from Sing Buri WWTP were the least abundant among other WWTPs. It is because Sing Buri WWTP has the lowest MP retention rate. The removal of MPs in a facultative pond depends solely on gravitational settling, unlike other WWTPs. MPs can be retained by the entrapment of sludge flocs and settling down at the clarification tanks.

MP abundance in the sludge of Bang Sue WWTP was the highest among studied WWTPs. Despite the highest removal rate by secondary treatment, the number of MPs captured in sludge was lower than Din Daeng WWTP, with a percentage removal of 59.77% after secondary clarification. It can be explained by removing MPs from settling and skimming in Bang Sue WWTP. MPs were not only reduced by settling at the clarifiers but also skimmed off by a scum skimmer at the clarification tanks. However, this study did not include the number of MPs captured in the scum.

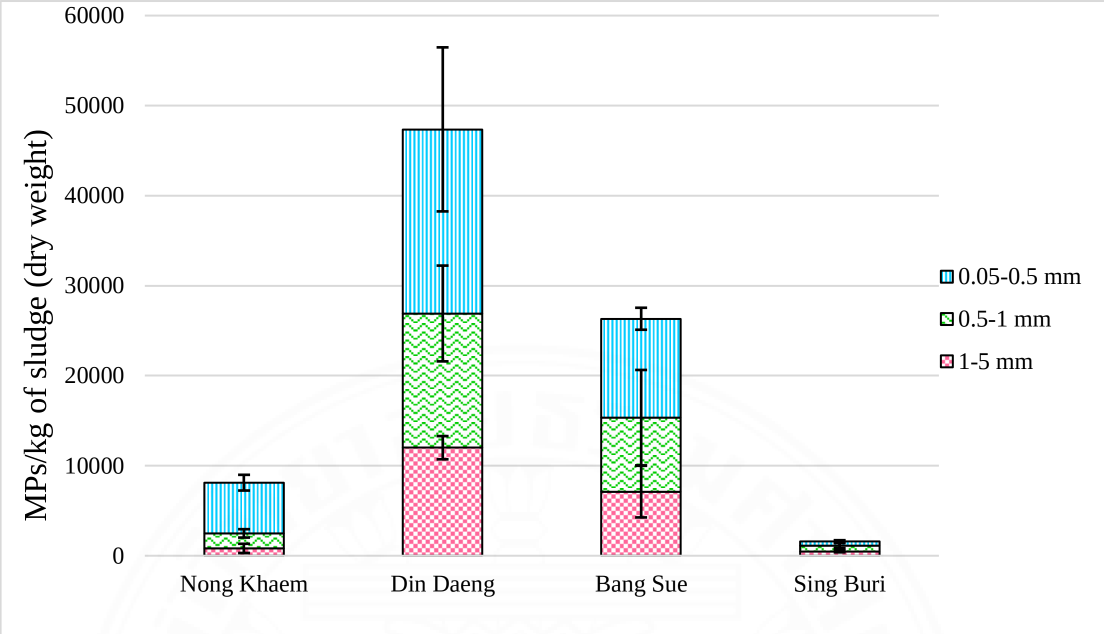


Figure 4.25 Size-based MP abundances in sludge from 4 WWTPs: Nong Khaem, Din Daeng, Bang Sue, and Sing Buri

Most of the excess sludge from the WWTPs in Bangkok is delivered to a sludge treatment facility for anaerobic digestion. The anaerobic digestion for sludge treatment potentially reduces MP concentration in sewage sludge (Mahon et al., 2016). This is due to specific enzymes from microorganisms that can break down the polymer structure of MPs into monomers (Othman et al., 2021). However, further study should focus on the microorganisms' breakdown of MPs into nano-plastics.

