Micro- and Nanoplastics Removal through Drinking Water Treatment Processes: Insights from Published Investigations and Development of a Possible Regulatory Approach

by

Jaita Saha

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Author’s Declaration

I hereby declare that I am the sole author of this thesis. This is a true copy of the thesis, including any required final revisions, as accepted by my examiners.

I understand that my thesis may be made electronically available to the public.

Abstract

Micro- and nanoplastics (MNPs) have emerged as significant environmental pollutants with potential implications for human health. The presence of MNPs in freshwater bodies, such as lakes, rivers, and sometimes groundwater, is primarily attributed to the extensive use and degradation of plastic products. This contamination raises concerns for human health, given the critical role of freshwater as the primary drinking water source, through which MNPs can be ingested. However, there is limited information available regarding human exposure and the associated toxicity of MNPs in freshwater, and the area of nanoplastics (NPs) in freshwater environments remains relatively unexplored. This thesis addresses the limited knowledge surrounding MNPs in freshwater sources, particularly in relation to their abundance, removal efficiencies during treatment, potential toxicity, and regulatory considerations. The thesis is focused on evaluating data from full-scale drinking water treatment plant (DWTP) surveys, assessing bench-scale studies on MNPs removal, understanding toxicological impacts, summarizing legislative standards, and developing a regulatory framework.

The research approach involves analyzing data from full-scale DWTP surveys and bench-scale studies to assess MNPs abundance and removal efficiencies of different categories of MNPs, considering their size, shape, and polymer type. Special attention is given to coagulation-flocculation-sedimentation (CFS) and filtration processes. The findings from full-scale DWTP surveys reveal significant variability in MNPs abundance and removal efficiencies. Advanced DWTPs generally exhibit higher removal rates compared to conventional plants. However, the variability in results highlights the differences in MNPs properties, analysis techniques, and treatment procedures, making it challenging to establish definitive conclusions. Bench-scale studies indicate the effectiveness of CFS and filtration processes in MNPs removal, although findings differ due to variations in experimental conditions and methodological inconsistencies. This thesis highlights the need for consistency in sampling, quantification techniques, and reporting standards to establish a uniform dataset. Further research is necessary to better understand MNPs removal efficiencies, especially for NPs, and to address the lack of standardized methods. Additionally, the thesis examines current legislative standards, if any, related to MNPs in drinking water sources. The potential toxicological impacts of MNPs and their distinct characteristics compared to other micropollutants are also evaluated. Based on the findings, a regulatory framework is proposed to address the presence of MNPs in drinking water and reduce potential health risks. The proposed regulatory framework draws inspiration from existing approaches, but

acknowledges the unique challenges posed by MNPs properties based on their classification and prevalence in the source water. Challenges in implementation include financial barriers and monitoring difficulties, along with the need for continuous assessment and further research.

This thesis contributes to the understanding of MNPs in freshwater sources and provides insights into their removal efficiencies. The proposed regulatory framework lays the groundwork for developing guidelines to mitigate human health risks associated with MNPs in drinking water. Further research and collaboration are essential to address the current knowledge gaps and establish effective strategies for managing MNPs in drinking water.

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List of Abbreviations

ABS: acrylonitrile butadiene styrene

AC: activated carbon

ADWTP: advanced drinking water treatment plant

ASA: activated silicic acid

ATR: attenuated total reflection

CA: cellulose acetate

CFS: coagulation-flocculation-sedimentation

DAF: dissolved air flotation

DI: deionized

DWTPs: drinking water treatment plants

EBCT: empty bed contact time

EDCs: endocrine disrupting chemicals

EPA: environmental protection agency

EVA: ethylene vinyl acetate copolymer

FTIR: fourier transform infrared spectroscopy

IL(1): 1-methyl-3-propylimidazolium chloride

IL(2): 1-decyl-3-methylimidazolium chloride

LDPE: low-density polyethylene

LOQ: limit of quantification

MAC: maximum acceptable concentration

MNPs: micro- and nanoplastics

MPs: microplastics

NPs: nanoplastics

OCPs: organochlorine pesticides

PA: polyamide

PA6: polyamide-nylon 6

PACl: poly-aluminum chloride

PAM: polyacrylamide

PAN: polyacrylonitrile

PBA: poly(butyl acrylate)

PBDEs: polybrominated diphenyl ethers

PC: polyamine-coated

PCBs: polychlorinated biphenyls

PE: polyethylene

PEG: polyethylene glycol

PEO: polyethylene oxide

PES: polyester

PET: polyethylene terephthalate

PFAS: per- and polyfluoroalkyl substances

PMMA: poly(methyl methacrylate)

PolyDADMAC: diallyldimethylammonium chloride

PP: polypropylene

PPCPs: pharmaceuticals and personal care products

PS: polystyrene

PTFE: polytetrafluoroethylene

PTT: polytrimethylene terephthalate

PVA: polyvinyl acetate

PVC: polyvinyl chloride

PVC-u: unplasticised polyvinyl chloride

PVDF: polyvinylidene fluoride

ROS: reactive oxygen species

RPM: revolutions per Minute

SA: sodium alginate

SDWTP: sub-drinking water treatment plant

SEM: scanning electron microscopy

TOC: total organic carbon

UF: ultrafiltration

UV: ultraviolet

VINYL: vinyl chloride-vinyl acetate copolymer

VINYON: vinyon

WPET: weathered polyethylene terephthalate

WT: water treatment

WTP: water treatment plant

WWTPs: wastewater treatment plants

Disclaimer

In several instances in this study, commercial products and/or trade names were mentioned. The use of commercial names does not constitute endorsement or recommendation of these products and/or instruments by the author.

Chapter 1
Introduction

1.1 Problem Statement

Micro- and nanoplastics (MNPs) have been considered as one of the emerging pollutants which have the potential to cause harm to the environment and human health. The main underlying reason behind the significant appearance of MNPs is the extensive use of plastic products, followed by their disintegration into plastic particles (González-Pleiter et al., 2019). Such degradation gives rise to the expansive presence of microplastics (MPs) and even smaller-sized nanoplastics (NPs), which is further augmented by their deliberate creation for various consumer and industrial applications (Yee et al., 2021). These MNPs eventually find their way into water bodies and accumulate in lakes, estuaries, open seas, or oceans (W. Luo et al., 2019). There have been extensive investigations in the field of marine environments regarding the existence of MNPs in seas and oceans for a very long time. But studies focusing on MNPs in freshwater have been limited. The contamination of MNPs in freshwater sources is of particular concern for human health due to the critical role of freshwater as the primary drinking water source. Humans can be easily exposed to MNPs from drinking water sources through ingestion or inhalation (Koelmans et al., 2019). Besides, MNPs in freshwater vary in abundance and various properties like composition, size, shape, polymer type, etc. These characteristics may influence the potential toxic effects associated with MNPs. But in response to the growing concern about MNPs in drinking water, there is very little information available about human exposure and potential toxicity related to these particles (Barbosa et al., 2020). Therefore, it is crucial to give significant attention to the potential adverse health impacts that may arise from the intake of MNPs by humans from freshwater sources. Moreover, NPs have been less explored in the aquatic environment than MPs, primarily due to the challenges in their detection caused by their incredibly small size (Arenas et al., 2022). But it has been postulated that the potential toxicity and risks are more associated with the smaller particles than the larger ones. So, research should be extended to put focus on both MNPs. Again, considering the fact that drinking water is predominantly sourced from freshwater and can be a potential exposure route to MNPs for humans, the focus of the study is limited to freshwater sources.

In response to the growing presence of MNPs in freshwater systems (i.e., rivers, lakes, surface water, and groundwater sources), there has been a surge in the number of research studies investigating the removal of MNPs. Full-scale surveys are looking into the removal of MPs during the usual operation of conventional or advanced drinking water treatment plants (DWTPs). Bench-scale studies are

optimizing their experimental condition to observe the MNPs removal. But as yet the available information from both types of studies is in disintegrated form and there is a lack of consistency regarding the quantification of MNPs and insufficient details in the reporting of the operational specifics of the treatment processes. In addition, MNP abundance and characteristics can vary widely depending on the source water. These diversities are reflected in the reported results of the various studies. Besides, there is no regulatory limit for MNPs in drinking water at the national and international level. The absence of defined priorities for the removal of MNPs from drinking water highlights the need to establish specific guidelines for their presence and removal through drinking water treatment processes.

1.2 Objective

In order to assess the available information on MNPs from the previously published studies, two broad objectives were defined. The first one aims to conduct a meta-analysis of existing data accessible through different research studies on the presence of MNPs in drinking water sources and their relevant removal efficiencies through various drinking water treatment processes. The second one aims to propose a regulatory approach to eliminate/reduce potential human health impacts from exposure to MNPs in potable water to an acceptable level.

To accomplish these broad objectives, we can establish some sub-objectives:

- Evaluate the data available from the full-scale DWTP surveys on the abundance of different categorized MNPs (size, shape, polymer type) in various drinking water sources and analyze their removal through different combinations of conventional or advanced DWTP processes, with a particular focus on coagulation-flocculation-sedimentation (CFS) and filtration processes.
- Assess the existing information derived from bench-scale studies that aim to identify effective conditions for the removal of MNPs with different properties by optimizing various factors through CFS and filtration processes.
- Summarize the current legislative standards, if any, regarding MNPs in drinking water sources.
- Develop an overview of the toxicological impacts associated with the presence of MNPs in potable water and acknowledge the accompanying human health concerns.
- Develop a framework for an approach to regulate MNPs should it be necessary to reduce the exposure of MNPs through drinking water.

1.3 Research Approach

The abundance and removal of MNPs have been investigated in various full-scale DWTP surveys and bench-scale studies, whereas for NPs have only been evaluated by bench-scale studies until now. The full-scale DWTP surveys have provided significant insights into the MPs abundance of different characteristics (size, shape, polymer type) in diverse raw water sources of various locations and their removal efficiency by DWTPs consisting of different combinations of treatment units. To observe if there is any relationship between the removal efficiency of MPs and their abundance and different characteristics, the existing information from the relevant published studies was extracted and analyzed. In studying the data from these full-scale surveys, a particular focus was put on CFS and filtration processes. Bench-scale studies have also been found evaluating the removal efficiency of MNPs for some specific characteristics by CFS and filtration methods separately. The available information from those published bench-scale studies was also analyzed to determine the extent of MNPs removal efficiency with different characteristics under optimized conditions. Then comparative analyses were conducted, and conclusions were drawn based on the findings. In addition, based on the available information about the potential toxicity and human health concerns associated with MNPs, as well as their distinctions from other micropollutants, the need for the development of regulations for MNPs in drinking water was considered. Existing legislation for related dissolved micropollutants and ongoing efforts for regulating MNPs in source water were also reviewed. Finally, a framework for the development of a regulation for MNPs presence in drinking water was suggested and possible steps for implementing such a regulation were considered.

1.4 Thesis Organization

This thesis comprises six chapters, along with a list of references, and appendices.

Chapter 1 introduces the research topic and provides a rationale for the study. It outlines the overall goal of the research and presents detailed objectives that will be addressed throughout the thesis.

Chapter 2 provides a general overview of the existing literature on plastics, MPs, and NPs, with a focus on their sources, pathways, and occurrence in drinking water sources. It also describes various treatment methods used in DWTPs and the importance of removing MNPs through these DWTPs. This chapter aims to identify knowledge gaps and provides background information to support the research objectives.

Chapter 3 outlines the available information from full-scale DWTP surveys and presents the analysis of the data extracted from those surveys. The data analysis is followed by discussions on the findings.

Chapter 4 outlines the available information from the bench scale studies and represents the analysis of the data collected from those studies. The data analysis is again followed by discussions on the findings.

Chapter 5 includes background information about the toxicity and human health concerns of MNPs, their difference from other micropollutants, and a review of the existing legislation and ongoing efforts for regulating MNPs and some other micropollutants. Then a proposal for a regulatory framework for MNPs in drinking water is presented, followed by discussions on its components and possible considerations for implementing MNP regulations.

Chapter 6 summarizes the key conclusions of the research, implications for the water treatment industry, acknowledgment of the limitations, and recommendations for further research.

Chapter 2
Background

2.1 Overview

2.1.1 Plastics

Plastics are one of the most ubiquitous materials in the modern world. The ambiguity of the term 'plastic' can be pervasive as it encompasses a wide range of materials with very different characteristics (Boyle & Örmeci, 2020). They consist of extended, repetitive sequences of polymers, exhibit remarkable structural stability, resilience, and flexibility, among other attributes, and can be easily molded into various shapes and sizes (e.g., Bening et al., 2021). Due to their cost-effectiveness, versatility, and adaptability, plastics have become a critical component in the manufacturing process. The industrial revolution, population growth, urbanization, and technological advancements have all contributed to the widespread availability and mass production of plastic items (Adegoke et al., 2023). Such production of plastics on a large scale and their growing utilization in various applications (see Figure 2.1) has led to a throw-away culture of single-use products, leading to a significant accumulation of plastic waste due to their non-biodegradability and intrinsic persistence (Barnes et al., 2009). This issue is compounded by poor recycling, slow degradation, and high production rates, resulting in alarming levels of plastic accumulation in the environment (Schwarz et al., 2019).

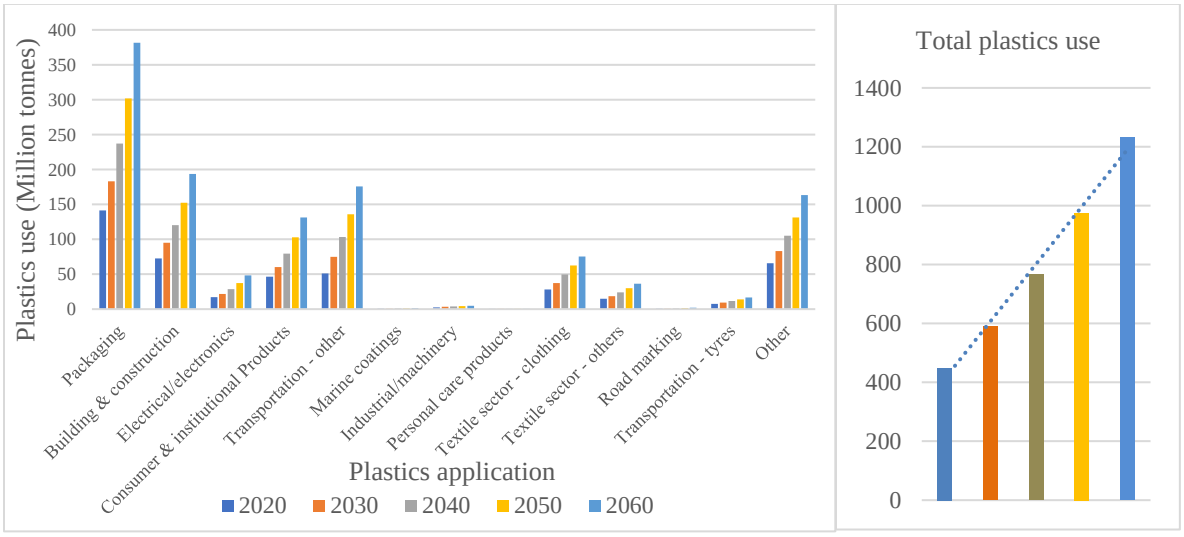


Figure 2.1: Plastics use by the application at the global level of 2020 and the projections for 2030, 2040, 2050, and 2060 (Generated the graph based on the data obtained from OECD ILibrary)

Plastics can break down into smaller-sized particles in a variety of pathways, either through biodegradation or non-biodegradation processes. Processes such as physical degradation, thermal degradation, thermo-oxidative degradation, photodegradation, and hydrolysis are involved in the non-biodegradation of plastics (e.g., Turner et al., 2020). These processes cause changes in the mechanical properties of polymeric structures and increase their surface area, which leads to greater physical-chemical reactions and interactions with microorganisms (Lucas et al., 2008). On the other hand, environmental bacteria and other microorganisms can facilitate the biodegradation process, where enzymes are secreted to cleave the plastic polymer chains through hydrolytic processes, reducing their molecular weight (da Costa et al., 2017). These degradation processes give birth to another environmental concern called micro- and nanoplastics (MNPs). Though MNPs may originate from a variety of sources, fragmentation from plastic particles is one of the most potent sources of their abundance (Karbalaei et al., 2018).

2.1.2 Microplastics (MPs)

MPs are a common form of plastics found in the environment and can exist in various shapes, sizes, polymer types, densities, colors, and along with some other properties. The definition of MPs has gone through many debates regarding the size limit, types, and shapes of MPs. Usually, plastic fragments with a main dimension lower than 5 mm are considered as MPs, where the lower boundary is typically set to 1 µm to separate NPs from MPs. But several authors considered different lower size limits (within 1 to 20 µm), while there was also uncertainty regarding the upper size limit either being 1 mm or 5 mm (Van Cauwenberghe et al., 2015). The most common types of MPs are fragments, fibers, and pellets, with microbeads, films, filaments, ropes, foams, sponges, and rubber also contributing to MPs pollution, albeit in decreasing order (Frias et al., 2018). It is worth noting that different countries may utilize varying terminology to categorize the same type of plastic. Color is not considered an essential aspect in defining MPs as it is subjective and cannot solely be relied upon for visual identification (Lusher et al., 2017). There have also been some attempts made to include physiochemical properties such as their chemical composition in the definition. MPs in the environment are also grouped according to their origin: primary and secondary, where the former ones are designed purposefully, and the latter ones are the results of the fragmentation of larger plastic particles (see Figure 2.2). Considering all these variances, the proposed definition for MPs is “Microplastics are any synthetic solid particle or polymeric matrix, with regular or irregular shape and with size ranging from 1 µm to

5 mm, of either primary or secondary manufacturing origin, which are insoluble in water" (Frias & Nash, 2019).

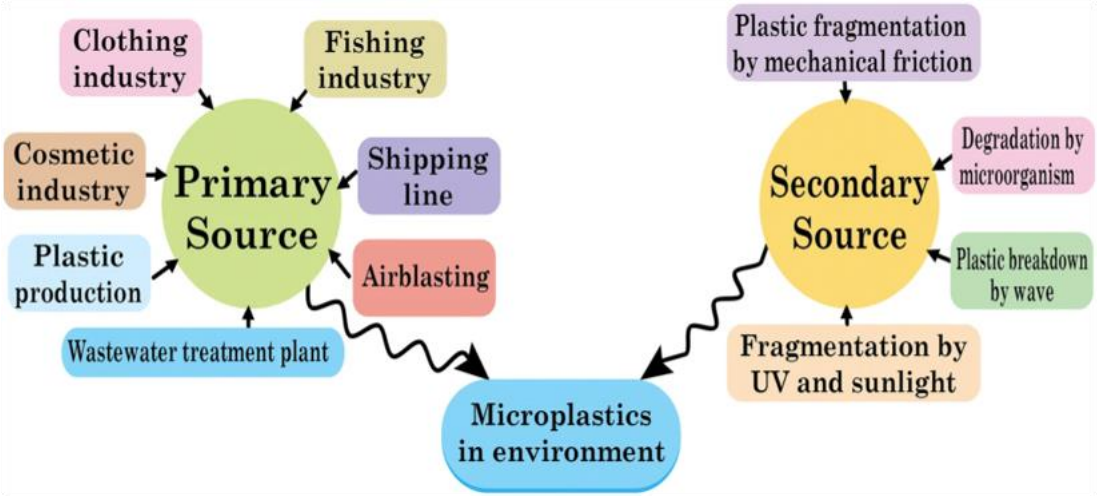


Figure 2.2: Origin of MPs
(Dey et al., 2021)

2.1.3 Nanoplastics (NPs)

The term NPs has been used to characterize plastic particles smaller than MPs. A clear definition of NPs has not been provided yet. The size limit for NPs is recommended as 1 nm to 1 µm (Hartmann et al., 2019). But some researchers follow the commonly used threshold (<100 nm) for nanotechnology (Wagner & Reemtsma, 2019). NPs are difficult to study and characterize due to their extremely small size, which has resulted in a lack of research in this area (Boyle & Örmeci, 2020). Most NPs are formed when larger plastic particles break down due to different environmental processes (physical, chemical, or biological) or when aged MPs are exposed to similar conditions (Bouwmeester et al., 2015). NPs are also directly manufactured and utilized in different consumer products like toothpaste, facial cleansers, cosmetics, waterproof coatings, and medicines (e.g., Chae & An, 2017; Koelmans et al., 2015). The assessment of the fate of NPs is inherently challenging because of their small size, but studying the fate of MPs can provide insight into their distribution in the environment (da Costa et al., 2016).

2.1.4 Occurrence of MNPs in Drinking Water Sources

The sources of MNPs can be categorized into two main groups: primary and secondary. The primary sources include intentionally engineered microbeads found in hygiene and beauty products (e.g., exfoliants), industrial abrasives, pellets for the manufacturing of larger plastic products (da Costa et al.,

2016), and emissions from 3D printers (Stephens et al., 2013). Typically, these sources exhibit well-defined sizes, shapes, and densities (Boyle & Örmeci, 2020). Plastic particles that disintegrate into smaller-sized particles whether in the terrestrial or aquatic environment are known as the secondary source (Karbalaei et al., 2018). The degradation of plastics results in the formation of MNPs with irregular sizes, shapes, densities, and different chemical compositions (Duis & Coors, 2016). Besides, plastic is a man-made material that is mostly produced on land. According to Yee et al. (2021), marine sources are responsible for less than 20% of MPs, while more than 80% are produced from land-based sources. So, MNPs can enter aquatic environments through these various sources such as environmental degradation of discarded plastic items, spills from industrial activities, and the physical wear and tear of plastic items in use (Chang, 2015; Hartline et al., 2016; Lambert et al., 2014; Lechner & Ramler, 2015). However, plastic particles can also be increasingly found in aquatic systems due to their accumulation in sewage sludge and treated effluents. While current wastewater treatment processes have been demonstrated to be effective in removing MPs (Carr et al., 2016), there is uncertainty regarding their effectiveness in removing smaller particles like NPs. In addition, the efficiency of removal may vary depending on the specific treatment processes employed, as some other studies have reported inefficient removal of MPs in WWTPs (Browne et al., 2011). Even if they receive good removals (e.g., >90%), WWTPs can still release substantial amounts of MNPs (depending on the MNPs load in the wastewater) into freshwater bodies through their large volume of effluent deposition (Rout et al., 2022). As MNPs are difficult to completely remove because of their small size, they enter the marine and surface water sources (Vance et al., 2015). The sludge from these WWTPs has been found to be commonly used for agricultural purposes providing another route for MNPs into surface water (Eerkes-Medrano et al., 2019). Moreover, MNPs can also be transported from soil to rivers and oceans through natural erosion processes (Horton et al., 2017). All these processes make WWTPs a substantial source of MNPs in freshwater sources. Researchers have also suggested that atmospheric transport and deposition by wind or precipitation may be a route for MNPs to enter aquatic environments, based on the detection of MPs in atmospheric samples (Dris et al., 2016; Free et al., 2014). However, a comprehensive overview of the sources and fate of MNPs in the environment can be found in Figure 2.3.

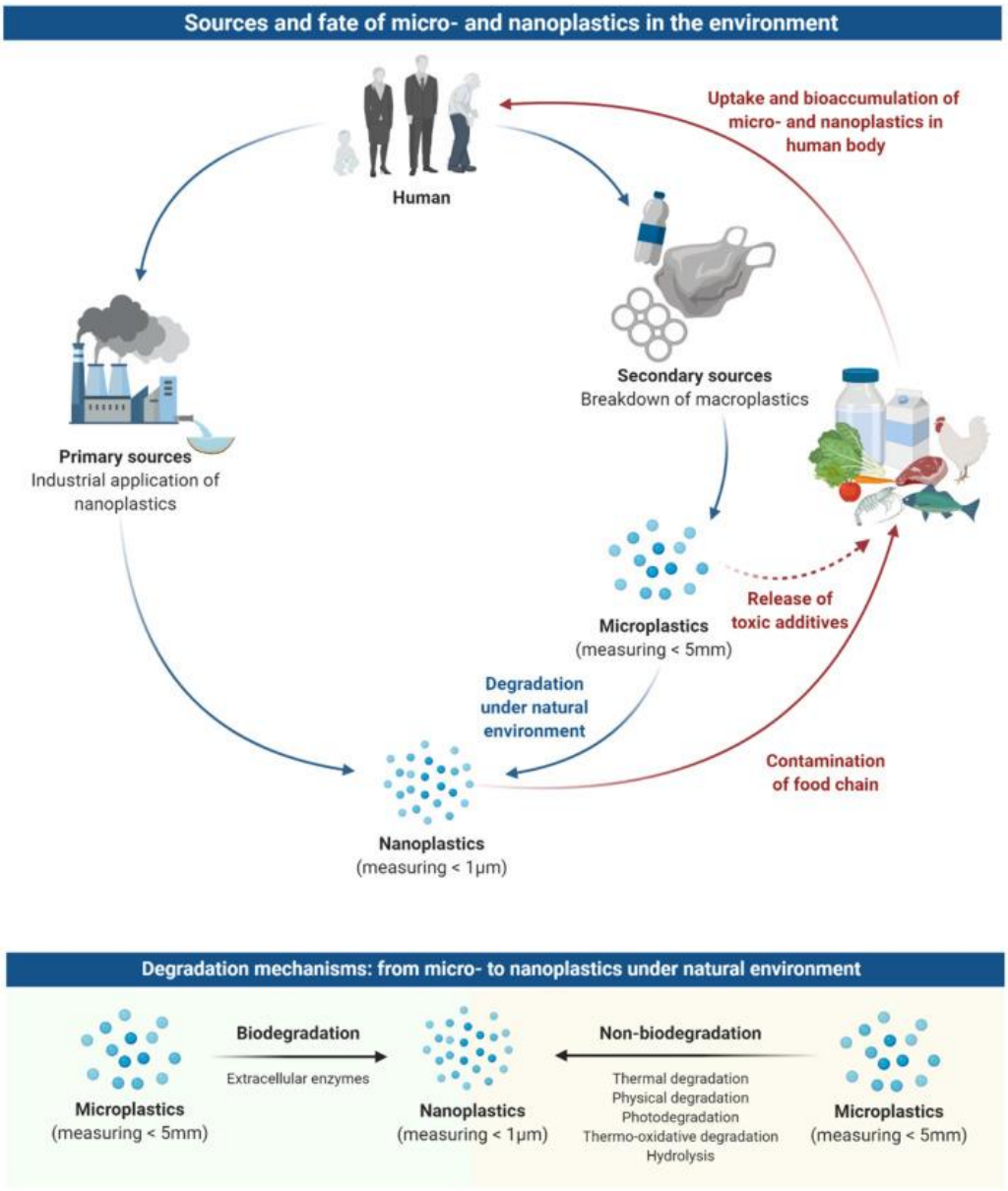


Figure 2.3: Sources and fate of MNPs in the environment
(Yee et al., 2021)

Research on the occurrence and fate of MNPs has predominantly focused on marine environments, with a limited amount of research conducted in freshwater environments. Moreover, research on NPs is less prevalent in both marine and freshwater environments when compared to MPs. Figure 2.4 presents a summary of the annual research publications on MNPs in marine and freshwater environments.

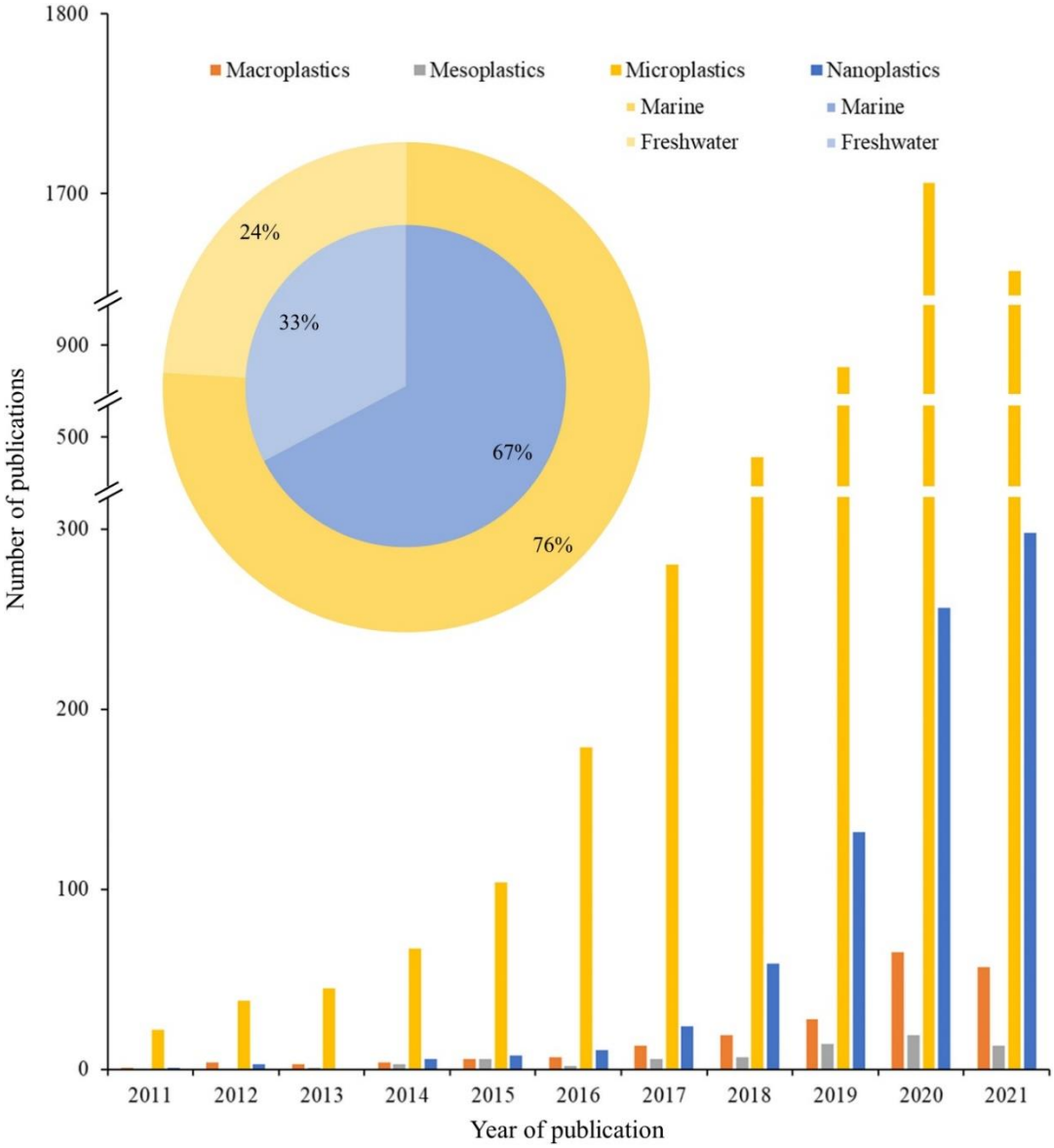


Figure 2.4: Annual number of publications on MNPs in marine and freshwater environments (Zaki & Aris, 2022)

The prime source of plastic pollution in the marine environment comes from terrestrial sources, coastal tourism, fishing, and other aquaculture activities (e.g., Browne et al., 2011). Marine environments serve as ultimate repositories for varied anthropogenic pollutants including MNPs (Oliveira & Almeida, 2019). Marine water is less commonly used as a source of drinking water, although in some locations it is the only source. With increasing population growth, urban development, and industrialization, there can be more need for freshwater than the sources available. Therefore, seawater desalination is

considered as an increasingly sought-after alternative for water supply. Seawater can be converted into freshwater through several methods such as reverse osmosis, distillation, or electrodialysis. So, based on this expensive and energy extensive technology, marine water can also be a potential source of drinking water, especially in areas with stressed and overdrawn freshwater resources.

The occurrence of MNPs in freshwater environments is not as widely recognized and documented as in marine environments. Freshwater, including surface water and groundwater, is broadly known as the main source of drinking water. Figure 2.5 provides an overview of the sources of drinking water and its transport routes for human consumption. MPs have been found to occur in freshwater at different levels ranging from almost zero to several million particles per liter (Shen et al., 2020). This distribution is influenced by multiple factors including the shape, size, and type of MNPs as well as the presence of other pollutants (Ding et al., 2021).

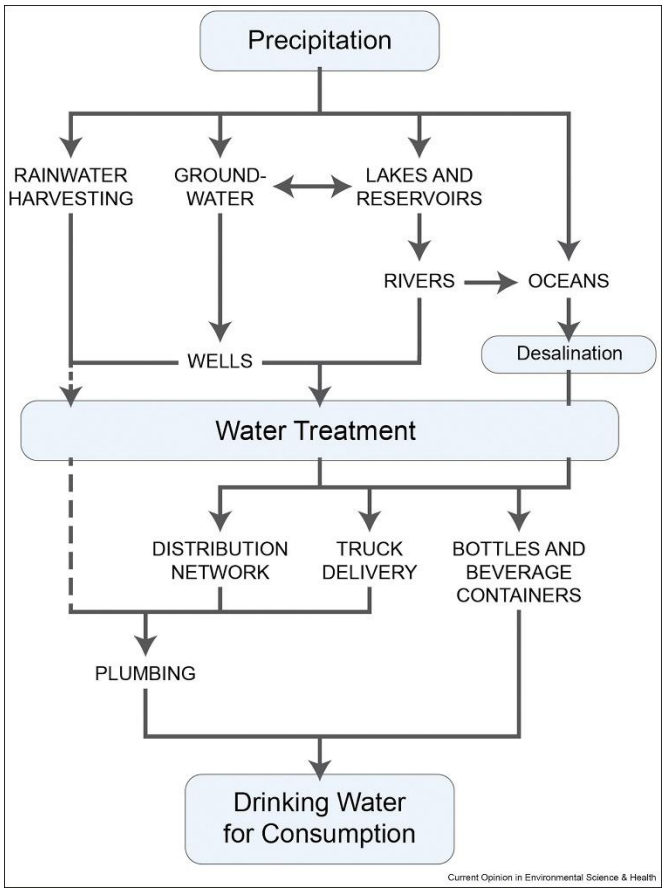


Figure 2.5: Sources of drinking water and transport routes taken towards human consumption (Eerkes-Medrano et al., 2019)

Surface water is accumulated above ground in a stream, river, lake, or reservoir. Surface water sources are vulnerable to contamination from multiple sources, including human and animal waste, as well as other forms of pollution. This susceptibility makes them particularly prone to the presence of harmful organisms such as bacteria, viruses, and parasites, which can pose significant health risks and lead to serious illnesses and diseases. But in many locations surface water is more easily accessible than groundwater, so surface water is commonly used for drinking purposes after appropriate treatment. Researchers worldwide have begun examining surface water as a potential source for MNPs uptake and have already found their significant presence.

Despite the lack of global attention on possible MNPs contamination in groundwater, a few studies have reported the presence of MPs (e.g., Mintenig et al., 2019; Samandra et al., 2022). They detected a comparatively lower number of particles in their groundwater samples. As groundwater sources are considered less likely to be contaminated with MNPs, the research in this area is still in the preliminary stage. More studies will be required to investigate the presence of both MNPs to identify whether groundwater contamination with MNPs is a matter of concern throughout the world like marine and surface water sources.

To mitigate possible MNPs exposure to humans through drinking water sources, DWTPs can play an important role. Drinking water sources can be contaminated with various types of pollutants including different microorganisms and harmful chemicals. DWTPs treat water by removing undesirable particles and dissolved contaminants from drinking water sources before supplying that to the distribution system for human consumption. So, to ensure drinking water quality, the evaluation of the removal of MNPs in DWTPs is essential.

2.1.5 Drinking Water Treatment Plants (DWTPs)

Water treatment is the process of removing various suspended and colloidal particles and dissolved substances which are generally considered harmful or undesirable. The DWTP is a facility designed to attain that goal and make the water safe for consumption. The purpose of this is to remove impurities and contaminants from the water source, which could include microorganisms, pathogens, chemicals, and particulate matter, ensuring that the public has access to clean and safe drinking water that is both visually and taste-wise pleasing. The process of water treatment involves purification and clarification of water, sometimes followed by softening and deodorizing, to make it potable and more useful. This

helps to protect the health and well-being of both people and the environment, providing reliable access to clean drinking water for the communities they serve.

Public drinking water systems employ various water treatment methods to ensure that the water supplied to their communities is safe for consumption. For surface water, these systems typically use a series of treatment steps, including coagulation, flocculation, sedimentation, filtration, and disinfection (see Figure 2.6). Sometimes they incorporate additional treatment measures such as pre- or post-oxidation, granular activated carbon filtration, membrane filtration, or adsorption processes. These steps work together to remove particles and organisms that can cause disease, remove dissolved contaminants, improve the taste and appearance of the water, and ensure that the water is safe and reliable for human consumption.

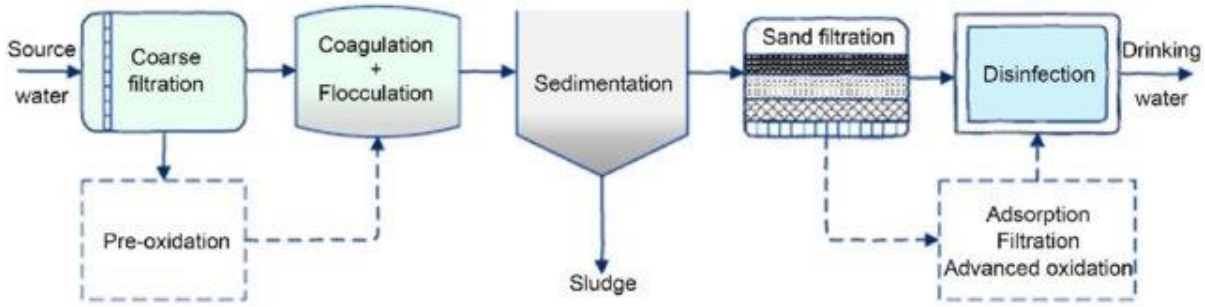


Figure 2.6: Overview of drinking water treatment
(Ghernaout et al., 2020)

Coagulation is a crucial chemical process in water treatment that is the first step in eliminating solids from water, by altering the electrostatic charges of suspended particles. This is achieved by introducing coagulants, which can form small, highly charged (generally opposite to those of particles) molecules, into the water to destabilize the charges on those dispersed particles or colloids in suspension. Coagulants can also perform “sweep coagulation”, whereby particles to be removed are incorporated into the flocs formed. The choice of type and concentration of coagulant varies depending on the conditions of the water and the DWTP. Choosing the right coagulant and its dosage is essential for improving the overall efficiency of the system, particularly in removing solids, which ultimately enhances the performance of filters and clarifiers. Most of the suspended particles in water carry a similar type of charge (negative) and so they cannot clump together because of electrostatic repulsion. When the charge on those particles is neutralized with the use of coagulants, they can stick together to form comparatively larger-sized particles called flocs. To achieve good performance in coagulation and

formation of flocs, a highly energetic rapid mixing is essential to properly disperse the coagulant throughout the water and facilitate particle collisions, as insufficient mixing may leave the coagulation step incomplete. In some cases, pre-ozonation can be used before coagulation to reduce the amount of coagulant added to the water and enhance the removal of many water constituents during coagulation followed by flocculation and sedimentation (Saunier et al., 1983).

Flocculation is a water treatment process that involves gentle stirring or agitating the water to encourage the particles to come together and form larger masses. This process leads to an enlargement of the particle size, transitioning them from the submicroscopic scale to the visible range. The small flocs which are formed during the coagulation process, come into contact with one another during the slow mixing phase, resulting in collisions that lead to the formation of larger flocs. Sometimes high molecular weight polymers named coagulant aids can be introduced during this step to improve the floc strength, weight, and settling rate. After reaching their optimum size and strength, the flocs can be easily removed from the water either through subsequent sedimentation, flotation, or less often direct filtration (Crittenden et al., 2012).

Coagulation and flocculation processes are generally followed by the sedimentation process and applied prior to the application of filtration and disinfection methods in conventional DWTPs. Sedimentation is a physical water treatment process that removes settable solids and flocs (created during the coagulation and flocculation processes) from the water under the influence of gravity. This process relies on the use of sedimentation tanks where the masses settle at the bottom of the tank due to their weight and are removed periodically. The combination of the CFS process is typically used to separate suspended particles and dissolved substances from the water. These procedures are relatively simple and cost-effective, as long as the required chemicals and dosages are adjusted according to the water composition (Worch, 2019).

After sedimentation, the water is filtered to further remove suspended materials and pathogens. Filtration is a physical water treatment process that removes the smaller particles which cannot be effectively removed during sedimentation. There are two primary categories of filtration processes: surface filtration and depth filtration. Depth filtration is the commonly used method, where the particles are separated from the water as it passes through a coarse-grained porous filter medium, such as sand, or anthracite. Throughout the depth of the filter bed, particles collide with and adhere to the filter media, resulting in their accumulation. Particles that are considerably smaller than the pore volume can also

be captured between the grains of the filter medium. Rapid sand filtration, one of the common forms of depth filtration, is usually adopted at the DWTPs, which can ensure the provision of safe drinking water with proper pre-treatment (CFS) as well as final disinfection processes (Crittenden et al., 2012).

Disinfection represents the ultimate stage in drinking water treatment before its distribution. After the filtration process, disinfectants like chlorine, chloramine, chlorine dioxide, ozone, or ultraviolet (UV) light are usually added to inactivate any remaining parasites, bacteria, or viruses to make the water safe for drinking (Worch, 2019).

The combination of coagulation, flocculation, sedimentation, filtration, and disinfection constitutes the extensively employed water treatment technology for the purification of significant volumes of drinking water. Sometimes DWTPs may employ some additional treatment steps which are known as advanced treatment processes such as membrane filtration, advanced oxidation process, activated carbon adsorption, and biofiltration. This advanced treatment usually provides a higher level of treatment than the conventional treatment.

Membrane filtration is one of the advanced water treatment processes that has become popular nowadays due to its excellent separation capabilities and decreasing costs of membranes. Common membrane processes include ultrafiltration, nanofiltration, microfiltration, reverse osmosis, and electrodialysis. This process is established based on the particle size, with water passing through the membrane, resulting in the separation of the particles. Essentially, only particles larger than the membrane's pore width are removed during this filtration process. This method ensures the production of high-quality treated water, as particles larger than the pore size are effectively filtered out (Crittenden et al., 2012).

Dissolved air flotation (DAF) has become widespread these days as an alternative clarification process to sedimentation. In this procedure, micro air bubbles are used to adhere to and float flocculated particles and suspended solids, facilitating their removal by bringing them to the water surface. On the other hand, sedimentation is employed to eliminate settled solids from the bottom. The main advantage of DAF is that it can achieve better removal of lightweight particles which are usually difficult to settle. Besides, this process does not require the production of heavy flocs for settling. Consequently, lower dosages of coagulant can be used and the time required for flocculation is reduced (Edzwald, 2006).

DWTPs are not explicitly designed to target MNPs, but their processes are intended to remove solids and colloids to produce safe drinking water, and MNPs fall under this category (Barchiesi et al., 2021).

MNPs properties like size, shape, polymer type, density, and other surface characteristics are required to understand the treatment efficiency of these methods. During the coagulation and flocculation process, negatively charged MNPs can interact with coagulants and other contaminants in water, similar to other particles and sweep coagulation may also occur. After this step, particles including MNPs can be removed by clarification processes, either by sedimentation or flotation. Subsequently, most of the remaining particles can be effectively removed through the media filtration process by accumulating them between the grains of the filter medium. Moreover, membrane filtration can be efficient in removing MNPs due to the smaller nominal pore size of the membrane compared to MNP size (Talvitie et al., 2017). In general, these drinking water treatment processes, both individually and combined, are expected to remove a certain amount of MNPs.