Air-dried sludge is usually used for soil application. Sewage sludge which contains a high number of MPs may lead to soil contamination because MPs were only transferred from the liquid phase to the solid phase. Fibers and microbeads were found in agricultural soil applied with sewage sludge (Chen et al., 2020). The characteristics of MP fibers remain the same as they were in the sludge for many years after the application (Zubris & Richards, 2005). MPs can spread to the surrounding area where there is no sludge application (Tagg et al., 2021). MPs were detected in agricultural plant roots (Yu et al., 2021). Some edible vegetables, for example, lettuces, could uptake small-sized polystyrene and transfer it to other plant parts (Li et al., 2019). MPs negatively impact fauna and flora in soil, and they pose a risk to human health when chemicals are transferred through the food chain (Kumar et al., 2020). Besides the risk of chemical leaching in soil, MPs can also transport from soil to water by surface runoff.

Incineration is expected to be a promising treatment for eliminating MPs from sewage sludge. On the contrary, MPs were detected in bottom ash and fly ash from the incinerator (Shen et al., 2021; Yang et al., 2021b). Another sludge treatment method, lime stabilization, increased the abundance of MPs due to shearing effects (Mahon et al., 2016). Hydrothermal liquefaction is a newly developed method for recycling sludge containing MPs. Bio-crude oil is produced from MP-rich sludge, and MP concentration is reduced in a residual product which can be further used as a fertilizer (Chand et al., 2022). Thus, sludge treatment for land application and disposal needs to be developed to prevent the further spread of MPs to terrestrial environments.

4.6 Limitations of sampling methods

Only grab sampling was performed in Bang Sue WWTP due to an underground system, and it results in the highest quantity of MPs in the influent. On the other hand, grab sampling and on-site filtration were performed parallelly in Sing Buri WWTP. Grab sampling results in about ten times higher number of MPs in the influent than 20-L on-site filtration. A significant portion of fibers can potentially be lost from the net and they may slip through a series of sieves (Watkins et al., 2021). Some of captured fibers may not be transferred to the container for lab processing. It results in some fiber loss during sample collection. Concerns of contamination for net-based sampling are air exposure time, MPs in rinse water, and MP contamination from the net (Barrows et al., 2017). On the other hand, grab sampling can only collect small-volume samples which might be largely contaminated by fibers (Green et al., 2018). However, grab sampling is simple and low-cost equipment demands which can be easily integrated into long-term or citizen science monitoring initiatives (Barrows et al., 2017). Grab sampling method can be utilized with existing environmental surveys to record patterns of MP contamination over space and time (Green et al., 2018). Thus, it has been suggested to use the combination of methods for an overall understanding of the concentrations (Barrows et al., 2017).

MPs can be overestimated by contamination from sampling vessels, researchers, surroundings, processing materials, and misidentification of non-plastic particles (Watkins et al., 2021). Moreover, factors that lead to underestimations of MPs include passing of small particles through mesh, low sample volume, net volume overestimated, full sample not transferred for analysis (Watkins et al., 2021).

4.7 Gaps in the current WWTPs

WWTPs are the ultimate barrier before MPs enter water bodies. The WWTPs in this study were designed to eliminate the contaminants of concern and meet the treated effluent standards. However, three conventional WWTPs are deprived of primary sedimentation, and two of them lack scum skimmers. The primary settlement has been suggested to remove fibers significantly (Talvitie et al., 2015), while skimming processes play a major role in removing lightweight MPs (Sun et al., 2019). Several studies have also suggested that the most important unit for MP removal is the primary skimming and settling stage (Carr et al., 2016; Murphy et al., 2016). The addition of primary sedimentation tanks and scum skimmers in conventional WWTPs may improve the removal rate of MPs and reduce the number of MPs released into the aquatic environment.

In addition, only one WWTP among WWTPs in Bangkok is currently equipped with tertiary treatment, and the UF unit in this WWTP is not fully operated. Tertiary treatment steps as a final polishing step can increase the overall removal rate of WWTPs (Talvitie et al., 2017a) and diminish MPs with specific properties and small-sized particles (Liu et al., 2021). WWTPs with tertiary treatment, especially MBR, achieved a higher percentage of MP removal, as reported by other studies (Table 2.1). It is necessary to consider tertiary filtration or advanced treatment to improve the quality of treated wastewater in terms of MPs.