2.1.6 Necessity of MNPs Removal through DWTPs

2.1.6.1 Health Impacts:

The increasingly ubiquitous presence of MNPs has aroused great concern regarding their potential effects on human health. Environmental MNPs exhibit varied characteristics because of different sizes, shapes, polymer composition, density, and additional constituents adsorbed on their surface due to various surface properties (Kooi & Koelmans, 2019). Their distinctive characteristics and long-lasting presence in the environment make them readily consumed by marine and freshwater organisms, posing potential threats to both ecological and human health (Peng et al., 2020). Furthermore, due to their small size and large specific surface area, MNPs have the capability to adsorb different organic and inorganic contaminants on their surface, which can potentially endanger human health and the ecosystem upon exposure (B. Zhang et al., 2021). MNPs can ultimately be taken into the human body through inhalation, ingestion, or skin contact (O'Neill & Lawler, 2021). A recent study suggested that humans may get exposed to 0.1-5 g of MPs per week through these various pathways (Senathirajah et al., 2021). Though the effects of MPs or NPs on human health are still under investigation, there has already been evidence of their potential toxicity. The details will be discussed later in Chapter-5.

2.1.6.2 MNPs Regulations:

Globally MNPs are not yet regulated in drinking water. As the presence of MNPs is becoming more apparent in drinking water day by day and this is one of the potential routes of exposure to MNPs, there is a need to develop strategies for regulating MNPs in drinking water. Although no direct human health

impacts have been reported to-date, precautionary measures should be taken. The existing situation regarding the established legislation in source water and ongoing regulatory initiatives concerning MNPs worldwide will be delineated in Chapter-5.

2.2 Observations, Knowledge Gaps, and Research Needs

While DWTPs are not specifically designed to remove MNPs, published research studies have shown that they can play a significant role in reducing the abundance of MNPs in water. Recent investigations have been conducted on the presence, behavior, and elimination of MPs through drinking water treatment processes at both full-scale and bench-scale levels. There are a limited number of studies at the bench-scale level examining the removal of NPs, but there is none at the full-scale level. The full-scale DWTP surveys investigated different types of MPs present in source water and their removal efficiency through DWTPs, consisting of different combinations of treatment units. In some cases, they also looked into the removal of MPs specifically by individual treatment units within DWTPs. The bench scale studies also experimented with MNPs of different properties to evaluate the removal efficiency pattern by different treatment processes commonly employed in DWTPs.

In full-scale surveys, it has been observed that the operational units within DWTPs can affect the distribution of MPs in treated water, with varying removal efficiencies. The abundance of MPs varied substantially in various water sources used for drinking water production and exhibited significant diversity in terms of their properties, including sizes, shapes, and polymer composition. Bench scale studies, utilizing various MNPs properties under different optimized conditions, have also demonstrated varying removal of MNPs by different treatment processes. A couple of studies (Tang et al., 2022; Xue et al., 2022) have attempted to summarize some of the existing findings in this regard, but a detailed analysis of the correlations has not yet been conducted. It would be beneficial to explore the different trends of results and identify potential influential factors. It is necessary to examine the relationship between removal efficiency and both MNPs abundance and their various properties throughout the overall removal process conducted by DWTPs. Besides, since CFS and filtration processes are involved in particle removal, there is a specific need to investigate their individual effectiveness in MNPs removal and search for potential correlations with the MNPs abundance and their properties during these specific treatment processes.

Besides, no information is currently available for NPs from full-scale DWTP surveys, while knowledge from bench-scale studies is also limited on NPs compared to MPs. Thorough investigations are needed

to understand MP pathways, their properties, and removal efficiency in treated drinking water, enabling insights into NPs and comparisons with existing limited bench-scale data.

Based on available information regarding the abundance and removal of MNPs in drinking water, there has been ongoing research to understand their potential toxic effects and impacts on human health. MNPs, characterized by their small size and large surface area, have demonstrated potential harm to humans. Considering drinking water as a potential source of MNPs exposure, there is a need to regulate their abundance in drinking water. However, MNPs represent a unique contaminant class of solid particles, which encompass a wide range of particles with different sizes, shapes, polymer compositions, densities, solubility, surface charges, etc. They also possess a highly persistent nature and ability to adsorb pathogens and other contaminants. Their unique and diverse characteristics pose challenges in integrating MNPs into conventional risk-based regulatory frameworks. Assessing the risk associated with MNPs is further complicated by the difficulty in determining the potential toxicity and health effects of each individual class of MNPs. Moreover, their persistent nature and continuous production contribute to their accumulation in surface water, presenting difficulties for their removal from drinking water. Considering all these facts, there have been already some discrete efforts made to regulate MPs in source water worldwide, but there is still no definite regulatory framework established for MPs or NPs in drinking water. The existing situation underscores the need for a unique and dedicated regulatory approach for MNPs. It is imperative to review the existing guidelines for particles with similar properties while acknowledging the unique aspects of MNPs. Consequently, this presents an opportune moment for starting to develop a comprehensive framework for regulating MNPs of different characteristics in drinking water.

Chapter 3
Analysis of Data from Full-scale Surveys

3.1 Introduction

With the exponential increase in the abundance of MNPs in freshwater sources, a growing number of research studies can be found evaluating the removal of MNPs by different full-scale DWTPs. As DWTPs hold an important role in ensuring the quality of drinking water before distribution, the optimization of their removal capability is essential. As a result, researchers all over the world are carrying out their own investigations utilizing their nearby DWTPs. But there is a lack of synchronization in the findings from such investigations because of different water sources, MNPs characteristics, treatment processes, and evaluation techniques. So, instead of executing another research study evaluating the performance of a full-scale DWTP for the removal capacity of MNPs, it has become more necessary to evaluate the existing findings from the published literature. To-date, all of these full-scale DWTP surveys have been mostly published over the last five years and looked into the presence and removal of MPs from surface water sources with different objectives, with little work done on groundwater. There were no surveys found looking into NPs from this full-scale drinking water treatment perspective. Recently, a couple of reviews (e.g., Tang et al., 2022; Xue et al., 2022) have summarized the existing information on MPs removal efficiencies from some of the full-scale DWTP surveys which are also included in this chapter. But further in-depth analysis of the data and interpretation of the results of the studies discussed in these reviews will be incorporated in this chapter. The findings from bench-scale studies will be discussed separately in the next chapter.

The main objective of this chapter is to analyze published literature on the availability and removal of MPs with discrete properties from various surface water sources through different full-scale DWTPs. To address this goal, the initial step is the selection of full-scale surveys to be included in the analysis from a very wide range of published literature. The next step involves categorizing the available information on MPs removal through different DWTPs, followed by the evaluation and comparison of the information on the availability of different characteristics of MPs in the influent and effluent of different full-scale DWTPs. After that, the removal of MPs with different properties through the full-scale DWTPs and individually by conventional treatment units, i.e., CFS and filtration processes will be investigated. Then conclusions will be drawn with respect to the presence and removal pattern of MPs based on the available statistics from the full-scale surveys.

3.2 Research Approach

To address the objectives of this chapter (detailed in section 3.1), two different types of analysis approaches were carried out.

Part A: Qualitative Analysis

3.2.1 Selection of Studies

There is a broad range of studies published on the trending topic MNPs. The initial main criterion for the selection of studies among the published peer-reviewed literature was set to focus on the full-scale DWTP surveys investigating the abundance and removal efficiency of MNPs from drinking water sources. This selection parameter limited the number of studies to be included here. The selection process went through certain steps, followed by which the number of selected studies for this chapter narrowed down to a few. So, at first, a list was produced with the identified studies fulfilling the main criterion. Certain keywords were used for the initial search process, i.e., 'microplastics/nanoplastics removal in drinking water', 'microplastics/nanoplastics removal through drinking water treatment plants', and 'microplastics/nanoplastics in full-scale drinking water treatment plants'. The options which were selected while searching for the relevant published articles in google scholar were 'any time', 'sort by relevance', 'any type', and 'include citations'. In this manner, some bench-scale/pilot-scale studies and review papers also came along with the full-scale surveys, but they were not included in this chapter. The literature published until December 2022 was screened and the published articles dated from 2022 to 2018 were mostly found fulfilling the principal requirement of this chapter. The references mentioned in these studies were also screened. Finally, 16 full-scale surveys were identified as satisfying the primary selection criterion. No full-scale DWTP surveys were found investigating the presence and removal of NPs. Moreover, groundwater was detected as the source of drinking water for only one full-scale DWTP survey. So later, due to the limitation of data, the selection benchmark was modified to focus exclusively on surface water sources.

Part B: Quantitative Analysis

3.2.2 Basis of Data Analysis

Different full-scale DWTP surveys investigated the removal of MPs under different circumstances, i.e., locations of source water, characteristics of MPs, treatment units of DWTPs, analytical methods to quantify MPs, and data evaluation techniques varied among the selected surveys. Therefore, all of

them reported varying findings and did not always provide all the information needed for the analysis of this chapter. Besides, some of the studies provided additional information, because of their unique approach to evaluating MPs removal. For the sake of the data collection process, a list of criteria was established, which was used as a guideline to select studies for data analysis purposes.

The main basis of the data collection for this chapter is based on the list below:

A. Irrespective of MP size, shape, and polymer type
    I. Abundance of MPs in the inlet and outlet of the DWTPs
    II. Total removal efficiency of MPs by overall full-scale DWTPs

B. Removal efficiency of
    I. Different size ranges of MPs by
        a. Overall full-scale DWTPs
        b. CFS unit of full-scale DWTPs
        c. Filtration unit of full-scale DWTPs
    II. Different shapes of MPs by
        a. Overall full-scale DWTPs
        b. CFS unit of full-scale DWTPs
        c. Filtration unit of full-scale DWTPs
    III. Different polymer types of MPs by
        a. Overall full-scale DWTPs
        b. CFS unit of full-scale DWTPs
        c. Filtration unit of full-scale DWTPs

The full-scale surveys which at least directly reported the value of criteria A (either I or both I & II) or provided enough data to calculate the values of criteria A (if value of I is provided, II can be calculated), were included in the analysis of this chapter. The required values mentioned in criteria B were not found in all of the selected full-scale surveys. Some surveys directly mentioned a few of the values in their published articles or supplementary information. Others were provided indirectly through figures,

from which the necessary values could be calculated. Among the initially selected 16 full-scale surveys, 5 of them could not meet the minimum criteria for the main basis of the data collection. Therefore, 11 surveys were selected for the analysis of this chapter. Table 3.1 has been prepared to represent which values from these criteria were available for each selected survey, along with their other findings. The above-mentioned numbering of the selection criteria for data collection has been also adopted in the table.

Table 3.1: Surveys with their availability of data under different selection criteria and their additional reported data

No.*	Reference	A (I/II)	B (I)			B (II)			B (III)			Additional reported data
			a	b	c	a	b	c	a	b	c	
1	Jung et al. (2022)	x	x	x	x				x	x	x	>MPs abundance over 12 months of a year in the inlet and outlet of DWTPs >Correlation of MPs concentration with water quality parameters (TOC, Manganese)
2	Wu et al. (2022)	x	x	x	x	x	x	x	x	x	x	>MPs abundance and removal efficiency of all their mentioned size ranges, shapes, and polymer types for each treatment unit of DWTP (besides CFS and filtration)
3	Radityaningrum et al. (2021)	x	x	x	x	x	x	x				>Abundance in raw and treated water and overall removal efficiency reported for different colors of MPs
4	Shen et al. (2021)	x										>The percentages of different size ranges, shapes, and polymer types of MPs in the raw and treated water sample in four different months
5	Babel & Dork (2021)	x	x	x	x	x	x	x				>The percentages of different polymer types of MPs in the inlet of the DWTPs
6	Dalmau-Soler et al. (2021)	x										>The variation of MPs concentration in seven different surface water samples along the river basin with shape, size, and polymer type >The percentages of MPs for different size ranges and shapes in the different treatment units of the DWTP
7	Sarkar et al. (2021)	x										>The percentages of different polymer types of MPs in different treatment units of the DWTP >Correlation of MPs abundance with different water quality parameters

8	Kankanige & Babel (2021)	x	x	x	x	x	x	x				>MPs concentration in two different seasons of a year >MPs abundance analysis in the sediment and polymer type identification of 300–≥500 μm MPs
9	Pivokonský et al. (2020)	x	x	x	x	x	x	x				>The percentages of different polymer types of MPs found for all water samples of both DWTPs
10	Wang et al. (2020)	x	x	x	x	x	x	x	x	x	x	/
11	Pivokonsky et al. (2018)	x	x			x			x			>The percentages of different polymer types of MPs in raw and treated water at three DWTPs

**References are organized from the most recent ones*
MPs-microplastics, DWTPs-drinking water treatment plants, TOC-total organic carbon

Table 3.1 provides a clear idea of the available information based on the listed criteria for each selected survey. It is important to note that some of the selected surveys provided additional data beyond the listed criteria, which came from their extension of research in different directions keeping their main focus on the MPs abundance and removal through full-scale DWTPs. The additional data may be helpful for explaining any differences in findings between studies. They also can provide valuable insights which may be useful for future research in this field.

Table 3.2 lists the full-scale surveys that were initially identified as meeting the main criterion for selection, but did not satisfy the minimum criteria for the basis of data collection. These surveys had objectives and findings that were different from the selected ones, or they lacked the necessary data for the analysis. The specific reasons for their exclusion are described in this table.

Table 3.2: List of the full-scale DWTP surveys which were not included in the analysis of this chapter

No.*	Reference	Objective	Reason for not including in the analysis
a.	Cherniak et al. (2022)	To compare the performance of full-scale conventional to advanced and pilot-scale biological treatment technologies regarding MPs removal	They did not differentiate between the number of MPs and other anthropogenic particles while reporting the values of abundance and removal efficiency through different treatment processes of DWTPs. They reported their values for overall anthropogenic particles, which included MPs and then they reported the relative percentages of different types of MPs
b.	Siegel et al. (2021)	To evaluate the presence of MP particles by analyzing the treatment sludge	They reported MPs abundance by analyzing the sludge sample from the DWTP instead of the water samples. They also did not focus on the removal by different treatment units for different properties of MPs.
c.	Kirstein et al. (2021)	To characterize MPs in the drinking water distribution system	They just sampled water from several places in the distribution system after being treated and did not report any value regarding the removal of MPs by the DWTP
d.	Johnson et al. (2020)	To assess the effectiveness of water treatment works in removing MPs through DWTPs	They did not mention any treatment process used for water treatment, collected raw water directly only from the sources and treated water from the tap. They identified only the presence of different MPs but did not provide any specific value needed for the analysis

e.	Mintenig et al. (2019)	To assess the presence of MPs from groundwater source to DWTP	The source of water was groundwater

***References are organized from the most recent ones*
MPs-microplastics, DWTP-drinking water treatment plant

The surveys listed in Table 3.2 did not meet the minimum criteria for inclusion in the database for this chapter due to their different goals and reported results. For instance, only Mintenig et al. (2019) analyzed groundwater instead of surface water as their source of water, but data analysis could not be performed based on data only from one survey for a specific category. Siegel et al. (2021) analyzed sludge samples instead of water, though this full-scale survey was based on a DWTP which treats surface water. The other three full-scale surveys did not report the specific value of MPs abundance in the influent or effluent of DWTPs or removal efficiency by the DWTPs. Therefore, these full-scale surveys could not be included in the analysis due to the lack of the required information. It is also important to note that the exclusion of these studies from this analysis does not necessarily imply that they are not valuable, rather it means that they could not fulfill the requirements for inclusion in this specific analysis. These studies can be useful for other research, which might have different objectives than those of this chapter.

3.2.3 Method of Data Extraction

The required data from the selected surveys were collected in a couple of ways. Initially, some values were directly reported in the literature or sometimes outlined in the supplementary information in the form of a table or chart. Those values were directly collected from there for analysis purposes. Then, in the other case, some values were represented in the form of different graphs or figures, which were difficult to read directly with bare eyes. In these cases, an online application named 'WebPlotDigitizer' was used to extract data from those figures. This web application can extract data from plots, images, and maps. This web tool requires the user to upload an image of the figure/graph from where the data needs to be extracted, then select the plot type and axis alignment, and finally align the required data points with a data extraction bar tool. After that, they will extrapolate the values from the figure for those given data points. The extracted data were further processed using Microsoft Excel, depending on the specific requirements. This may involve calculating removal efficiency from abundance data, generating averages, rounding values to establish a uniform dataset, and other necessary operations. This method of data extraction can be considered more reliable than just visually approximating values.

3.2.4 Limitation of the Data Extraction Process

The data collected directly from the literature have their own inherent limitations based on the methods used in the respective full-scale surveys. The direct data usage from those surveys did not include any additional uncertainty factors. However, the data extracted from different figures using 'WebPlotDigitizer' may not be 100% accurate. As some assumptions were made for setting the data extraction bar tool on the required value points only with bare eyes, it may have some refractive errors. After the data collection process, sometimes the values needed to be rounded to follow a continuous pattern in the dataset for the convenience of the analysis. For instance, some surveys reported their results using more than two decimal points, whereas several values were found in whole figures. In order to bring uniformity to a particular dataset, rounding was performed when needed. Again, during the data extraction using the 'WebPlotDigitizer' and then data processing in Microsoft Excel, the values were mostly rounded to either two decimal points or whole numbers for the convenience of usage. In these cases, it may cause small round-off errors.

3.2.5 Data Analysis

Microsoft Excel, Microsoft Word, Particle Graphing Tool, and IBM SPSS Statistics were used for the data analysis processes. The 'Particle Graphing Tool' was developed by Sina Golchi for the NSERC Chair in Water Treatment group at the University of Waterloo. This tool was designed to graphically represent removal efficiencies for different particle size ranges of MPs from different surveys and was used, for example, in Figure 3.9. Microsoft Excel or other available graphing tools could not be applied due to the complex nature of the plot. The code which was used to generate this tool is available in Appendix A as well as in an open repository on GitHub and it is under the MIT license. For IBM SPSS Statistics, a licensed version (29.0.0.0) was used for constructing the relationship graphs between removal efficiency and abundance of MPs with respect to the inlet and outlet points of different DWTPs. For all other analysis works, Microsoft Excel and Microsoft Word were used.

3.3 Sources of Information

The chosen full-scale DWTP surveys exhibit a wide range of geographic diversity as they were conducted at different times and locations around the world. Accordingly, they also vary in terms of MPs abundance and properties (size ranges, shapes, polymer types), treatment processes of the DWTPs, etc. A list has been prepared (Table 3.3) for the selected full-scale surveys with their respective locations and showed all DWTPs involved in each survey including their water sources, hydraulic

capacity, and treatment processes. The numbering of these full-scale surveys in Table 3.3 follows the same order as in Table 3.1 and will be consistent throughout the entire chapter for other tables.

The selected 11 full-scale DWTP surveys were conducted by researchers from 8 different countries and included 19 different DWTPs in total. More than half of the surveys (6) investigated MPs removal capacity through only one DWTP, while 5 surveys explored the efficacy of multiple DWTPs for the removal of MPs, i.e., 3 surveys looked into 2 DWTPs and the remaining 2 surveys looked into 3 and 4 DWTPs individually. The hydraulic capacity of 6 DWTPs among 4 surveys varied within the range of 1000-2000 L/s (considering 1 m^3/day ≈ 0.0116 L/s). The ADWTP from Wang et al. (2020) had the highest hydraulic capacity, while the DWTP of Kankanige & Babel (2020) also had a relatively higher hydraulic capacity than the others. The hydraulic capacities for the DWTPs from Pivokonsky et al. (2020) and Pivokonsky et al. (2018) were comparatively low, except for one (WTP1) from Pivokonsky et al. (2018). Only two surveys did not mention the hydraulic capacity of their DWTPs. Sarkar et al. (2021) mentioned the capacity of their pulsator clarifier as being less than 1000 L/s, as they used pulse clarification instead of sedimentation after coagulation and flocculation. Almost all of these DWTPs used river water as their raw water source, with only 3 DWTPs collecting water from lakes and 2 from water reservoirs. The treatment processes also varied among the DWTPs. 9 DWTPs among 19 employed conventional drinking water treatment processes, while the remaining 10 utilized advanced treatment processes. In general, the conventional treatment processes for the selected DWTPs include screening as a pre-treatment step, coagulation followed by flocculation, sedimentation, granular media filtration, and disinfection as a post-treatment step. The advanced treatment processes involve the incorporation of additional advanced technology such as pre or post-ozonation, biological or granular activated carbon filtration, membrane filtration, and UV adsorption, along with conventional treatment steps.

Table 3.3: List of the selected surveys with their locations, treatment units across different DWTPs, and hydraulic capacity of those DWTPs

No.	Reference*	Location	DWTPs	Hydraulic/WT capacity of the DWTPs	Source of water	Treatment processes**
1	Jung et al. (2022)	Busan, South Korea	DWTP-A	_	Nakdong river, South Korea	pre-ozonation, coagulation/ sedimentation, sand filtration
			DWTP-B	_	Nakdong river, South Korea	pre-ozonation, coagulation/ sedimentation, sand filtration
			DWTP-C	_	Lake 1, South Korea	pre-ozonation, coagulation/ sedimentation, sand filtration
			DWTP-D	_	Lake 2, South Korea	coagulation/sedimentation, sand filtration
2	Wu et al. (2022)	Jiaxing, China	_	usual capacity: 150 thousand m^3/day	Yangtze river, China	pre-ozonation, biological pretreatment, coagulation, sedimentation, ozonation, biological activated carbon filtration, micro-flocculation, sand filtration, disinfection
3	Radityaningrum et al. (2021)	Surabaya city, Indonesia	SDWTP-I	1450 L/s	Surabaya river, Indonesia	aeration, pre-sedimentation, coagulation, flocculation- sedimentation, filtration, disinfection
			SDWTP-II	2000 L/s	Surabaya river, Indonesia	aeration, pre-sedimentation, coagulation, flocculation- sedimentation, filtration, disinfection
4	Shen et al. (2021)	Changsha, China	_	100000 m^3/day	XiangJiang river, China	aeration, coagulation/sedimentation, sand filtration, GAC filtration, disinfection

5	Babel & Dork (2021)	Phnom Penh city, Cambodia	Chroy Chongva (WTP1)	150,000 m^3/day	Mekong River, Cambodia	coagulation, flocculation, sedimentation, rapid sand filtration, disinfection
			Phum Prek (WTP2)	140,000 m^3/day	Tonle Sap Lake, Cambodia	coagulation, flocculation, sedimentation, rapid sand filtration, disinfection
6	Dalmau-Soler et al. (2021)	Catalonia, NE Spain	Sant Joan Despí DWTP	_	Llobregat river, NE Spain	coagulation/flotation, sand filtration, two parallel treatment lines [(upgraded conventional line - ozonation followed by GAC filtration) & advanced treatment - ultrafiltration followed by reverse osmosis)], disinfection
7	Sarkar et al. (2021)	Kolkata, India	Indira Gandhi WTP	5 modules of pulsator clarifier and purification capacity of 20 million gallons/day each	Ganga river, India	pre-disinfection, coagulation-flocculation, pulse clarification, sand filtration, post-disinfection
8	Kankanige & Babel (2021)	Thailand	_	maximum capacity: 4,400,000 m^3/day	Chao Phraya river, Thailand	screening, clarification (coagulation followed by flocculation and sedimentation), dual media filtration (rapid sand/anthracite coal), chlorination
9	Pivokonsky et al. (2020)	Czech Republic	DWTP Milence	usual/maximum capacity: 180/400 L/s	Úhlava River, Czech Republic	coagulation, flocculation, sand filtration, disinfection
			DWTP Plzeň	usual/maximum capacity: 400/1000 L/s	Úhlava River, Czech Republic	coagulation, flocculation, sedimentation, sand filtration, ozonation, GAC filtration, UV adsorption, disinfection

10	Wang et al. (2020)	China	ADWTP	usual/maximum capacity: 120/150 million m^3/day	Yangtze river delta, China	coagulation/flocculation, sedimentation, sand filtration, ozonation combined with GAC filtration
11	Pivokonsky et al. (2018)	Czech Republic	WTP1	usual/max capacity: 3700/7000 L/s	Large valley water reservoir, Czech Republic	coagulation/flocculation, sand filtration
			WTP2	usual/max capacity: 100/200 L/s	Smaller water reservoir, Czech Republic	coagulation/flocculation, sedimentation, sand filtration, granular activated carbon filtration
			WTP3	usual/max capacity: 90/150 L/s	River, Czech Republic	coagulation-flocculation, flotation, sand filtration, granular activated carbon filtration

**The references are organized from the most recent ones and divided for the different DWTPs where there is more than one DWTP in a study*

***The presence of screening and disinfection processes was not specified in several full-scale DWTP surveys. But it is assumed that all DWTPs employ these two fundamental treatment processes regardless of whether they were mentioned in the surveys or not. Furthermore, some surveys did not mention the flocculation process in the list of treatment steps, but flocculation is an integral part of the coagulation process. So, it is assumed that every coagulation process is followed by flocculation process in these DWTPs.*

MPs-microplastics, DWTPs-drinking water treatment plants, WT-water treatment, WTP-water treatment plant, SDWTP-sub-drinking water treatment plant, ADWTP-advanced drinking water treatment plant

The selected full-scale DWTP surveys vary not only in locations and drinking water treatment processes, but also in the methods used to identify MPs. All these factors contribute to the differences in the observed properties of MPs such as size ranges, shapes, polymer types, and other surface properties. The size, shape, and polymer type are the most frequently reported MP properties in these full-scale DWTP surveys. But other surface properties of MPs such as color, zeta potential, weathering condition, etc. are not typically reported. Table 3.4 presents a list of the size ranges, shapes, and polymer types of MPs identified in these selected full-scale DWTP surveys, along with the corresponding analysis techniques used for their quantitative and qualitative analysis.

The identified size ranges, shapes, and polymer types of MPs varied significantly across the selected full-scale DWTP surveys. All of them separated the MPs into several size ranges, which were different from each other. Every survey created these size ranges for the benefit of identifying and reporting the abundance of MPs. Here 6 out of 11 surveys detected MPs as small as 1 µm, while others had higher detection limits. Jung et al. (2022) determined MPs abundance starting from 10 µm, Wu et al. (2022) from 5 µm, Babel & Dork (2021) and Kankanige & Babel (2021) from 6.5 µm and Dalmau-Soler from 20 µm. Though the upper size limit of MPs was considered 5 mm for all surveys, they represented that in different ways. Some surveys specified the definite size ranges until 5 mm, while others mentioned it as > 100 µm or >500 µm. With respect to the shape of MPs, fibers and fragments were the most commonly identified shapes, with some surveys also detecting spheres, films, and pellets. All these surveys revealed many different polymer types found in their water samples. Among them, PE, PP, and PET were the most frequently reported polymer types in almost all these surveys. For quantitative analysis of MPs, most of the selected surveys (8) adopted automated image analysis techniques using software, where chemical images were obtained via different micro-FTIR, micro-Raman spectroscopy, SEM, fluorescence microscope, and stereomicroscope. 2 surveys [Babel & Dork (2021) and Kankanige & Babel (2021)] conducted manual counting through visual observation under a microscope for larger sized MPs and automated analysis technique for smaller sized MPs. Only one survey [Radityaningrum et al. (2021)] used the manual counting method for all MPs. For qualitative analysis, all of them used either micro-FTIR or micro-Raman spectroscopy to characterize MPs. 3 surveys [Shen et al. (2021), Kankanige & Babel (2021), and Pivokonsky et al. (2018)] analyzed comparatively smaller MP particles using Raman spectroscopy and all other particles by using FTIR.

Table 3.4: MPs properties which are identified in the influent and effluent of these full-scale DWTPs

No.	Reference	Identified MP properties			Analysis techniques for quantitative analysis of MPs	Analysis technique for qualitative analysis of MPs
		Size range	Polymer type	Shape		
1	Jung et al. (2022)	>10 µm (10-20, 20-50, 50-100, >100)	PE, PP, PET, PMMA, PS, PA, PU, PVC-u, sbr-red, ABS	–	Micro-FTIR spectroscopy*	Micro-FTIR spectroscopy
2.	Wu et al. (2022)	>5 µm (5-10, 10-20, 20-100, 100-500, 500-1000, 1000-5000)	PP, PE, PVC, PA, VINYL, VINYON	Granular	Micro-Raman spectroscopy*	Micro-Raman spectroscopy
		>100 µm (100-500, 500-1000, 1000-5000)	PET, VINYL, others	Fibrous		
3.	Radityaningrum et al. (2021)	>1 µm (1–100, 101–350, 351–1000, 1001–5000)	PE, PP, LDPE, PA 6, PA 6.6, PES, polytrimolithicamide, cellophane	Fibers, fragments, films, pellets	Dissecting microscope**	FTIR spectroscopy
4.	Shen et al. (2021)	>1 µm (1–10, 10–50, 50–100, > 100)	PE, PET, PMMA, PP, PS, PVC, PA, others	Fragments, fibers, spheres	SEM and micro-FTIR spectroscopy*	Micro-FTIR spectroscopy and Raman spectroscopy (for <20 µm)
5.	Babel & Dork (2021)	>6.5 µm (6.5-20, 20-53, 53-500, >500)	PET, PE, PP, PA, PVC, PS, PES, HDPE, PMMA, Cellophane, others	Fiber, fragment	Optical microscope** (for MPs>53 µm) and fluorescence microscope* (for 6-53 µm)	FTIR spectroscopy
6.	Dalmau-Soler et al. (2021)	>20 µm (20-200, 200-500, 500-1000, 1000-2000, 2000-5000)	PP, PES, PS, PAN, ABS, PE, PVA, others	Fragments, fibers	Stereomicroscope*	Micro-FTIR spectroscopy

7.	Sarkar et al. (2021)	>1 µm (<25, 25–50, 50–100, >100)	PE, PP, PET, PS, others	Fibers, films/ fragments	Fluorescence microscope*	ATR FTIR spectroscopy
8.	Kankanige & Babel (2021)	>6.5 µm (6.5-53, 53-300, 300-500, >500)	PE, PP, PA, PVC, PET, PS, others	Fibers, fragments, films, pellets	Biological system microscope** (for MPs>300 µm) and fluorescence microscope* (for 6.5-300 µm)	ATR FTIR spectroscopy (for ≥300 µm) and Confocal Raman spectroscopy (for <300 µm)
9.	Pivokonsky et al. (2020)	>1 µm (≥1–<5, ≥5–<10, ≥10–<50, ≥50–<100, ≥100)	CA, PET, PVC, PE, PP, EVA, PBA, PTT, PTFE, PS, PA6, PEO+PEG, others	Fragments, fibers	SEM*	Micro-Raman spectroscopy
10.	Wang et al. (2020)	>1 µm (1–5, 5–10, 10–50, 50–100, >100)	PET, PE, PP, PAM, others	Fibers, fragments, spheres	SEM*	Micro-Raman spectroscopy
11.	Pivokonsky et al. (2018)	>1 µm (1–5, 5–10, 10–50, 50–100, >100)	PET, PP, PE, PS, PMMA, PBA, PTT, PVC, others	Fragments, fibers, spheres	SEM*	FTIR spectroscopy (for >10 µm) and Micro-Raman spectroscopy (for 1-10 µm)

**Automated image analysis using software obtained through Micro-FTIR/ Micro-Raman spectroscopy/ SEM/ Fluorescence microscope/ Stereomicroscope*

***Manual counting through visual observation under a dissecting microscope/ optical microscope/ biological system microscope*

PE-polyethylene, PP-polypropylene, PVC-polyvinyl chloride, PA-polyamide, VINYL-Vinyl chloride-vinyl acetate copolymer, VINYON-vinyon, PET-polyethylene terephthalate, PS-polystyrene, PES-polyester, PMMA-poly(methyl methacrylate), PAM-polyacrylamide, PVC-u-unplasticised polyvinyl chloride, LDPE-low-density polyethylene, PAN-polyacrylonitrile, ABS- acrylonitrile butadiene styrene, PVA-polyvinyl acetate, CA-cellulose acetate, EVA-ethylene vinyl acetate copolymer, PBA-poly(butyl acrylate), PTT-polytrimethylene terephthalate, PTFE-polytetrafluoroethylene, PA6-polyamide-nylon 6, PEO-polyethylene oxide, PEG-polyethylene glycol, FTIR-Fourier Transform Infrared, SEM-Scanning Electron Microscope

Now, for the convenience of the analysis of data, these full-scale surveys have been categorized according to the publication year, identified MPs properties, and treatment processes.

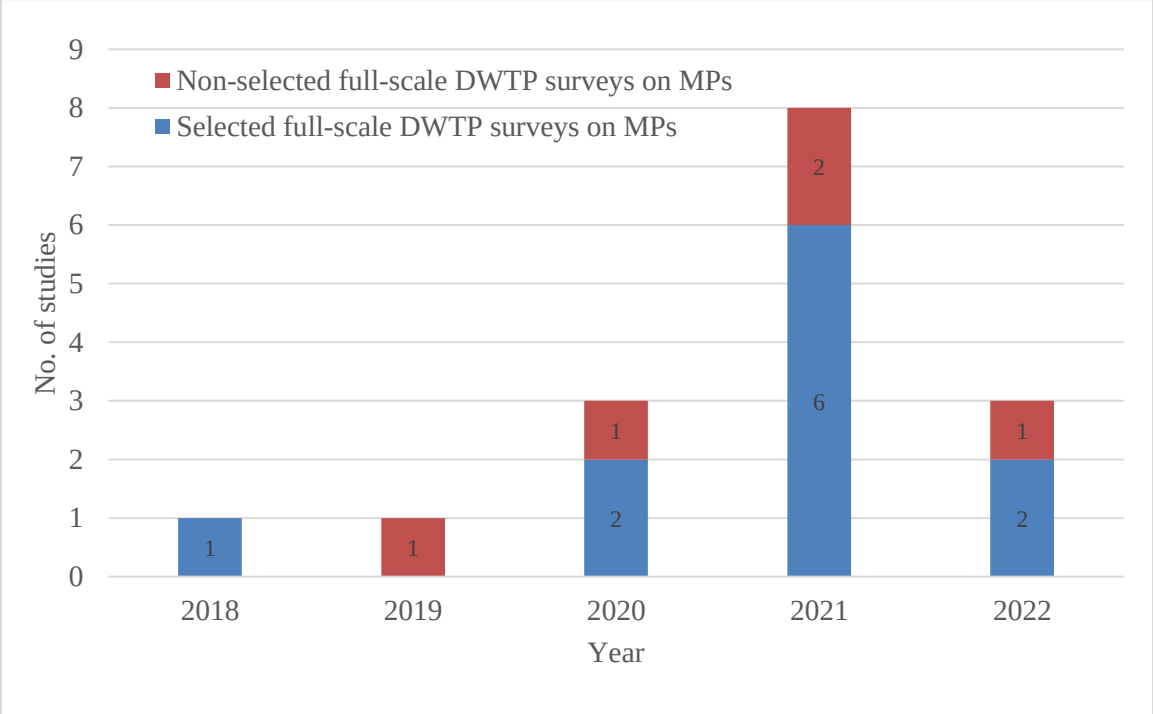


Figure 3.1: The number of full-scale DWTP surveys looking into the removal of MPs in the last five years
(DWTP-drinking water treatment plant, MPs-microplastics)

All of the 16 full-scale surveys, initially identified, were published within the last 5 years. Half of the studies were published in 2021, while 2020 and 2022 had an equal number of published full-scale surveys, with 3 in each year. There was one publication each in 2018 and 2019. The highest frequency of publication for these full-scale DWTP surveys was observed in 2021.

Figure 3.2 (a) classifies the surveys according to the frequency of reporting the overall MP removal efficiency by DWTPs irrespective of MP properties (size, shape, and polymer type), as well as the individual removal efficiency for different MP properties including sizes, shapes, and polymer types. Then Figure 3.2 (b) categorizes the surveys based on the frequency of reporting MPs removal for different properties (size, shape, and polymer type) by overall DWTPs and individually by CFS and filtration units within those DWTPs.

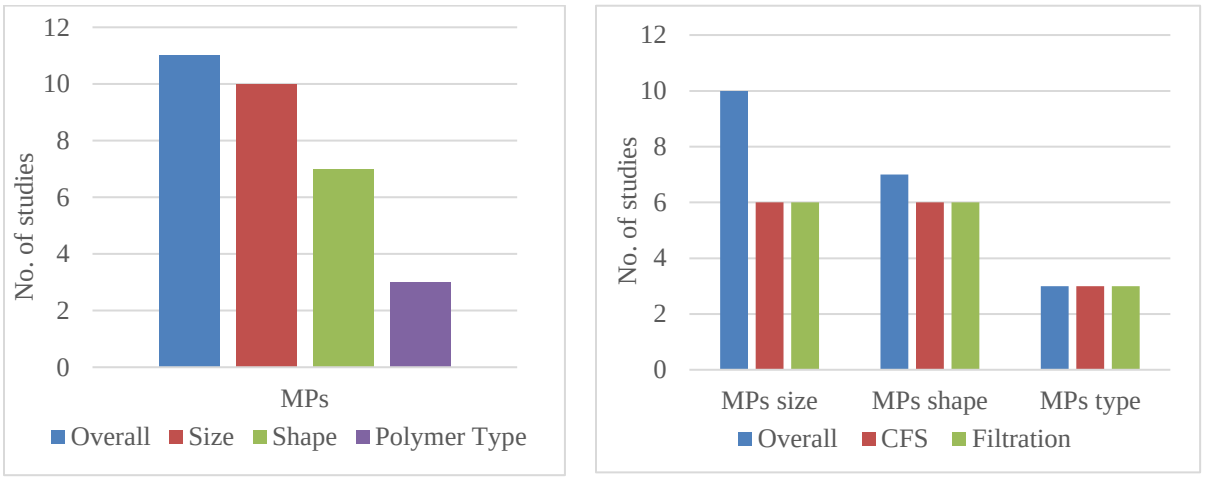


Figure 3.2: The number of full-scale DWTP surveys reporting the removal efficiencies for (a) all MPs in general irrespective of different properties and individually for MP size, shape, and polymer type (b) MPs with different properties by overall DWTPs and individually CFS and filtration units of DWTPs *(MPs-microplastics, CFS-coagulation-flocculation-sedimentation)*

In Figure 3.2 (a), all of the surveys (11) reported the overall removal efficiency of MPs (irrespective of MP size, shape, and polymer type) by the full-scale DWTPs. Among them, 10 surveys investigated the removal efficacy of MPs for different size ranges, while 7 and only 3 of them examined the removal efficiency of MPs for different shapes and polymer types, respectively. In Figure 3.2 (b), out of the 10 surveys looking into MPs removal for different size ranges by DWTPs in general, 6 of them investigated the removal efficacy individually by CFS and filtration processes. There was only one survey that reported overall removal for different MP shapes but did not investigate the removal by individual drinking water treatment processes. Surveys investigating overall MPs removal for different polymer types also provided data for individual drinking water treatment processes (CFS and filtration).

In general, these graphs present a comprehensive overview of the relevant obtainable information from the selected full-scale surveys in various areas of analysis to be shown in this chapter. While all the designated surveys provide overall MPs abundance in the influent and effluent of DWTPs and overall removal efficiency by those DWTPs (irrespective of size, shape, and polymer type), not all of them include all other important information. The dataset used for the analysis in this chapter is built based on the available information from these surveys.

3.4 Results and Discussion

3.4.1 Presence of Different Categories of MPs in Different DWTPs

All of the selected full-scale surveys determined the abundance of MPs for their raw water sources at the inlet point of the DWTPs. They also enumerated the MPs abundance in the treated water at the outlet point of those DWTPs after going through all the treatment units. The presence of MPs was reported regardless of the variations in MPs properties (size, shape, polymer type, or any other characteristics) and was expressed as the total number of MP particles per liter of water (MPs/L). Figures 3.3 and 3.4 illustrate the abundance of MPs in the inlet and outlet points of the DWTPs respectively. To best represent the data, a logarithmic scale is used. The actual values of MPs abundance are shown in Appendix A (Table A1). Almost all of these surveys provided mean and standard deviation values for the total number of all MP particles found in the raw and treated water of these DWTPs. In the figures, the rectangular bar represents the mean value, and the error bar represents the standard deviation in both positive and negative directions. But the error bars will not be equidistant on either side of the mean because of the use of a logarithmic scale to accommodate the extensive range of MPs abundance data. It should be noted that Radityaningrum et al. (2021) did not provide the standard deviation value or any dataset from which that can be calculated. For Shen et al. (2021), both mean and standard deviation were calculated from their given range of data. They outlined 4 sets of mean and standard deviation values for four different months and the average of them was taken here. The standard deviation value for Kankanige & Babel (2021) was derived from the figure where the error bar was depicted. In the case of the raw water source reported by Jung et al. (2022), which included four DWTPs, two of them were sourced from a river, and the other two were from two different lakes. But Jung et al. (2022) reported an average mean value with standard deviation for total MP number as there were no distinct differences. Besides, the influent concentration for Kankanige & Babel (2020) was reported for two different (dry and rainy) seasons.

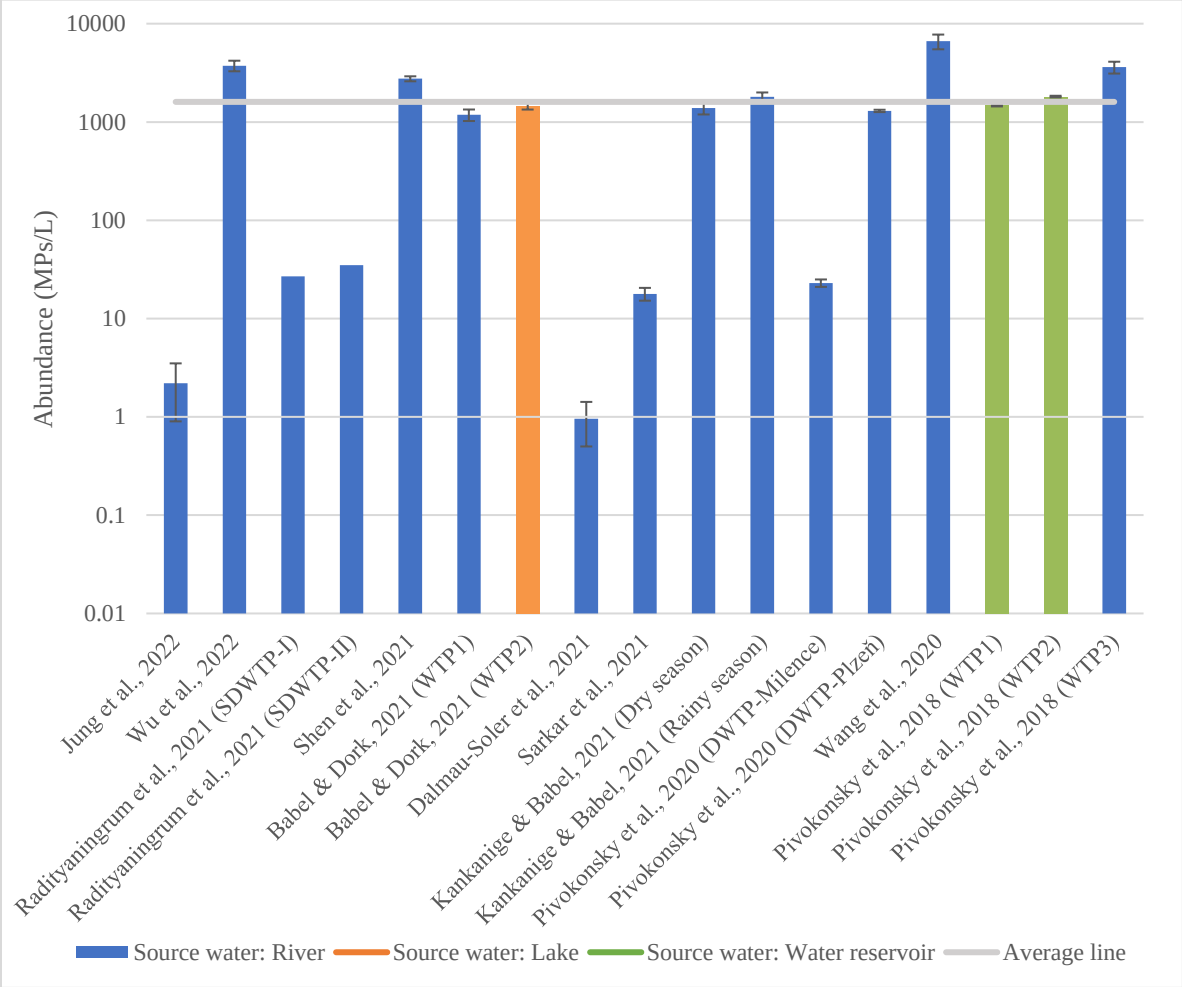


Figure 3.3: MPs abundance as the total number of MP particles per liter of water in the influent of different DWTPs from the selected full-scale surveys

A high variation can be observed in the total number of MPs in the raw water of these DWTPs, ranging from about 1 MPs/L to over 5000 MPs/L, with an average value of around 1700 MPs/L. Among these surveys, Wang et al. (2020) reported the highest MPs abundance of 6614 MPs/L in the inlet of the DWTP, whereas Dalmau-Soler et al. (2021) reported the lowest value of 0.96 MPs/L. Several possible factors can contribute to such significant differences. One factor is the variation in the size ranges of MPs considered in each survey. For instance, Wang et al. (2020) identified MPs ranging from as small as 1 µm up to 5 mm, while Dalmau-Soler et al. (2021) focused on MPs starting from 20 µm. Though in both of these cases, river water is their raw water source, Wang et al. (2020) collected raw water

from the Yangtze river estuary and Dalmau-Soler et al. (2021) collected from the Llobregat river basin. The collection point of raw water along with the geographical location and surrounding condition of the source water can make a significant contribution to the difference in the number of MP particles present in the water. Different detection techniques employed by each study can also lead to varying results. These reasons are responsible for the differences in the other reported findings as well. Babel & Dork (2021) collected their raw water from two types of sources (river and lake) and found higher MP numbers in lake water compared to river water, suggesting that the lake was influenced by the residential wastewater discharge more than the river because of the location. In contrast, Jung et al. (2022) observed no significant difference in the number of MP particles between the lake and river. They also noted the variation of MP abundance with the season and reported comparatively higher amounts of MPs in the river during January-April and November, whereas the opposite trend was observed for the lake having a higher number of MPs in the rest of the months. Kankanige and Babel (2021) collected water from the same river in two different seasons (dry and rainy) and reported higher MP numbers in the rainy season. The raw water for two DWTPs of Pivokonsky et al. (2018) was collected from two different water reservoirs, and they reported close to average number of MPs compared to the others. In general, most DWTPs exhibited MPs level exceeding 1000 MPs/L. The wide variation in the reported number of MP particles can be attributed to numerous contributing factors. MPs loading can differ among water sources depending on their geographical location, surrounding condition, characteristics of the deposited waste and stormwater in the surface water. Additionally, variations in sampling time, location, and volume, as well as the use of different detection methods and analysis processes, can also have an impact on the findings.