4.8 Potential solutions

Fibers for laundering activities are the major type of MPs found in wastewater effluent. It is important to control the amount of fiber emission at the source. Some studies have suggested mitigation measures for MP fiber reduction at sources.

McIlwraith et al. (2019) investigated two technologies to prevent fiber emission: a laundry ball that can capture fibers from flowing water and an external filter for washing machine discharge. The study found that both technologies significantly reduced MP fibers in washing machines (McIlwraith et al., 2019). The external filter with a pore size of 60 µm reduced 78% of microfibers from the washing machine effluent (Napper et al., 2020). Erdle et al. (2021) studied the efficiency of washing machine filters at a community level. Installing filters on a larger scale effectively lowers the concentration of MP fibers in the connected WWTP (Erdle et al., 2021). These technologies have been suggested to control fiber emissions from laundering activities.

As the combined sewer system is employed in Bangkok Metropolitan Area, WWTPs receive both domestic and stormwater runoff wastewater. During heavy rainfall, combined sewer overflows (CSOs) are discharged directly into water bodies without treatment. Untreated CSOs might contain a high concentration of MPs and become a significant source of MPs in the aquatic environment. Therefore, measures to prevent MP load from CSOs require attention.

Filters can be installed at the end of the pipes to remove a portion of MPs. Using retention soil filters effectively removes micropollutants in CSOs (Brunsch et al., 2020). The high-rate filtration technology is an alternative to CSO treatment which has a lower investment than separating sewage and stormwater runoff (Helness et al., 2019). This filtration approach can reduce suspended solids in the CSOs by 50% (Helness et al., 2019). Moreover, the constructed wetland is another solution to reduce MP concentration and other pollutants. CSOs treatment by constructed wetlands effectively removes conventional, emerging, and pathogenic pollutants (Rizzo et al., 2020). Studies have suggested that constructed wetlands are a promising approach for MP removal (Lu et al., 2022; Xu et al., 2022; Zhou et al., 2022). MP retention rate of sand-filled constructed wetlands on lab-scale was more than 98.8% (Wang et al., 2021b).

In the case of sub-urban and urban provinces with no WWTP, constructed wetlands can be utilized to prevent the spread of MPs into the aquatic environment. The MP removal efficiency of more than 89% by constructed wetlands has been reported in China (Zhou et al., 2022). Constructed wetlands utilized as tertiary treatment step has the average MP removal rate of 88%, and it has been suggested as an efficient filter for preventing MPs transport to aquatic systems (Wang et al., 2020). However, constructed wetlands require spaces for construction. Therefore, rapid sand filtration is one of wastewater treatment methods which showed high removal efficiency of 97% (Talvitie et al., 2017a). It is also considered cost-effective method when compared to MBR system (Iyare et al., 2020). Rapid sand filter can be an alternative for MP removal before they enter the aquatic environment.

CHAPTER 5
CONCLUSIONS AND RECOMMENDATIONS

5.1 Conclusion

WWTPs are one of the important land-based sources of MPs in the aquatic and marine environment. Four WWTPs investigated in this study showed different removal as all four plants differ. Bang Sue WWTP, constructed as a closed underground system, achieved the highest MP removal percentage among studied WWTPs. The underground system prevents airborne contamination, and MPs were removed by the scum skimmer at the clarifiers and settled with the sludge. This WWTP is equipped with the UF unit, which increases the overall removal to 96.97%. The advanced filtration further removed larger-sized MPs remaining in the secondary effluent. However, the operation of the UF unit in this WWTP is still on a pilot scale. The removal efficiency attained by a WWTP with a waste stabilization pond was minimal. MPs from atmospheric deposition may increase the abundance of MPs in the pond system. Moreover, sampling methods (on-site filtration and grab sampling) influence the quantity of MPs.