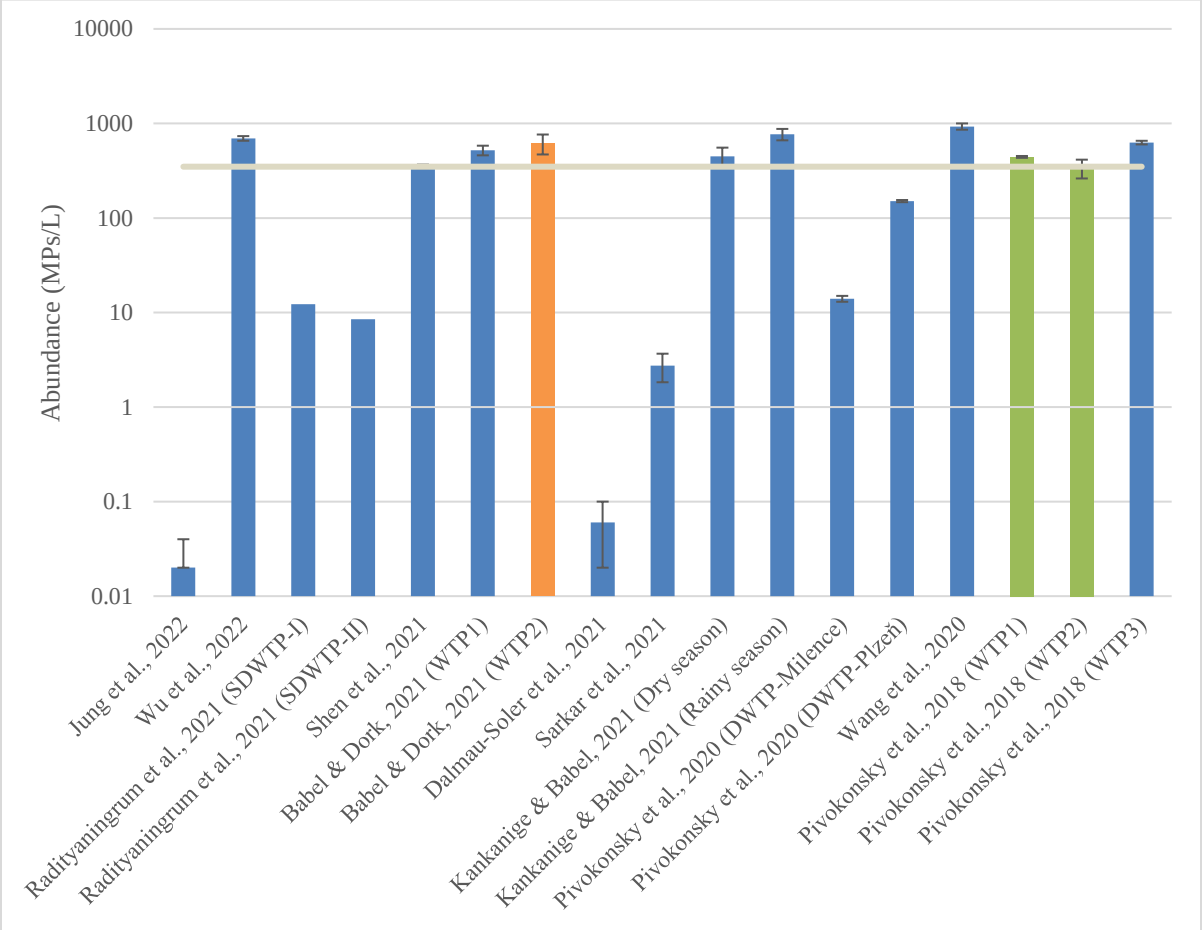


Figure 3.4: MPs abundance as the total number of MP particles per liter of water in the finished water of different DWTPs from the selected full-scale surveys

Figure 3.4 demonstrates a similar kind of variation for the total MP number in the outlet point of different DWTPs after undergoing treatment. The highest MP number (930 MPs/L) in the effluent was reported by Wang et al. (2020), which is consistent with the highest MPs abundance (6614 MPs/L) in their influent as seen in Figure 3.3. The lowest MPs abundance (close to 0.01 MPs/L) was reported by Jung et al. (2022) which also had the second lowest MPs number in the inlet of DWTPs. Dalmau-Soler et al. (2021) had the lowest MPs abundance in the influent and kept that lowest number in the effluent as well. For all of these surveys, the abundance of MPs seems to be under 1000 MPs/L in the effluent of these DWTPs, indicating that a certain percentage of MPs was removed by these DWTPs irrespective of the variations in their different characteristics such as size, shape, and polymer type. After going

through the treatment system of these DWTPs, the water is sent to the distribution system. Currently, the MP particle numbers are not monitored in the drinking water and so there exists no regulation on the allowable particle numbers of MPs in the treated water from DWTPs. But it has been postulated that human health can be impacted chronically by the long-term intake of MPs (Wright & Kelly, 2017). Therefore, there is a critical need for the development of guidelines as MPs are considered contaminants of emerging concern.

Along with this varying range of MPs numbers in the influent and effluent of these DWTPs, MPs also tend to be very divergent in terms of their properties i.e., size, shape, and polymer type. Figures 3.5, 3.6, and 3.7 illustrate the raw and finished water quality in terms of the percentage distribution of MPs across different size ranges, shapes, and polymer types respectively.

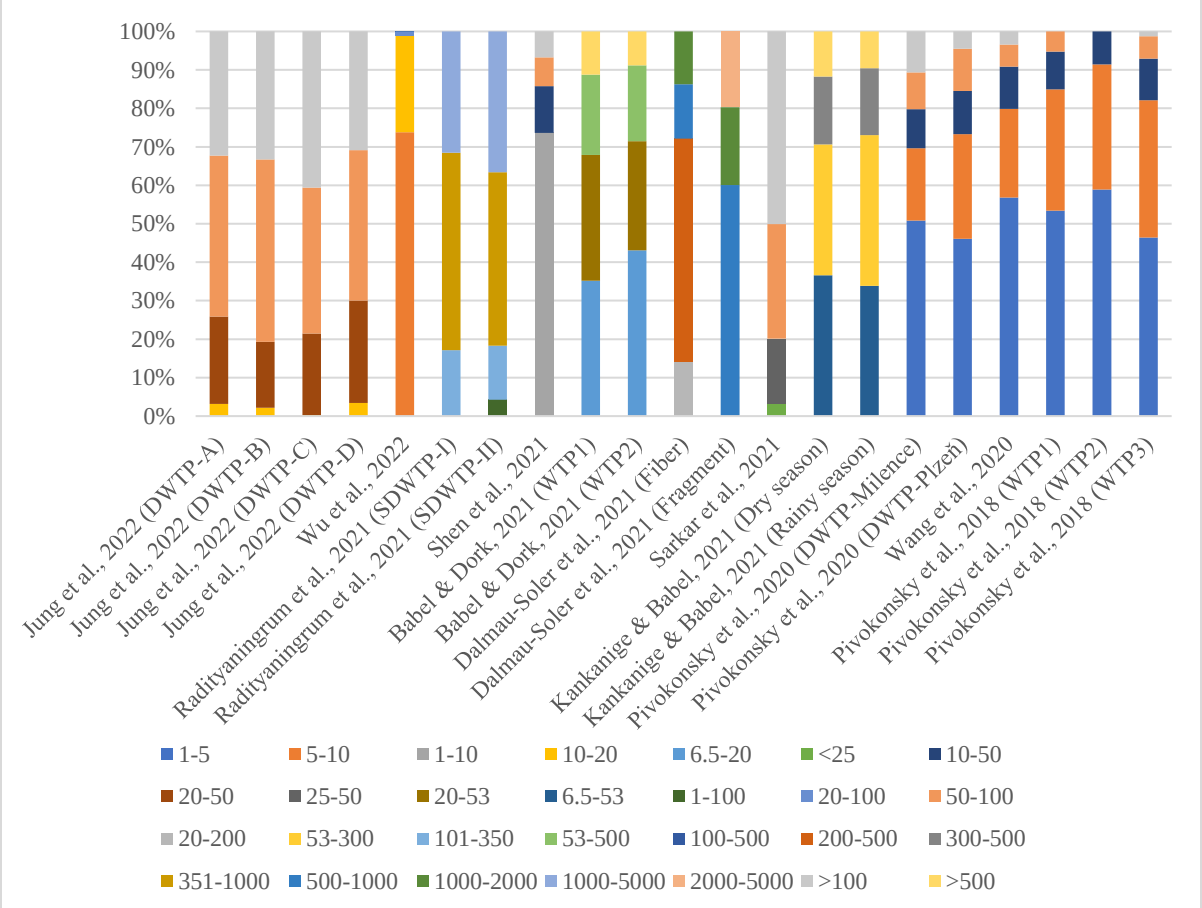


Figure 3.5 (a): Raw water quality in terms of the MPs size range

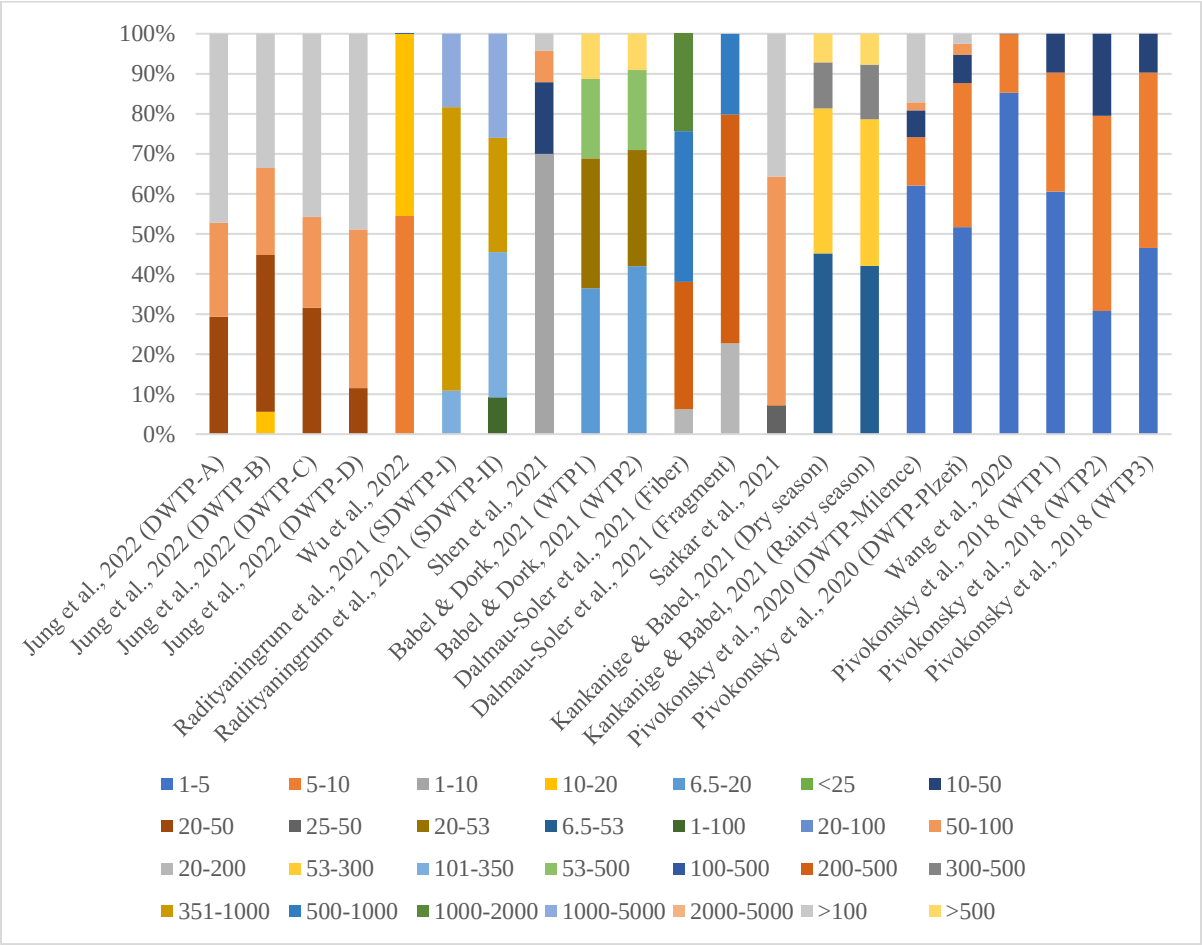


Figure 3.6 (b): Treated water quality in terms of the MPs size range

The size ranges of MPs show significant variation both in the influent and effluent of the DWTPs. Different surveys reported MPs abundance based on the specific size ranges they identified. In general, there were more size groups reported for MPs <100 µm across most of these surveys. MPs<100 µm were found to be the most common in the influent of the DWTPs irrespective of the difference in location of water sources and also in the finished water regardless of the treatment processes employed.

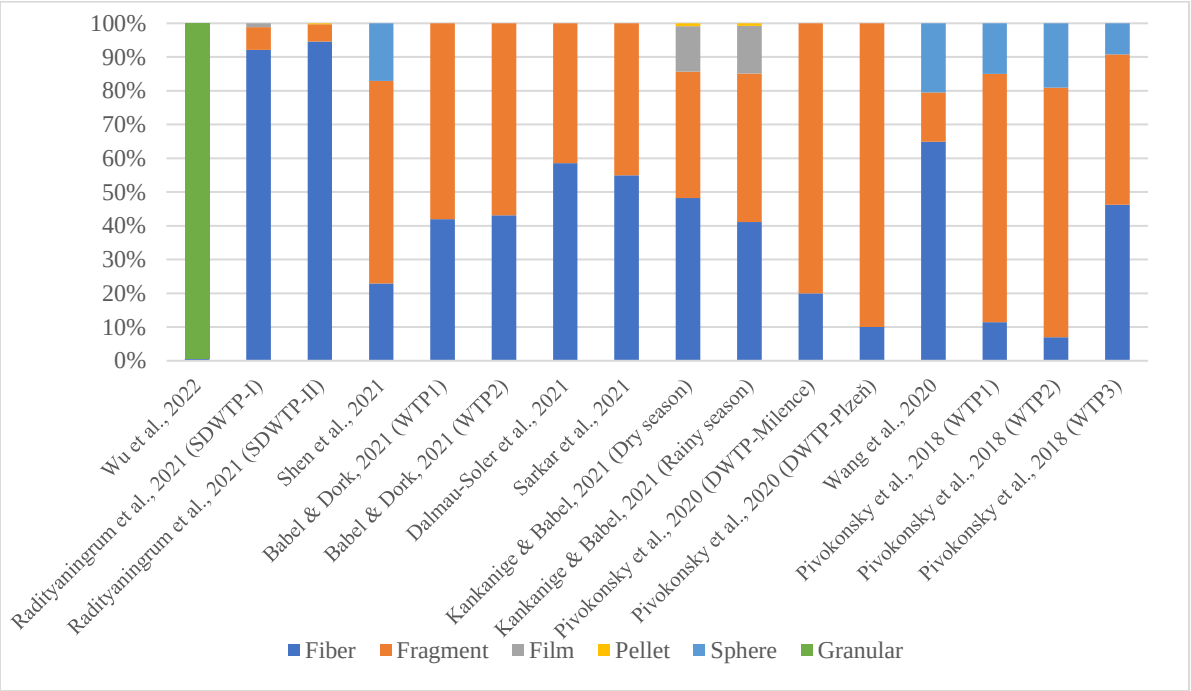


Figure 3.7 (a): Raw water quality in terms of the MPs shape

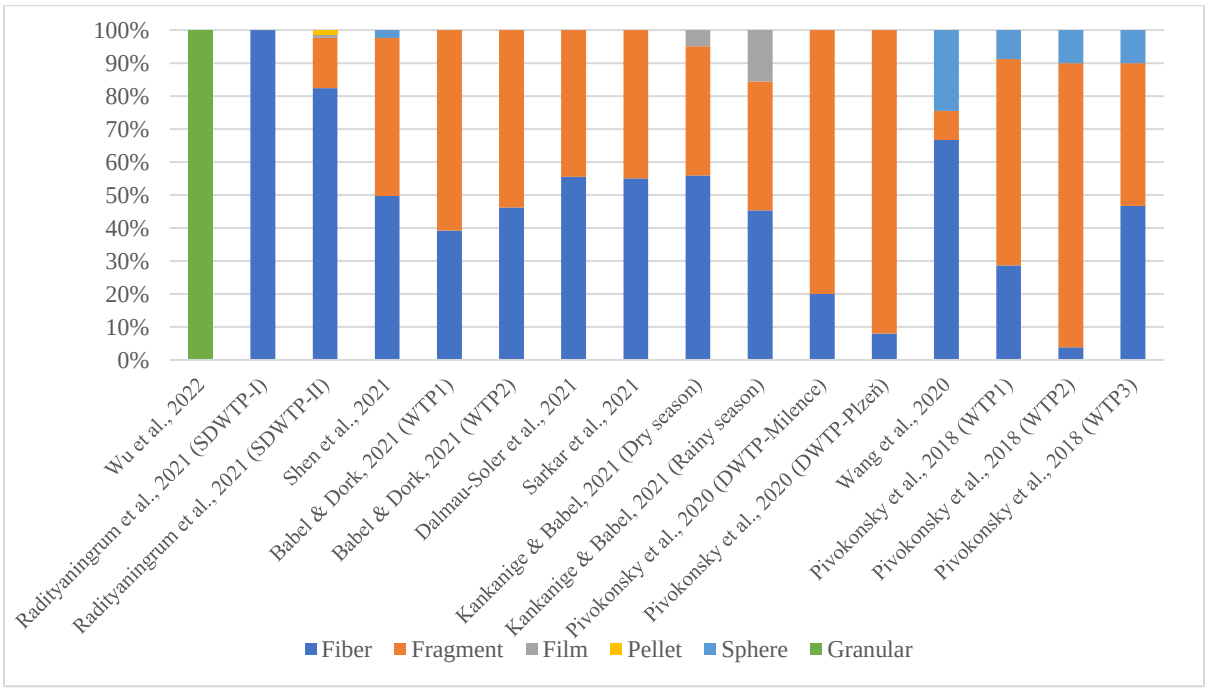


Figure 3.8 (b): Treated water quality in terms of the MPs shape

The shape of MPs was also examined in terms of the influent and effluent quality of these DWTPs. Fibers and fragments were found to be the most commonly detected MPs shapes both in the raw and treated water. Other shapes were rarely reported by these DWTPs. Though the actual numbers of MP particles varied between the influent and effluent of the DWTPs, the percentages of different MP shapes appeared to remain relatively consistent. This suggests that all MP shapes are being removed almost to the same extent by these DWTPs.

In addition, the water quality was assessed based on the distribution of MPs polymer type. PE, PP, and PET were found to be the prevailing polymer types both in influents and effluents. The percentages of different polymers reported in the surveys remained somewhat similar in both influents and effluents of the DWTPs, with a little variation in the polymer types. Besides, PAM was only detected in the effluent of four DWTPs, which can be attributed to the usage of PAM-containing coagulants during the coagulation process of those DWTPs.

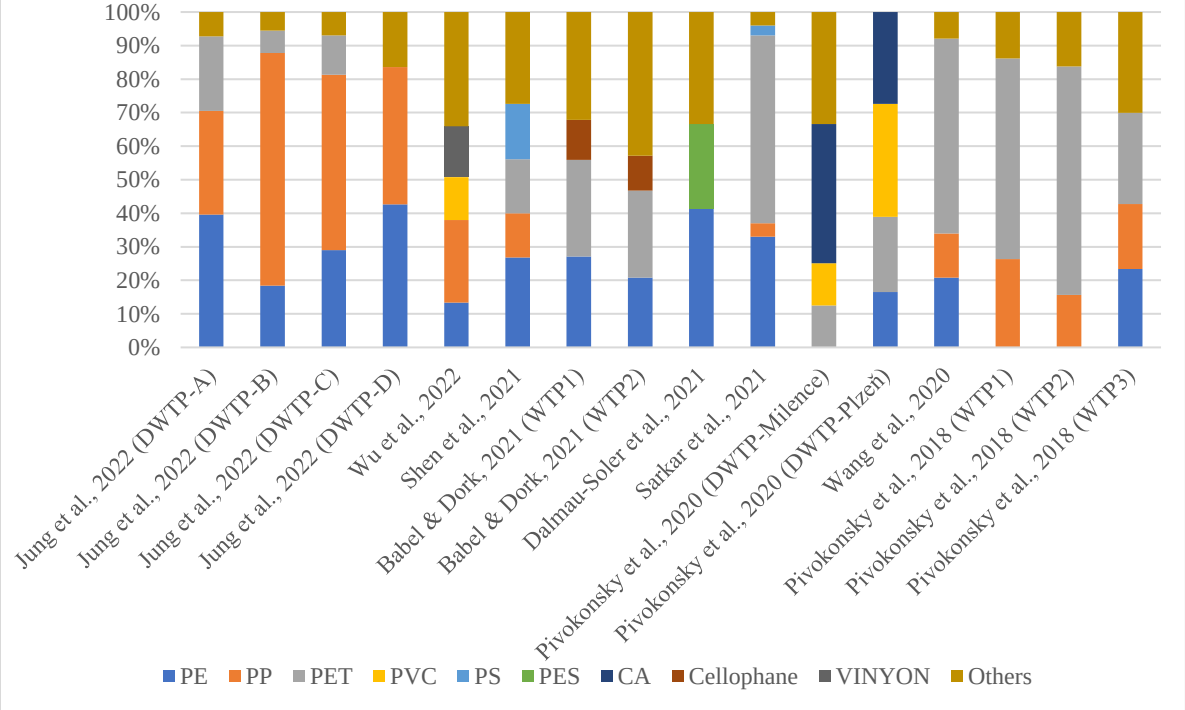


Figure 3.9 (a): Raw water quality in terms of the MPs polymer type

(PE-polyethylene, PP-polypropylene, PET-polyethylene terephthalate, PVC-polyvinyl chloride, PA-polyamide, PES-polyester, CA-cellulose acetate, PAM-polyacrylamide, Others-polymer types individually comprising <10% are included)

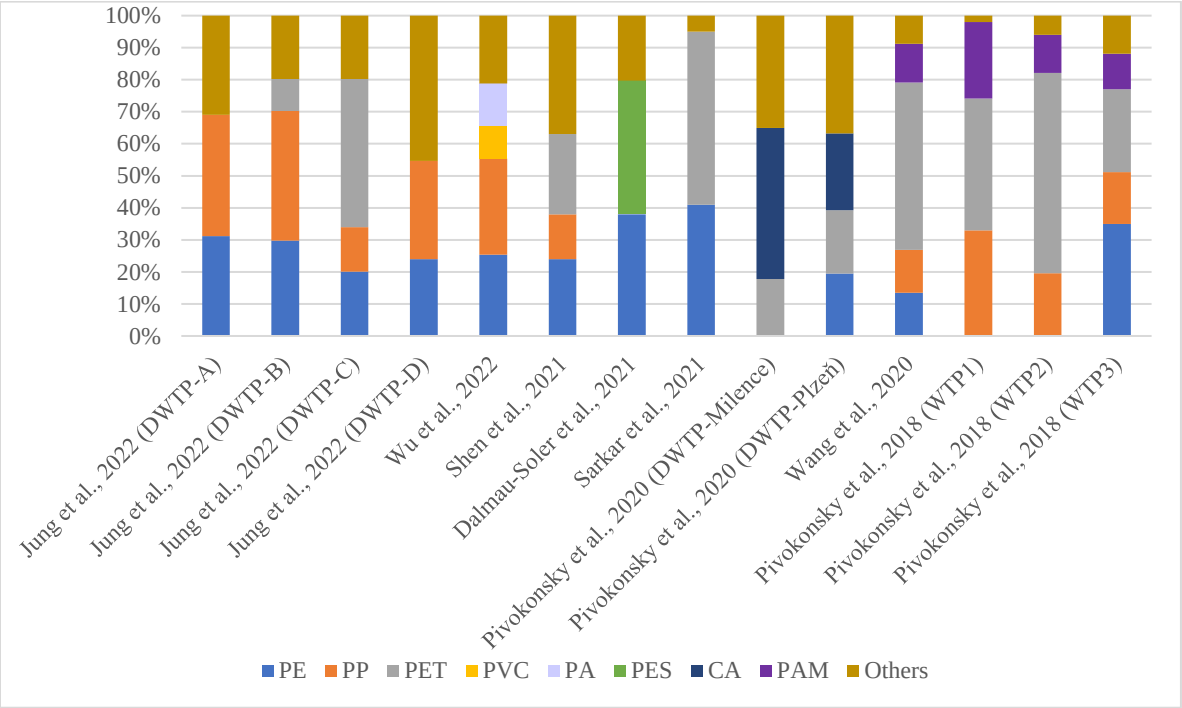


Figure 3.10 (b): Treated water quality in terms of the MPs polymer type

The comparison of Figures 3.5 (a,b), 3.6 (a,b), and 3.7 (a,b) depicts that MPs with different size ranges, shapes, and polymer types are widespread in both the influent and effluent of DWTPs. While some properties of MPs were occasionally completely removed during the treatment processes, most properties remained similar in both raw and treated water, albeit in varying percentages among different DWTPs. The reduction in total MPs particle number from the surface water sources by DWTPs is evident from the comparison of Figures 3.3 and 3.4. Then Figures 3.5, 3.6, and 3.7 demonstrate a comparable distribution of MP properties in terms of their abundance in the inlet and outlet points of the DWTPs. These DWTPs had their distinct combination of treatment processes, leading to differences in MPs removal.

Figure 3.8 provides an overview of the overall removal efficiency of MPs in full-scale DWTPs irrespective of their size, shape, polymer type, and initial abundance in the raw water. It also highlights the individual contributions of the CFS and filtration processes to the overall removal efficiency, which were calculated based on the MPs particle number in the inlet and outlet points of these respective treatment units. But not all full-scale surveys provided the exact values of individual removal efficiency

or the MPs abundance in the influent and effluent of the CFS and filtration units. Some of the surveys directly reported the removal efficiency value by CFS or filtration process or both. In cases where the removal efficiency was not directly reported by the author, it was calculated based on the difference in MP particle number per liter between the inlet and outlet of the treatment unit. The removal efficiency of MPs by CFS and filtration system could be obtained from 7 and 6 full-scale surveys, respectively. Wang et al. (2020) provided a range of overall removal efficiency values by DWTP, and the average value was used for the figure. Dalmau-Soler et al. (2021) outlined the average removal efficiency value with the standard deviation, and only the mean value was considered for the graph.

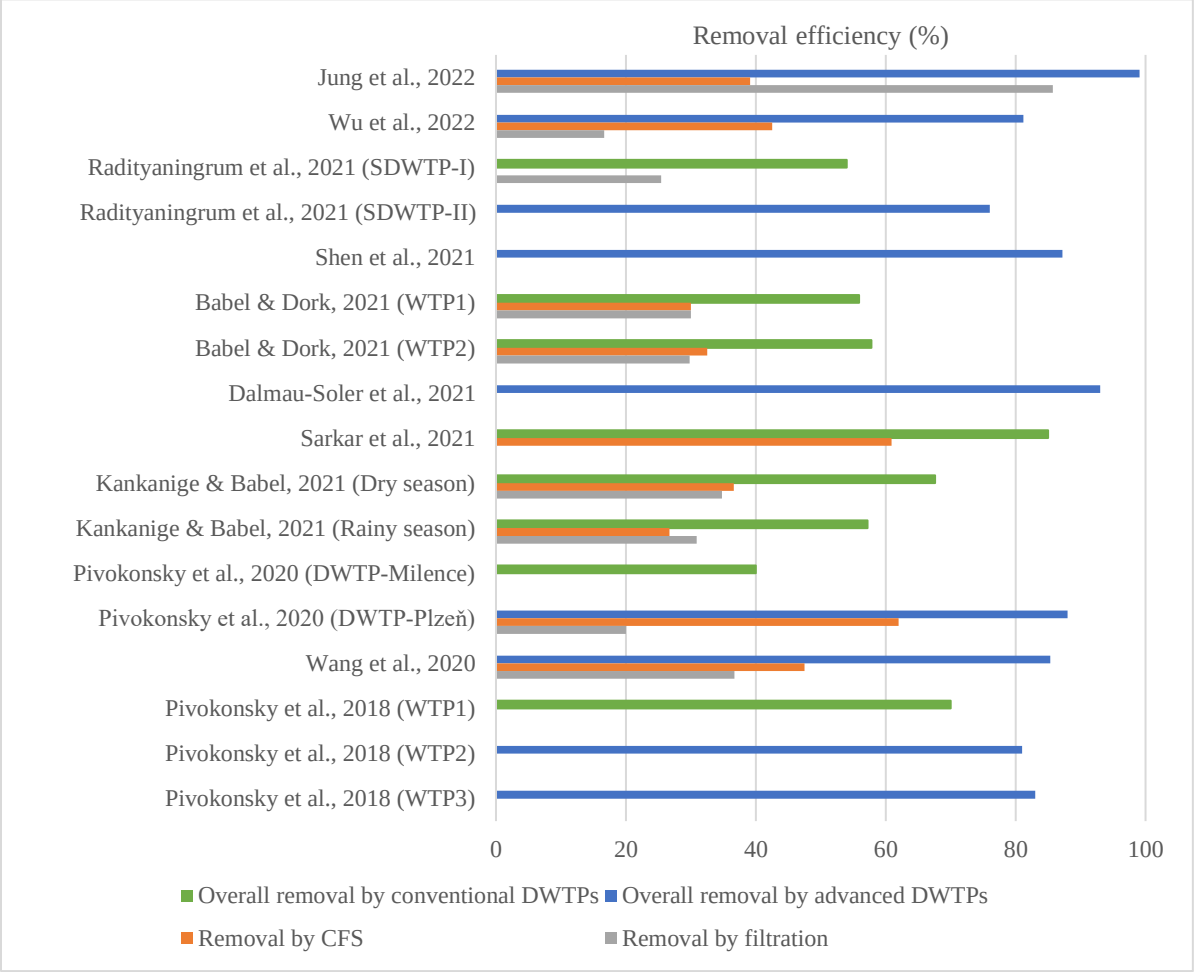


Figure 3.11: MPs removal efficiency of different DWTPs from different locations

(Removal by CFS and filtration are calculated based on the difference of MPs abundance in the influent and effluent of the respective treatment unit)

The removal efficiency of MPs in DWTPs, irrespective of their abundance in the influent and other properties (size range, shape, and polymer type), varied from around 40% to almost 100%, with an average value of 74%. The highest removal efficiency (99%) was reported by Jung et al. (2022), while the lowest removal efficiency (40%) was reported by Pivokonský et al. (2020) for one of the DWTPs. This removal percentage is significant considering the fact that these DWTPs were not specifically designed to remove MPs, although surface water treatment plants are designed to remove particles. In general, the DWTPs which were incorporated with any advanced technology along with conventional treatment processes achieved higher removal efficiencies than other ones, surpassing 75%. But exceptions were found, such as the WTP-1 in the study of Pivokonsky et al. (2018), which gained a relatively higher removal efficiency with only conventional treatment units. Again, Sarkar et al. (2021) utilizing pulse clarification instead of sedimentation, reported removal of MPs as high as 85%. In terms of the contribution of the CFS process to this overall removal of MPs by DWTPs, it varied from about 27 to 62%, highlighting its significance as a treatment technique for the removal of MPs from source water. The highest removal efficiency by the CFS unit was reported by Pivokonsky et al. (2020), potentially due to the wide range of MPs sizes (from 1 µm to >100 µm) considered in their survey. Other studies, such as Wang et al. (2020) and Sarkar et al. (2021), focusing on similar wide size ranges, reported comparable removal efficiencies of about 48% and 61% respectively. Sarkar et al. (2021) used pulse clarification instead of the sedimentation process after coagulation and flocculation, but the abundance of MPs was comparatively lower than in the other surveys. The lower removal efficiencies were reported by Babel & Dork (2021) and Kankanige & Babel (2021), which examined a narrower size range from >6.5 µm to >500 µm. For the surveys reporting removal by the CFS process, the abundance of MPs mostly ranged from 1000 to 2000 MPs/L, except for Jung et al. (2022), Wu et al. (2022), and Wang et al. (2020). Wang et al. (2020) reported an exceptionally high number of MPs compared to the others, while Jung et al. (2022) reported the opposite. In case of the filtration process, all DWTPs employed granular media filtration and the removal efficiency (calculated based on filter influent concentrations) usually varied from about 17 to 37%, with an exceptional value of 86% reported by Jung et al. (2022). The high removal efficiency only in this case may be attributed to the lower abundance of MPs in the inlet of the filtration unit (<2 MPs/L). However, in general, the filtration process achieved lower removal of MPs compared to the CFS process across these DWTP surveys.

In summary, this figure conveys an important message that DWTPs, despite not being specifically optimized for MPs removal, can effectively remove a significant number of MPs from source water using their diverse treatment techniques. Though advanced DWTPs have the tendency to better remove MPs, the two conventional treatment processes, CFS and filtration, can play crucial roles in MPs removal. The findings reflect overall the efficacy of existing treatment systems in reducing MP contamination, regardless of variation in MP abundance and characteristics, and highlight the varying performance of DWTPs in different situations.

3.4.2 Removal of Different Size Groups of MPs by Different Treatment Units of DWTPs

As noted previously, different full-scale DWTP surveys have taken different size ranges into consideration within the size limit of 1 µm to 5 mm based on the methodology used for MP quantification. As a result, it has become quite difficult to compare the findings of one survey with another. The selected surveys either directly reported MP removal efficiency values or provided MP particle numbers in the inlet and outlet of the DWTPs for their specific size ranges of MPs irrespective of other characteristics. In some cases, MP particle numbers and removal efficiency values were calculated based on the information from the literature and supplementary information given in the form of a table or figure. The overall removal efficiency values for different size ranges reported by these full-scale surveys have been separated here for conventional and advanced DWTPs. Furthermore, these figures are constructed without considering variations in other characteristics of MPs (i.e., shape and polymer type) and treatment processes employed by the DWTPs.

Figures 3.9 (a) and (b) are based on the dataset presented in Table A4 (which can be found in Appendix A), where they showcase the overall removal efficiency of MPs in different size ranges by conventional and advanced DWTPs, respectively. The figures display the particle size range in a logarithmic scale to encompass the wide size range from 1 to 5000 µm in a single graph. Each color represents a different DWTP from the selected full-scale surveys, with the horizontal bar in each color indicating the size ranges considered in that particular survey. Some surveys did not specify their upper size detection limit. For instance, Jung et al. (2022), Pivokonsky et al. (2020), Wang et al. (2020), and Pivokonsky et al. (2018) expressed their upper size limit as >100 µm, while Babel & Dork (2021) and Kankanige & Babel (2021) outlined it as >500 µm. For the convenience of understanding the graphs and keeping consistency, the upper size limit was shown as 5000 µm for all these surveys. Additionally, similar

graphs with dotted trend lines have also been drawn for the convenience of illustrating the trend of MP removal by each DWTP (Figures A1 & A2, in Appendix A). Those lines connect the mid-values of each size range within a survey, although the mid-values may not seem to align precisely in the middle of the lines due to the use of a logarithmic scale for plotting the size range. Those graphs may appear complex, but they provide valuable insights into the removal pattern of MPs within specific size ranges across the surveyed DWTPs.

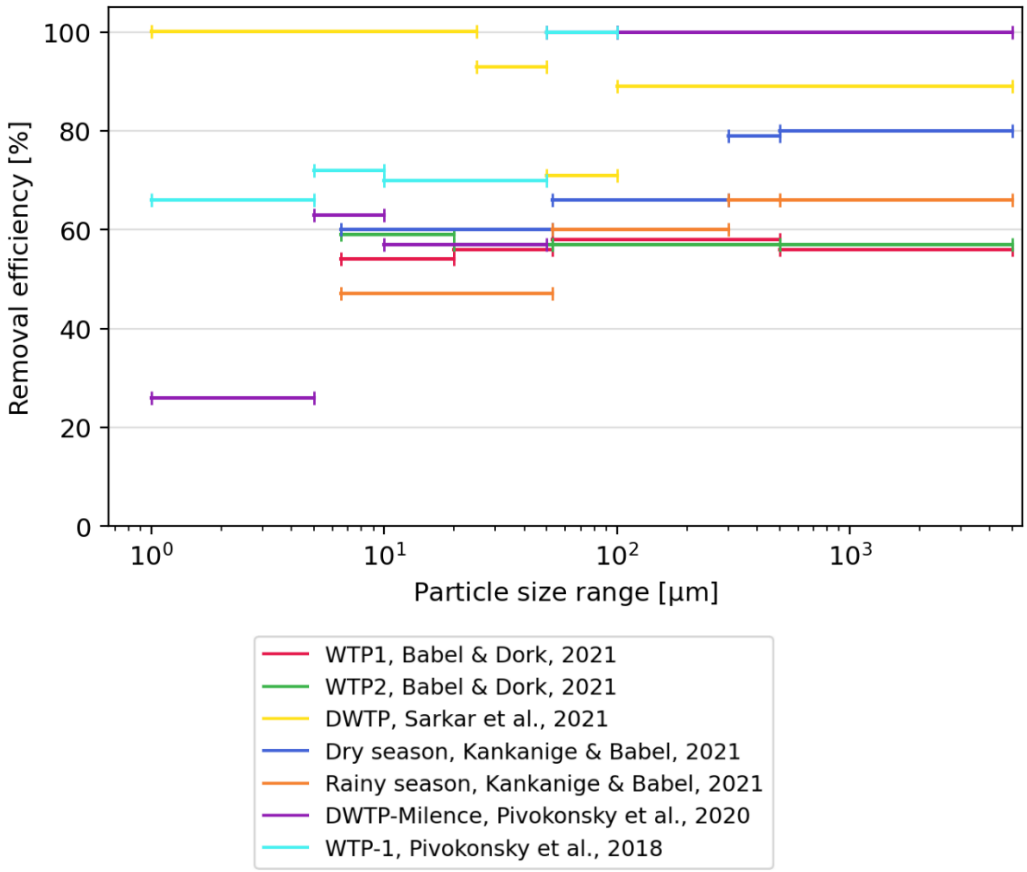


Figure 3.12 (a): Overall removal efficiency of MPs for different size ranges by conventional DWTPs

The overall removal efficiency values of MPs for different size ranges were reported for 6 conventional DWTPs of 5 full-scale surveys. The overall value across the surveys ranged between about 25 to 100%, regardless of the different size ranges considered. But the majority of the reported removal efficiencies fell within the range of approximately 50 to 80%. Notably, MP particles >100 µm consistently achieved more than 50% removal efficiency in all surveyed DWTPs. Babel & Dork (2021) and Kankanige &

Babel (2021) focused on MPs ≥6.5 µm, and the rest of them detected MPs as small as 1 µm. The trend of the removal efficiency across the size ranges was not consistent for all of these DWTPs. Babel & Dork reported almost similar removal efficiency for all size ranges, while Kankanige & Babel (2021) observed a slight increase in removal efficiency with increasing size ranges. Pivokonsky et al. (2020) and Pivokonsky et al. (2018) showed an overall increase in removal efficiency, particularly for higher size ranges, with minimal variations. On the other hand, Sarkar et al. (2021) exhibited a fluctuating pattern of removal efficiency as size ranges increased. Therefore, no definite pattern can be drawn for the removal efficiency of MPs across different size ranges based on the available data.

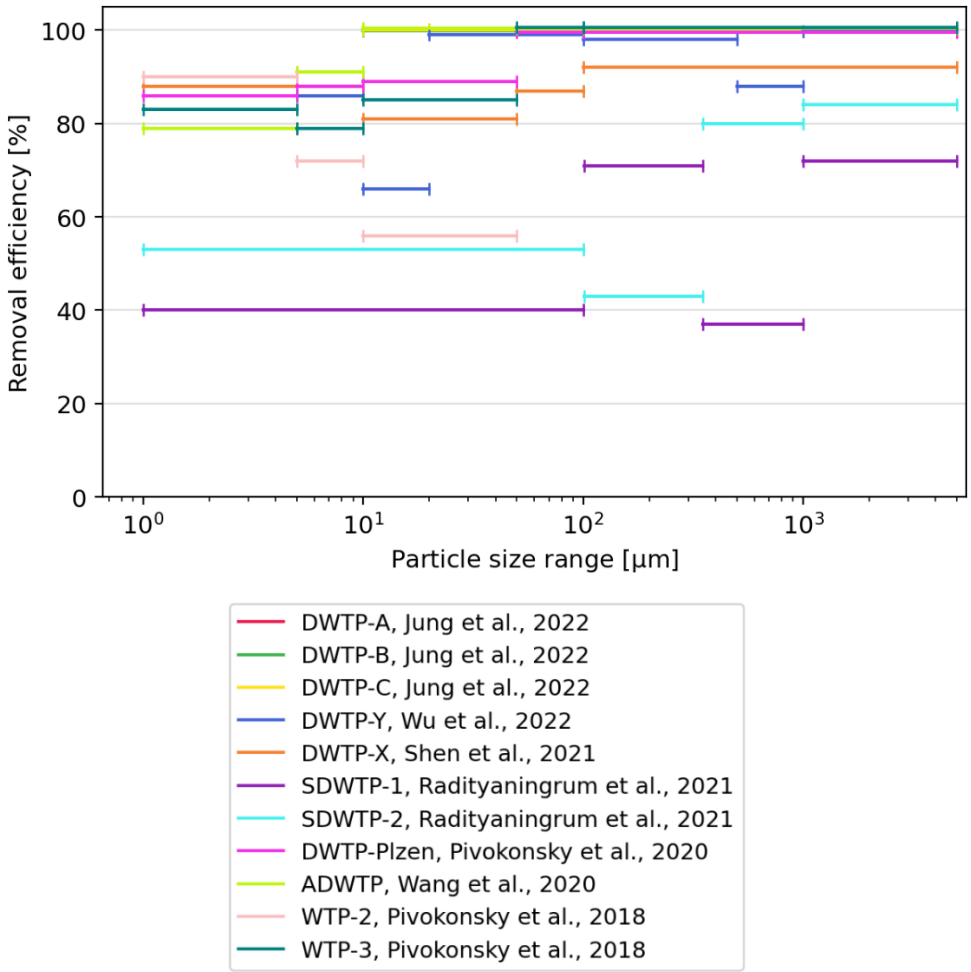


Figure 3.13 (b): Overall removal efficiency of MPs for different size ranges by advanced DWTPs

Again, the overall removal efficiency values of MPs for different size ranges were reported for 11 advanced DWTPs of 7 full-scale surveys. The reported values varied from around 40 to 100%, irrespective of those specific size ranges. In this case, most of the removal efficiencies were found to be within the range of about 80 to 100%, where only 4 out of the 11 DWTPs showed removals lower than 75% over the entire MP size range. These DWTPs incorporated advanced treatment units alongside their conventional treatment processes, which contributed to their higher removal efficiencies for all detected size ranges of MPs compared to the conventional treatment. In particular, Jung et al. (2022) attained 100% removal for all the size ranges. It can be because of the significantly lower amounts of MPs detected in this survey, which is less than even 10 MP particles per liter of the raw water. Again, the size detection limit in this survey started from 10 µm, which is higher than the lower size limit in the other surveys. Wu et al. (2022) detected MPs >5 µm, while all other surveys focused on MPs >1 µm. A few of the surveys observed an upward trend in removals with increasing size range, while more fluctuating removal patterns are evident for others.

Indeed, the results indicate a general tendency for advanced DWTPs to exhibit better overall removal efficiency for all size ranges of MPs. However, it is worth noting that conventional DWTPs also demonstrated good removal capabilities for all different size ranges of MPs, indicating that advanced technologies are not always necessary for getting effective removal of MPs. The performance of each DWTP varied and the specific size range of MPs did not exhibit a clear and consistent pattern of removal efficiency due to the wide variation in data reporting. But it is important to highlight that most of the surveys categorized MPs into multiple groups for sizes <100 µm. It is crucial to pay special attention to smaller-sized MPs in terms of the study of drinking water contamination, as these smaller-sized particles are often associated with potential toxic health impacts (Novotna et al., 2019).

Figures 3.10 (a) and (b) follow a similar approach to Figure 3.9 which show the removal efficiency of MPs for different size ranges by the CFS unit of these DWTPs. Here, the removal efficiency values have been separated based on the observed trend in the reported values. Figure 3.10 (a) displays the removal efficiency of MPs by the CFS units in DWTPs that demonstrated higher removal with increasing size ranges. Conversely, Figure 3.10 (b) represents the results showing the opposite trend. Due to the lack of data availability from the selected surveys, these graphs contain fewer data points compared to Figure 3.9 for overall removal. The removal percentage of MPs for these different size

ranges by the CFS unit is calculated based on the MP abundance measured at the inlet and outlet points of these CFS units. These values are reported irrespective of the variation in MPs shape, polymer type, and their abundance. Besides, similar figures with dotted trend lines can be found in Appendix A (Figures A3 & A4).

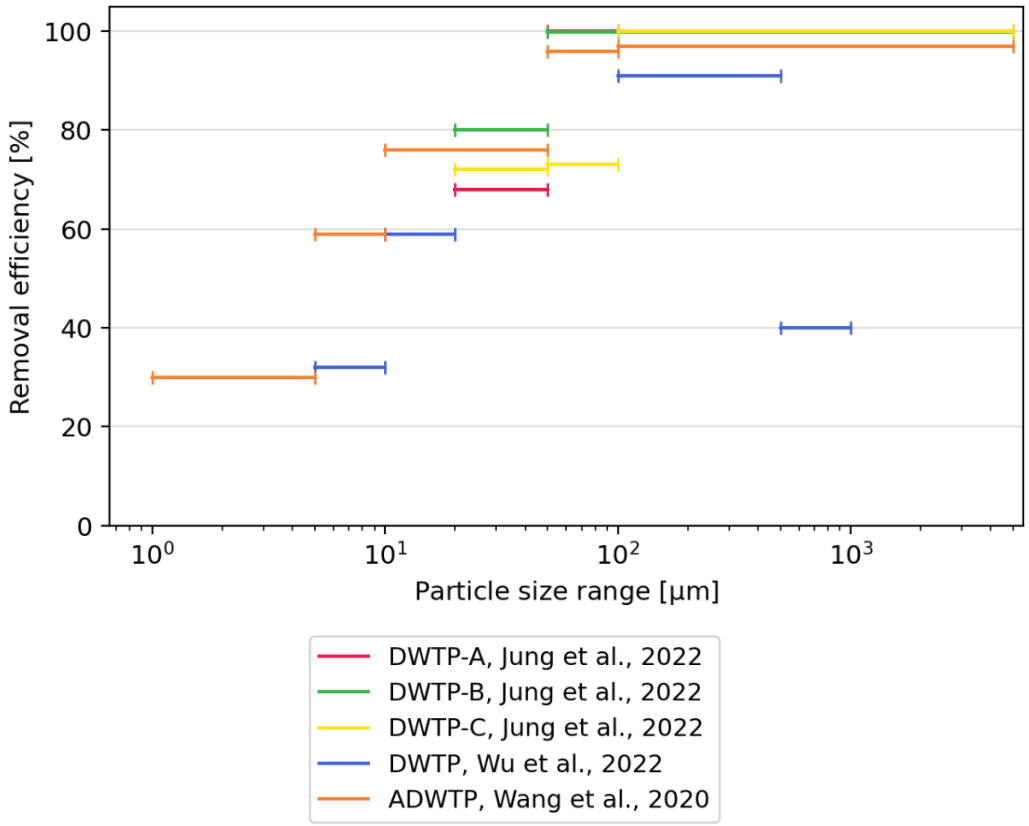


Figure 3.14 (a): Removal efficiency of MPs for different size ranges by CFS unit of different DWTPs (upward trend)

3 full-scale surveys comprising 5 DWTPs exhibited an upward trend in the removal of MPs for different size ranges, where the larger the MP size, the higher the removal efficiency was. However, Wu et al. (2022) showed a decline in the trend for the 500-1000 µm size range. Despite the similar removal trend, the abundance of MPs varied among these DWTPs. Jung et al. (2022) reported the lowest MP abundance in the influent, whereas Wang et al. (2020) reported the highest MP abundance among these selected surveys. The particle types were also investigated, but no significant common properties could be found, although there were some shared polymer types and shapes. The surveys had different size

detection ranges, but all of them employed automated image analysis techniques using three different microscopic systems. The full-scale surveys did not provide information about the specific details and operating conditions of the CFS processes, preventing a detailed analysis of their influence on MPs removal efficiency.

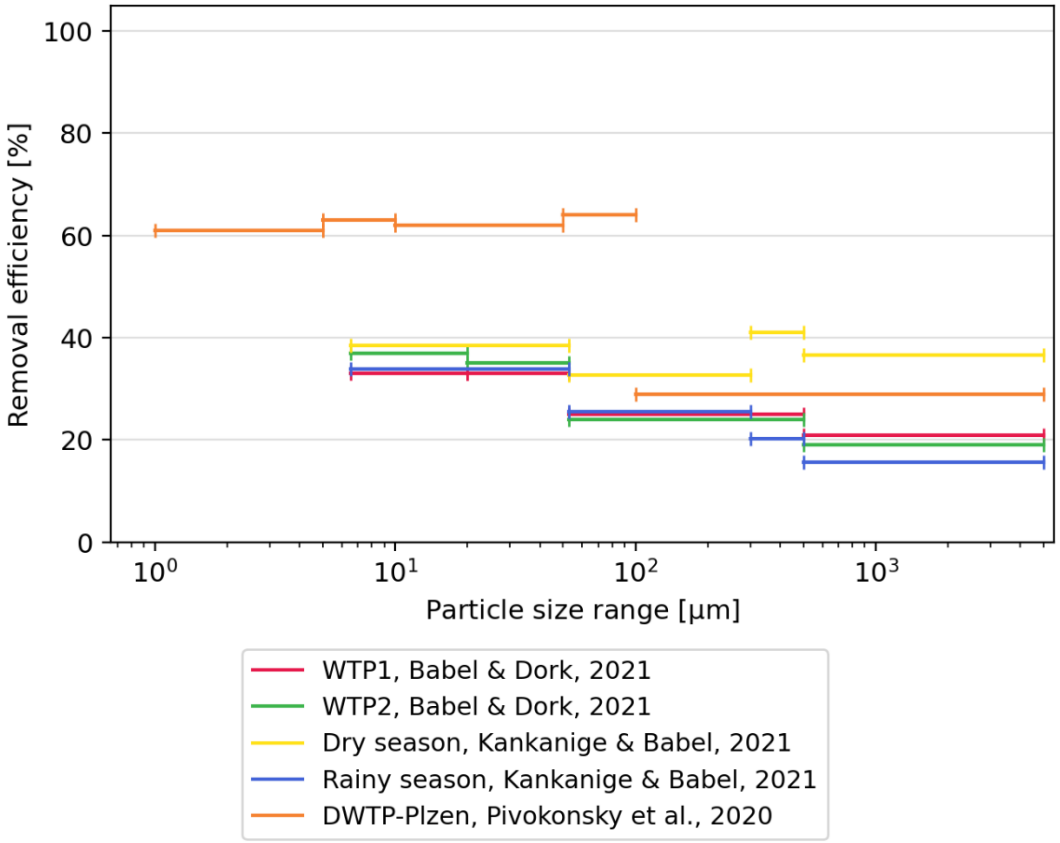


Figure 3.15 (b): Removal efficiency of MPs for different size ranges by CFS unit of different DWTPs (downward trend)

On the other hand, the opposite result was also observed for 4 DWTPs of 3 full-scale surveys, where the removal efficiency decreased with the increasing size of MPs. These surveys indicated that the CFS process achieved better removal for the small-sized MP, with the highest reported removal efficiency at around 60%. The removal efficiencies for all other larger size ranges were lower than that value. The influent concentration of MPs for these DWTPs ranged from approximately 1200 to 1800 MPs/L. These surveys reported relatively lower removal efficiencies (within 20-40%) for all their size ranges (≥6.5 µm), except for Pivokonsky et al. (2020), which focused on MPs>1 µm and showed the highest removal

efficiency among these surveys. Additionally, the surveys had differences in their reported MP shapes and polymer types. No specific reason could be determined for their downward trend in the removal with increasing size range. Besides, insufficient details on the specifics of the CFS process and their operating conditions in the surveys hindered the assessment of its impact on removal efficiency.

Figures 3.11 (a) and (b) are established in the same way as Figures 3.10 (a) and (b) to show the removal efficiency of MPs for different size ranges by the filtration units of DWTPs. The reported removal efficiency values were segregated again based on the observed trends. Figure 3.11 (a) represents the trend of higher removal efficiency with increasing size ranges, while Figure 3.11 (b) showcases the opposite trend. Limited surveys provided the necessary information for these graphs, so again resulting in fewer data points compared to the overall removal graph. The removal efficiency values were calculated based on the difference in influent and effluent concentration for each size range of MPs in the filtration units of these DWTPs. These values are also reported regardless of the differences in MPs shape, polymer type, and their abundance. Additionally, corresponding figures with dotted trend lines can be found in Appendix A (Figures A5 & A6).

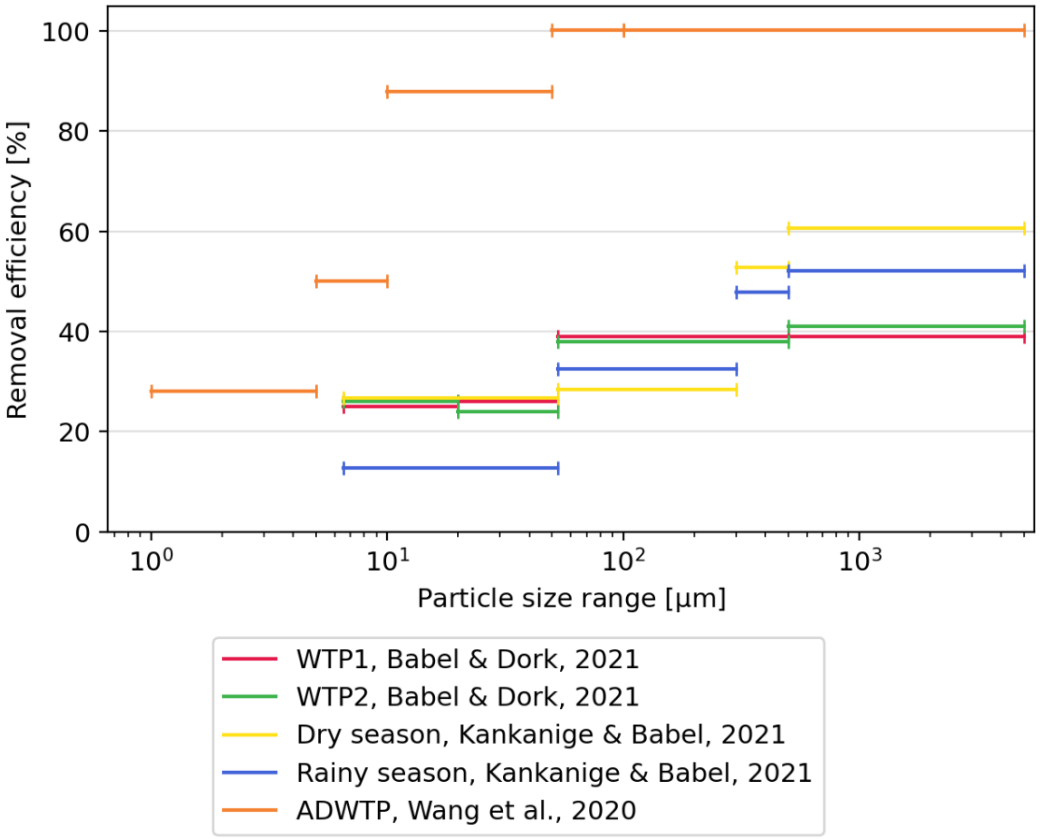


Figure 3.16 (a): Removal efficiency (based on filter influent MP numbers) of MPs for different size ranges by filtration unit of different DWTPs (upward trend)

4 DWTPs from 3 full-scale surveys showed higher removal with increasing size ranges. Except for Wang et al. (2020), the other surveys reported their removal within about 15-60% for size ranges 6.5-5000 µm. Wang et al., 2020 had the highest removal efficiency trend overall for MPs of all of the size ranges, where they focused on MPs from 1 µm and their MPs abundance in the influent was also the highest among the selected surveys.

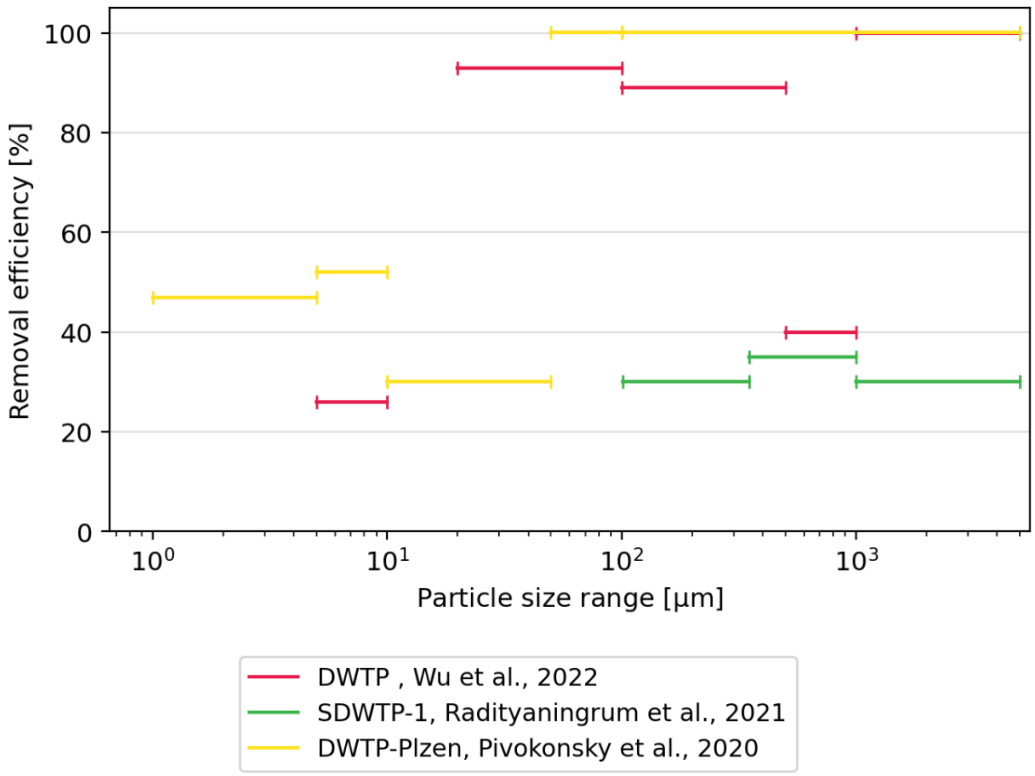


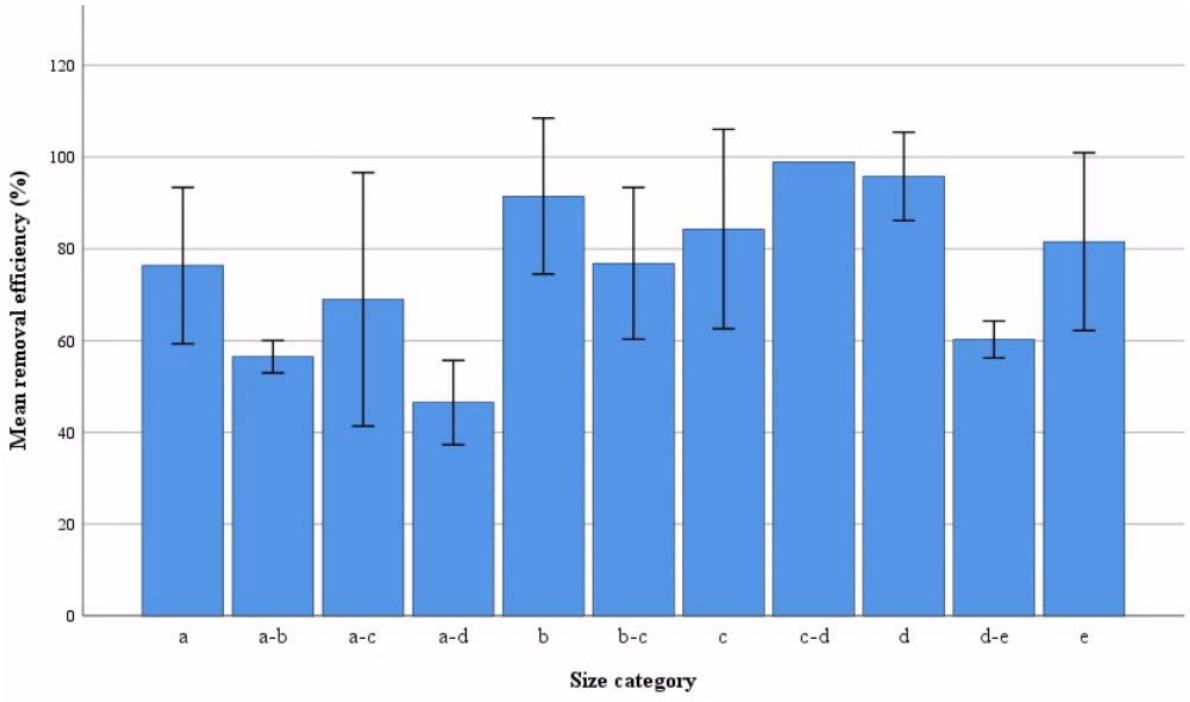
Figure 3.17 (b): Removal efficiency (based on filter influent MP numbers) of MPs for different size ranges by filtration unit of different DWTPs (downward trend)

The removal efficiencies depicted in Figure 3.11 (b) exhibit irregularities in their reported values. They do not show any specific trend in their change of removal efficiency with respect to size ranges. For instance, the 5-10 µm and 500-1000 µm size ranges of Wu et al. (2022) and the 10-50 µm size range of Pivokonsky et al. (2020) had lower removal than the other size ranges. Additionally, Radityaningrum et al. (2021) reported low removal efficiencies (below 40%) for all their size ranges. The MPs abundance and their other characteristics were also different among these surveys. Though the observed trend in the removal efficiency of MPs varied within the size ranges, larger-sized MPs generally experienced better removal in the filtration unit. However, the absence of specific information regarding media filtration conditions such as media size and surface properties, filtration rate, etc., limits the ability to establish a definitive reason or conclusion.

In summary, the CFS and filtration units of the DWTPs demonstrated a certain percentage of removal efficiency for all different size ranges of MPs within the range of 1-5000 µm. Though these processes

were not specifically optimized for MPs removal, they exhibited good removal capabilities, regardless of the differences in MPs abundance and their properties. The variations in removal efficiencies also did not appear to be consistent with regard to the other characteristics of the MPs studied in these surveys including shape and polymer type. Besides, the lack of information regarding the treatment processes properties, such as coagulant name and dosage for the CFS process or filter size and surface properties for the filtration process, and their operating conditions, hinders the identification of potential factors contributing to the observed differences in the reported results. Moreover, the full-scale surveys did not provide data on turbidity removal, which would have been valuable in understanding the overall particle removal efficiency and the specific removal of MPs under normal operating conditions.

Since different full-scale surveys investigated the removal of MPs for different size ranges, it has become quite difficult to comment on the pattern of removal efficiency for a definite size range. In order to alleviate the situation and to utilize the existing dataset in a way that can help to manifest some uniform decisions, five size categories have been formed within the size range of 1-5000 μm. These size categories, labeled as a, b, c, d, and e, help group MPs based on their size range. Any MPs falling in the 1-10 μm size range, would go under size category ‘a’, then following the same pattern, any MP size range falling in between 10-20 μm, 20-50 μm, 50-100 μm, and 100-5000 μm size ranges, would be designated under the size category ‘b’, ‘c’, ‘d’, and ‘e’ respectively. If there is any size group that falls into more than one category, it can be conveyed by a joint version of the categories. For example, in the study of Radityaningrum et al. (2021), the first size range is reported as 1-100 μm. So, for this one, the size category would be modified as a-d, as it includes all the size ranges in between that. All those joint size categories have been arranged following the same pattern. The size categories have been prepared keeping the main focus on the smaller size ranges, as most of the surveys divide their detected MPs <100 um into multiple size ranges. The size category for each of the size ranges of all the selected full-scale surveys can be found individually in Table A4. This approach may enable a more organized and consistent analysis of data.



n value: a: 14, b: 4, c: 6, d: 10, e: 24, a-b: 2, a-c: 3, a-d: 2, b-c: 7, c-d: 1 and d-e: 4

Figure 3.18: Mean removal efficiency of MPs by size category
(where a: 1-10 µm, b: 10-20 µm, c: 20-50 µm, d: 50-100 µm, e: 100-5000 µm)

Figure 3.12 provides an overview of the average removal efficiency of MPs for each size category, with n values for each category and error bars representing standard deviation. This figure may help to understand the general trend of removal for different size categories in DWTPs. Here, the highest removal efficiencies can be seen for the size categories 'c-d' and 'd', where the size ranges are 20-100 and 50-100 µm respectively. But there was only one data point for the 'c-d' category, indicating that only one survey reported a higher removal value for this size range. The 'd' category, with 10 different values from different surveys, attained a higher average removal efficiency (>90%) for MPs within the 50-100 µm size range. Then the second highest average removal efficiency (>80%) is observed for size category 'b', which has only 4 data points. Size category 'c', with only 6 data points, also shows a higher mean removal efficiency value (>80%). Then, 'a-b' and 'a-d' size categories had only two data points each and showed lower mean removal efficiency than the other categories. 'b-c', 'd-e', and 'a-c' size categories had 7, 4, and 3 data points respectively. The majority of the data points fall into the size category 'e' portraying the wide size range of 100-5000 µm, where the average removal efficiency

value based on 24 data points is about 80%. So, in general, MPs falling in the size range of >100 µm were more frequently reported to be better removed than the MPs of < 100 µm. Indeed, it is crucial to explore the removal pattern of smaller-sized MPs due to their potential human health concerns.

It is also essential to portray if there is any relation between the removal efficiency of MPs for different size categories and their concentrations at the inlet and outlet points of the DWTPs. Figures 3.13 and 3.14 are prepared to assess this, while they provide visual insights into the variations in removal efficiency and MPs abundance across different size categories and surveys. These figures utilize different colors to represent various full-scale DWTP surveys, while different sizes of circles indicate different size categories, starting with the smallest circle representing the smallest size category. It is worth noting that certain circles of the same size and color may appear multiple times, since several size ranges of MPs may fall into the same size category within a particular survey.

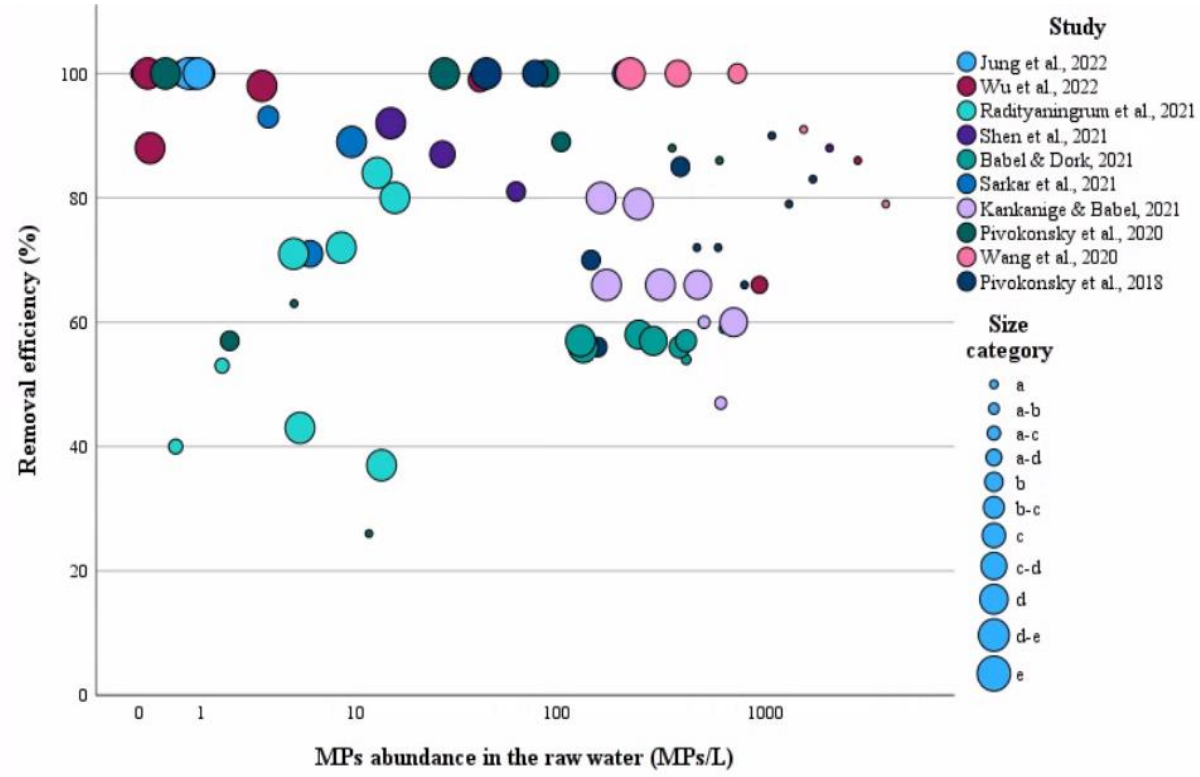


Figure 3.19: Relation between removal efficiency and MPs abundance in the inlet points of DWTPs for different size categories from the selected full-scale surveys

(where a: 1-10 µm, b: 10-20 µm, c: 20-50 µm, d: 50-100 µm, e: 100-5000 µm)

Despite the scattered nature of the observed data, several general inferences can be drawn. It is evident that the abundances of larger size categories are typically lower than 1000 MPs/L in the influent of the DWTPs. Smaller-size categories, on the other hand, tend to exhibit higher abundances. Generally, higher MPs abundances seem to correspond to higher removal efficiency. Additionally, the higher abundances of larger size categories tend to attain comparatively better removal than the higher abundances of smaller size categories. Apparently, some higher-size categories exhibit complete removal, regardless of whether their MPs abundance is lower or higher.

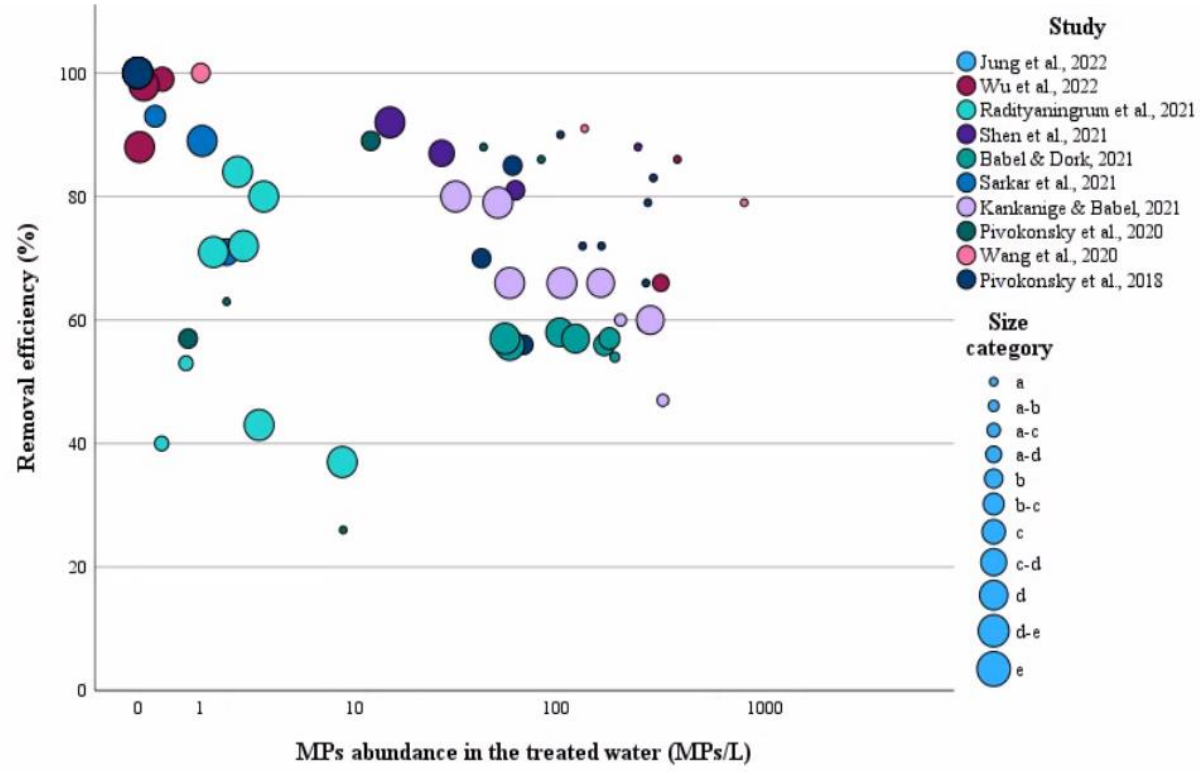


Figure 3.20: Relation between removal efficiency and MPs abundance in the outlet points of DWTPs for different size categories from the selected full-scale surveys

(where a: 1-10 µm, b: 10-20 µm, c: 20-50 µm, d: 50-100 µm, e: 100-5000 µm)

In the effluent of the DWTPs, the abundance of MPs for the majority of size categories is below 500 MPs/L. As most of the MPs from larger size categories were completely removed, MPs abundances of smaller size categories tend to have a more frequent presence in the effluent. Most of the MP

abundances for all size categories are observed to be close to 100 MPs/L and their removal efficiencies range from over 40% to 90%.

The relationships between the removal efficiency of MPs within the defined size categories and their abundance in the raw and treated water of different DWTPs facilitate comparisons of widely scattered data. The use of size categories helps in organizing the data and understanding the patterns across different surveys. Despite the wide variations in reported results due to differences in water sources, MP properties, and detection techniques, these findings offer a general understanding of the overall scenario.

Additionally, the relationship between MPs removal efficiency within the specified size categories and their abundance in the influent and effluent of CFS and filtration units was analyzed. Detailed figures presenting these findings can be found in Appendix A (Figures A7-A10). However, the observations did not reveal any significant associations between the removal efficiency of CFS and filtration processes and the abundance of MPs in the influent or effluent of the respective treatment units across the selected surveys.

3.4.3 Removal of Different Shapes of MPs by Different Treatment Units of DWTPs

Several full-scale surveys among the selected ones have delineated information on the removal efficiency of MPs based on their different shapes, irrespective of other factors such as MPs abundance, size ranges, and polymer types. As noted previously, MPs can be of various shapes like fiber, fragment, film, granule, sphere, pellet, etc. Not all the studies have reported on all of these shape categories, however, some of them provided enough information to find out the removal percentages for various shapes of MPs. Figure 3.15 (a), (b), and (c) depict the removal efficiency of MPs for various shapes by the full-scale DWTPs and individually by CFS and filtration units of those DWTPs respectively. The overall removal efficiency values were calculated by comparing the MPs abundance in the influent and effluent of DWTPs. Similarly, the removal efficiency for CFS and filtration units was calculated based on the MPs abundance in the inlet and outlet of those respective treatment processes individually. These figures provide insights into the effectiveness of the DWTPs in removing MPs of different shapes.

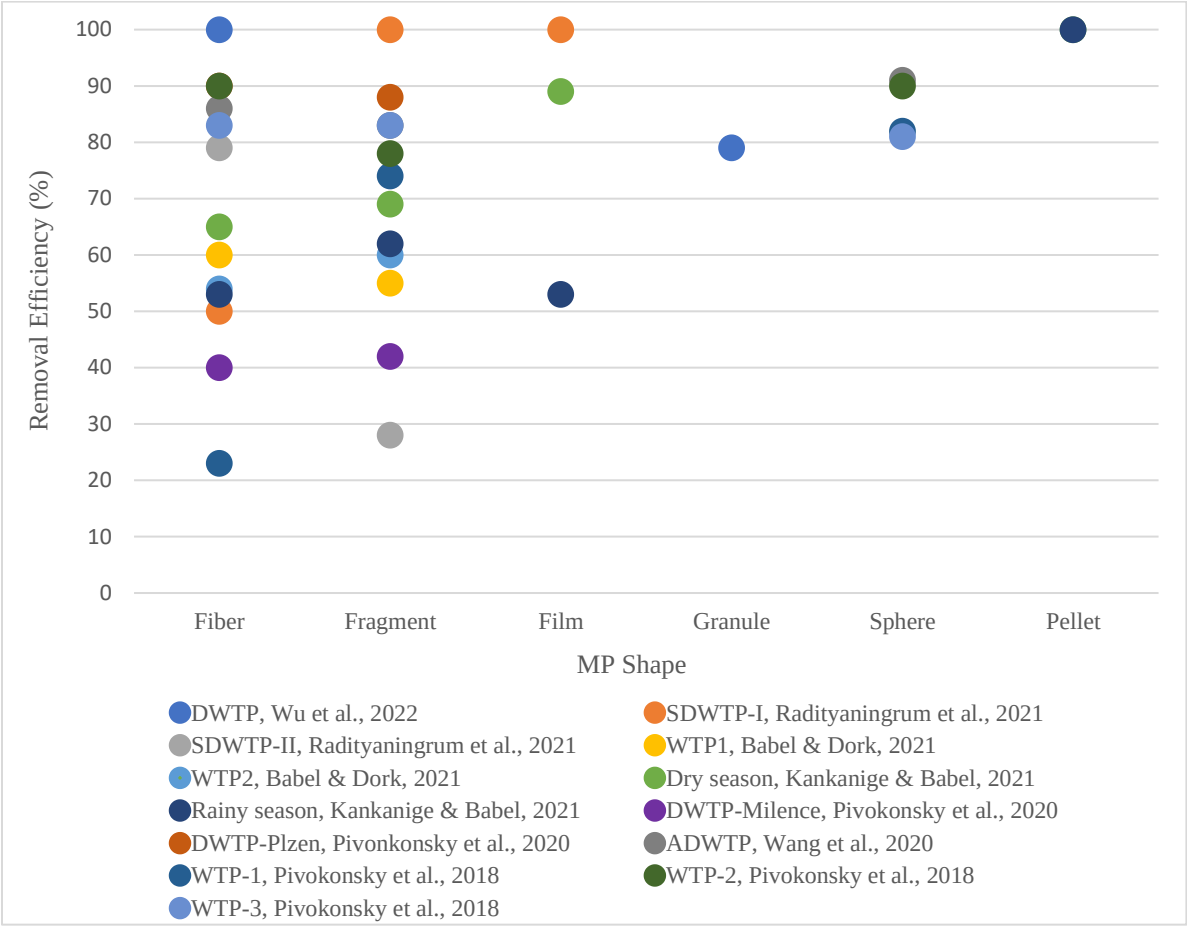


Figure 3.21 (a): Removal efficiency of MP for various shapes by full-scale different DWTPs

Although Figure 3.15 (a) shows the removal efficiency of MPs for various shapes, irrespective of differences in abundance, size ranges, and polymer type, most of the surveys report removal efficiency primarily for fibers and fragments. Fibers and fragments were found to be the most commonly observed MP shapes in the raw water sources. Nevertheless, there are variations in the reported removal efficiency values for the same shape of MPs. For fibers, the removal efficiency varied from about 20 to 100% among these DWTPs. The lower removal efficiencies in the range of 20-70% were reported by the DWTPs which had only conventional treatment processes. The DWTPs which had advanced treatment units achieved higher removal of fibers, in the range of 80 to 100%. Similarly, for fragments, the removal efficiency ranged from around 30 to 100%. As with fibers, the conventional DWTPs achieved comparatively lower removal of fragments (<80%), except the SDWTP-I of Radityaningrum

et al. (2021). That DWTP had 100% removal of fragments despite having only conventional treatment processes. The advanced DWTPs achieved >80% removal for fragments. Due to the limited availability of data for other MP shapes, no definitive removal patterns can be discerned.

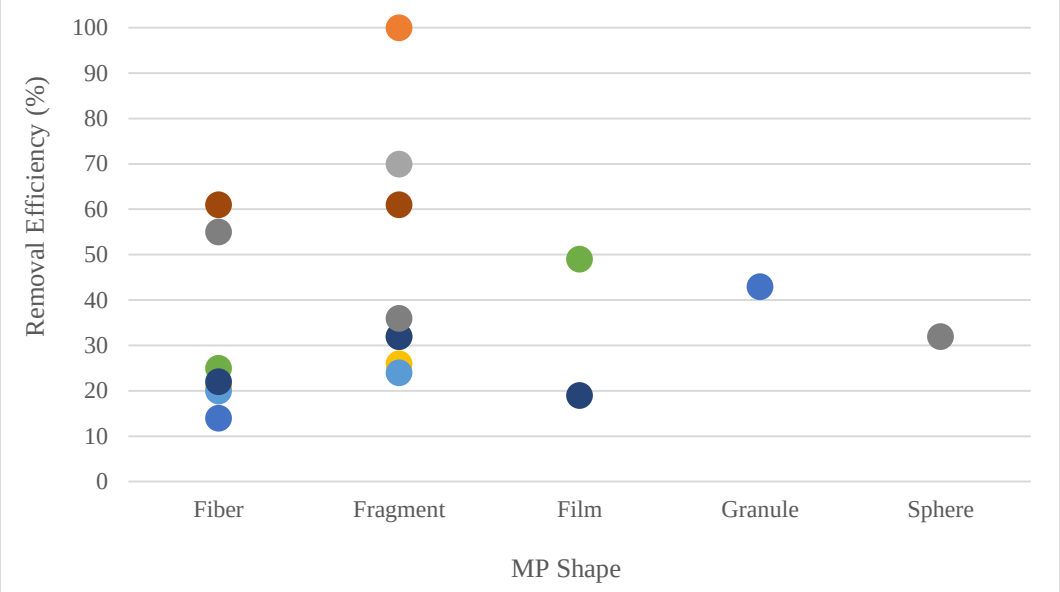


Figure 3.22 (b): Removal efficiency of MPs for various shapes by CFS unit of different DWTPs

In Figure 3.15 (b), the removal percentages for fibers appear to be clustered in two groups, one in the range of 10-30% and the other one close to 60%. The lower range of removal was mostly reported by conventional DWTPs, except in the survey by Wu et al. (2022). For fragments, some removal efficiencies ranged between 20-40%, while others had higher removal. The removals in the range of 60-70% were primarily reported by advanced DWTPs. Notably, one conventional DWTP achieved 100% removal of fragments, where the MPs abundance in the influent was lower than the other DWTPs.

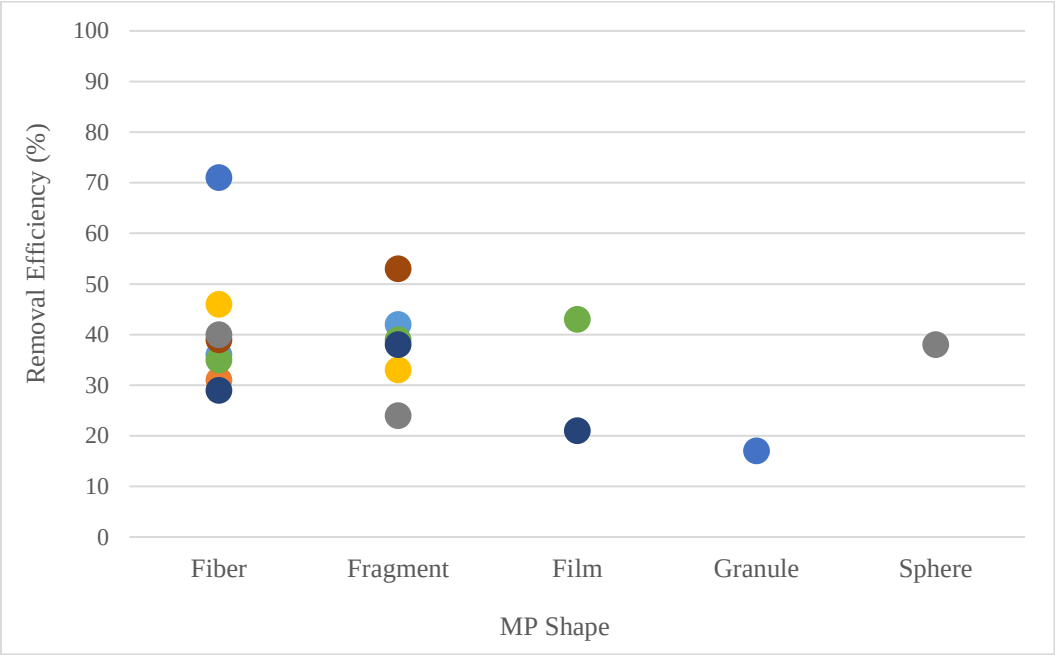


Figure 3.23 (c): Removal efficiency of MPs for various shapes by filtration units of different DWTPs

In Figure 3.15 (c), the removal percentages for fibers and fragments seem to fall within a range of values from >20% to <60%. On average, the removal achieved by the filtration unit is lower than the removal by the CFS unit. But both situations have some exceptions. This small dataset of MPs removal efficiency for various shapes from these studies only allows a preliminary assessment of a pattern in the results. An important inference however is that the removal percentages are scattered over a wide range of values. However, the differences in detection methods may contribute significantly to these large variations, and other factors such as MP particle numbers and properties (size ranges, polymer types) can also influence the value of removal efficiency.