Three conventional WWTPs lack a primary settling tank which has been suggested to remove some types of MPs efficiently. The addition of membrane filtration enhances the removal rate. Despite the effective removal by the UF, a considerable number of MPs are discharged daily to the recipient water due to a large volume of treated wastewater. Therefore, the elimination of MPs should not rely only on the wastewater treatment system. Reducing plastic waste at sources, such as filters at washing machines to prevent fiber loss, is another solution to consider.

MPs are retained in sewage sludge after they are removed from wastewater. Sewage sludge from conventional WWTPs in Bangkok is treated with anaerobic digestion before land application. Introducing sludge containing MPs in the soil can spread microplastics to the terrestrial environment. Moreover, the accumulation of MPs was found in some edible plants, which can be transferred to human bodies. Therefore, the management and technology of MPs-rich sludge should be developed.

This study shows that the WWTP design and treatment technologies influence MP removal. Final-stage technologies, such as membrane filtration, play an essential role in removing MPs.

5.2 Recommendation for future study

In this study, even though there is an absence of primary sedimentation and the skimming process is applied in only one conventional WWTP, the discharge quality of treated water meets the standard (e.g., pH, total suspended solids, BOD, and COD). To improve the removal of this emerging pollutant (MPs), WWTPs need an upgrade. Including primary settling and skimming in conventional WWTP may reduce the concentration of MP released to water bodies. Moreover, the removal of MPs in conventional WWTPs is involved with biological processes in secondary treatment. The effects of biofilms on MP surface and the fate of MPs in aeration tanks should be further investigated. In addition, a lab-scale experiment can be set up to verify the most effective method for MP removal and improve current wastewater treatment systems for MP removal.

This study found that UF as tertiary filtration is effective in removing MPs. The addition of advanced treatment must consider many factors, such as cost-effectiveness, plant design, area availability, and population. Fibers are the most abundant type of MPs in the wastewater treatment system and can escape the small pore size of UF. Thus, fibers should be reduced not only by the treatment but also at sources by filtering at washing machine discharge. The design of tertiary treatment needs to consider the complexity of wastewater being processed and the contaminants of concern to improve the quality of treated wastewater. The investment in advanced technologies should consider the long-term effects of MP pollution.

Only ten spots of the smaller size range of MPs on filter papers were randomly selected and identified by micro-FTIR. The number of spots selected for micro-FTIR should be increased to enhance the success rate. As MPs in the size range of 0.05-0.5 mm were investigated in this study, smaller-sized MPs (down to 1 μm) should be further focused. Due to degradation, small-sized MPs are more abundant in the environment than larger MPs.

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APPENDICES

APPENDIX A

TYPES OF RESIDENTS IN THE SERVICE AREA OF THREE WASTEWATER TREATMENT PLANTS IN BANGKOK

Table A.1 the number of different types of residents in the service area of three WWTPs in Bangkok in this study.

Types of residents	Community housing	Suburban housing	Urban housing	Housing estate	High-rise building	Slum
Nong Khaem WWTP	0	27	48	34	2	34
Din Daeng WWTP	2	4	-	73	31	64
Bang Sue WWTP	0	10	1	35	29	91