3.4.4 Removal of Different Polymer Types of MPs by Different Treatment Units of DWTPs

Only a couple of surveys have provided information about the removal of MPs with respect to different polymer types, regardless of their differences in abundance, size range, shape, etc. As these full-scale surveys were conducted in different regions, they collected water from different sources and at different times. The presence of different polymer types in the water was influenced by the surrounding environment and the type of plastic materials used and deposited. Different properties of MP materials are used for different purposes, leading to the reporting of diverse polymer types in the surveyed DWTPs. Figure 3.16 (a), (b), and (c) represents the removal efficiency of MPs for different polymer types by full-scale DWTPs, as well as individually by CFS and filtration units within those DWTPs respectively. It should be noted that these figures are based on information derived from only 3 full-scale surveys. The removal efficiencies were calculated based on the MPs abundance in the influent and effluent of those DWTPs, CFS units, and filtration units.

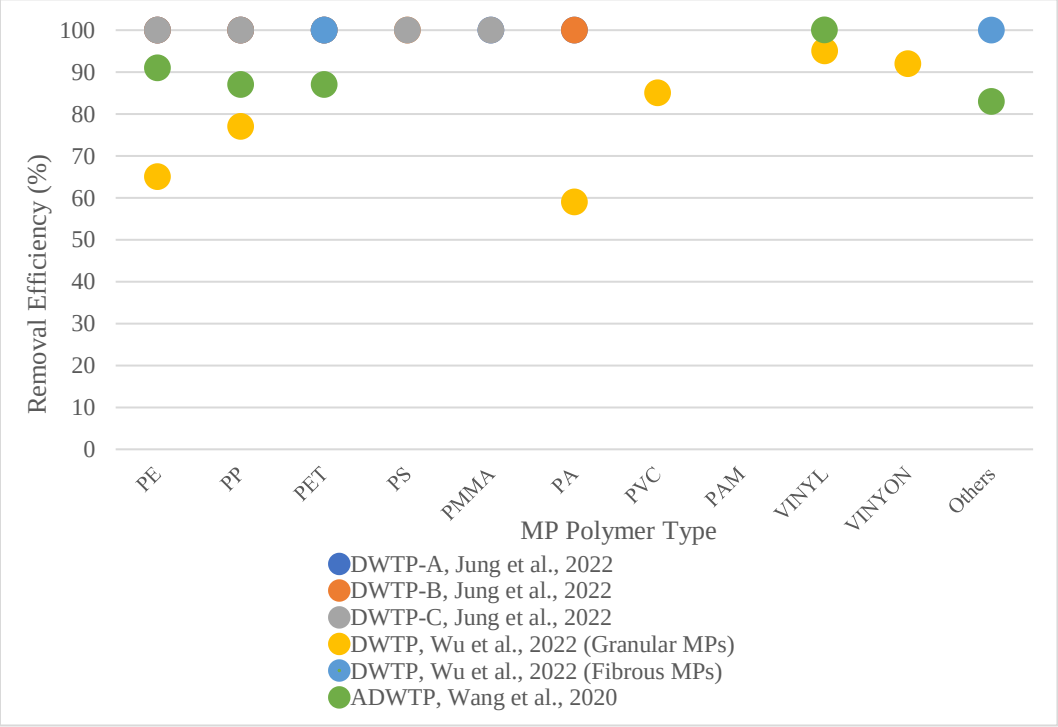


Figure 3.24 (a): Removal efficiency of MPs made from various polymer types by full-scale DWTPs

Though various polymer types are reported here, the data are very limited, with only a single value available for some polymer types. In this small dataset, almost all types of polymer are getting an overall good percentage of removal (>60 to 100%). All of these DWTPs had advanced treatment units along with their conventional ones.

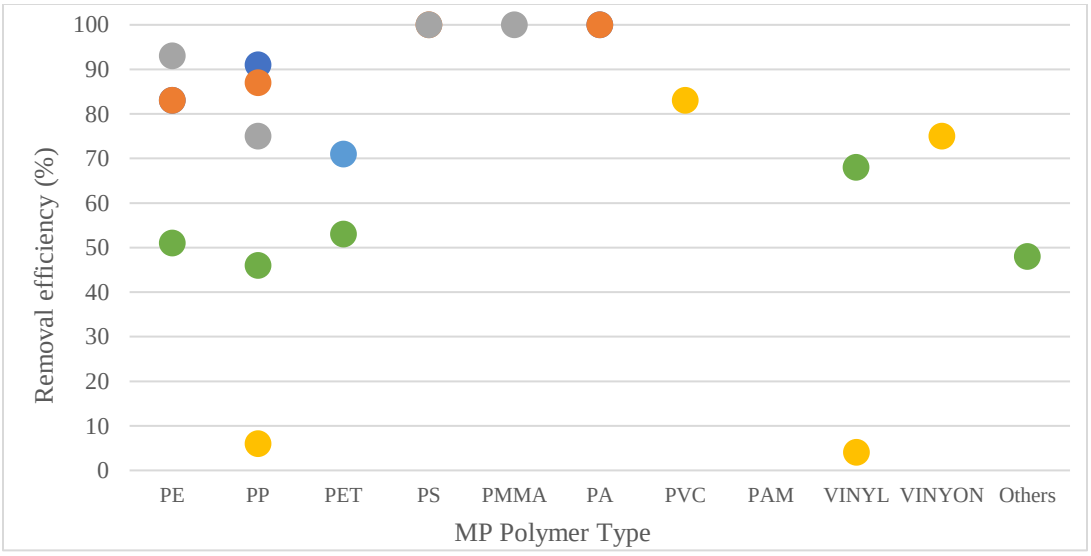


Figure 3.25 (b): Removal efficiency of MPs made from various polymer types by the CFS unit of different DWTPs

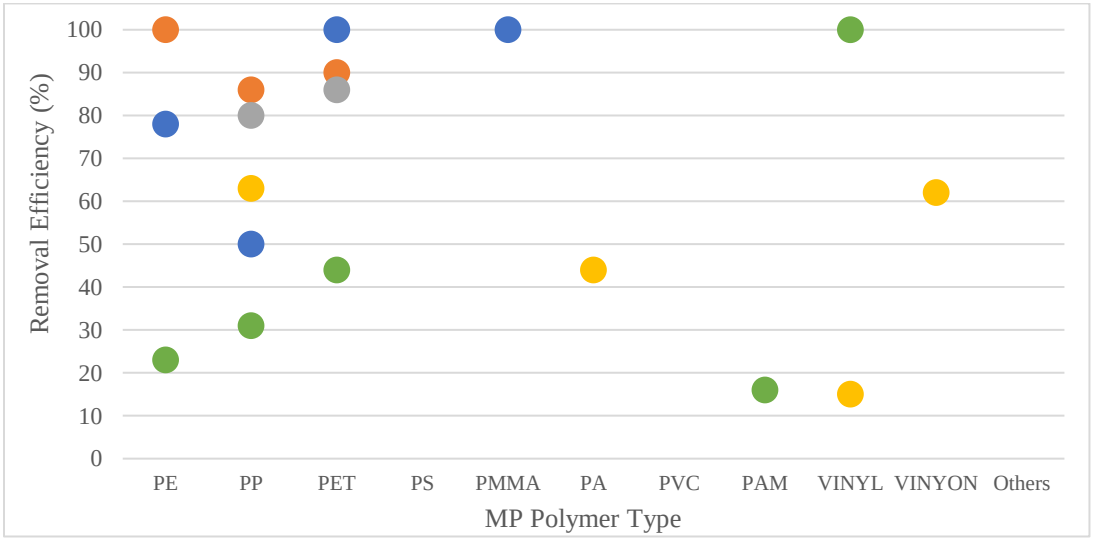


Figure 3.26 (c): Removal efficiency of MPs made from various polymer types by the filtration unit of different DWTPs

In Figures 3.16 (b) and (c), the data points are distributed over the range of 10 to 100%. The MP polymer types reported by Wang et al. (2020) had higher removal efficiency by CFS compared to the filtration process. The other data points are widely scattered, indicating significant variations in removal efficiencies across the surveyed DWTPs. PE, PP, and PET are among the more widely reported polymer types, and in some cases, they also received good removal by the DWTPs. The findings from this limited dataset suggest that the polymer type of MPs does not appear to have an influence on their removal. More investigation is needed to understand the reasons for the widely varying removal efficiencies for different polymer types by the full-scale DWTPs.

3.5 Conclusions

The ubiquity of MPs in drinking water sources has prompted many researchers around the world to investigate MPs removal by full-scale DWTPs. Despite being an emerging field of research, there exists the issue of limited data availability. In addition to that, variations in the reported MPs abundance, properties, sampling location, process, and detection techniques pose challenges, resulting in inconsistencies in the existing information. Inspired by this situation, this chapter has focused on synthesizing available datasets from these full-scale surveys to analyze the MPs abundance for different size ranges, shapes, and polymer types, as well as their removal in full-scale DWTPs.

- MPs abundance varied by a wide range both in the influent and effluent of the full-scale DWTPs. In spite of the huge variation in the reported size ranges, shapes, and polymer types, smaller-sized MP, specifically fibers and fragments shapes, along with PE, PP, and PET polymers were commonly observed both in the raw and treated water. Although each DWTP employed a unique combination of treatment processes, they all were able to remove MPs by a certain amount, with varying percentages among the surveys. Advanced DWTPs consistently achieved higher removal rates compared to the conventional ones irrespective of the characteristics of MPs. While CFS and filtration processes were not specifically optimized for MPs removal, they played an important role in the overall removal within the DWTPs.
- Because of the high abundance and potential toxic effects, the small-sized MPs (<100 µm) were prioritized, while establishing size ranges for data reporting in these surveys. MPs of all size ranges were found to be removed to some extent by these DWTPs, with better removal observed for larger-sized MPs in general. In case of both CFS and filtration, higher removal

rates were observed for higher size ranges in some surveys, while other surveys showed the opposite pattern. Attempts were made to identify any possible relation between MPs removal efficiency for these size ranges and their abundance in the influent or effluent of the respective treatment processes. No definitive relation could be established due to their wide variation in abundance and other properties.

- Although there was limited data on the removal of MPs in terms of different shapes and polymer types, all MP shapes and polymer types were removed in some measure by the DWTPs. The removal percentages of MPs varied widely, but both CFS and filtration processes contributed well to their removal.

MPs in the influent to DWTPs can differ in terms of their abundance, size, shape, polymer type, and any other properties, and these can vary depending on spatial and temporal factors. Though different researchers around the world are investigating MPs, inconsistency in sampling, detection and quantification methodologies is one of the factors contributing to the absence of uniform conclusions in this field of research. Besides, there is a lack of consistency in categorizing the size groups, shapes, and polymer types of MPs among the studies, which also leads to the deviation in results. This is one of the major problems in not being able to form any overall conclusion based on the available data from different research works. There is a severe lack of detailed information regarding the actual treatment processes and their operating conditions. The inclusion of parallel reporting of turbidity values would be valuable in providing a comparative context for understanding the MPs results within the framework of routinely monitored parameters at DWTPs. Another major barrier is the limited number of studies. More studies are necessary to investigate the removal of MPs by full-scale DWTPs to better understand the relation with their occurrence, particle size, shape, polymer type, etc. Moreover, no study could be found portraying the information about NPs at the full-scale level because of the difficulties in identifying and quantifying smaller-sized particles. It is necessary to make significant changes to this detection technique so that NPs can be easily investigated at the full-scale level. Thus, although the great variability in reported removals is undoubtedly due in part to site-specific performance differences, the measurement factors discussed above are certainly a major contributor. The broader significance of this great variability is that it creates substantial difficulties for the development of regulations.

Consistency will be the key solution to most of the problems. If consistency can be brought in terms of the identification of MNPs for different size groups, shapes, and polymer types, then it would help to establish a uniform dataset. That will facilitate comparison and trend analysis among the research findings. It is high time to establish standards for the detection of MNPs for their different properties including size ranges, shapes, and polymer types. Then, standardization has to be brought for studying the removal of MNPs at the full-scale level. Uniform sampling procedures and identification techniques can be the initial steps in terms of bringing consistency in this area of research. Besides, there is a need for improved reporting on the specifics of the treatment processes and their operating conditions. After that, to the extent that clear removal trends exist, they will be found using the approach taken in this chapter by observing the removal efficiency by different full-scale DWTPs for different characteristics of MNPs. Additionally, the concurrent reporting of turbidity removal would offer insights into the effectiveness of the processes in removing particles and allow for a comparative analysis in relation to the MNPs removal. Lastly, more studies are needed in this field to support the existing analysis and to better understand the MNPs removal of various abundance and different properties (size range, shape, polymer type) by overall DWTPs and also individually by conventional treatment units (especially CFS and granular media filtration/membranes, as the major particle removal steps) of DWTPs.

Chapter 4
Analysis of Data from Bench-scale Studies

4.1 Introduction

The growing concern about the presence of MNPs in surface water sources has promoted many researchers worldwide to investigate the abilities of various drinking water treatment processes for the removal of these pollutants. DWTPs have been identified as playing a considerable role in removing MPs of different characteristics from various drinking water sources. The conventional drinking water treatment techniques, CFS and filtration have also been found to perform a significant role in removing a certain percentage of MPs at the full-scale level. To optimize these treatment processes, researchers are increasingly turning to bench-scale studies to test and refine their methods under controlled laboratory conditions. Currently, bench-scale studies investigating the removal of MNPs outnumber the published full-scale surveys. At the bench-scale level, different studies are investigating various removal techniques to observe the removal percentages of MNPs of various categories. In this chapter, the focus will be on the prominent treatment techniques of CFS and filtration, which are conventionally used for particle removal and are incorporated into almost every full-scale DWTP. Despite the increasing number of published bench-scale studies investigating the removal of MNPs, the findings are still dispersed and a comprehensive analysis is required to bring them together. Though a couple of studies (e.g., Tang et al., 2022; Xue et al., 2022) have provided an overview of some of the existing information, a thorough analysis and interpretation of the results would indeed be valuable for advancing further research in this field. It is worth noting that the number of bench-scale studies looking into the removal of MPs is much higher than that of NPs.

The major purpose of this chapter is to analyze published information on the abundance and removal of MNPs by conventional treatment methods (CFS, filtration) of drinking water treatment systems at the bench-scale level. To attain that goal, the preliminary step is the selection of relevant bench-scale studies that have conducted experiments in this certain arena. Then the necessary information needs to be extracted from their extensive range of findings and categorized according to the need of this thesis. Subsequently, synthesis would be carried out based on the available data in order to observe the removal pattern of MNPs with different characteristics (size, shape, polymer type) by CFS and filtration

processes at the bench-scale level. Lastly, conclusions will be drawn by comparing the results obtained from analyzing the data of these bench-scale investigations.

4.2 Research Approach

4.2.1 Selection of Studies

The selection of the studies for this chapter has been centered on the removal of MPs or NPs either by CFS or filtration techniques at the bench-scale level. The keywords 'microplastics/nanoplastics removal from drinking water', 'microplastics/nanoplastics removal at bench-scale level', and 'microplastics/nanoplastics removal by coagulation-flocculation-sedimentation/filtration' have been used to search for relevant literature in google scholar. The selection process has been focused on peer-reviewed studies published between 2019 and 2022 that investigated the removal of MPs or NPs through CFS or filtration treatment techniques. No previous studies could not be found that specifically addressed this particular occurrence. The selected studies are listed in Table 4.1.

At the bench-scale level, CFS or filtration treatment techniques are carried out by different researchers to observe the removal of MPs or NPs of some specific properties. But in most cases, observing the removal of MNPs is just a small part of their investigation, they often explore other aspects besides that. These include the influence of different water quality properties, the mechanism of the treatment processes, causes of variations for different types of observations, and methods to improve the removal processes. So, the peer-reviewed bench-scale studies that have reported on the removal of MPs or NPs of a certain size, shape, polymer type, or other properties by CFS or filtration or both treatment processes as one of their findings have been selected for this chapter's analysis and are listed in Table 4.1.

4.2.2 Data Extraction, Analysis Process, and Their Limitations

The data collection procedure for the bench-scale studies followed the same procedure as described in Chapter 3 for the full-scale surveys. Some required data was obtained directly from the published articles or supplementary information. The necessary values from different figures were extracted using the same online application named 'WebPlotDigitizer' and the data extraction process is similar to the

procedure described in section 3.2.3. Subsequently, the data processing was performed using Microsoft Excel.

The limitations of the data extraction process for bench-scale studies are also similar to those of full-scale surveys as the data extraction is the same for both categories of investigations. This can be found in section 3.2.4 of Chapter 3.

In terms of the data analysis process, a part of it remains the same as well for both bench-scale studies and full-scale surveys. Microsoft Excel and Particle Graphing Tool have been used for the data analysis in this chapter, as described in section 3.2.5 of Chapter 3. The graphing tool has been used to generate graphs showing the removal efficiency of MNPs for different size ranges and Microsoft Excel has been used for other analytical tasks.

4.3 Sources of Information

This section will provide an overview of the selected studies for this chapter, with a primary focus on the removal efficiency of MPs or NPs using CFS and filtration treatment techniques at the bench-scale level. The outline of the selected studies is provided in Table 4.1, which lists the main objective together with the type of MNPs, treatment processes, and water quality parameters that are studied in those investigations. This table summarizes the number of polymer types, shapes, and sizes of MNPs used, as well as the number of coagulant and coagulant aid (if used) for the CFS process and the type of filter media for the filtration process employed in these studies. It also lists the name of the water properties analyzed to determine their relationship with MNPs removal. Lastly, it contains the main focus of these studies, which are related to the chapter's topic.

This table is divided into two main groups, where Table 4.1 (a) and (b) are prepared for MPs and NPs respectively. There are 17 and 10 bench-scale studies in total looking into the removal of MPs and NPs respectively either by CFS or filtration treatment techniques. Among them, 4 bench-scale studies investigated the removal of both MNPs. 16 out of 17 and 7 out of 10 studies examined the removal of MPs and NPs respectively by CFS treatment process. Then again, 3 of those 16 studies and an additional study, so 4 studies in total evaluated the removal of MPs by the filtration process. In the case of NPs, 2 from those 7 studies and 3 additional studies, so 5 studies in total evaluated the removal of NPs by the filtration process. The removal by CFS category is divided into 4 different sub-categories namely Al-

based, Fe-based, Al & Fe-based, and other coagulants for both MNPs. The majority of the studies for MNPs fall under the Al-based coagulant category, i.e., 10 and 4 studies for MPs and NPs respectively. The remaining categories have 1-3 studies each. Therefore, Al-based coagulants are the most commonly used in the bench-scale studies for both MNPs. Then removal by filtration category is again divided into 2 sub-categories namely media and membrane filtration. Both MNPs have 3 studies investigating media filtration, while only 1 and 2 studies looked into membrane filtration for MPs and NPs respectively. The sub-categories are further divided based on the source of water used in their experiments, with three sections for studies using synthetic water, surface water, or both. In the case of MPs, 11 and 4 studies used synthetic and surface water respectively, and 2 studies used both types for comparison. For NPs, 5, 2, and 2 studies used synthetic, surface, and both types of water respectively. So, it is apparent that most researchers use laboratory-based water for bench-scale experiments.

Some selected studies labeled particles less than 1 μm as MPs, but in this thesis, all plastics particles less than 1 μm are categorized as NPs. From the column of particle properties for both MNPs, 4 and 7 studies focused on only one size for MPs and NPs respectively, while the rest of them studied multiple sizes or size ranges. Besides, most studies examined one polymer type and shape of MNPs, with fewer looking into more than one. Then, from the column of water properties, not all of the selected studies reported on them. But a few analyzed source water properties and examined relationships between MNPs and water properties. Then from the column of the removal process studied, it was found that only 7 studies utilized coagulant aid with coagulants to improve the CFS process. Lastly, all of these selected studies had different objectives along with one that aligns with the focus of this chapter. So, those related to the focus of this chapter are included in the table.

In this table, numbers are used to designate each study and the same number is repeated every time the study is mentioned under different groups and categories. The numbering in Table 4.1 will be maintained throughout Chapter 4.

Table 4. 1 (a): Overview of the bench-scale studies investigating MPs removal by CFS and/or filtration

	No.	Reference	MP properties	Removal process studied	Water properties studied	Main focus of the study*
CFS (Al-based coagulant)						
(a) Synthetic water source	1	Zhang et al. (2022)	1 polymer type 1 shape 1 size	1 coagulant 1 coagulant aid	pH	>Effect of different dosages of coagulant and coagulant aid on MPs removal >Effect of particle size and pH on the MPs removal
	2	Zhang et al. (2021)	1 polymer type 1 shape 3 size ranges	1 coagulant 3 coagulant aids	pH, typical ions (Cl^-, SO_4^{2-}, HCO_3^-), dissolved organic matter	>Effect of different coagulant aids on MPs removal under different dosages of both coagulant and coagulant aids. >Mechanisms (effect of double layer compression, adsorption, and enmeshment by a precipitate) of coagulation and enhanced coagulation. >Impact of some influencing factors (particle size, pH, typical ions, and dissolved organic matter) for MPs removal by coagulation
	3	Shao et al. (2020)	1 polymer type 1 shape 1 size range	1 coagulant	N/A	>Influence of pre-oxidization on MP removal by CFS process
	4	Shahi et al. (2020)	1 polymer type 2 shapes 4 size ranges	1 coagulant 1 coagulant aid	N/A	>Effect of different doses of coagulant and coagulant aid on MPs removals in relation to their size, shape, and surface morphology
	5	Xia et al. (2020)	1 polymer type 1 shape 5 sizes	1 coagulant	pH	>Influence of surfactants on the coagulation removal of PS MPs. >Effect of pH, coagulant dosage, and MPs size on the removal by coagulation under specific conditions
	6	Skaf et al. (2020)	4 polymer types 2 shapes several sizes	1 coagulant	Turbidity	>Evaluation of MPs behavior during coagulation treatment optimized for kaolin under different alum concentrations >Effect of surfactants on MPs stabilization evidenced by zeta potential measurement under different pH
(b) Surface water source	7	C. Li et al. (2021)	1 polymer type 1 shape 1 size	1 coagulant	pH	>Effect of initial water pH, flocculation stirring intensity, flocculation time, sedimentation time, floc-breakage, and regrowth on MPs removal
	8	Xue et al. (2021)	1 polymer type shape 5 sizes	1 coagulant	pH, turbidity, dissolved organic carbon, LC-OCD fractions	>Effect of different MP sizes and water sources under different doses of coagulant for MPs removal >Change in surface charge and hydrophobicity for different-sized MPs with different properties of water.

	9	Lapointe et al. (2020)	3 polymer types 2 shapes 3 sizes	1 coagulant 1 coagulant aid	Turbidity	>Impact of coagulant & flocculant type and dosages on the removal of MPs. >Impact of weathering on MPs removal during CFS process
	10	Zhang et al. (2020)	2 polymer types 1 shape 4 different sizes	1 coagulant	N/A	>Effect of the CFS process (with or without the use of coagulant and coagulant aid) on MPs removal.
(a) + (b)	11	Bayarkhuu & Byun (2022)	1 polymer type 1 shape 2 sizes	1 coagulant	pH	>Optimization of the CFS process conditions by turbidity measurement for MPs removal
CFS (Al-based & Fe-based coagulant)						
(a)	12	Zhou et al. (2021)	2 polymer types 1 shape 3 size ranges	2 coagulants	pH, typical ions (Cl^-, SO_4^{2-}, CO_3^{2-})	>Effect of various dosages of two different coagulants on the removal of two distinct MP types
	13	Ma, Xue, Hu, et al. (2019)	1 polymer type 1 shape 4 size ranges	2 coagulants 2 coagulant aids	pH, turbidity, HA, ionic strength	>Effect of various dosages of coagulants under different conditions including with or without the addition of coagulant aids on MPs removal
(a) + (b)	14	Na et al. (2021)	1 polymer types 1 shape 4 sizes	2 coagulants	pH, typical water components	>Effect on MPs removal by two different types of coagulants for two types of water. >Effect of settling time, solution pH, and water components on the removal of 20 μm MPs.
CFS (Fe-based coagulant)						
(a)	15	Ma, Xue, Ding, et al. (2019)	1 polymer type 1 shape 4 size ranges	1 coagulant 2 coagulant aids	pH	>Effect of various dosages of the coagulant with or without the addition of coagulant aids on MP removal
CFS (Other coagulant)						
(a)	16	B. Li et al. (2022)	1 polymer type 1 size range	1 coagulant 1 coagulant aid	N/A	>Effect of using magnesium hydroxide combined with PAM on MPs removal >Effect of different types, dosages, and dosing time of PAM on coagulation performance for MPs removal
Filtration (Media filtration)						
(a)	3	Shao et al., 2020	1 polymer type Shape size	Granular media (Sand)	N/A	>Influence of pre-oxidization on MP removal by the filtration process, which was preceded by the CFS process

(b)	10	Zhang et al. (2020)	2 polymer types 1 shape 4 sizes	Granular media (Sand)	N/A	>Effect of filtration on MPs removal. >Impact of interactions between biofilms and MPs on removal efficiency
(a)+ (b)	14	Na et al., 2021	1 polymer types 1 shape 4 sizes	Granular media (Sand)	pH, typical water components	>Effect of the filtration process for MPs removal in DI water.
Filtration (Membrane filtration)						
(a)	17	J. Li et al., 2021	1 polymer type 1 shape 3 sizes	Ultrafiltration	N/A	>Effect of MP concentration and size on membrane fouling

Table 4.2 (b): Overview of the bench-scale studies investigating NPs removal by CFS and/or filtration

	No.	Study	NP properties	Process studied	Water properties studied	Main focus of the study*
CFS (Al-based coagulant)						
(a)	1	Zhang et al. (2022)	1 polymer type 1 shape 3 sizes	1 coagulant 1 coagulant aid	pH	>Effect of different dosages of coagulant and coagulant aid on NPs removal >Effect of particle size and pH on the NPs removal
	18	Lu et al. (2021)	1 polymer type 1 shape 1 size	1 coagulant	pH	>Removal of NP in pristine and weathered condition and change of zeta potential under different dosages of coagulant at different pH
(b)	10	Zhang et al. (2020)	1 polymer type 1 shape 1 size	1 coagulant	N/A	>Effect of CFS process (with or without the use of coagulant and coagulant aid) on NPs removal
(a)+ (b)	11	Bayarkhuu & Byun (2022)	1 polymer type 1 shape 1 size	3 coagulants (1 Al-based & 2 others)	turbidity	>Optimization of CFS process conditions by turbidity measurement for NPs removal
CFS (Al-based & Fe-based coagulant)						
(a)	19	Gong et al. (2022)	1 polymer type 1 shape 1 size	3 coagulants (2 Al-based & 1 Fe-based)	pH, turbidity	>Effect of different coagulant and coagulant dosages on the removal of the NP >Effect of pH, temperature, salinity, and inorganic colloid on the NP removal

CFS (Fe-based coagulant)						
(a)+ (b)	20	Hofman-Caris et al. (2022)	1 polymer type 1 shape 2 sizes	1 coagulant	N/A	>Removal of NPs by CFS under different water conditions
CFS (Other coagulant)						
(a)	21	Peydayesh et al. (2021)	1 polymer type 1 shape 1 size	1 bio-flocculant (natural)	pH, turbidity, TOC	>Effect of flocculant dosage and suspension pH on the NP removal under different conditions
Filtration (Media filtration)						
(a)	22	X. Li et al. (2023)	1 polymer type 1 shape 1 size	Granular media (Sand & sand-limestone)	N/A	>Effect of granular limestone amended sand filters for enhanced removal of NPs
(b)	20	Hofman-Caris et al. (2022)	1 polymer type 1 shape 2 sizes	Granular media (Sand)	N/A	>Removal of NPs by sand filtration under different water conditions
	10	Zhang et al. (2020)	1 polymer type 1 shape 1 size	Granular media (Sand)	N/A	>Effect of filtration on NPs removal
Filtration (Membrane filtration)						
(a)	17	J. Li et al. (2021)	1 polymer type 1 shape 1 size	Ultrafiltration	N/A	>Effect of NP concentration and size on membrane fouling
	23	Enfrin et al. (2020)	1 polymer type 1 shape 1 size range	Ultrafiltration	N/A	>Impact of NPs on ultrafiltration membrane performance and membrane fouling

**The focus of these studies which are directly related to the focus of this thesis work are only mentioned here. Additional findings of these studies are not included.*
MPs-microplastics, NPs-nanoplastics, CFS-coagulation-flocculation-sedimentation

4.4 Results and Discussion

4.4.1 Synthesis of MNPs Properties

The selected bench-scale studies are performed by different researchers all over the world to test the feasibility of a process in a controlled laboratory environment. As there is a wide range of characteristics available for MNPs, these studies used MPs or NPs of different properties for their experiments. In these experiments, they chose a specific type of MPs or NPs and spiked them in their source of water. So, the size ranges, shapes, polymer types, and other properties of MNPs are different among these selected studies under each category. The initial spiked concentrations of MNPs are also different in these studies. Table 4.2 has been prepared to show a summary of the different properties of MNPs that have been used throughout these studies. The table is divided into two sections, with Table 4.2 (a) and (b) describing the properties of MPs and NPs respectively. The numbers are maintained the same as they were for Table 4.1, and they are also subdivided into categories based on the removal process studied and the source of water as in Table 4.1.

Table 4.2 (a): List of the studies with different properties of MPs that have been used for the experiments

	No	Study	MP properties			Additional properties			Initial conc. (mg/L)	Analysis techniques for determining removal
			Polymer type	Shape	Size (µm)	Density (g/cm^3)	Surface property (mV)	Others		
CFS (Al-based coagulant)										
(a) Synthetic water source	1	Zhang et al. (2022)	PS	Sphere	1	1.05	-ve	Green fluorescent	N/A	IA
	2	Zhang et al. (2021)	PET	Fragments	400–500, 100–400, <100	1.38	-6.17	N/A	100	WM
	3	Shao et al. (2020)	PE	Sphere[a]	250-300	N/A	N/A	Oxidized	5000	WM
	4	Shahi et al. (2020)	PE	(73%) Spherical and (27%) elongated	10-30, 30-50, 50-70 & 70-100	0.91	-14.2	N/A	5	IA2
	5	Xia et al. (2020)	PS	Spheres	1, 2, 3, 4 & 5	N/A	-21.1	Fluorescent	10	IA
	6	Skaf et al. (2020)	N/A	Spheres	1-5	1.3	N/A	Fluorescent red	5	IA3
			PE	Fibers	5 & 15	0.96		N/A		
			Rayon		8.7 or 20.6	1.5				
			PES		17.5 to 50.6	1.38				
(b) Surface water source	7	C. Li et al. (2021)	PS	Sphere[a]	100	1.04–1.06	N/A	Pristine	10	VC
	8	Xue et al. (2021)	PS	Sphere	3, 6, 25, 45, 90	~1.05	-11.6 to -60.1	Fluoresbrite yellow-green carboxylated, refractive index of 1.6	0.003, 0.026, 2.054,4.447,24.294	CM using epifluorescence microscope
	9	La pointe et al. (2020)	PE	Spheres	15 & 140	N/A	N/A	Pristine & weathered	500 MPs/L	CM using stereomicroscope
			PS	Spheres	140					
			PES	Fibers	90					

	10	Zhang et al. (2020)	PS	Spheres[a]	1.2	~1.0	N/A	Fluorescent, hydrophobic	9500 particles/L	IA2
			PE		10–20, 45–53 & 106–125				7000, 2000 & 1800 particles/L	
(a)+ (b)	11	Bayarkhuu & Byun (2022)	PS	Sphere	1 & 10	N/A	-ve	N/A	0.01-30	WM
CFS (Al-based & Fe-based coagulant)										
(a)	12	Zhou et al. (2021)	PS	Fragments[a]	<500, 500-1000, 1000-5000	1.05	-15.77	N/A	500	WM
			PE			0.91	-14.55			
	13	Ma, Xue, Hu, et al. (2019)	PE	Fragments[a]	<500, 500-1000, 1000-2000 & 2000-5000	N/A	N/A	N/A	N/A	WM
(a)+ (b)	14	Na et al. (2021)	PS	Sphere[a]	10, 20, 45, 90	N/A	N/A	N/A	54	WM
CFS (Fe-based coagulant)										
(a)	15	Ma, Xue, Ding, et al. (2019)	PE	Fragments[a]	<500, 500-1000, 1000-2000 & 2000-5000	N/A	N/A	N/A	N/A	WM
CFS (Other coagulant)										
(a)	16	B. Li et al. (2022)	PE	Sphere[a]	270 ± 20	N/A	N/A	N/A	50	WM
Filtration (Media filtration)										
(a)	3	Shao et al. (2020)	PE	Sphere[a]	250-300	N/A	N/A	Oxidized	5000	WM
(b)	10	Zhang et al. (2020)	PS	Spheres[a]	1.2	~1.0	N/A	Fluorescent, hydrophobic	9500 particles/L	IA2
			PE		10–20, 45–53 & 106–125				7000, 2000 & 1800 particles/L	

(a)+ (b)	14	Na et al. (2021)	PS	Sphere[a]	10, 20, 45, 90	N/A	N/A	N/A	54	CM using Raman microscope
Filtration (Membrane filtration)										
(a)	17	J. Li et al., 2021	PS	Sphere[a]	1, 10, 18	~1.05	-0.2	N/A	0.01-1	IA

Table 4.2 (b): List of the studies with different properties of NPs that have been used for the experiments

	No	Study	Properties			Additional properties			Initial conc. (mg/L)	Analysis techniques for determining removal
			Polymer type	Shape	Size (µm)	Density (g/cm^3)	Surface property (mV)	Others		
CFS (Al-based coagulant)										
(a)	1	Zhang et al. (2022)	PS	Sphere	0.05, 0.1, 0.5	1.05	-ve	Green fluorescent	N/A	IA
	18	Lu et al. (2021)	PET	Sphere[a]	0.53	N/A	N/A	Pristine	50	WM
			WPET		0.22			Weathered		
(b)	10	Zhang et al. (2020)	PS	Sphere[a]	0.18	~1.0	N/A	Fluorescent, hydrophobic	9500 particles/L	IA2
(a)+(b)	11	Bayarkhuu & Byun (2022)	PS	Sphere	0.1	N/A	-ve	N/A	0.01-30	WM
CFS (Al-based & Fe-based coagulant)										
(a)	19	Gong et al. (2022)	PS-COOH	Sphere	0.05	1.05	-ve	Refractive index 1.59	50	TOC
CFS (Fe-based coagulant)										
(a)+(b)	20	Hofman-Caris et al. (2022)	PS	Sphere	0.05 & 0.2	N/A	Uncharged & negatively charged	Either plain or modified with a carboxyl group	10	IA
CFS (Other coagulant)										
(a)	21	Peydayesh et al. (2021)	PS	Sphere[a]	0.5	N/A	-ve	Carboxylated	10000	TM

Filtration (Media filtration)										
(a)	22	X. Li et al. (2023)	PS	Sphere[a]	0.1	N/A	-32.50±2.5	Fluorescent	5	IA
(b)	20	Hofman-Caris et al. (2022)	PS	Sphere	0.05 & 0.2	N/A	Uncharged & negatively charged	Either plain or modified with a carboxyl group	10	IA
	10	Zhang et al. (2020)	PS	Sphere[a]	0.18	~1.0	N/A	Fluorescent, hydrophobic	9500 particles/L	IA2
Filtration (Membrane filtration)										
(a)	17	J. Li et al. (2021)	PS	Sphere[a]	0.1	~1.05	N/A	N/A	0.01-1	IA
	23	Enfrin et al. (2020)	PE	Sphere[a]	0.013-0.69	N/A	-31.2 ± 0.7	N/A	10	IA

[a]The shape was inferred from the material and methods section/ literature as they provided the diameter of the particles
IA-MPs/NPs concentration was determined in the suspension through analysis by fluorescence spectrophotometer
IA2-Imaging was performed using a microscope and then image analysis was done by using software to quantify MPs/NPs on the membrane
IA3-Imaging was performed using a microscope and then images were analyzed visually using photographs to determine the amount of MPs/NPs
WM-Weighing method was used to calculate MPs/NPs removal by comparing the mass change on the membrane
VC-MPs/NPs concentration was determined in the suspension by visual counting under an optical microscope
CM-MPs/NPs removal was calculated by manual particle counting on the membrane using a microscope
TOC-Total organic carbon analyzer was used to determine the concentration of NPs
TM-Turbidity meter was used to determine the removal of NPs in terms of turbidity
MPs-microplastics, NPs-nanoplastics, PS-polystyrene, PET-polyethylene terephthalate, WPET-weathered polyethylene terephthalate, PE-polyethylene, PES-polyester

The properties of MNPs including polymer type, shape, and size were reported by all these selected bench-scale studies. In the case of polymer type, PS was mentioned to be used in 10 studies among the enlisted ones for MPs and in 7 studies for NPs. Then, PE was used in 9 studies for MPs and only in 1 study for NPs. So, the use of both PE and PS dominated in case of doing experiments with MPs. Other than these two types, PES was utilized twice, while PET and Rayon were found to be used only once for experiments with MPs, and PET was employed in only one of the studies for NPs. So, PE and PS - these two polymer types are found to be the most frequently used MNPs among these selected studies. PE can be considered as one of the representative polymer types found in source water, as almost all of the full-scale surveys reported the presence of PE in the influent of the DWTPs. Another important characteristic of MNPs is their shape. A sphere is the most common shape reported by all the selected studies for both MPs and NPs. All of the investigations with NPs reported the particle shape as spheres. Besides, 13 studies mentioned spheres as the shape of their MPs. Fragments and fibers were mentioned to be used 4 and 3 times respectively while experimenting with MPs, whereas they were the most prevalent MPs shape found in the source water with spheres being rarely found. Then comes the size of MNPs as one of the most important characteristics. In the case of MPs, different studies used different-sized MPs over the wide range of 5 mm to 1 µm. 7 studies defined the size in terms of some size ranges and others used distinctly sized particles. The majority of the full-scale surveys were able to detect MPs across the entire size range (5 mm-1µm), while some surveys could not detect MPs as small as 1 µm due to certain detection limitations. In bench-scale studies, NPs were found to be used in the size range of 0.013 to 0.53 µm and most of them used only one size of particles. The size of these particles in a given study is very different from the other studies. None of the full-scale surveys investigated the presence of NPs in either their raw or treated water.

Other than these three properties, the bench-scale studies were found to report on some additional properties of MNPs as well. They were, however, not consistently reported by all of them. Those properties are summarized under the additional properties category in Table 4.2. Ten out of 17 studies and 4 out of 10 studies mentioned the density of the particles used in their experiment for MPs and NPs respectively. The density of MPs range between 0.91 – 1.38 g/cm^3, but most MPs had density $\geq$ 1 g/cm^3. The density of NPs is about 1 g/cm^3 for all the selected studies which reported the particle density. Compared to the density of water which is 1 g/cm^3, some of the used MNPs were slightly lighter than the water and most of them were slightly heavier. In terms of surface properties, the zeta potential was mentioned in some of the studies. 8 out of 17 studies reported the zeta potential for MPs as negatively

charged. Then 7 out of 10 studies for NPs reported their surface to be negatively charged and one of them mentioned using uncharged particles at the same time. Although all of them did not specify the exact value of the charge, they reported their state of charge. Moreover, a few studies described some other initial conditions of the used MNPs, which are listed in the 'others' column of Table 4.2. The important one among them is the weathering condition of the particles, as weathering can alter the physicochemical properties of the MNPs impacting their performance during treatment processes. A couple of studies narrated whether the used MNPs were in pristine or weathered condition. Most of the experiments used commercially available pristine particles. Two bench-scale studies used particles both in pristine and weathered conditions to evaluate the difference in properties. Almost all the studies for MPs (14 out of 17) and NPs (9 out of 10), reported the initial concentration of MNPs which were spiked in the water samples before carrying out the experiments. The spiked concentrations also varied among these studies. Most of the studies reported their value in the low range starting from lower than 1 to about 50 mg/L, then there are some studies which used higher initial concentrations, i.e., 100, 500 and even 5000 mg/L was reported to be spiked by one study each. The initial concentrations were mostly reported in mg/L, however, two studies reported their initial concentration in particles per liter of water. The reported MPs abundance in source water from full-scale surveys, measured in particle number per liter, poses a challenge when comparing them with spiked concentrations used in these experimental studies. Particle counting can be challenging due to detection limitations, although it would enable direct comparisons with the values obtained from full-scale surveys. On the other hand, the mass concentration provides a direct measure of the total amount of the MPs present, but it can make it difficult to compare different types or sizes of MPs. Lastly, for determining the removal efficiency of MNPs by CFS and filtration processes, various analysis techniques were employed in the selected studies. These include the use of weighing methods, fluorescence spectrophotometer, different image analysis methods, and visual counting methods. 9 studies used a weighing method where the particles were weighed after filtering them on the membrane surface and drying them in the oven, which was followed by comparing the mass change. 6 studies used a fluorescence spectrophotometer for determining their concentration in the suspension and subsequently calculated the difference. 3 studies used image analysis where they generated images using different microscopes. Among them, 2 studies analyzed the image afterward using software to quantify the number of MPs/NPs and one study analyzed the pictures by visually quantifying the MPs/NPs. Then three studies determined the MPs/NPs

concentration by visually counting them on the membrane and one study enumerated them in the suspension under various microscopes. Moreover, one study used a total organic carbon analyzer, and one study used a turbidity meter to determine the removal of MPs/NPs in terms of other units. While the weighing method was commonly used in many studies, the presence of impurities in the water may introduce inaccuracies in the results, as the removal efficiency is calculated based solely on weight. Alternatively, fluorescence and other optical microscopic analysis methods can be considered more reliable and convenient for quantitatively measuring MNPs compared to visual counting methods. However, the use of TOC and turbidity measurement techniques as reliable indicators of MNPs will require further investigation and validation.

The information in Table 4.2 can help to understand the impact of MNPs characteristics, their concentrations, and analysis techniques on removal efficiency in the selected bench-scale studies. When comparing the selected MNPs from the bench-scale studies with MPs properties found in the source water, it is important to consider that full-scale surveys only reported the size, shape, and polymer types of MPs present. The selected MNPs in bench-scale studies may represent one of the prevalent polymer types found in the source water. The shape of the MNPs studied may not be representative of the overall distribution of shapes observed in the source water, and the different sizes of MNPs investigated may only represent a fraction of the total range of MPs present in the source water. Besides, as different sizes, shapes, and polymer types of MNPs can interact differently with coagulants, flocculants, or filtration media, they can affect their removal efficiency. The density of particles can affect their settling velocity, which in turn can affect their removal in sedimentation or filtration processes. Surface properties can affect the interactions between the particles and the treatment processes, which can influence the removal efficiency. Overall, various characteristics of MNPs can contribute differently during the removal process by CFS and filtration. In addition, the initial spiked concentration of MNPs can also affect the derived results in bench-scale studies. Higher initial concentrations of MNPs can lead to saturation of the removal system, which can result in lower removal efficiencies. On the other hand, higher or lower initial concentrations may not accurately represent real-world conditions and may not provide reliable information on the performance of the removal system. Lastly, it is important to consider the different analysis processes used in determining removal efficiency as this can affect the accuracy and reliability of the results obtained. Comparing and understanding the differences in results obtained from different techniques can help to improve the accuracy and reliability of the data, which

can lead to better insights and conclusions. In summary, all of this information can facilitate understanding the removal pattern of MNPs by CFS and filtration processes.

4.4.2 Removal of MNPs by CFS Process

The CFS process is the most commonly used method for the removal of colloidal, suspended, and dissolved organic matter from surface water. In the coagulation process, chemical coagulants and sometimes coagulant aids are added to condition the particulate matter for subsequent processing by flocculation. Different types of coagulants such as Al-based, Fe-based, or other varieties are added to the water being treated and they rapidly hydrolyze to form insoluble precipitates. These precipitates and soluble complexes destabilize particles by adsorbing to their surfaces, neutralizing their charges, and forming a bridge between them. Flocculation refers to the agglomeration of those destabilized particles and precipitation products formed by the addition of those coagulants, resulting in the formation of larger particles referred to as flocs. Then the aggregated floc can be removed by settling them from water through subsequent sedimentation. However, since particles in water can vary in size, shape, type, distribution, and concentration, these properties and their interactions with the water and coagulant can impact the efficiency of the CFS process for removing them.

CFS is the most widely tested conventional treatment mechanism for the removal of MNPs. MPs removal via the CFS process has received more attention in the last few years than the removal of NPs. As mentioned earlier, 16 and 7 bench-scale studies were found to report on the removal of MPs and NPs respectively by the CFS process, whereas 3 studies were looking into the removal of both MNPs. Almost all of these selected studies provided some initial information on the bench-scale coagulation process, including the type and dosage of coagulant, jar test conditions, and the use of coagulant aids. Optimal conditions were determined through their experiments involving MNPs with different characteristics, various dosages of different coagulants, coagulant aids, and jar test conditions. These parameters may affect the removal of MNPs to a certain extent. Table 4.3 provides an overview of the bench-scale coagulation conditions investigated and the maximum MNPs removal obtained under optimal conditions. The table is divided into two parts: MPs (4.3 (a)) and NPs (4.3 (b)).

Table 4.3 (a): Overview of coagulant conditions and results of published bench-scale studies for MPs

	No.	Study	Coagulant [name and dosage (mg/L)]	Jar test conditions (mixing speeds and settling times)	Coagulant aid [name & dosage (mg/L)]	Removal efficiency at optimal condition (Particle property-Dosages-Removal)
Al-based coagulant						
(a) Synthetic water source	1	Zhang et al. (2022)	PACl (100, 200, 400, 600, 800, 1000)	300 rpm for 1 min 100 rm for 14 min 30 min for settling	PAM (1, 8, 20, 40, 80)	1 µm - PACl 400 mg/L - 89.3 ± 1.1% 1 µm – PACl 400 mg/L & PAM 20 mg/L - 98.5% (pH=8)
	2	Zhang et al. (2021)	PACl (20, 50, 100, 200)	500 rpm for 1 min 100 rpm for 15 min 30 min for settling	Anionic PAM, SA, and ASA (5, 10, 20, 50, 100)	1-500 µm – >> PACl 20 mg/L - 35.5% PACl 20 mg/L & PAM 5 mg/L - 44.47% PACl 20 mg/L & PAM 100 mg/L - 79.35% PACl 200 mg/L & PAM 100 mg/l - 91.45% PACl 20 mg/L & SA 5 mg/L - 45.52% PACl 20 mg/L & SA 100 mg/L - 69.9%, PACl 200 mg/L & SA 100 mg/l - 73.35% PACl 20 mg/L & ASA 5 mg/L - 54.70% PACl 20 mg/L & ASA 100 mg/L - 69.87% PACl 200 mg/L & ASA 100 mg/l - 77.60%
	3	Shao et al. (2020)	PACl (30)	300 rpm for 1 min 100 rpm for 14 min 30 min for settling	N/A	250-300 µm ->> (without oxidization)– PACl 30 mg/L - 15.8 ± 0.9% (oxidized with $KMnO_4$)– PACl 30 mg/L - 16.4 ± 1.9% (oxidized with NaClO)– PACl 30 mg/L - 12.5 ± 3.5% (oxidized with O_3)– PACl 30 mg/L - 18.2 ± 2.2% (pH=7)
	4	Shahi et al. (2020)	Alum (10, 20, 30, 40, 50)	200 rpm for 1 min 50 rpm for 10 min 30 min for settling	PC Sand (300, 400, 500)	10-100 µm ->> Alum 30 mg/L - 70.7% Alum 20 mg/L & PC sand 500 mg/L – 92.7% (pH=7)
	5	Xia et al. (2020)*	$AlCl_3 \cdot 6H_2O$ (100, 200, 300, 400, 500)	150 rpm for 1 min 30 min for settling	N/A	1-5 µm - $AlCl_3 \cdot 6H_2O$ 250 mg/L - 75% (pH=7)
	6	Skaf et al. (2020)*	$Al_2(SO_4)_3 \cdot 18H_2O$ (0-40 Al) (185-988)	100 rpm for 1 min 30 rpm 30 minute Settling for 120 min	N/A	5-50 µm - 5 mg Al/L (185 mg/L) - 97% NTU removal

(b) Surface water source	7	C. Li et al. (2021)	$Al_2(SO_4)_3 \cdot 18H_2O$ (3.4 Al) (283.5)	300 rpm for 1 min 50, 100, 150, 200, 250 rpm for 7 min 30 min for settling	N/A	100 µm - $Al_2(SO_4)_3 \cdot 18H_2O$ at 3.4 mg Al/L (283.5 mg/L) - 98.9 ± 0.94% (pH=5)
	8	Xue et al. (2021)	$Al_2(SO_4)_3$ – (48% liquid Alum) (10, 20, 30, 40, 50)	122 ± 1 rpm for 20 ± 1 s 30.5 ± 0.5 rpm for 20 min 15 min for settling	N/A	≤25 µm – Alum 30 mg/L - > 80% 45 & 90 µm – Alum 30 mg/L – < 80% (pH=8)
	9	Lapointe et al. (2020)	Alum (0.45-3.64 Al) (11.2-90.4)	600 rpm for 2 min 300 rpm for 4 min 1 min for settling	PAM (0.05-0.30)	Alum 2.73 mg Al/L (67.6 mg/L) & PAM 0.3 mg/L –>> 90 µm PES fibers (Pristine) - 99%, 140 µm PE (Pristine) - 82% 140 µm PE (Weathered) - 99% 15 µm PE (Pristine) - 89% 140 µm PS (Pristine) - 84% (pH=7)
	10	Zhang et al. (2020)	$Al_2(SO_4)_3$ (20)	200 rpm for 1 min 70 rpm for 2 min 15 min for settling	PolyDADMAC (0.5)	1.2, 10-20, 45-53, 106-125 µm - $Al_2(SO_4)_3$ 20 mg/L - <2% 45-54 µm - $Al_2(SO_4)_3$ 20 mg/L & PolyDADMAC 0.5 mg/L - 13.6 ± 6.8% 1.2, 10-20, 45- 106-125 µm ->> $Al_2(SO_4)_3$ 20 mg/L & PolyDADMAC 0.5 mg/L - <2%
(a) + (b)	11	Bayarkhuu & Byun (2022)	PACl (0.5 for 1 µm & 9 for 10 µm) PACl (15 for both 1 & 10 µm)	300 rpm for 1 min 100 rpm for 14 min 120 min for settling	N/A	(a) 1 µm PS – PACl 0.5 mg/L - 10% 10 µm PS – PACl 9 mg/L - 60% (b) 1 µm – PACl 15 mg/L - 80% 10 µm – PACl 15 mg/L - 90%
Al-based & Fe-based coagulant						
(a)	12	Zhou et al. (2021)	PACl and $FeCl_3$ (30, 60, 90, 120, 150, 180)	500 (PACl) or 400 ($FeCl_3$) rpm for 1 min 100 rpm for 15 min 30 min for settling	N/A	1-5000 µm PS - PACl 90 mg/L - 77.83% 1-5000 µm PE - PACl 90 mg/L - 29.70% 1-5000 µm PS - $FeCl_3$ 90 mg/L - 63% 1-5000 µm PE - $FeCl_3$ 90 mg/L - 17%

	13	Ma, Xue, Hu, et al. (2019)	$AlCl_3 \cdot 6H_2O$ (0.1-405 Al) (0.9-3600) & $FeCl_3 \cdot 6H_2O$ (0.1-278 Fe) (0.5-1347)	300 rpm for 1 min 100 rpm for 14 min 30 min for settling	Cationic and anionic PAM (0-15)	In general, Al-based coagulant performed better than Fe-based coagulant. At 13.5 mg/L Al, < 500 µm - 8.28% ± 1.06% 500 -1000 µm - 2.71% ± 0.84% 1000-2000 µm - 1.68% ± 0.63% 2000-5000 µm - 1.02% ± 0.37% At low dosages (13.5 mg Al/L), both cationic and anionic PAM had little influence. At higher dosages (135 mg Al/L), anionic PAM showed better removal efficiency, especially for smaller particles than cationic PAM. (pH=7)
(a) + (b)	14	Na et al. (2021)	$AlCl_3.6H_2O$ and $FeCl_3.6H_2O$ (10, 20, 30, 40, 50)	160 rpm for 2 min 60 rpm for 30 min 1 h for settling	N/A	Al-based coagulant showed better performance compared to Fe-based coagulant. At 10 mg/L dosage, 10 µm - 33.0-41.1% 20, 45, and 90 µm - 77.4-95.3% (pH=6)
Fe-based coagulant						
(a)	15	Ma, Xue, Ding, et al. (2019)	$FeCl_3 \cdot 6H_2O$ (0.1-278 Fe) (0.5-1347)	300 rpm for 1 min 100 rpm for 14 min 30 min for settling	Cationic and anionic PAM (0-15)	1-5000 µm ->> Low removal efficiency (below 15%) for even high dosages of $FeCl_3.6H_2O$. At low dosages (11.11 mg Fe/L), both cationic and anionic PAM had little influence. At higher dosages (111.12 mg Fe/L), anionic PAM showed better removal efficiency, especially for smaller particles than cationic PAM. (pH=7)
Other coagulant						
(a)	16	B. Li et al. (2022)	$MgCl_2.6H_2O$ (40)	300 rpm for 2 min 60 rpm for 5 min 5 min for settling	PAM (5)	270±20 µm - $MgCl_2.6H_2O$ 40 mg/L->> Anionic PAM - 84.9 ± 3% Cationic PAM - 54.6 ± 2% Non-ionic PAM - 77.6 ± 1.7% Atmospheric PAM - 62.9 ± 1.75%

**Surfactants were used*

PACl-poly-aluminum chloride, PAM-polyacrylamide, SA-sodium alginate, ASA-activated silicic acid, PC-polyamine-coated, PolyDADMAC-Diallyldimethylammonium Chloride, rpm-revolutions per minute, PE-polyethylene, PS-polystyrene, PET-polyethylene terephthalate, PES-polyester

Table 4.3 (b): Overview of coagulant conditions and results of published bench-scale studies for NPs

	No.	Study	Coagulant [name and dosage (mg/L)]	Jar test conditions (mixing speeds and settling times)	Coagulant aid [name & dosage (mg/L)]	Removal efficiency at optimal condition (Particle property-Dosages-Removal)
Al-based coagulant						
(a)	1	Zhang et al. (2022)	PACl (100, 200, 400, 600, 800, 1000)	300 rpm for 1 min 100 rm for 14 min 30 min for settling	PAM (1, 8, 20, 40, 80)	0.05 µm - PACl 400 mg/L - 85.6 ± 0.2% 0.1 µm - PACl 400 mg/L – 86.3 ± 0.2% 0.5 µm - PACl 400 mg/L - 91.0 ± 0.7% 0.05, 0.1, 0.5 - PACl 400 mg/L & PAM 20 mg/L - 98.5% (pH=8)
	18	Lu et al. (2021)	Al_{13} (0.27, 0.81, 1.35, 0.10, 2.7, 5.4)	200 rpm for 90 s 40 rpm for 10 min 30 min for settling	N/A	0.22 & 0.53 µm PET – 0.27 mg/L - ~100% 0.22 & 0.53 µm WPET – 0.27 mg/L - ~88% (at pH 6,7 & 8)
(b)	10	Zhang et al. (2020)	$Al_2(SO_4)_3$ (20)	200 rpm for 1 min 70 rpm for 2 min 15 min for settling	PolyDADMAC (0.5)	No significant removal was observed
(a) + (b)	11	Bayarkhuu & Byun (2022)	PACl, IL(1) & IL(2) (0-18)	300 rpm for 1 min 100 rpm for 14 min 120 min for settling	N/A	(a) 0.1 µm – PACl 3 mg/L - 78.3%, 0.1 µm – IL(2) 6 mg/L - 71.3%, 0.1 µm - PACl 3 mg/L & IL(2) 6 mg/L - 75.3% IL(1) was not effective due to the low coagulating power.
			PACl, IL(1) & IL(2) (0-50)	300 rpm for 1 min 100 rpm for 14 min 120 min for settling	N/A	(b) 0.1 µm – PACl 15 mg/L - 78.9%, 0.1 µm – IL(2) 12 mg/L – 72.2%, 0.1 µm – PACl 15 mg/L & IL(2) 12 mg/L – 87.7% IL(1) was not effective due to the low coagulating power.
Al-based & Fe-based coagulant						
(a)	19	Gong et al. (2022)	$AlCl_3$, $FeCl_3$ & PACl (2.5, 5, 10, 20, 40)	70 rpm for 1 min 180 rpm for 30s 40 rpm for 15 min 30 rpm for 15 min 20 rpm for 15 min 15 rpm for 25 min 30 min for settling	N/A	0.05 µm - $AlCl_3$ 10 mg/L - 96.6% 0.05 µm - $FeCl_3$ 10 mg/L – 95.8% 0.05 µm – PACl 5 mg/L - ~ 98% (pH=6.5)

Fe-based coagulant						
(a) + (b)	20	Hofman-Caris et al. (2022)	$FeCl_3$ (12 and 18)	400 rpm for 10 s 70 rpm for 15 min 30 min for settling	N/A	0.2 µm – $FeCl_3$ 12 & 18 mg/L - 79-86% 0.05 µm - $FeCl_3$ 12 & 18 mg/L - 6-47%
Other coagulant						
(a)	21	Peydayesh et al. (2021)	Flocculant - lysozyme amyloid fibrils (0.1 - 62)	300 rpm for 5 min 50 rpm for 15 min 30 min for settling	N/A	0.5 µm - 9.4 to 20 mg/L - 98.2% turbidity reduction from 51 x 10^3 to 914 NTU (at pH 7)

PACl-poly-aluminum chloride, PAM-polyacrylamide, IL(1)-1-Methyl-3-propylimidazolium chloride, IL(2)-1-decyl-3-methylimidazolium chloride, PolyDADMAC-Diallyldimethylammonium Chloride, rpm-revolutions per minute, PET-polyethylene terephthalate, WPET-weathered polyethylene terephthalate

In Table 4.3 (a), out of the 11 studies that solely used Al-based coagulants for their experiments on MPs, 5 of them employed polyaluminum or aluminum chloride, and the rest were found to use aluminum sulphate. Among them, 5 utilized coagulant aid to enhance their coagulation. A couple of studies experimented with very high dosages of the coagulant, such as Zhang et al. (2022) and Skaf et al. (2020), starting from 100 mg/L up to 1000 mg/L. Zhang et al. (2022) received a very high removal efficiency of about 90% for 1 µm sized MPs at 400 mg/L and Skaf et al. (2020) received about 97% removal in terms of turbidity reduction for 5-50 µm sized MPs at about 200 mg/L dosage of the coagulant. The additional optimum dosage of PAM increased the removal percentage by about 8-9% in Zhang et al. (2020). Xia et al., (2020), Zhang et al. (2021), and C. Li et al. (2021) also tested with very high dosages up to 500, 200, and 283.5 mg/L respectively. Xia et al. (2020) received about 75% removal for 1-5 µm sized MPs at 250 mg/L, while C. Li et al. (2021) received almost complete removal for 100 µm sized MPs. Here larger sized MPs got better removal at a similar coagulant dosage. Zhang et al. (2021) received comparatively lower removal (below 40%) for a wide range of MPs (1-500 µm). Then they assessed the removal performance with three different coagulant aids, where removal performance improved with increasing dosages of the coagulant aids. They achieved over 90% removal for the highest dosage of PACl and PAM and the other two coagulant aids brought above 70% removal at their highest dosage. These high ranges of coagulant dosages used in the mentioned studies were significantly higher than the typical dosages employed in full-scale DWTPs. On the other hand, Bayarkhuu & Byun (2022) experimented with very low dosages of PACl, i.e., 0.5 to 15 mg/L, where they used comparatively lower dosages for testing in synthetic water and higher dosages for surface water. In synthetic water, they reported only 10% removal for 1 µm and 60% removal for 10 µm. In surface water, they found 80-90% removal for both 1 and 10 µm. Besides, all other studies utilized coagulant dosages within the range of 10-50 mg/L, with one study (Lapointe et al. (2020)) going up to about 90 mg/L. Shao et al. (2020) received very low removal (about 12 to 20%) for 250-300 µm sized MPs at 30 mg/L of dosage. But at the same dosage, Shahi et al. (2020) received about 70% removal for 10-100 µm sized MPs, which was also improved to above 90% by the use of PC sand as a coagulant aid. At the same dosage, Xue et al. (2021) received more than 80% removal for MPs>25 µm and less than 80% removal for <25 µm. Zhang et al. (2020) also received very low removal (below 2%) at 20 mg/L dosage of the coagulant for 1 to 125 µm sized MPs. The addition of coagulant aid (PolyDADMAC) did not improve the removal efficiency, except for 45-53 µm, they were removed by about 14%, which is still very low compared to others. Three other studies compared the performance

of Al-based and Fe-based coagulants, with one of them also using a coagulant aid to enhance coagulation. Based on their experiments, all studies concluded that Al-based coagulant performed better than Fe-based coagulant under the conditions investigated. Na et al. (2021) used comparatively lower coagulant dosages ranging from 10-50 mg/L and achieved removal rates of about 77-95% removal for 20, 45, and 90 µm sized particles and lower removal (33-41%) for 10 µm sized MPs. Zhou et al. (2021) used higher coagulant dosages ranging from 30-180 mg/L and achieved a removal rate of 77% for all PS MPs at an optimal dosage of 90 mg/L, with low removal for PE MPs. Ma, Xue, Hu, et al. (2019) tested a broad range of coagulant dosages and found that the removal efficiency remained stable after 298 mg/L of $AlCl_3.6H_2O$ but achieved very low removal rates for all sized MPs. The addition of cationic and anionic PAM as coagulant aid had little influence on the removal efficiency. Only one study used a Fe-based coagulant with very high dosages but achieved very low removal (<15%) for all sized MPs. Additionally, they used coagulant aid, but that did not improve the removal efficiency significantly. Moreover, there is only one study that tested a different coagulant named magnesium chloride with various types of PAM as a coagulant aid. They received above 50% to about 85% removal for 270 µm sized MPs with four different kinds of PAM along with the coagulant.

Of the studies that examined the CFS process for removing of NPs, enlisted in Table 4.3 (b), 5 of them used Al-based coagulant exclusively, while the remaining sub-categories had only one study each. Except for Zhang et al. (2020), the others reported good removal efficiency (>70%) at both low and high dosages of the coagulants. Zhang et al. (2022) received almost complete removal with the addition of coagulant aid. Gong et al. (2022) tested 3 coagulants including 2 Al-based and 1 Fe-based ones and obtained above 90% removal for 0.05 µm sized particles in each case. Hofman-Caris et al. (2022) received lower removal for 0.05 µm sized NPs than 0.2 µm. Only one study utilized a different coagulant: Peydayesh et al. (2021) examined the potential of a natural bio-flocculant named Iysozyme amyloid fibrils to enhance the performance of CFS process and received about 98% reduction in terms of turbidity.

In terms of the jar test conditions of the CFS process, they varied among the studies. In general, most of these jar tests used a faster initial mixing speed to simulate the coagulation condition and then a slower speed to simulate floc formation, which was followed by varying lengths of settling times. Sedimentation time varied from 15 minutes to 2 hours, and 30 minutes is the most frequent one among

these studies. In these jar tests, small-scale simulations of the CFS processes are carried out under controlled conditions. So, mixing speed, mixing time, pH of the water, coagulant and flocculant dosages are required to be controlled to optimize the treatment process. Because of the short settling distance, sedimentation times may overestimate the removals that would be achieved at full scale.

Particle size is one of the crucial factors to consider during the removal of MNPs by the CFS process. Larger particles or particles forming heavy flocs settle faster than smaller and lighter ones during sedimentation, which makes their removal easier using the process. It is also an important consideration to find the optimal dosage of the coagulant, i.e., smaller-sized particles may require a higher dosage of the coagulant to be effectively removed. Mixing conditions during the process can also impact particle size. For instance, aggressive mixing can break larger-sized particles into smaller ones while insufficient mixing can cause inadequate agglomeration of particles into flocs. Therefore, in order to observe the removal of MNPs for different sizes, Figures 4.1, 4.2, and 4.3 have been plotted. Figures 4.1 and 4.2 represent the removal of different-sized MPs irrespective of other properties (including shapes, polymer types, concentrations, and surface properties) by various optimized dosages of Al-based and Fe-based coagulants respectively. Figure 4.1 is divided into (a) and (b) to show the results of the MP removal, where experiments were conducted using synthetic water and surface water respectively. Figure 4.2 could not be divided as only one bench-scale study used a surface water sample while coagulating with Fe-based coagulant. Figure 4.3 is drawn to show the removal of different-sized NPs by using various optimized dosages of different coagulants irrespective of other properties of NPs. The plot for NPs was not separated based on coagulant type as each type of coagulant has limited data. Moreover, to enhance visual clarity, Figure 4.1 (a) is further partitioned into two distinct plots, namely (i) and (ii), to effectively highlight the observed trends in the reported values. On the other hand, Figures 4.1 (b), 4.2, and 4.3 contain fewer data points, allowing the visual trends to be adequately displayed within a single graph, eliminating the need for separation. In all these figures, the x-axis represents the particle size range in a logarithmic scale and the y-axis represents the removal efficiency in percentage. Different colors have been used to identify different coagulation conditions in these studies. The horizontal bar in different colors represents the size ranges considered in those studies and the markers represent the mid value of the size ranges and the definite value for individual-sized particles. Additionally, to facilitate the depiction of the trend of MNPs removal, similar graphs have been generated employing dotted trend lines connected by the markers (see Figures B1-B4, Appendix B).

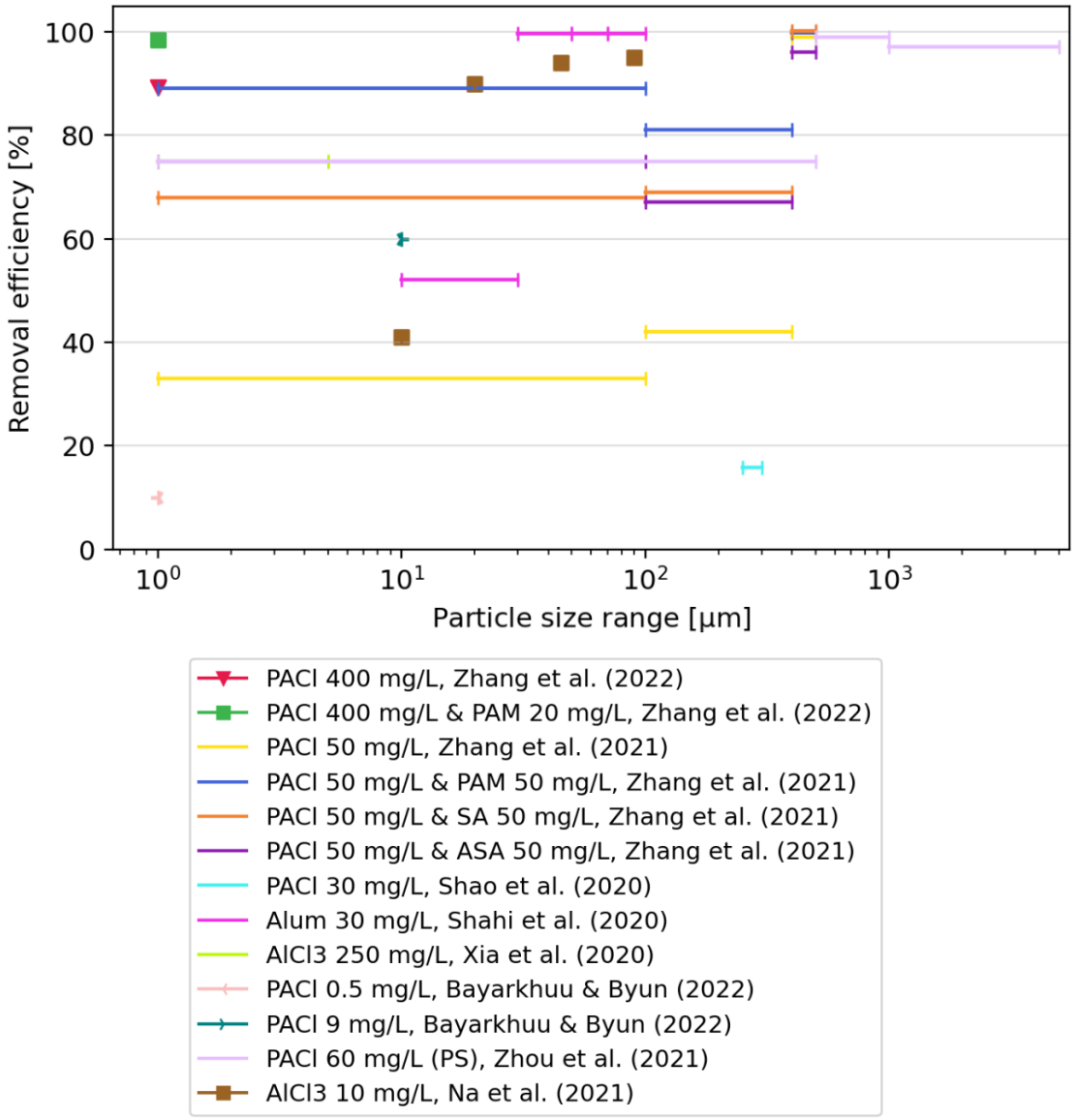


Figure 4.1 (a) (i): Removal efficiency of different-sized MPs by CFS using optimized dosages of Al-based coagulant in synthetic water irrespective of MPs polymer types, shape, and other properties (upward trend)

(PACl-poly-aluminum chloride, PAM-polyacrylamide, SA-sodium alginate, ASA-activated silicic acid, PS-polystyrene)

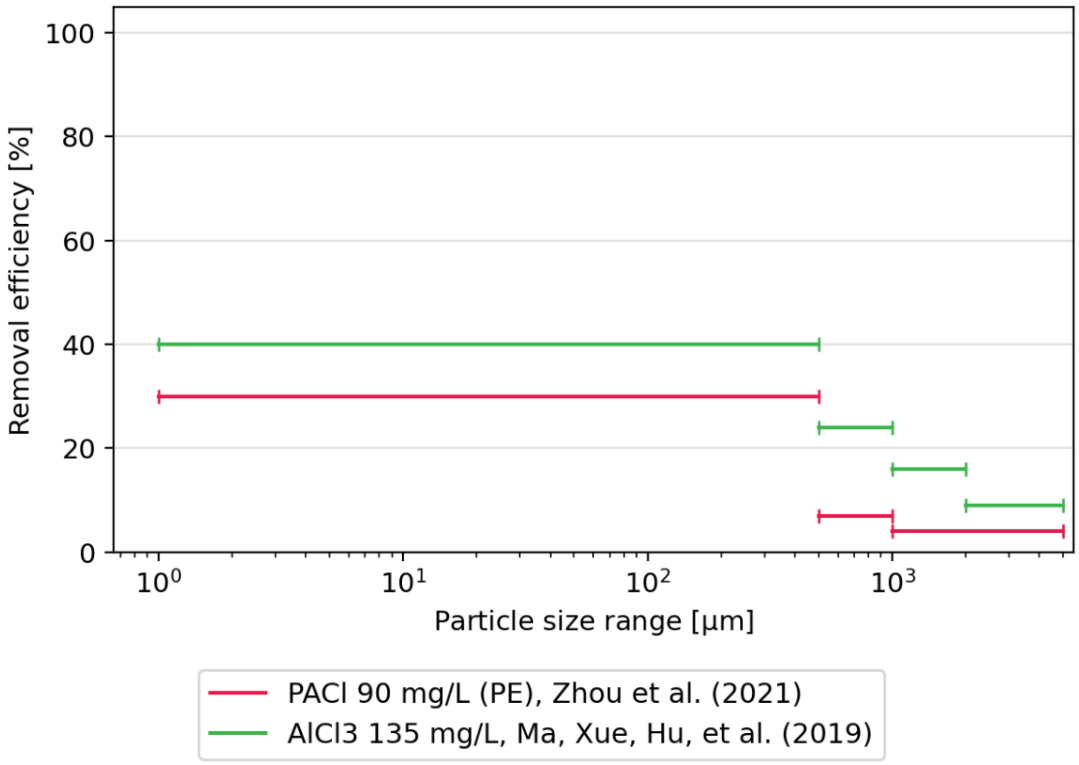


Figure 4.2 (a) (ii): Removal efficiency of different-sized MPs by CFS using optimized dosages of Al-based coagulant in synthetic water irrespective of MPs polymer types, shape, and other properties (downward trend)
(PACl-poly-aluminum chloride, PE-polyethylene)

In Figure 4.1 (a) (i), the values of removal efficiency for different-sized MPs are scattered across the entire range. Several studies, such as Zhou et al. (2021) for PS MPs, Zhang et al. (2021), Shahi et al. (2020), Bayarkhuu & Byun (2022), and Na et al. (2021) have indicated that there is a positive correlation between the size of the MPs and their removal efficiency, meaning that larger sized MPs tend to be removed more effectively. However, in Figure 4.1 (a) (ii), Zhou et al. (2021) for PE MPs and Ma, Xue, Hu, et al. (2019) (where experiments were also done for PE MPs) reported a different trend, showing lower removal for higher size ranges of MPs. In this case, the highest removal is about 40% for even smaller-sized particles. Zhou et al. (2021) identified the higher density of PS MPs as the main reason for receiving higher removal of PS MPs than PE MPs.

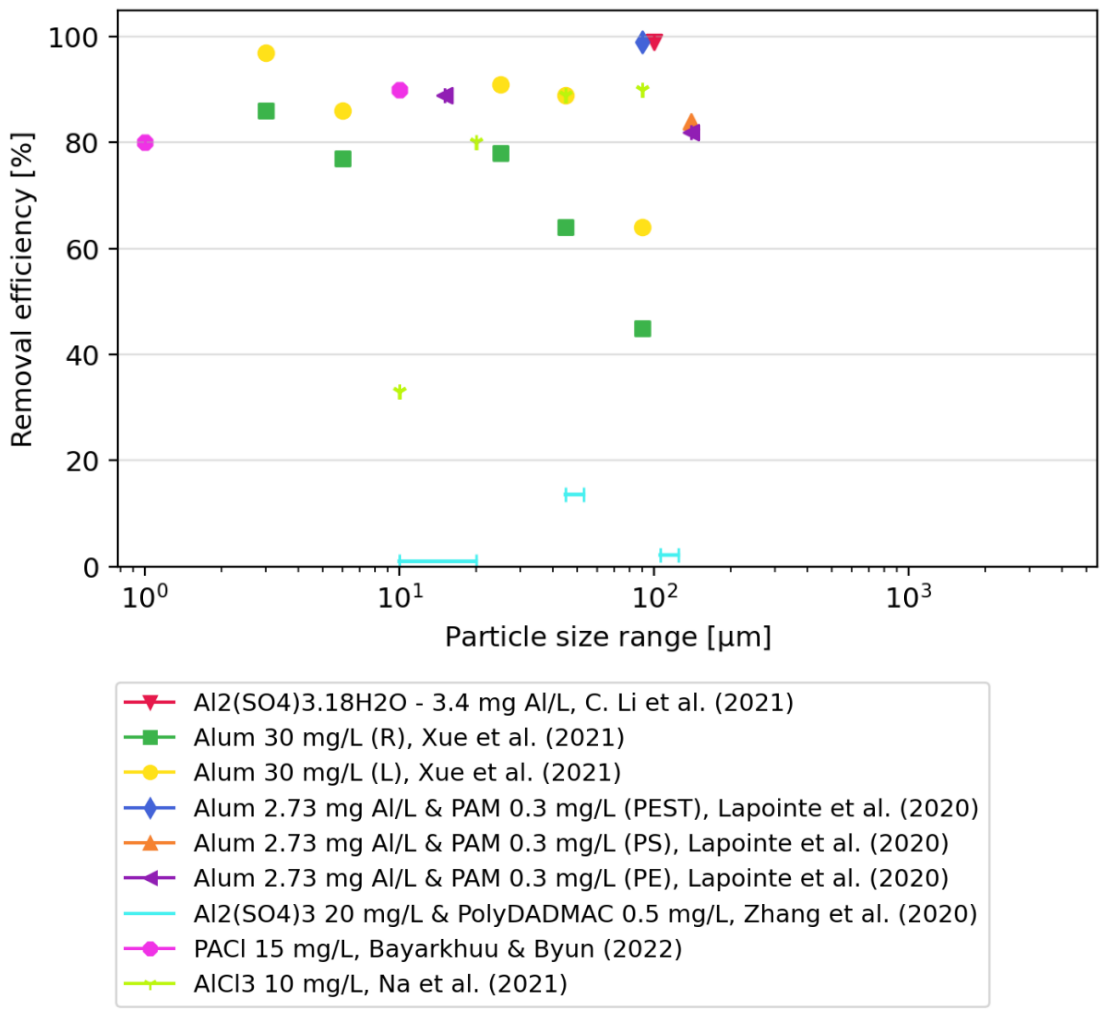


Figure 4.3 (b): Removal efficiency of different-sized MPs by CFS using optimized dosages of Al-based coagulant in surface water irrespective of MPs polymer types, shape, and other properties *(PACl-poly-aluminum chloride, PAM-polyacrylamide, PolyDADMAC- Diallyldimethylammonium Chloride, PE-polyethylene, PS-polystyrene, PES-polyester)*

In Figure 4.1 (b), Xue et al. (2021) and Lapointe et al. (2020) reported comparatively higher removal efficiency for smaller-sized particles. Zhang et al. (2020) found lower removal of all-sized MPs, while the remaining studies indicated higher removal for relatively larger-sized particles. Especially, Lapointe et al. (2020) demonstrated higher removal efficiency for three different polymer types of MPs. Besides, from the comparison of Figures 4.1 (a) and (b), no significant difference can be drawn regarding the impact of using synthetic or surface water on the removal efficiency of MPs.

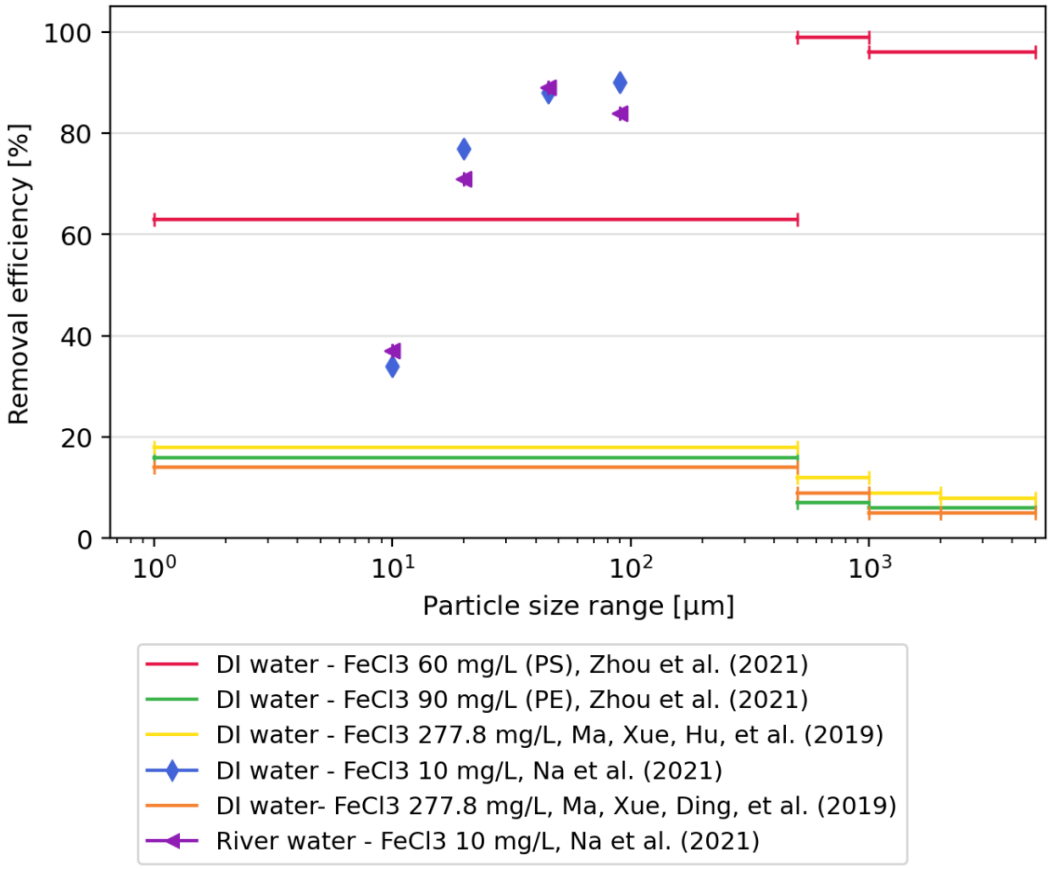


Figure 4.4: Removal efficiency of different-sized MPs by CFS using optimized doses of Fe-based coagulant in both synthetic and surface water irrespective of MPs polymer types, shape, and other properties

(DI-deionized, PE-polyethylene, PS-polystyrene)

Figure 4.2 illustrates the removal of MPs with different sizes using Fe-based coagulants at various doses. A comparison of Figures 4.1 and 4.2 reveals that Fe-based coagulants achieved lower MPs removal compared to the same dosages of Al-based coagulants. Interestingly, higher dosages of coagulants, as observed in Ma, Xue, Hu, et al. (2019) and Ma, Xue, Ding, et al. (2019), did not lead to increased removal of MPs compared to the other studies. Additionally, in these two studies and in Zhou et al. (2021) for PE MPs, larger-sized MPs were removed less than the smaller ones, where the highest removal is less than 20%. The other two studies reported the opposite trend, showing higher removal for relatively larger-sized MPs, though their MPs size is different. Moreover, only one study, Na et al.

(2021), conducted experiments using both synthetic and surface water samples, and almost similar removal efficiency was observed for MPs.

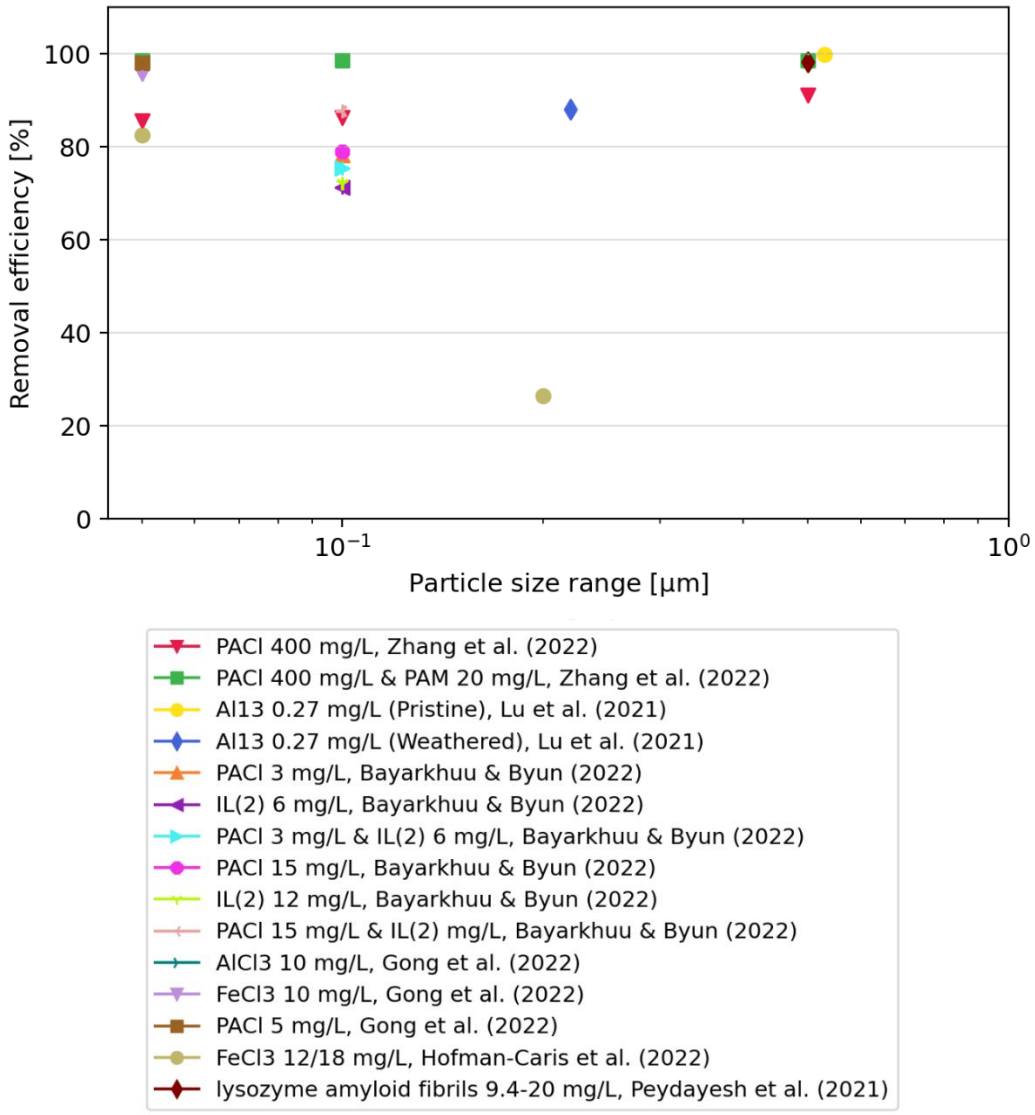


Figure 4.5: Removal efficiency of different-sized NPs by CFS using optimized doses of coagulants in synthetic water irrespective of NPs polymer types, shape, and other properties

(PACl-poly-aluminum chloride, PAM-polyacrylamide, IL(2)-1-decyl-3-methylimidazolium chloride)

In Figure 4.3, the focus is on NPs with sizes <1 µm. Despite variations in coagulation conditions and the properties of the NPs, they all had a good removal percentage. All of the studies achieved about 70-100% removal in those experiments, except the one reported by Hofman-Caris et al. (2022) for 0.2 µm

sized NPs (about 30%). For the 0.05 μm sized NPs in Hofman-Caris et al. (2022), they found around 80% removal, as did the ones reported by other studies. Hofman-Caris et al. (2022) hypothesized that the relatively larger size of the 0.2 μm NPs (compared to 0.05 μm) prevented their entrapment within the formed floc, leading to the observed lower removal.

It can be derived from Table 4.3 that the higher coagulant dosage does not necessarily mean better removal of MNPs for different size ranges. There is an optimal dosage of coagulant that maximizes the removal and an increase in coagulant dosage beyond that point does not bring any notable change in the removal efficiency of MNPs. That optimal dosage of coagulant depends on several factors such as the characteristics of the particles being removed, the type of the coagulant used, and the water properties. Though the tables and graphs were categorized according to the coagulant type and source of water, the properties of MNPs varied widely among the selected studies. So, the reported optimal coagulant dosages and the removal of MNPs varied among these studies. In terms of actual drinking water treatment practice, the coagulant dosage for Al-based or Fe-based coagulants is usually below 20 mg/L (calculated as Al and Fe, respectively) (Ma, Xue, Hu, et al., 2019). Besides, it has been observed that Al-based coagulants are more commonly employed and have shown greater efficacy in removing MNPs with different properties compared to Fe-based coagulants. The variations are assumed to be a consequence of the differences in the strength of hydrophobic and electrostatic interactions between the MNPs and coagulant flocs (Xue et al., 2022). Further investigation is required to fully understand and characterize these interactions and their impact on removal efficiencies. Moreover, based on the limited dataset available, no significant difference has been observed in the removal efficiency of MNPs when using different types of water samples (synthetic or surface water).

The effectiveness of the CFS process in MNPs removal is significantly influenced by various properties of the MNPs. One important factor is the particle size of MNPs, which affects the collision efficiency and settling behavior during their removal by the CFS process (Zhang et al., 2022). Though larger-sized MPs are supposed to get better removal in the CFS process, several studies reported opposite results where smaller MPs and NPs received higher removal. In cases where charge neutralization is the prevailing coagulation mechanism, larger-sized particles exhibit greater susceptibility to compressive double-layer effects, leading to better removal (Zhang et al., 2021). Conversely, when sweep flocculation dominates, removal efficiency decreases with increasing particle size, as smaller

particles have a higher likelihood of being captured within the floc, given a specific floc size (Lapointe et al., 2020). However, the specific coagulation mechanism dominant in each study may vary due to changes in coagulant dosage, pH, and other factors (Tang et al., 2022). The polymer type of MNPs may also cause some differences in the removal efficiency, i.e., Zhou et al. (2021) reported higher removal efficiency for PS MPs compared to PE MPs and they indicated the higher density of PS particles as the reason. The shape of MPs also can affect their removal, i.e., Shahi et al. (2020) reported higher removal for fibers than for spheres and they specified the larger surface area of fibers leading to higher adsorption and interception of MPs to the coagulated flocs as the cause. However, most of the selected studies experimented with similar polymer types and shapes, with a few exceptions.