APPENDIX B

POLYMER TYPE IDENTIFICATION OF MPs

B.1 Analysis of MPs by ATR-FTIR

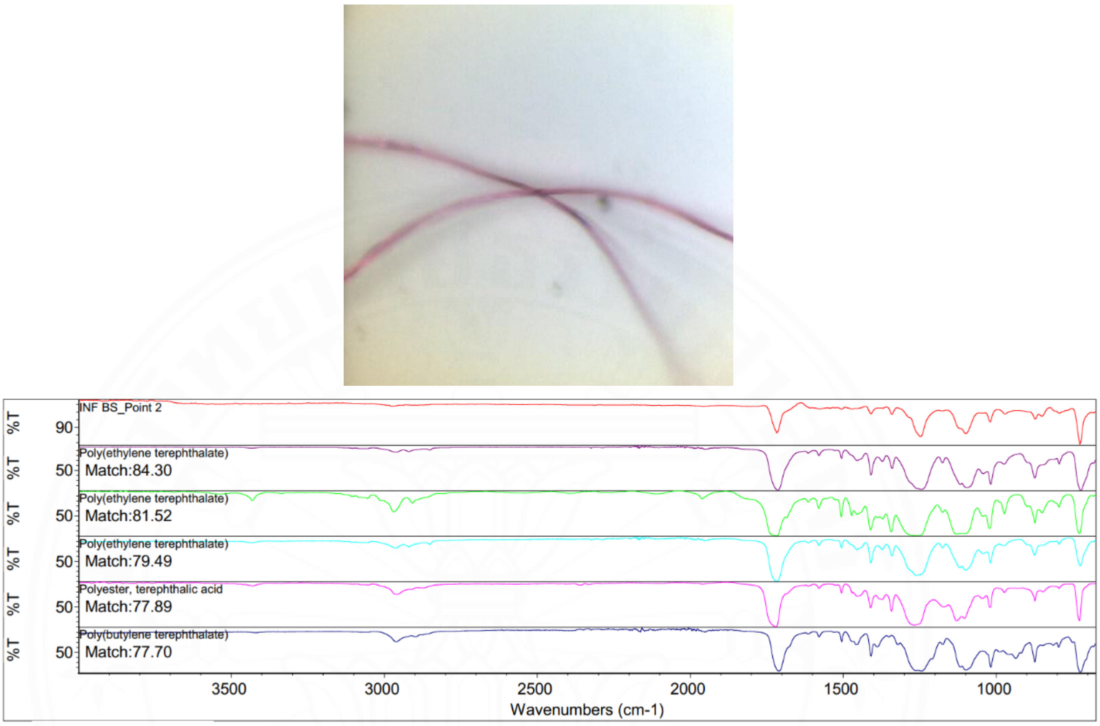
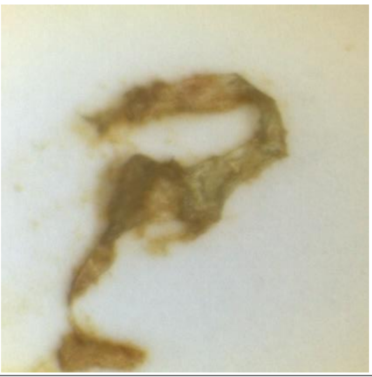


Figure B.1 Red fiber from the influent of Bang Sue WWTP identified by ATR-FT-IR connected to microscope as polyethylene terephthalate (PET)



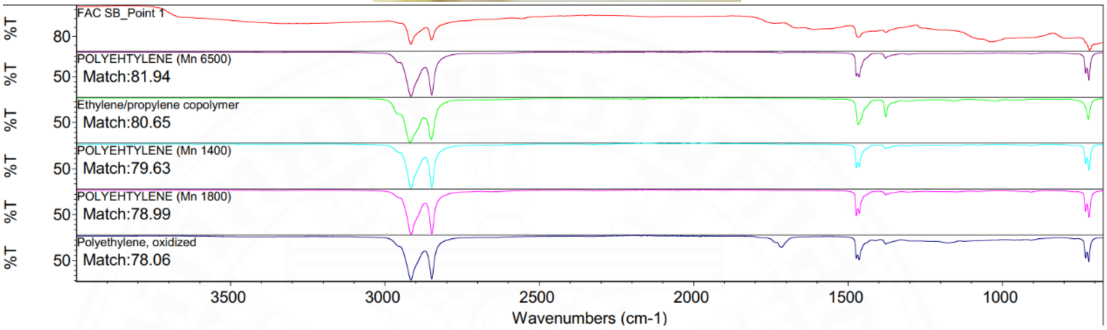


Figure B.2 Fragment from the outlet of facultative pond from Sing Buri WWTP identified by ATR-FT-IR connected to microscope as polyethylene (PE)

BIOGRAPHY

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