Besides the size, shape, and polymer type of MNPs, the other properties are not that much reported by the selected studies, but they can also contribute to the removal of MNPs by a CFS process. The particles which are comparatively lighter and close to the density of water may have greater possibilities of staying in suspension or floating in the water than the others and pose a hindrance to removal (C. Li et al., 2021). The low density of MNPs can be considered as a contributing factor behind the unsatisfactory removal of MPs by conventional coagulation (Y. Zhang et al., 2021). So, the density of MNPs may pose a challenge during the CFS process. Most of the MNP particles were reported to be negatively charged in the selected studies. The addition of a metal salt coagulant to water leads to the hydrolysis of the salt, resulting in the formation of various positively-charged complexes. This causes the compression of the double layer and charge neutralization of the negatively-charged MNP particles by a certain dose of the coagulant (Zhang et al., 2021). That results in the removal of MNPs by forming or being incorporated into large-sized flocs. Positively charged PS NPs were found to be more difficult to coagulate and eliminate in surface and mineral waters (Ramirez Arenas et al., 2020). Moreover, the physical condition of the particles also has an impact on MNPs removal, i.e., Lapointe et al. (2020) reported higher removal for the weathered MPs than the pristine particles. They indicated the formation of new functional groups as the reason which encouraged the interaction between MPs and coagulants by increasing the active sites available. Though all of the studies did not report the weathering impact of MNPs in their investigations, experimental results may differ for the naturally weathered irregularly shaped particles which may behave differently during coagulation. Furthermore, most of the bench-scale studies reported the initial mass concentration of MNPs. The variation in the values of their initial

spiked concentrations does not seem to bring any significant effect on the removal of MNPs, which was also supported by Zhou et al. (2021).

Lastly, in terms of the different detection techniques used for analyzing the removal of MNPs in the selected bench-scale studies, each of them has its advantages and disadvantages. For instance, the reliability of the methods with visual and manual counting is poor when the sample number is large and its also time-consuming. The weighing method might be affected by impurities of water which can affect the accuracy of the result. The automatic counting method will have more precision in the reported value. Besides, fluorescence analysis has been proven to be more effective than other conventional visual methods in terms of ease and precision. In general, these analysis methods can have an impact on the reported results of MNPs by the CFS process (Tang et al., 2022).

In summary, the CFS process can be an effective method for MNPs removal, where the variations in MNPs characteristics and different analytical techniques can bring a notable effect on the removal efficiencies observed in the bench-scale studies.

4.4.3 Removal of MNPs by Filtration

Filtration is another widely used method of water treatment that involves passing the water through a porous medium to remove suspended particles, sediments, and other organic and inorganic matter. One common approach is granular media filtration, where the water is passed through a thick bed of granular material such as sand (Crittenden et al., 2012). The effectiveness of this filtration technique depends on the properties of the filter media and the filter bed. Another increasingly popular filtration technology is membrane filtration, where generally different types of pressure-driven membranes are used in water treatment (Worch, 2019). During this filtration, water is pushed through a thin wall of porous materials which can be made of different polymers, ceramics, metals, etc. The main difference between these two filtration systems lies in the requirement of CFS facilities for effective particle removal in granular media filtration, while membrane filtration does not rely on CFS. In granular media filtration, the effectiveness of removal depends on destabilizing the particles through the use of a coagulant, which facilitates their attachment to the media. On the other hand, membrane filtration does not require destabilization as the void spaces in the membrane are smaller, allowing for the straining of particles from the water. Besides, both filtration processes consider factors like size, density, and charge of the particles to achieve efficient removal.

The CFS system is always followed by filtration in full-scale DWTPs. Filtration enhances the clarity of the water by eliminating the remaining suspended particles. It has already been seen in Chapter 3 that the filtration process can cause the removal of MPs to a certain degree. There are only 8 studies among the selected ones which looked into the removal of MNPs through filtration. Six of them investigated granular media filtration and 2 of them evaluated the performance of membrane filtration. Among the 6, there are 3 studies for MPs and 3 for NPs. In each case, one of them had media filtration and the other one had membrane filtration. Tables 4.4 (a) and (b) outline the properties of the filtration systems and the removal of MNPs by media and membrane filtration respectively.

Table 4.4 (a): List of the studies with different properties for **media filtration** system which have reported on MNPs removal by filtration

	No.	Study	Properties for media filtration				Maximum removal efficiency at optimal condition (Particle property – Removal)
			Filter media properties	Filtration rate	L/d ratio	CFS before filtration?	
MPs							
(a)	3	Shao et al. (2020)	Sand [EBCT-5 min]	9 m/h	N/A	Yes	250-300 µm - Sand filtration after CFS resulted in 100% removal
(b)	10	Zhang et al. (2020)	Anthracite [porosity 0.54, media length 38 cm]	N/A	46 cm/4 cm	No	1 µm – 94.9±0.4% 10-20 µm - 86.9% ± 4.9% 45-53 µm – 97.0±3% 106-125 µm – 99.9±0.1%
(a) + (b)	14	Na et al. (2021)	Quartz sand [effective grain size range 0.3–0.45 mm, porosity 0.45, uniformity coefficient < 2]	9 m/h	30 cm/3 cm	No	10 µm – 83.4% 20 µm – 98.8% 45 and 90 µm – 100%
NPs							
(a)	22	X. Li et al. (2023)	Quartz sand [sieve size 20-30 mesh (0.6-0.85 mm) and 40-50 mesh (0.3-0.425 mm)]	3 & 21 ml/min	16.2 cm/2.5 cm	No	By sand-packed column: Almost all NPs passed through the filter regardless of different sand sizes and flow velocities. By sand-limestone-packed column: The removal efficiency increased from 1.55% to 85.04% and reached 94.94 % in 40-50 mesh sand and from 2.54% to 77.98% and finally 85.67% in 20-30 mesh with limestone thickness increasing from 0 to 5 cm and for 7.5 cm at the lower flow rate. The higher flow rate had lower removal efficiencies.
(b)	20	Hofman-Caris et al. (2022)	Clean sand [d_{50} = 1016 µm]	N/A	70-83 cm/3.5 cm	No	0.05 µm (negatively charged) - 63% 0.05 µm (uncharged) & 0.2 µm (negatively charged & uncharged) - 4-22%
	10	Zhang et al. (2020)	Anthracite [porosity 0.54, media length 38 cm]	N/A	46 cm/4 cm	No	0.18 µm - 98.9% ± 0.7%

MNPs-micro- and nanoplastics, MPs-microplastics, NPs-nanoplastics, EBCT-empty bed contact time, CFS-coagulation-flocculation-sedimentation

Table 4.4 (b): List of the studies with different properties for **membrane filtration** system which have reported on MNPs removal by filtration

	No.	Study	Properties for membrane filtration				Maximum removal efficiency at optimal condition
			Filter media properties	Filter membrane pore size (µm)	Filtration rate	CFS before filtration?	
MPs							
(a)	17	J. Li et al. (2021)	PVDF hollow fiber membrane [maximum allowed trans-membrane pressure (TMP) of 80 kPa]	0.03	N/A	Yes	All particles (1, 10, and 18 µm sized) were rejected during the filtration owing to the small pore size (0.03 µm) of the UF membrane used
NPs							
(a)	23	Enfrin et al. (2020)	Poly (sulfone) porous membrane	pore size distribution from 29 ± 6 to 43 ± 12 nm and mean flow pore diameter 31±7 nm	N/A	No	All NPs received 80-50% rejection rate. Rejection percentage decreased with time and 41±2 nm sized particles passed through the membrane over 48 h of filtration

MNPs-micro- and nanoplastics, MPs-microplastics, NPs-nanoplastics, PVDF- Polyvinylidene fluoride, UF-ultrafiltration

Sand filtration is considered as the predominant form of media filtration used in water treatment. Most of these selected bench-scale studies used sand as their filter media, except one Zhang et al. (2020) who used anthracite, and all of them employed simple tests with small-scale columns. Only in the study of Shao et al. (2020), the filtration process was preceded by CFS, which resulted in complete removal of 250-300 µm sized MPs. This study also included pre-oxidation as a pre-treatment step to determine the change in physiochemical properties, which did not seem to influence the removal. Zhang et al. (2020) and Na et al. (2021) reported higher removal of 80-100% for 1 to 125 µm sized MPs. Na et al. (2021) found that some smaller-sized MPs (10 & 20 µm) passed through the filter media which caused comparatively lower removal than 45 and 90 µm sized MPs. Zhang et al. (2020) received about 99% removal for the 0.18 µm sized NPs as well. Hofman-Caris et al. (2022) experimented with two different-sized NPs, with different functional groups and surface charges. Negatively charged NPs with a carboxyl group received higher removal than the others. X. Li et al. (2023) reported no removal of NPs through a sand-packed column. But with a sand-limestone-packed column, the removal efficiency improved as the thickness of the limestone filter bed increased.

Ultrafiltration is generally used in drinking water treatment for the removal of contaminants which are not easily removed by other filtration processes. J. Li et al. (2021) reported that the UF membrane with its small pore size was able to reject all MPs during the filtration process. Enfrin et al. (2020) outlined 50-80% removal for all NPs with increasing rejection rate with time and after 48 h of filtration they detected 41 ±2 nm sized NPs in the permeate. They also demonstrated the potential negative impact of interactions between NPs and UF membranes on the overall filtration performance.

The filtration process including both media and membrane filtration systems reported a significant amount of MNPs removal irrespective of different sizes, shapes, polymer types, concentrations, other properties, and analysis techniques for determining the removal. The relationship between particle size and removal efficiency in media filtration for different-sized MNPs was not consistent. The removal efficiency did not necessarily increase or decrease proportionally with changes in the particle size of MNPs. As a result, smaller-sized particles did not necessarily mean a lower removal efficiency. Overall, the results showed that media filtration can be an effective process to provide at least some removal of all sizes of MNPs. But the reported removal efficiencies of MPs by granular media filtration in full-scale DWTP surveys were much lower compared to the results observed in these bench-scale studies. The lack of detailed information regarding the filtration process and operating conditions in the full-scale surveys makes it difficult to identify a definitive reason for this difference. Moreover, complete

rejection of MNPs was possible in membrane filtration, but this can also cause membrane fouling by particle deposition which may become severe with increased contact and time. In general, the results showed that membrane filtration technology can be an important way of removing MNPs from drinking water irrespective of their different properties, as long as the MNPs size is larger than the pore size of the membrane.

To date, there is very limited research on the removal of MNPs with varying properties using granular media and membrane filtration methods. The impact of various granular media filtration conditions, such as filter bed depth, filtration rate, surface properties, media size, temperature, and backwashing cycles, on the removal of MNPs remains largely unknown due to the limited research and the wide variety of factors involved. These conditions may have diverse effects on the removal of MPs with different sizes, shapes, surface morphologies, and properties. Understanding the interactions between MNPs and membranes, as well as their influence on fouling mechanisms, is crucial for mitigating membrane fouling and optimizing the performance of membrane processes. Furthermore, exploring how MPs interact with different components present in water, including NOM, colloids, and varying ionic strengths, within the context of filtration processes is also an important aspect to consider. The research in the field of filtration process for MNPs removal is still developing and requires further investigation.

4.5 Conclusions

The synthesis of available data from the published bench-scale studies on the removal of MNPs by CFS and filtration treatment processes is a challenging, yet important task. With growing concerns regarding the ubiquitous presence of MNPs in drinking water sources, research on finding suitable removal techniques for MNPs with varying properties is ongoing. However, there is a high variability in approaches and procedures in these bench-scale studies, resulting in variable findings which makes it difficult to draw definitive overall conclusions regarding removals. The general findings from this chapter and key associated limitations and recommendations are outlined below:

- MNPs employed in the bench-scale studies may not always accurately represent the MPs found in the source water as reported by the full-scale surveys. The majority of MNPs used in the bench-scale studies have spherical shapes, which do not reflect the diverse shapes of MPs observed in the source water. In terms of polymer types, the use of PE aligns with the prevalence of one of the polymer types found in the source water. Additionally, the different-

sized particles investigated in the bench-scale studies represent only a part of the MPs reported by the full-scale DWTP surveys.

- Obtaining any conclusion on the removal efficiency for any specific category of MNPs is complex since the properties of MNPs (various shapes, sizes, densities, initial concentrations, and other surface properties) vary widely. The analysis techniques used for the determination of the removal efficiency of MNPs are also diverse among the studies. These differences in MNPs properties and analysis techniques have an effect on the reported results of MNPs removal by CFS and filtration processes.
- Al-based coagulants are most frequently used to investigate the removal of MNPs by CFS, and they generally exhibit higher removal efficiency compared to Fe-based coagulants. Increasing dosages of coagulants do not always bring a significant increase in the removal of MNPs, irrespective of their size. The optimal dosage of coagulant that maximizes the removal, depends on the properties of MNPs, the coagulant type, and the properties of the water being treated. Furthermore, the variation in the water matrix (synthetic or surface water) used in the studies does not seem to make any significant difference in the reported results. The interpretation should be taken into consideration due to limited datasets with a wide range of variations in certain processes, which is similar to the observations made for the CFS process in the full-scale DWTP surveys.
- The limited number of bench-scale studies have demonstrated that granular media filtration can achieve high removal efficiencies for MNPs regardless of the size or other properties studied. These bench-scale granular media filtration reported much higher removals compared to the filtration results observed in full-scale surveys.
- Membrane filtration has also achieved higher removal efficiencies in the bench-scale studies as long as the studied MNPs were larger than the pore size of the membrane. This filtration has brought significant removal for MNPs irrespective of their other properties and different operating conditions. But there are only very few studies available on filtration, emphasizing the need for further investigations to support these preliminary findings.

The bench-scale studies reviewed in this chapter highlight several gaps that need to be addressed in future research. A key area for improvement is the development of standardized sampling methods and uniform analytical techniques to facilitate the comparison among studies on the removal of MNPs with

varying properties by different drinking water treatment processes. It is crucial for bench-scale studies to employ MNPs that are more representative of the properties found in the source water, leading to the standardization of MNPs used in the experiments. Consistency should be brought in terms of reporting the MNPs properties and their subsequent removal efficiencies to better understand their removal mechanisms and to facilitate the further analysis process using existing data from published studies. Besides, while most of the existing experiments are focused on MPs, more attention should be directed toward also investigating NPs. This broader approach will help to develop a more comprehensive understanding of the fate and removal of MNPs in drinking water treatment processes.

Chapter 5
Development of a Regulatory Framework

5.1 Introduction

MNPs have become a concern as they have been found in drinking water due to their potential impacts on human health. The relevance of this problem is evidenced by the exponential growth in the number of research papers on MNPs. There have been ongoing investigations to determine the possible health effects of the presence of MNPs in drinking water, but further studies are required for a comprehensive understanding of the potential risks. However, some studies have already suggested that exposure to MNPs may have negative health impacts. Currently, there are no regulations for MNPs in drinking water, so DWTPs are not mandated to conduct monitoring for MNPs in treated water. But ongoing monitoring and assessing their presence by different full-scale surveys and bench-scale studies have shown that existing treatment processes of the DWTPs can remove MNPs by a certain percentage. This kind of assessment may aid in forming regulations and guidelines to ensure safe and clean drinking water. To assure that levels of these particles in the water are safe for human consumption, MNPs in drinking water may have to be regulated. The development of regulations regarding the presence of MNPs will involve a range of factors, but human health concerns are one of the most crucial aspects. The biggest challenge in regulating MNPs lies in their diverse nature, as they are comprised of a wide range of particles varying in size, shape, and surface properties. The specifics of the regulations may depend on the risks and challenges associated with MNPs of different characteristics.

The main objective of this chapter is to initiate the development of a framework to regulate MNPs in drinking water. To fulfill that goal, the first step would be to provide an overview of the effects of MNPs on human health with their associated potential toxicity. Then a comprehensive investigation has to be performed regarding the progress made to date on regulating MNPs in general and also specifically in drinking water. Some concepts of regulations for micropollutants and microorganisms will also be summarized, from which ideas can be obtained for the development of a framework to regulate MNPs. After that, the main basis for the construction of this framework has to be elaborated and discussed in terms of its elements, implications, and challenges. Lastly, conclusions have to be drawn based on the next potential step concerning the development of this framework.

5.2 Background

5.2.1 Toxicity and Human Health Concerns related to MNPs

The rise of global plastic production and their constant usage contributes significantly to the abundance of MNPs as plastics degrade down to smaller-sized particles until they reach micro- and nano-sized particles (Kokilathasan & Dittrich, 2022). These plastic materials ultimately find their way into aquatic bodies mainly through surface runoff and wastewater discharge (Alimi et al., 2018). MPs have already been found at every level of the food chain starting from producers (e.g., algae, phytoplankton) to top consumers (e.g., fish, zooplankton) in aquatic ecosystems (Badola et al., 2022). In contrast to MPs, there is considerably limited knowledge about the abundance of NPs in this environment (Materić et al., 2022). Although scientists have been investigating MPs since the 1960s, there is still no international consensus on the definition of MPs (Hartmann et al., 2019). The occurrences of NPs are not as much reported as MPs due to the lack of appropriate analytical methods to detect particles <1 μm (Hartmann et al., 2019). Though there have been a growing number of studies on NPs, there is still no consensus on the definition of NPs (Kokilathasan & Dittrich, 2022). Despite that fact, the pervasiveness of MNPs across the food chain in oceanic life forms, birds, plants, and humans has become a serious issue (D. Zhou et al., 2022). With the growing human population, the frequency of releasing MNPs into aquatic environments increases as human activities increase (Elgarahy et al., 2021). Such a high rise in abundance of MNPs raises a key concern about their potential risk to human health. Due to limited data on the exposure and impact of MNPs, there is considerable uncertainty surrounding this issue. Even though plastic is generally regarded as inert, MNPs possess various attributes like their chemical composition, shape, size, and hydrophobicity, that might be potentially harmful and affect the toxicity of the MNP particles to cells and tissues (Wright & Kelly, 2017). The adverse effects caused by exposure to MNPs could be separated by two groups: physical effects and chemical effects. The physical effects are relevant to the particles' concentration, shape, and size, whereas the chemical effects are attributed to the presence of hazardous chemicals that can be adsorbed onto MNPs (Campanale et al., 2020).

The physical effects of MNPs are directly affiliated with the particle properties. One of the primary challenges for assessing health risks due to the physical effects is quantifying and monitoring them in the environment. Uncertainties regarding the effects of small-sized MPs and NPs still exist because of unidentified hazards and experimenting bias toward comparatively larger-sized MPs which are thought

to be less harmful (Adam et al., 2019). Analytical and technological challenges involved in extraction, characterization, and quantification have limited the usage of small-sized MPs and NPs in experiments (Campanale et al., 2019). Most of the studies concerning health effects associated with MNPs are focused on marine life. Compared to that, there are only a few studies that have looked into the impacts of MPs on freshwater and terrestrial life (Boyle & Örmeci, 2020). The impacts of MNPs on human health are far less investigated than the other ecological aspects (Y. Zhang, Pu, et al., 2020), and so there is a limited number of studies published in this field. Most of the studies conducted to date examined the impacts of MPs on model organisms and then extrapolated the results to human health (Blackburn & Green, 2022). The three main routes for the exposure of the human body to MPs are ingestion, inhalation, and direct contact (C. Wang et al., 2021). The increased presence of MPs in drinking water can be one of the sources for direct and indirect ingestion of MPs by humans. The ingestion of food and beverages including fruits, vegetables, alcohol, etc. can serve as another major pathway for MNPs to enter the human system (Toussaint et al., 2019). When MNPs result from atmospheric depositions, agricultural erosions, road dust, aerosol, etc., and are carried by the wind, they can also enter the human body through inhalation (Gasperi et al., 2018), including inhalation of vaporized water during showering. Washing with water and using scrubs or cosmetics that contain MNPs can give exposure through the skin, where NPs absorption is more probable than MPs through the skin (Revel et al., 2018). Hence, MPs can enter the human body unintentionally throughout life and they do not have an immediate harmful effect on the human body, but they can exert a subtle effect after accumulating in the body (L. Wang et al., 2021). Various human systems such as the digestive system, respiratory system, nervous system, and placental barrier have been reported to be affected at different levels by high concentrations of MPs (Elizalde-Velázquez & Gómez-Oliván, 2021). Though the impacts of NPs on human health are still unknown due to some limitations, they have the potential to pose a more significant threat to biota than MPs because of their increasingly small size (Boyle & Örmeci, 2020). Studies have shown the potential of NPs to pose an ecotoxicological risk in aquatic environments, and possible human health risk via their presence in drinking water production (Ramirez Arenas et al., 2020b). Smaller-sized MPs or NPs can enter human tissue and release toxic additives in the process, damage cells, and cause inflammation and local immune responses (B. Zhang et al., 2021). Recent in-vitro investigations have predominantly focused on employing engineered NPs (intentionally produced NPs with specific properties for a definite purpose) to assess their effects on the human body, which may affect the generation and translocation of reactive oxygen species (ROS) in relation to the

NPs' size, shape, and charge (e.g., Lehner et al., 2019). Besides, many studies have already assessed the impact of engineered nanoparticles, especially on human health, owing to their extensive use in various applications (Kumah et al., 2023). But the extrapolation of results for potential toxicity from engineered nanoparticles to NPs is very delicate (Revel et al., 2018). As NPs are still less investigated than the MPs in terms of exposure to human health, more research is required to understand their impacts clearly.

Chemical effects of MNPs arise from the release of chemicals from their matrixes or absorbed from the environment. There are two different types of chemicals which can be associated with MNPs: (i) additives and polymeric raw materials originating from plastics, and (ii) chemicals adsorbed onto MNPs from the surrounding environment. Plastic products are often manufactured with various kinds of chemical additives to enhance their performance for different commercial and industrial applications. The additives include plasticizers, lubricants, dyes, flame retardants, stabilizers, antioxidants, inert or reinforcing fillers, curing or blowing agents, etc. (Campanale et al., 2020). Despite their ability to enhance the polymeric products' properties, many of the additives are found to pose a significant risk of contaminating water, air, and soil (Hahladakis et al., 2018). Most of these additives are not chemically bonded to the polymer structures, so they can be easily released from the plastic particles (Adegoke et al., 2023). These compounds can come into contact with the human body and may contribute as a factor to chronic health effects. Besides, both MNPs exhibit small size and increased surface area/volume ratio compared to other particles, which contribute to their strong affinity towards adsorbing a broad range of contaminants (Hanif et al., 2022). Various studies have also reported the ability of MNPs in adsorbing different types of chemicals like hydrophobic pollutants, persistent organic and inorganic pollutants, pathogens, antibiotics, heavy metals, etc. which can enter the human body by MNPs uptake (Campanale et al., 2020). Then again, because of their high adsorption capacity, more pollutants can easily accumulate in organisms via MNPs and reach tissues and organs affecting them toxicologically (B. Zhang et al., 2021). As a result, MNPs can serve as potential vectors for exposure to different types of contaminants (Kokilathasan & Dittrich, 2022). The health risks associated with these chemical contaminants can be considered as indirect health risks attributable to MNPs.

5.2.2 Difference between MNPs and Micropollutants

MPs are generally referred to as synthetic solid particles or polymeric matrices that exhibit insolubility in water with sizes falling within the range of 5 mm to 1 µm, which can be either of regular or irregular shape and either of primary or secondary manufacturing origin (J. P. G. L. Frias & Nash, 2019). As NPs are less studied, there is still no universal definition, though several studies set their size limit as < 1 µm or <0.1 µm (Hartmann et al., 2019). MNPs are generally grouped into primary and secondary categories, where the primary ones get directly released into the environment whereas the secondary ones originate from the decomposition of larger-sized plastics.

Micropollutants are not particles themselves, but they include a diverse range of natural or anthropogenic organic and inorganic pollutants, including pharmaceuticals, antibiotics, personal care products, heavy metals, pesticides, and industrial chemicals (Sharma et al., 2023). They are mostly found dissolved in water or sometimes adsorbed onto particulate matter. Their concentrations may vary in the environment, but usually are present in trace amounts in water from a few ng/L to several µg/L (Y. Luo et al., 2014), and sometimes up to a few hundred µg/L. They can comprise both individual compounds and technical mixtures. Atrazine, a selective and most widely used herbicide, is one of the examples of the organic micropollutant which falls into the category of individual compounds (Shirmardi et al., 2016). Polychlorinated biphenyls (PCBs) are a group of chlorinated aromatic organic compounds recognized as persistent organic pollutants because of their thermal and chemical stability (Ficko et al., 2010). Again, pharmaceuticals are one of the examples of complex mixtures of molecules with different functionalities, physicochemical and biological properties (Kümmerer, 2004). Organochlorine pesticides (OCPs), polybrominated diphenyl ethers (PBDEs), and per- and polyfluoroalkyl substances (PFAS) are also known as persistent organic pollutants which have bioaccumulation and biomagnification potential (Verhaert et al., 2017). The impacts of these wide varieties of micropollutants have been proven to be difficult to find at their low exposure concentrations and ever-changing compositions in different mixtures (Eggen et al., 2014). Several micropollutants are currently subject to regulatory measures based on a risk assessment approach, while certain emerging ones have not yet been regulated due to their complex nature.

Though MNPs themselves are also known as emerging pollutants, one of the major differences between these micropollutants and MNPs is in terms of solubility, where micropollutants are generally soluble and MNPs are insoluble solids, which brings distinct physicochemical characteristics resulting in different toxicological behaviors.

Table 5.1: Summary of differences between MNPs and micropollutants

Feature	MNPs	Micropollutants
Physical state	Insoluble solids	Dissolved
Source/Origin	Primary source (manufactured and released as small particles); Secondary source (degraded from larger plastic particles)	Anthropogenic sources (agriculture, pharmaceuticals, steroid hormones, food products, pesticides, etc.)
Size	Solid particles ranging from 5 mm to 1 µm (MPs); <1 µm (NPs)	Individual molecules ranging from approximately one hundred Da to kDa in size
Type	Synthetic polymer	Both natural and synthetic organic and inorganic compounds
Abundance/concentration in surface water	Vary with source and location from zero to thousands of particles per liter	Trace amounts up to high µg/L in some cases
Adsorption	They can adsorb micropollutants	They can adsorb to solids including MNPs
Persistence/biodegradability	MNPs are persistent, not biodegradable, and can bioaccumulate	Some compounds or derivatives have partial or incomplete biodegradability, depending on the chemical structure

MNPs-micro-and nanoplastics, MPs-microplastics, NPs-nanoplastics

Some of the micropollutants are known as the contaminants of emerging concern because of their persistence, bioaccumulation, and life-threatening potential, as they may cause abnormal growth, reduce fertility and reproduction, delay neurodevelopment, inhibit wildlife species, degrade aquatic ecosystems, and possibly harm the human immune system (Arman et al., 2021). They are made from diverse molecules, have varying molecular structures, and come from various sources of applications, which are like MNPs. But the unique property of MNPs which makes them different than micropollutants is that MNPs are particles of different sizes, shapes, polymer compositions, colors, etc. MNPs are comprised of diverse polymer types characterized by their unique backbone structures, which are formed by repeating monomers (Rochman et al., 2019). This variation in polymer composition

accounts for the fundamental differences observed among different types of MNPs, influencing their distinct physical and chemical properties. The addition of chemical additives during production gives the polymers different physical and surface properties even when derived from the same polymer type. Besides, the fragmentation process of polymers generates particles in a wide size range and differing shapes. Moreover, they have the potential ability to sorb different persistent organic pollutants and trace metals, which depends on the MNPs' diffusivity, crystallinity, hydrophobicity, and surface area (Bergmann et al., 2015). This unique property of MNPs makes them more complex than micropollutants in terms of characterization and regulation. So, all of this diversity should be taken into consideration while developing regulatory frameworks (Rochman et al., 2019).

5.2.3 Review of Existing Legislation for MNPs

As the emerging presence of MNPs is a key concern, efforts are underway to better understand the toxicity associated with these particles and to develop new strategies to reduce their abundance in drinking water, because drinking water is one of the potential ways to get exposed to MNPs. Though no direct human health impact of MNPs has been reported to-date, precautions should be taken beforehand to tackle the problem. Notwithstanding the absence of any definite regulatory framework for MNPs in drinking water so far, several guidelines and initiatives have been taken with the aim of reducing and regulating MP pollution in the environment. Seventeen years after the first use of the term MPs, various policies around the world have started to take them into account to prevent and mitigate their presence (Munhoz et al., 2022). The majority of these efforts have concentrated on the minimization of production and use of plastics and primary MPs, which will ultimately lead to the reduction of the abundance of MNPs in the environment and consequently in drinking water sources.

Plastics make a substantial contribution to the presence of MNPs in the environment, with approximately 69-81% of MPs originating from them (Singh et al., 2022). Since the industrial revolution, plastic use has increased with population increase, urbanization, and technological advancement (Adegoke et al., 2023). So, in the current world, plastic use cannot be completely prohibited, but other sustainable solutions can be found (Badola et al., 2022). Starting from 1991, many countries restricted the use of plastics by imposing different legislation (Lam et al., 2018). The early policies mostly focused on reducing plastic waste by imposing bans, fines, and taxes on plastic use. Single-use plastic bans have proven effective in reducing plastic consumption and waste generation in several countries including Italy, Ireland, England, Israel, and the Netherlands (Schnurr et al., 2018).

In some cases, biodegradable plastics have been suggested as an alternative to traditional plastics (Shruti & Kutralam-Muniasamy, 2019). In 2015, the seven major developed countries of the world (Canada, the United States, the United Kingdom, Germany, France, Italy, and Japan) as well as the European Union, discussed possible substitutes to reduce plastic pollution and are now working on a plan of action to address marine debris, including plastic waste (Singh et al., 2022). In the following years, several countries started to include MPs in their policy and they initiated this action either by proposing new rules or amending some existing ones regarding plastics to control the use of MPs. The United Nations Conference on Sustainable Development addressed MPs as an emergent environmental issue in 2012 (Munhoz et al., 2022). Afterwards, the United Nations included MPs under the 2030 Agenda for Sustainable Development by establishing the need for actions to reduce MP pollution. Certain regulatory organizations in the US and Europe have classified certain chemicals including MPs as 'non-threshold contaminants' considering any release to the environment a plausible risk (ECHA, 2019; Lin et al., 2018). Besides, various industrial sectors along with government agencies have been trying to implement different mitigation actions to protect human health and the environment from the known and unknown effects of MPs. A summary of various national initiatives taken around the world to reduce the production and use of MPs is provided in Table C1 in the appendix. All these existing preventative responses and mitigation measures can eventually make a marginal impact on reducing the load of MPs in source water and highlight the necessity of reducing NPs pollution. It can also be expected that the global acknowledgement for the regulation of both MNPs will continue to grow in the upcoming years.

At present, there are no specific guidelines for regulating MPs or NPs in drinking water at the international and national level. Despite a huge scientific knowledge gap regarding the available information on MPs, California took the first step towards setting the world's first health guidelines for MPs in drinking water. In acknowledgement of preliminary evidence of extensive contamination of drinking water with MPs (e.g., Novotna et al., 2019) and significant uncertainties associated with human health risks, the California Legislature enacted SB 1422 in 2018. This legislation mandated the establishment of a definition for 'MPs in drinking water' by July 1, 2020, and the adoption of a standard methodology for detecting MPs in drinking water by July 1, 2021 by the State Water Resources Control Board (Coffin et al., 2021). This bill also required a 4-year period for conducting tests and providing reports on the prevalence of MPs in drinking water, publicly releasing the findings, potentially establishing a health-based guidance level for interpreting the results, and authorizing laboratories in

California to analyze MPs (California Code - HSC, 2019). Besides, California passed SB 1263 at the same time to address MPs in the marine environment which required the California Ocean Protection Council to develop a comprehensive statewide strategy (California Legislative Information). The timeline outlining the implementation of California Senate Bills 1422 and 1263 can be found in Figure 5.1.

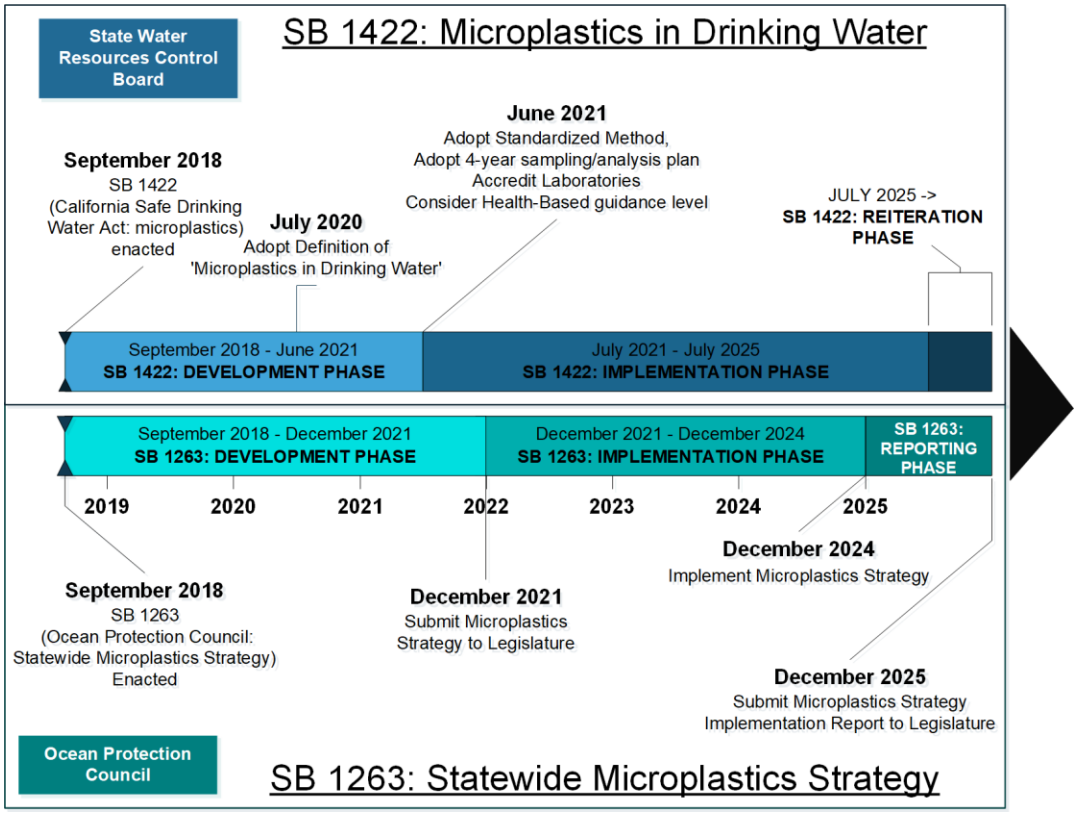


Figure 5.1: Timeline for implementation of California Senate Bills 1422 and 1263 (Coffin et al., 2021)

To confront unparalleled challenges in tackling possibly the most intricate and diverse group of contaminants (plastics; including MPs), California is collaborating with a global network of researchers, federal, state, and local agencies, non-governmental institutions, water providers, and committed citizens (Coffin et al., 2021). In 2018, the California State Water Board hosted annual, multiday, immersive “Trash Data Dives” (trash includes MPs) in which researchers along with other collaborators actively participate to create a “trash management picture informed by open and accessible data, to identify and understand trends, data gaps, and priorities” (California State Water Resources Control Board, 2019). The execution of California’s legislative requirements faced

challenges because of the lack of a globally accepted definition for MPs resulting from the absence of standardized techniques and regulations (Coffin et al., 2021). In June 2020, an official definition for "MPs in drinking water" was adopted by the California State Water Resources Control Board, which may evolve depending on the available information (California State Water Resources Control Board, 2022). The definition is stated as "'Microplastics in Drinking Water' are defined as solid polymeric materials to which chemical additives or other substances may have been added, which are particles which have at least three dimensions that are greater than 1 nm and less than 5,000 micrometers (μm)". Then in August 2022, they published a policy handbook introducing a standard approach for testing and reporting MPs in drinking water (California Water Boards, 2022). They standardized two analytical methods (Infrared spectroscopy and Raman spectroscopy) through an inter-laboratory comparison study for extracting and analyzing MPs in drinking water. Raman spectroscopy and infrared spectroscopy showed excellent performance in average recovery and chemical identification for particles >20 μm and 50 μm respectively, with the possibility of better accuracy and precision through further development (De Frond et al., 2022). However, they could not provide satisfactory performance in the determination of particles <20 μm. At that time, there were only a few commercial or utility laboratories equipped to monitor MPs as no government mandated the monitoring for MPs. The State Water Board has also started assessing the appropriateness of sampling methodology as a part of the pilot phase in terms of feasibility and quality control between the sampling method described in ASTM D8332-20 (ASTM, 2020) and the sampling method in Yuan et al., 2022. After that, they will release a comprehensive guidance manual and offer the necessary training for sampling and analysis. Then the State Water Board laid out a timeline consisting of two phases (see Figure 5.2), which will involve an iterative approach to monitor MPs. This approach aims to gather the necessary information to assess the potential risks associated with exposure to MPs through drinking water. The first two years of monitoring in the first phase will be followed by six months of evaluation to determine the extent of contamination of MPs and the necessity of evaluating drinking water in the next phase which will be two years with more sophisticated testing methods.

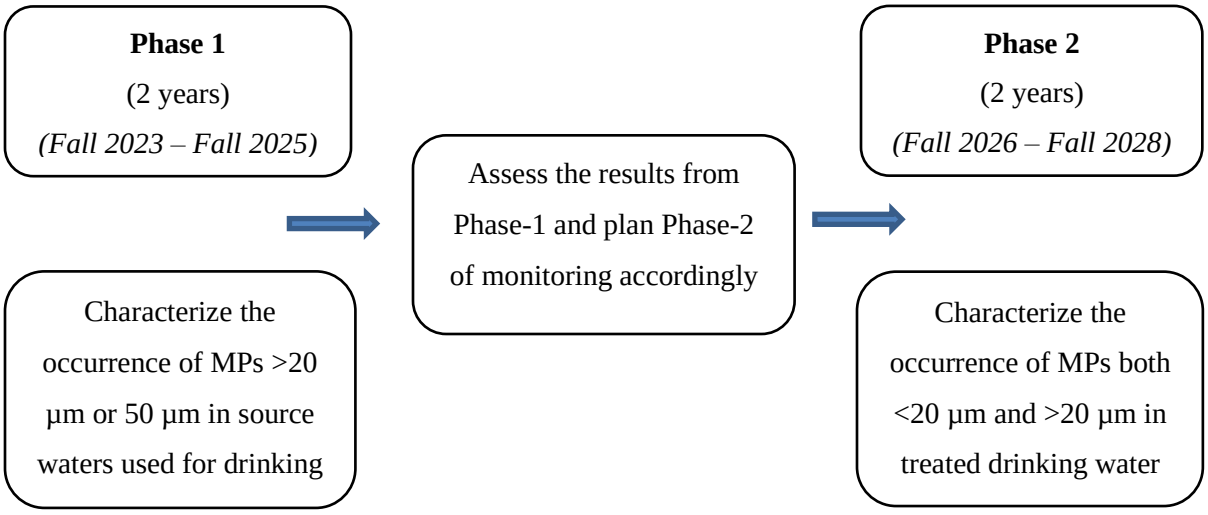


Figure 5.2: Two-phase monitoring plan for MPs prepared by California State Water Board

The State Water Board is working to ensure data harmonization and reporting consistency by building an open-source reporting tool. They are also conducting additional research to address logistical and scientific challenges associated with monitoring outside of the four years commitment to monitoring and reporting. In the case of the public water system selection for monitoring of MPs in drinking water, they selected a small number of sources that serve a large population while fulfilling other requirements of treatment capabilities, geospatial representation, and primary water sources (e.g., surface water, groundwater, groundwater under the direct influence of surface water) (California Water Boards, 2022). Based on the evaluation of findings from Phase I, the locations for Phase II will be determined. All these initiatives will serve as a foundation and be helpful for the comparison of findings between studies, resulting in reducing the variability in the risk assessment. As a result of this whole deliberative approach, California became the first jurisdiction in approving the world's first requirements for testing MPs in drinking water sources.

Though California launched the first requirement for MPs testing in drinking water, there are some challenges associated with it. According to their published report (California Water Boards, 2022), two of those standardized analytical methods showed excellent performance for particles larger than 20 and 50 µm. The smaller size range of MPs < 20 µm was not reported to be well identified and quantified by these methods. But based on the research published to-date, most of the health risks are associated with smaller-sized particles. Again, in the process of standardization, round-robin tests were conducted using certain specific types of MPs as reference materials. Later when measuring environmental MPs

in water samples these established standardized methodologies may not be able to identify certain varieties of environmental MPs as they may differ substantially in their properties from those of the reference MPs used in method development and round-robin testing. So, there will be a continuous need to improve these detection methodologies with time and update these standardized methods as they evolve. Besides, NPs are not considered in this approach and they have a high potential for toxicity and hazardous effects on the environment and human health. The above-mentioned standardized methods would not be able to identify NPs as they are even smaller in size than MPs. So, there is also a need to incorporate NPs in the monitoring plan and formalize the detection techniques accordingly.

Chowdhury (2021) developed a conceptual framework to identify the indirect health risks (attributable to chemical contaminants sorbed on and in MPs) from the ingestion of waterborne MPs. Although direct health risk assessment from waterborne MPs themselves is not yet possible because of the scarcity of data on human exposure, there is data available regarding the potential health effects of the chemical contaminants that may sorb on MPs. So, they wanted to support the drinking water industry for risk assessment and decision-making purposes by providing this indirect health risk assessment tool. They established a framework to calculate "Threshold MPs Concentration" (TMC) based on the available scientific data regarding MP size, shape, type, contaminant adsorption/desorption capacity, the extent of MPs removal during DWT, human exposure to MPs through drinking water, and drinking water contaminant standards. TMC is defined as the number of MPs particles per liter of water which can cause any potential health risk from exposure to chemical contaminants sorbed in or on those MPs surfaces. This TMC value is analogous to the maximum acceptable concentration (MAC) value that is used to ensure safe water by managing risk from chemical drinking water contaminants in the US and Canada (Health Canada, 2019; USEPA, 2018). Based on this research, in a well-operated DWTP, the existence of MPs in treated water will not be considered as a significant threat if the concentration is below the TMC value. If the concentration of MPs is above the TMC, it does not mean that there is an immediate threat to human health, instead, it indicates that a thorough inspection of MPs type, concentration, adsorbed contaminants, and treatment methods present in that system is necessary to better evaluate the potential health risk.

This framework has only considered the potential health impacts which can be caused by the chemical contaminants absorbed in or adsorbed on MPs, they did not consider the health risks which can be caused by the MPs themselves. Moreover, the concept of NPs is also not considered in this framework, and NPs have more adsorption capacity per unit mass than MPs because of their smaller size and

therefore higher surface area. So, still, there remains the need for a regulatory framework to control the abundance of both MNPs in drinking water considering the potential health impacts caused by the MNPs themselves.

5.2.4 Existing Regulatory Concepts for Micropollutants and Microorganisms

A substance or microorganism is typically regulated through a risk assessment approach, evaluating its potential hazards and the level of risks to human health and the environment. The amount of possible exposure is determined by its concentration in the finished drinking water. The compound's properties and potential adverse effects are identified through scientific research, followed by assessing human and environmental exposure, which helps to determine the overall risk associated with the contaminant. The regulatory measures and strategies are developed and implemented to mitigate the identified risk. The regulatory agency establishes standards that can limit the levels of a specific compound in drinking water. The regulatory values are established to reduce exposure and associated risk to an acceptable level. The utility must monitor regulated compounds in the finished drinking water to ensure compliance. But the application of a conventional risk assessment approach is not suitable for certain contaminants of emerging concern, including endocrine disrupting chemicals (EDCs), pharmaceuticals and personal care products (PPCPs), etc. because of the challenges associated with monitoring them and accurately assessing their potential risks.

PPCPs are widely used in soaps, lotions, toothpaste, fragrances, sunscreens, etc. in high quantities throughout the world (Liu & Wong, 2013). Given the extensive range of PPCPs in use, the estimation of the environmental risk for each compound is impractical due to the insufficiency of data (Mo et al., 2022). Additionally, new PPCPs are continuously being introduced into the market and they have metabolites and transformation products, further complicating the assessment process. Prioritizing methods might be used to determine which PPCPs are likely to cause the highest concerns by evaluating them based on factors like exposure, risk, or hazard. This facilitates the process of resource allocation in an effective way for ecotoxicological investigations along with environmental monitoring programs in particular geographical areas.

There are also limitations and difficulties in testing and regulating EDCs. They can be found in various sources such as water, food and food packaging, household goods, personal care products, fabrics, detergents, pesticides, etc. which can lead to illness and impairment over the lifetime by interfering with hormone action (Kassotis et al., 2020). The process of prioritizing substances for detailed

assessment of the EDC properties has been hindered by incomplete identification due to insufficient data (Michel, 2019). This lack of data highlights the difficulty in effectively regulating EDCs following the traditional risk assessment approach. These complexities associated with regulating PPCPs and EDCs by the traditional approach are also valid for MNPs, such as a large number of diverse substances, data limitations, and the continuous emergence of new compounds.

One of the emerging contaminant classes is PFAS, which have been found to have some similarities with MPs in terms of persistence, diversity, toxicity, unknown composition, and bioaccumulative potential (Sunderland et al., 2019). The fate and transport of PFAS could not be well studied using traditional models as they are found inadequate due to the diversity in class and unique characteristics of PFAS (Naidu et al., 2020). In that sense, MNPs are even less understood because of their insolubility and unique synthetic polymer characteristics. Efforts are underway to understand the transport behavior of MNPs by leveraging existing knowledge of particle transport. Besides, significant challenges are associated with regulating MPs because of difficulties in determining the toxicity of dispersed particles with different concentrations and lack of standardization in assessment methods (Backhaus & Wagner, 2020). Hence, there is a need for the development of novel and alternative approach to determine their exposure and identify the threats.

In that case, reviewing existing regulatory efforts for certain micropollutants or microorganisms can be helpful before drawing on the proposed regulatory framework for MNPs. Among the available regulatory concepts, the Swiss approach in reducing micropollutants in wastewater and the health-based treatment guideline for *Giardia* and *Cryptosporidium* are reviewed here and were considered in developing the pattern of the proposed regulatory framework for MNPs.

- Regulations for micropollutants in wastewater (Swiss Policy):

Micropollutants i.e., pesticides, pharmaceuticals, personal care products, etc. enter WWTPs usually in measurable amounts and these substances are usually not fully removed by existing treatment steps. As a result, they get released from WWTPs into surface water bodies. Though they are typically found in water in varying amounts, they can have adverse impacts on health and the environment. Switzerland first addressed the issue in the early 2000s and took a step ahead of others by targeting these dissolved organic micropollutants for removal in wastewater treatment plants. Later their initial step resulted in a Swiss Water Protection Act in 2016 (Bourgin et al., 2018). As a part of this program, they initiated a 20-year commitment for investment to reduce the release of micropollutants from WWTPs into the

aquatic environment so that the country's primary drinking water sources are free from persistent and artificial substances irrespective of their toxicity level (Federal Council, 2015). Their main goal was to achieve an 80% reduction of micropollutants through advanced treatment by the year 2040. But monitoring and developing uniform regulations for the diverse array of micropollutants at the same time would be complicated. So, the Swiss strategy developed a list of indicator compounds for regulating micropollutants in wastewater. The selected compounds are readily analyzable, allowing for regular detection and comparison with influent and effluent samples in treatment plants. Besides, they set a list of criteria for selecting those compounds and in that way, they came up with 12 representative compounds.

Table 5.2: List of the selected compounds with their classification and the criteria for their selection

Selected compounds	Classification	Selection criteria
Amisulpride	Pharmaceutical	✓ Only parent compounds (no transformation products) ✓ Can be easily and routinely measured in one analytical method (at cantonal or private labs) ✓ Occurring in bigger WWTPs at a measurable concentration (influent concentration 10xLOQ in effluent) ✓ Degraded to less than 50% in biological treatment ✓ Similar abatement in advanced treatment (not favoring ozone or AC) ✓ Continuous discharge into WWTPs
Benzotriazole	Industrial	
Candesartan	Pharmaceutical	
Carbamazepine	Pharmaceutical	
Citalopram	Pharmaceutical	
Clarithromycin	Pharmaceutical	
Diclofenac	Pharmaceutical	
Hydrochlorothiazide	Pharmaceutical	
Irbesartan	Pharmaceutical	
Methylbenzotriazole	Industrial	
Metoprolol	Pharmaceutical	
Venlafaxine	Pharmaceutical	

LOQ-limit of quantification, AC-activated carbon, WWTPs-wastewater treatment plants

The Swiss strategy established this set of 12 indicator compounds for regulating micropollutants, which do not have to be high-risk chemicals and are not generally removed by conventional processes. 10

pharmaceuticals fulfilled these criteria and were included in the list of indicator compounds, with 2 industrial compounds used as corrosion inhibitors. Pharmaceuticals are usually complex compounds with different physicochemical and biological structures, leading to uncertainty regarding their impact (Miarov et al., 2020). Those pharmaceutical indicator compounds could be evaluated through a single laboratory experiment, which offers a relatively efficient monitoring option. They started monitoring these representatives for organic micropollutants in several aquatic media through their comprehensive environmental program. In 2016, the Swiss program launched new regulations to upgrade around 120 municipal WWTPs over the next 20 years targeting an overall 80% removal of the micropollutants across the plant (Miarov et al., 2020). This regulation requires that these WWTPs be equipped with an advanced treatment stage. The WWTPs are chosen for upgrades by taking into account two factors: the size of the plant and the sensitivity of the receiving water (Ort et al., 2009). In March 2014, the first full-scale plant started its operation by using ozone as an advanced step for wastewater treatment (Spurenstoffe et al., 2021). The average elimination performance for indicator compounds across the entire plant was around 84%, which is more than the average value required by the law (ARA Neugut, 2016). At the beginning of 2020, a total of ten plants were equipped with an additional treatment stage and another 29 were either in the planning stage or under construction (Spurenstoffe et al., 2021). Besides, this strategy involved the demonstration of two alternative processes, either ozonation or granular/powdered activated carbon as advanced treatment steps. These processes are technically proven to be feasible, can be integrated at the existing plants, and are sufficiently cost-effective. Subsequent full-scale trials have verified that the necessary personnel with the required expertise to operate advanced WWTPs can effectively manage them after undergoing specialized training. Following this strategy, the estimated cost of wastewater treatment expense increased by 12% (Federal Council, 2015). The financing of this strategy follows the ‘polluter pays’ principle by introducing a wastewater tax for WWTPs, where the upgraded WWTPs are exempt from the tax but have higher operating costs, and 75% of the investment costs for WWTP upgrades are covered by the federal government (EurEau, 2019).

This Swiss policy serves as an example of establishing legislation for a vast variety of contaminants by a comprehensive approach of monitoring, reducing, and removing micropollutants in wastewater before releasing them into the environment. The selection of representative compounds among the complex and diverse contaminants class is one of the vital ideas which can be considered while developing a regulatory approach and framework for MNPs. This approach may ultimately enhance the efficiency

and effectiveness of the regulatory measures. As MNPs encompass a diverse range of polymeric particles with varying physical and chemical properties, including size, shape, polymer composition, etc., this approach would be beneficial for their efficient assessment and regulation. Then, initiation of the removal goal for those representative compounds and fulfilling that by an additional advanced treatment step can be considered as a potential approach for regulating a diverse class of contaminants. In short, this approach targeting the dissolved compounds can work as a roadmap for regulating solid particles like MNPs.

- Regulation for *Giardia* and *Cryptosporidium*:

Giardia and *Cryptosporidium* are protozoan pathogens, which are widely recognized as waterborne disease-causing microorganisms due to their significant effect on human health (e.g., Efstratiou et al., 2017). Their capacity to generate (oo)cysts has enabled their dissemination and potential to cause illness. The U.S. Environmental Protection Agency's (EPA) Method 1623/1623.1 is the predominant method for detecting *Giardia* and *Cryptosporidium* in water, both at the national and international level, due to the validation of their simultaneous detection (USEPA, 2012). Because of their high infection rates, a MAC would have to be set at quite a low level. Besides, considering the difficulty and expense of the quantification methods for detecting (oo)cysts at that low level in water, it was deemed not feasible to implement a MAC (Ligda et al., 2020). Therefore, a risk management approach i.e., a source to tap water approach was adopted to decrease the occurrence of these disease-causing organisms in drinking water prior to its distribution to the first consumer (O'Connor, 2002). The utilization of the combined approach of monitoring source water for *Giardia* and *Cryptosporidium* and physical removal and inactivation technologies has been found to be an efficient way for reducing (oo)cysts in drinking water. Physical removal and inactivation barriers are assigned "log removal" and "log inactivation" credits respectively to quantify their effectiveness in reducing the level of (oo)cysts (Health Canada, 2012). The overall "log-reduction" is determined by combining these credits for the treatment process being evaluated. The existing guidelines for drinking water are health-based treatment goals that encourage the water treatment authorities to reduce the risk of illness as much as possible. The removal credits assigned for *Giardia* and *Cryptosporidium* in Canada and the United States are summarized in Table 5.3.

Table 5.3: Regulation for *Cryptosporidium* and *Giardia* in Canada and the United States

Country	Treatment barrier	*Cryptosporidium* removal credit	*Giardia* removal credit	Reference
Canada	Conventional filtration	3 log	3 log	Health Canada, 2012
	Direct filtration	2.5 log	2.5 log	
United States	Conventional filtration	3 log	2.5 log	Federal Register., 1989, USEPA, 2005
	Direct filtration	2.5 log	2 log	

Health Canada stipulates a minimum of 3-log (99.9%) removal and/or inactivation of *Giardia* and *Cryptosporidium* and the U.S. EPA requires 3-log (99.9%) removal or inactivation of *Giardia* and a minimum 2-log (99%) removal or inactivation of *Cryptosporidium*. The establishment of these requirements had to go through years-long monitoring program for assessing the extent of treatment necessities (Betancourt & Rose, 2004). Adequate characterization of the source water is also necessary in order to find out the required treatment level. In some instances, further elimination or inactivation might be required to achieve the treatment objectives depending on the source water quality, especially for some surface waters used as sources of drinking water (Health Canada, 2012). In such scenarios, site-specific assessments would be needed to determine the extent of reduction required for (oo)cysts in that particular source water. If direct measurements (source water sampling and analysis) are not feasible, then the data derived from the routine assessment of the system and other water quality parameters can be utilized for assessing the level of risk in the source water (Health Canada, 2012). This type of information can assist in determining whether a higher level of treatment is necessary beyond the minimum requirement.

This approach shows promise when regular monitoring results seem hardly possible to achieve for some contaminants. Defining the level of those contaminants in the incoming water by an initial monitoring program and then ensuring the DWTPs can remove a definite percentage of those contaminants – can be useful to apply for a diverse class of contaminants where constant monitoring is not a suitable option.

5.3 Proposition of a Regulatory Framework by Identifying Representative MNPs in Drinking Water

Regulatory frameworks are often constructed using the traditional risk assessment method, which involves comparing the exposure amounts of contaminants with known toxic thresholds and setting up maximum contaminant levels in drinking water (USEPA, 1986). Due to the extremely diversified nature of MNPs, they will not readily fit the conventional risk-based regulatory frameworks (Coffin et al., 2021). MNPs include a wide range of particles with different sizes, shapes, polymer compositions, densities, solubility, surface charges, etc. with an immensely persistent nature, which may lead to high-level uncertainty in associated impacts. So, the main challenge in assessing the risk associated with MNPs lies in the difficulty of determining the potential toxicity and human health impacts of each individual class of MNPs. Then due to the extremely persistent nature, their continuous production may lead to their accumulation in surface water, making their removal from drinking water increasingly challenging (Cousins et al., 2019) Another critical challenge in the risk assessment of MNPs is that they can be associated with different chemicals of various possible hazards. Most of the time the properties of plastics-additives remain hidden from the consumers or are provided with inadequate documentation in the accessible database (Z. Wang, Walker, et al., 2020). So, the identification of possible toxic effects of MNPs become more complicated owing to the risk associated with additional chemical components or pathogens adsorbed to MNPs. Moreover, insufficient data on the occurrence of MNPs in drinking water, along with the absence of standardized methods leading to inconsistent occurrence data, further contributes to the challenges in adopting a risk assessment approach. As NPs are barely investigated to date, there have been no regulatory efforts for their control and management. While substantial research is being conducted on nanoparticles, the scope of direct transferability of knowledge may be limited due to the unique properties of NPs, which will influence their fate, behavior, and potential health impacts. Besides, NPs possess a higher risk potential because of their smaller size range compared to MPs, while their detection poses greater challenges, making the risk assessment process even more complex for NPs.

The primary objective of this chapter is to develop a framework for the regulation of MNPs in drinking water. To achieve that goal, a regulatory framework has been proposed. The main basis of this framework has been inspired by the Swiss policy undertaken for controlling (dissolved) micropollutants in wastewater. In this section, the elements of this proposed framework will be outlined and the feasibility of establishing these components will be discussed. Because this framework represents a

first step, it is focused only on MNPs themselves and does not encompass chemical contaminants or microorganisms that might be absorbed in or on MNPs. Additionally, surface water is considered as the source of drinking water. An overview of the proposition for the development of a regulatory framework for MNPs in drinking water can be found in Figure 5.3.

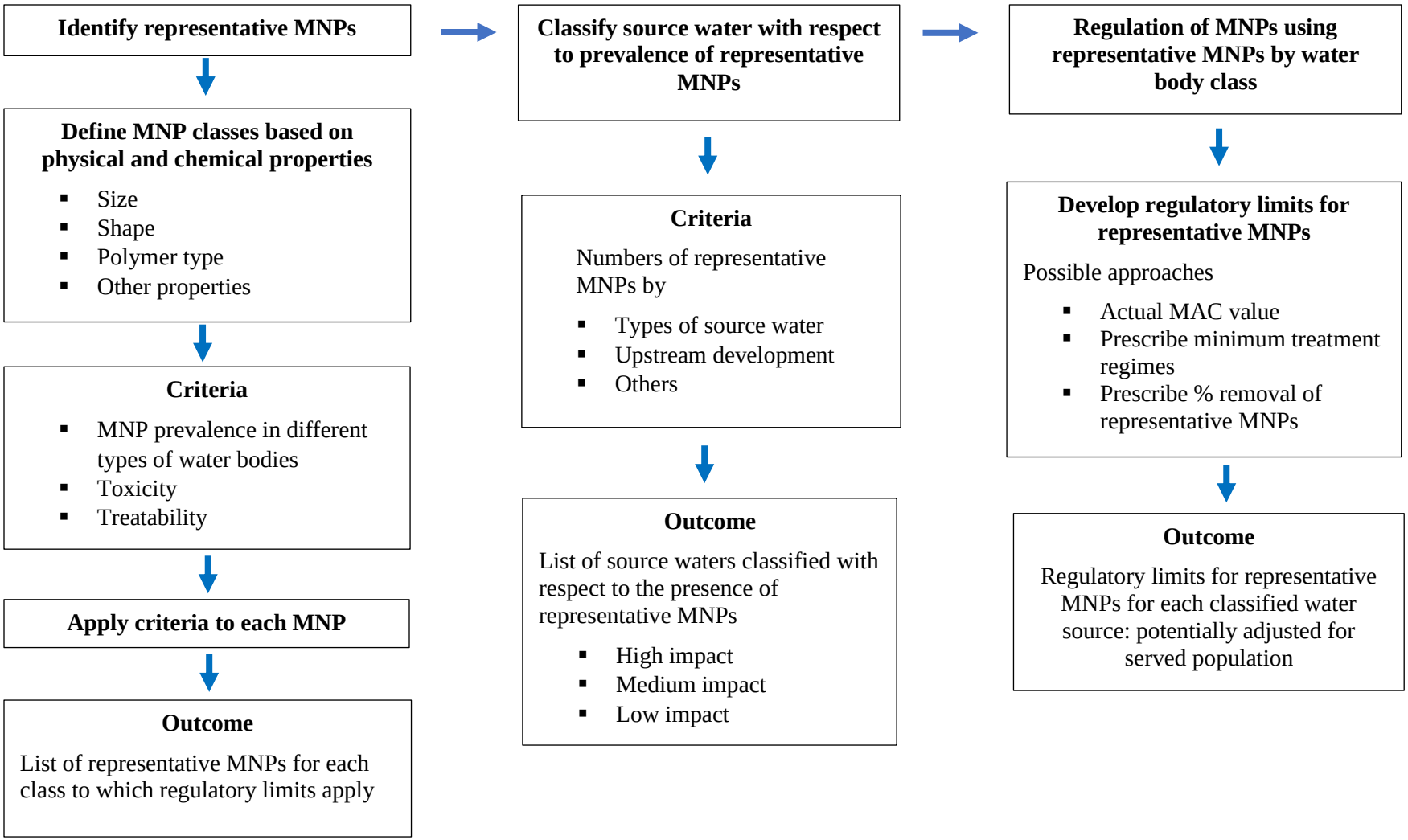


Figure 5.3: Development of a regulatory framework for MNPs in drinking water

There are three principal sequential elements of this framework which start with the initial step of identifying representative MNPs, then move to classify source waters with respect to the prevalence of those identified representative MNPs, and end with applying regulation for MNPs using the selected representative MNPs by water body class. The main three elements of this regulatory framework, their subsequent criteria, and possible outcome are explained below:

- Identification of representative MNPs

The wide variety of MNPs and their versatile nature have made it difficult to monitor this whole class of contaminants and interpret the potential risk associated with them. The concept of finding indicator compounds that will represent the vast number of MNPs is analogous to the principle of the Swiss policy undertaken for controlling dissolved micropollutants in wastewater. Swiss policymakers chose micropollutants that can serve as indicators for the broader classes, resist removal by conventional processes and enable efficient assessment. So, the selection of representative MNPs can be an effective approach to addressing the complex and diverse nature of the MNPs. This would make it feasible to monitor and regulate representative MNPs instead of a wide range of diversified MNP particle types. The MNPs, which are the most abundant, toxic, less treatable, and therefore posing the highest risk hazard, can be prioritized as the representative MNPs.

Representative MNPs can be selected from some broader classes formed based on their physical and chemical properties. Adopting a class-based approach would be beneficial for managing an extensive variety of MNPs and ensuring representative coverage. This system would allow for comprehensive and efficient monitoring of MNPs. The classes can be determined by the physical and chemical properties of MNPs. There is a wide range of varieties in terms of those properties, including size, shape, polymer type, color, surface properties, etc. First, they can be classified based on one major characteristic and then divided into different sub-classes as necessary. Then the MNPs which are predominant, potentially most harmful, and resistant to treatment in each class can be selected as representative MNPs. The selected representative would be able to vouch for the diversified nature of the contaminants that fall in each class. These representative MNPs are chosen such that they provide an indication of the overall presence, characteristics, and potential risks associated with each class of MNPs.

In order to develop such a list of representative MNPs, a list of criteria has to be set up which can be used to identify those representative compounds. The prevalence of MNPs in different water bodies,

associated potential toxicity, and resistance to treatment or treatability can be considered as some of the potential criteria for the process of selection of those representative MNPs.

i. The prevalence of MNPs in different types of water bodies can be one of the potential factors when choosing representative MNPs. This criterion will be helpful in identifying the likelihood of exposure for different types of MNPs in each defined class. Then monitoring efforts, research studies, and regulatory actions would be prioritized based on the prevalence. That would save a lot of time and resources in terms of undertaking a risk management strategy for MNPs. MPs have already been detected in various surface water sources including rivers, lakes, water reservoirs, etc. but their abundances are heterogeneous across different studies, depending on the surrounding environmental conditions. Due to the lack of appropriate analytical methods, the abundance of NPs is not that much investigated until now. But in general, the abundance of MNPs tends to be higher in water bodies that are in proximity to pollution sources, such as industrial areas, landfills, or densely populated regions. Upstream development structures can be one of the important considerations to comprehend the MNPs prevalence. For instance, WWTPs located upstream of water bodies can significantly contribute to the higher abundance of MNPs in water. The type of WWTPs and the quality of water discharged also play a role in the amount and properties of MNPs present. The quality of effluent from the upstream WWTPs having tertiary/advanced treatment steps would be better than the discharge from WWTPs having secondary treatment steps only. Again, smaller-sized MPs and NPs can pass through these WWTPs and enter the water bodies in the treated effluent. So, the higher risk is likely to be associated with the water bodies which have upstream structures with higher discharge potential of low-quality effluent. Furthermore, the construction of other development structures, such as roads, buildings, various industries, parking lots, etc., can also increase the deposition rate of MNPs in water bodies through stormwater run-off. Run-off also can transport MNPs from different land-based sources. Moreover, other upstream development activities such as dredging, channelization, or construction of dams can alter the condition of water bodies, impacting the number of MNPs present. Hydrodynamic conditions, such as strong wave action or stagnant water, may also influence MNPs distribution and accumulation in the water bodies. In addition, seasonal variations, such as rainfall, can affect the presence of MNPs. Again, the prevalence of MNPs would be different for various properties of MNPs. If the abundance for smaller-sized particles

is higher comparatively, then the representative should be chosen prioritizing those smaller ones. Moreover, as the number and characteristics of MNP particles present in different water bodies are not uniform, the selection criteria for representative MNPs would differ depending on the water body type. In short, the MNPs which are highly prevalent and easily identified in different types of water bodies should be prioritized as representative compounds from each class.

ii. Toxicity can be a crucial factor while identifying representative MNPs. Selection of representative MNPs with known or suspected toxicity would help to address the hazardous contaminants, and eventually protect human health and the environment. It would ease the process of risk prioritization, hazard assessments, and informed decision-making to mitigate potential adverse impacts. There has been an increasing amount of research going on to find the potential toxic effects of various MNPs on human health. MNPs with the highest risk of exposure and health effects should be included in the list of indicator compounds. The characteristics of MNPs such as size, shape, polymer composition, and other surface properties can influence their toxicity level and possible health impacts. For instance, most of the potential toxic effects are associated with smaller-sized MNPs. So, comparatively smaller-sized particles should be considered in the selection process. Representative compounds can also be chosen based on their ability to enter the human body through ingestion, inhalation, or dermal contact. The prevalence of MNPs is also an important consideration as higher prevalence increases the likelihood of exposure and accumulation in the body over time due to their persistent nature, leading to potential health hazards. Although human health impacts may not be immediately visible, they may become apparent over time with continuous unintentional ingestion. Therefore, both the short and long-term effects of exposure should be taken into account while discussing toxicity and its potential health impacts. Overall, MNPs with higher toxicity and a greater risk of causing health hazards should be prioritized as representative MNPs for each class.

iii. Treatability can be another significant factor when choosing representative MNPs. It will enable the prioritization of the particles that are less treatable, leading to the enforcement of regulatory efforts to ensure their effective removal and minimize their negative impacts. Some MNPs may show resistance to available treatment processes, which increases the possibility of finding those compounds in the treated water leading to potential negative impacts. On the

other hand, some MNPs are more likely to be effectively removed by water treatment processes, reducing the chance of their presence in drinking water. MNPs that are found to be less treatable should be prioritized in the selection process of representative MNPs. Previous research suggests that larger-sized MPs are comparatively easier to remove. Smaller-sized MPs or NPs may require additional or advanced treatment for better removal. The MNPs which can be removed by using existing conventional or advanced treatment methods do not need to be given emphasis for the list. Representative MNPs should be chosen in a way that addresses the risk of being present in the treated water and also are associated with any possible health hazard. The selection process should also consider other properties of MNPs such as shape, polymer composition, density, and surface properties, which can affect their treatability. In general, the selection process of representative MNPs for each class should aim to minimize the risk of finding compounds in the treated water that may pose potential risks to human health by ensuring the removability of representative MNPs to an acceptable degree.

These are just some of the proposed criteria to be taken into consideration while identifying representative MNPs for each defined class. This would help to ensure a comprehensive understanding of the occurrence of MNPs so that resource allocation for monitoring and risk assessment can be efficient. This might also help to interpret the condition without directly monitoring MNPs in every case. The outcome from all of these criteria would vary significantly with different characteristics of MNPs including size, shape, polymer composition, density, surface properties, etc. So, the selection of representative MNPs for each defined class would prioritize the MNPs properties. The most prevalent, vulnerable, toxic, and difficult-to-treat MNPs can be selected as the representative MNPs so that the preventative and reduction measures taken for them would provide conservative results for all other MNPs in that class. Due to the infinite variability of MNPs in different water bodies, identifying representative MNPs for each defined MNP class is recommended. This will aid in tracking their abundance, movement, and behavior in the water system. This concept will eventually be helpful in evaluating their presence in different water bodies and their removals in different full-scale and bench-scale studies. Monitoring representative MNPs will also be cost-effective compared to monitoring all MNPs. This will lead to the development of strategies to regulate and remove representative MNPs instead of trying to regulate all types of MNPs, which is impractical.

- Classify source water with respect to prevalence of representative MNPs

The framework's subsequent significant element involves characterizing source water based on the occurrence of representative MNPs. This characterization of source water can ease the process of implementing regulatory measures to mitigate risks associated with the presence of MNPs. By comprehending the prevalence of representative MNPs across different water body classes, regulators can assess the need for regulation and allocate appropriate resources for monitoring, mitigation, and treatment. It allows for targeted regulatory measures to be implemented in specific areas where it is most necessary, ensuring more efficient use of resources and better protection of water sources. It also helps in the assessment of the overall magnitude of the issue, identification of trends, and development of strategies to minimize the introduction and impact of MNPs in water sources. When direct monitoring is not feasible, classification provides guidance for implementing operational monitoring and necessary treatment measures based on the conditions of the water body class. The concept of classifying source water draws inspiration from the regulatory approach applied for *Giardia* and *Cryptosporidium* in Canada and the USA. Regulators established a minimum health-based treatment goal and conducted site-specific assessments to ascertain the required level of reduction for a given source water. In cases when direct measurements are unavailable, previous assessments can provide insights for estimating the associated risk level in source water and inform the need for additional treatment. Therefore, the classification of source water bodies based on the prevalence of representative MNPs simplifies the process for the application of regulations.

There are various types of water available as drinking water sources, and the abundance of MNPs can vary depending on several factors. Therefore, a list of criteria can be set up for classifying the source water based on its type, associated factors like upstream development, etc.

i. Type of source water: The abundance of representative MNPs can vary across different types of source water bodies including lakes, rivers, reservoirs, and streams. These surface water bodies receive inputs from various pollution sources such as urban and rural runoff, industrial discharge, and wastewater effluents, also from atmospheric deposition. The amount of pollution inputs and the proximity to these pollution sources play a role in the variation of the abundance for representative MNPs. For example, surface water bodies located near densely populated areas are more likely to exhibit higher levels of representative MNPs due to increased pollution inputs from various human activities. The hydrological characteristics of surface

water bodies, such as flow rate, water depth, and residence time, can also influence the distribution and accumulation of representative MNPs. For instance, stagnant water bodies might tend to have a higher abundance of representative MNPs compared to fast-flowing systems. Moreover, environmental conditions including seasonality, weather patterns, and temperature variations can impact the presence of representative MNPs by affecting their transport, deposition, and degradation patterns in different surface water bodies. Considering these factors, the classification of source water based on the prevalence of those representative MNPs becomes crucial for effective and targeted regulation. This approach may allow for resource allocation, monitoring efforts, and implementation of regulatory measures in an efficient way.

ii. Upstream development: The upstream development of source water bodies can also be another separate factor of consideration in relation to the abundance of representative MNPs. Upstream development refers to the human activities and infrastructure located upstream of a water body, such as urbanization, agriculture, industrial operations, and residential areas. This is already considered in terms of source protection measures for other contaminants in raw water sources, in various jurisdictions. Surface water bodies act as a sink for MNPs transported from these upstream sources. The presence and quantity of representative MNPs can vary based on the type and frequency of upstream development. The land use practices and activities in these upstream can contribute to the pollution and contamination of surface water bodies. For example, urban areas are prone to stormwater runoff carrying pollutants including plastic waste into nearby rivers or streams, while agricultural activities can introduce plastic contaminants through irrigation systems or runoff from fields. Industrial operations may release plastic particles and pollutants directly into the water. The nature of upstream development also differs depending on the location of the water source, whether it is situated in an urban or rural area. The characteristics of WWTP effluents can also vary depending on the adjacent environment. Industrial areas are likely to have more adverse effluent quality compared to areas without industrial facilities. Surface water bodies receiving effluent discharge from urban or industrial areas are more likely to exhibit a higher abundance of MNPs due to the discharge of treated or untreated wastewater. On the other hand, source water bodies located in remote or sparsely populated areas are expected to have lower levels of MNPs. Therefore, the extent of upstream

development plays a significant role in influencing the abundance of representative MNPs in the source water.

These proposed criteria can offer a foundation for identifying and characterizing water sources based on the prevalence of representative MNPs. To initially establish a classification of the water bodies based on the risk assessment, thorough monitoring efforts are required. This process will provide insights into the extent of the impact of representative MNPs on specific water sources. Then by evaluating the abundance of representative MNPs in those water bodies, they could, for example, be classified into three categories: high impact, medium impact, and low impact. Water bodies exhibiting a substantial and comparatively higher abundance of the representative MNPs can be considered as highly impacted, indicating a higher risk of exposure to potential toxicity. Then the water bodies with relatively lower levels of representative MNPs will fall into the medium impact category. Lastly, the low-impact category will encompass water sources with significantly fewer representative MNPs, thereby indicating a lower level of associated risk. This, or a similar classification of source water will be beneficial for subsequent regulatory approaches to be tailored to address the risk with prioritization.

- Regulations of MNPs using representative MNPs by water body class

After the selection of representative MNPs and classification of source water bodies based on their abundance, regulatory limits should be established. The development of regulations tailored to the representative MNPs by water body class will enable a focused strategy in mitigating the risks associated with the diverse range of MNPs. These regulations would establish standards, guidelines, and requirements to minimize the presence and potential harm of MNPs in finished drinking water. These will presumably lead to the implementation of control measures, treatment technologies, and best practices that are suitable for the unique characteristics of the pollutant and vulnerabilities of the respective water body class. By adopting regulations, a consistent and standardized approach is ensured in addressing the issue across different scenarios, thereby promoting the provision of safe drinking water. So, the implementation of regulations would impose responsibility on stakeholders to prevent, control, and reduce MNPs in water sources, fostering accountability and adherence to the prescribed standards for finished drinking water.

A number of approaches can be considered to develop regulatory limits for representative MNPs.

- First, one possible approach can be to establish a MAC value for each representative MNP. The MAC value signifies the threshold value that should not be exceeded in the distributed

water. It would have to be established whether it referred to a single measurement or an average over a period of time. The purpose of a MAC is to minimize the risk to human health from contaminants that are known or suspected to have adverse health effects. In cases where the abundance of representative MNPs in raw water sources exceeds the MAC value, appropriate treatment measures must be provided to ensure safe drinking water. However, implementing such an approach may present challenges, especially for a divergent contaminant class like MNPs, as their properties and concentrations vary considerably. It would require the regular monitoring of the representative MNPs in finished drinking water, which may be challenging due to the complexity and diversity of MNPs.

- Another approach can be prescribing a minimum treatment regime based on the classification of the source water bodies concerning the presence of representative MNPs. This approach is particularly suitable when achieving complete removal of the contaminants may not be practical or continuous monitoring of the contaminants in drinking water is not feasible. The adoption of this approach draws inspiration from the regulatory strategies employed for *Giardia* and *Cryptosporidium*. These microorganisms pose challenges in terms of assessing their infectivity, requiring specialized personnel, and incurring substantial costs for monitoring. Similarly, the analysis of MNPs is much more complicated due to their diverse properties, making the regular assessment in finished water a costly endeavor. By opting for this approach, constant monitoring of treated water concentrations would not be required. Instead, the minimum treatment level has to be designed in such a way that it can deal with the anticipated peak levels of representative MNPs. In instances where the level of representative MNPs found in the sources exceeds the anticipated peak levels, the implementation of advanced treatment techniques may be required to reduce exceptionally high concentrations to an acceptable limit. The regulatory limits can be adapted according to the classified source water with respect to the presence of representative MNPs. The adjustments in treatment requirements would ensure the appropriate management of MNPs which has to be strictly adhered to by DWTPs responsible for treating that water. In the case of highly impacted water bodies, more stringent treatment techniques would need to be implemented compared to others to minimize the associated risk. For water bodies classified as medium-impacted, less stringent treatment steps may be deemed sufficient to address the contamination. Conversely, for water bodies categorized as low impacted, the existing treatment processes may adequately manage the

situation and in that case, no additional treatment technique might be necessary. By aligning the regulatory limits with the classification of source water, the treatment requirements can be appropriately customized by ensuring the protection of treated water quality.

- An alternative approach can involve prescribing a minimum percentage removal for the representative MNPs. This approach can be adaptable when the complex characteristics of the contaminants make it difficult to determine specific concentration limits. By prescribing a minimum percentage removal, a consistent level of protection can be ensured while allowing for flexibility in choosing appropriate treatment methods and addressing limitations in monitoring capabilities. The period over which the average required removals would need to be achieved would have to be determined. The incorporation of this strategy draws inspiration from the Swiss policy adopted for regulating micropollutants in wastewater. Due to the complexities associated with identifying emerging micropollutants, they prescribed an 80% removal target for indicator compounds, with the option of implementing advanced treatment as necessary. Like dissolved micropollutants, MNPs exhibit complex characteristics and unique properties, posing challenges for regular monitoring of the wide range of variation. So, the analysis process can be factored only for the representative MNPs. Moreover, they lack a well-defined health-based threshold, so by focusing on achieving a minimum percentage removal, utilities can navigate the complexities associated with monitoring and tailor required treatment technologies accordingly to ensure effective reduction of the pollutant. Various levels of percentage removal can be implemented to address the classified water bodies according to the prevalence of representative MNPs. For example, in highly impacted water bodies, DWTPs could be required to achieve a minimum of 80% removal comprehensively. In the case of medium-impacted water bodies, a minimum removal of 50% of the representative MNPs could be required by the DWTPs. Lastly, for water bodies classified as low impact, no additional action might need to be mandated for the DWTPs. To meet these regulatory requirements, DWTPs must employ all necessary treatment processes and ensure their effectiveness in achieving the desired removal.

In general, regulatory limits can be accommodated to address the presence of representative MNPs in each classified water source, and these limits could potentially be further adjusted based on the size of the population being served. So, it is not necessary for all treatment plants to implement the same treatment techniques. Rather, treatment plants handling water from the classified water sources need to

adapt their processes according to the specific requirements. Large treatment plants are generally better equipped to execute these strategies and achieve the goal of the regulatory approach. As the requirement of removal techniques is contingent upon the quality of source water including the abundance of representative MNPs, DWTPs need to be adjusted accordingly to address the issue and ensure the provision of safe drinking water.

5.4 Implementation and Associated Challenges

The main challenge lies in the implementation of a proposed regulatory framework. This proposition would require a lot of work to bring this proposed framework into action.

- To implement the process of identifying representative MNPs, the required step would be to define the classes of MNPs complying with the criteria. Various approaches can be considered to define classes. One possible method is to classify MNPs based on their shape, such as fibers, fragments, and others (all shapes except fibers and fragments). These classes can be further sub-divided into different size groups under each class, including <1 µm, 10-20, 20-50, 50-100, and 100-5000 µm. Prioritizing smaller size ranges can be given importance as they are mostly associated with greater potential toxicity. Other properties should also be considered within each class and subclass. The selection of representative MNPs should fulfill specific criteria, requiring extensive research and monitoring to determine their prevalence. Monitoring would be required to be conducted over a minimum period (for example perhaps 2 years) to determine the abundance of the MNPs. Toxicological data must be established to assess their health impacts, prioritizing those with the most severe consequences. Additionally, years of data on MNPs treatment would have to be collected to identify the MNPs that are less treatable. So, sufficient research is necessary to establish their toxicity and treatment characteristics, focusing on identifying the MNPs which are more toxic and less treatable. With the accumulation of these data, representative MNPs can be chosen accordingly.
- To classify water sources with respect to the prevalence of representative MNPs, a program has to be set up where different types of watersheds would be monitored for a period such as 2 to 3 years. Extensive sampling across different water sources and analysis of the collected data using standardized techniques would be required to quantify the abundance and characteristics of MNPs present. Then categorization of the water sources would be needed by setting

threshold values or ranges that distinguish the different impact levels based on the selected representative MNPs.

- To establish regulations for MNPs using representative MNPs by different water body classes, different requirements must be met for the implementation of a specific approach. While developing a MAC value, it would require an assessment of the available scientific literature and data to understand the potential health risks (any known or suspected) associated with the representative MNPs. Exposure assessment would also be necessary to evaluate the duration and level of exposure to MNPs. Then based on the risk characterization, a MAC value is determined to ensure acceptable risk levels for humans. Regulatory agencies set this value by considering scientific evidence, public health considerations, and other factors upon consistent monitoring data. Regular monitoring of the contaminant levels in drinking water sources and periodic reviews of the MAC value are also essential afterwards. While implementing a minimum treatment regime or minimum percentage removal of representative MNPs, understanding existing treatment barriers is crucial. Performance evaluation of treatment technologies is necessary to ensure effective removal. Specific performance criteria are established to meet the prescribed minimum treatment requirements for each classified source. A robust monitoring program has to be implemented to assess treatment performance and achieve the desired goals. Adjustments of regulatory approaches are needed over time based on the latest scientific evidence, advancement in analytical techniques, and emerging knowledge to ensure the continued protection of public health.

To successfully implement the proposed regulatory framework, it is also imperative to address the challenges associated with it. Some of them are:

- One of the major challenges is the lack of standardized, reliable, yet accessible analytical quantification methods for MPs, especially for NPs. Consequently, there is insufficient comprehensive data on the prevalence, toxicity, and treatment characteristics of various MNPs. The existing dispersed dataset are not enough for data comparison and informed decision-making processes.

- MNPs can exhibit diverse particle characteristics, including shapes, sizes, surface properties, and chemical compositions. Classifying MNPs based on these wide attributes is complex and challenging, and the emergence of new compounds makes it more difficult.

- Conducting long-term monitoring to assess the prevalence of different MNPs in source water bodies is logistically demanding. Gathering sufficient data necessitates extensive research, in-depth studies, regular monitoring, and testing, which can be time-consuming and resource intensive.
- Implementing the proposed framework would require significant resources, including funding, laboratory facilities, qualified personnel, and a long-time commitment to future research. Adequate support and collaboration from various stakeholders would be essential to overcome these challenges.

5.5 Conclusions

The proposed framework for developing regulatory limits for MNPs in drinking water diverges from the traditional approaches used for other micropollutants. The main inspiration has been drawn from the Swiss model approach applied to control dissolved micropollutants in wastewater, as well as from guidelines for *Giardia* and *Cryptosporidium*. The unique properties of MNPs, such as their small size, complexity in chemical composition, persistent nature, and uncertainty around their health effects, cause significant challenges in the development of regulations. The proposed regulatory framework attempts to address some of these unprecedented uncertainties by selecting representative MNPs that can capture the diversity of these contaminants. They are expected to be prevalent in source water, usually resistant to treatment, and persistent in the environment. Classifying the diversity of source water allows for the development of simplified regulations targeting these representative MNPs. However, the implementation of the framework in the future entails challenges and risks, such as the emergence of products with unknown chemical composition, financial obstacles, and difficulties in monitoring and establishing new regulations. The research regarding the abundance of MNPs in different water sources, the presence of MNPs with varying characteristics, and their associated health concerns are still emerging. Moreover, technical limitations pose challenges in obtaining sufficient evidence of the abundance of NPs and also smaller-sized MPs. Further qualitative and quantitative research is essential to address these knowledge gaps.

The proposed regulatory framework represents a novel approach in the field of regulating micro-and nano contaminants in drinking water since no existing legislation or framework acknowledges the presence of both MPs and NPs in this context. However, the indirect health risk which incorporates the adsorbed chemical contaminants as well as microbial pathogens associated with MNPs is currently not

considered in the proposed framework. Including this aspect in future work would enhance the comprehensiveness of the framework. Additionally, while the framework is focused on surface water sources, it is recommended to consider groundwater sources in future iterations. Moving forward, it is crucial to provide special attention to formulating some universal guidelines that encompass the monitoring of MNPs abundance, characteristics, toxicological impacts, and dedicated removal techniques from drinking water sources. It is imperative to recognize that a one-size-fits-all solution for MNPs pollution in drinking water is not feasible, as diversity pertaining to specific types, sources, and pathways should be considered. This proposition for the development of a regulatory framework for MNPs in drinking water can be a significant step towards establishing legislation for MNPs in drinking water.

Chapter 6
Conclusions

6.1 Summary of Findings

In response to the growing concern regarding the presence of MNPs in drinking water sources, there were two primary objectives of the thesis. The first one entailed conducting a comprehensive meta-analysis by synthesizing existing data from various research studies that investigate the occurrence of MNPs in drinking water sources and evaluate the effectiveness of different treatment processes in removing them. The second one sought to propose a regulatory approach aimed at mitigating the potential human health impacts associated with MNP exposure in potable water.

The followings are the key findings of this research:

- The presence of MPs in drinking water sources exhibits a wide range of characteristics, including diverse size ranges, shapes, and polymer types. Their abundance varied significantly in influents and effluents across different full-scale DWTPs, with varying MP removal percentages observed among surveys. Despite DWTPs not being optimized for MPs removal, they demonstrated considerable removal capabilities. Advanced DWTPs tended to achieve higher removal rates for all types of MPs (different sizes, shapes, and polymer types) compared to conventional plants, with CFS and filtration processes playing significant roles in MP removal. MPs with varying properties were removed by the treatment processes, but no distinct trends were observed between properties such as size, shape, or polymer type and MP removals. Larger-sized MPs (>1000 μm) generally had higher removal rates. However, no clear relationship could be established between removal percentages through the overall DWTPs and abundances of different size categories of MPs in the influent or effluent of the DWTP. The same was observed for CFS and filtration units in those DWTPs. Limited available data on MPs removal suggests that the shapes and polymer types of MPs did not influence their removal throughout the DWTPs, as well as during CFS and filtration processes.
- Drawing a conclusive statement on the removal efficiency of specific categories of MNPs based on the dataset derived from bench-scale studies is challenging due to the diverse polymeric properties, shapes, sizes, densities, initial concentrations, and surface properties. Moreover, variations in analysis techniques employed among studies can significantly impact reported results of MNP removal by CFS and filtration processes. Al-based coagulants were

generally more effective than Fe-based ones in removing MNPs, although increasing coagulant dosages may not always lead to a substantial increase in removal across different MNPs sizes. Optimal coagulant dosages varied across different bench-scale experiments, depending on MNP properties, coagulant type, and water characteristics. MNPs can be effectively removed regardless of size, shape, polymer type, or other properties, by granular media filtration, based on various small-scale column tests. Limited bench-scale data on membrane filtration demonstrated the effective removal of all types of MNP particles smaller than the membrane's pore size. However, reported bench-scale MP removals by granular media filtration were much higher compared to those observed for full-scale filters in various DWTP surveys.

- The proposed framework for developing regulatory limits for MNPs in drinking water differs from traditional approaches used for other contaminants. Drawing inspiration from the Swiss model for controlling micropollutants in wastewater and drinking water guidelines for *Giardia* and *Cryptosporidium*, the framework recognizes the unique challenges posed by MNPs, including their particulate nature, a wide range of sizes, shapes, and other properties, persistence, and uncertain health effects. To address these challenges, the framework proposes selecting representative MNPs that reflect their diversity and prevalence in source water, resistance to treatment, and persistence in the environment. Simplified regulations can then be developed for representative MNPs, potentially based on the classification of source waters. However, implementing the framework in the future will encounter challenges and risks, such as the emergence of new varieties of MNPs, financial obstacles, and difficulties in monitoring and establishing regulations. Research on MNP abundance, diverse characteristics, and associated health concerns is still emerging, and technical limitations hinder obtaining sufficient evidence, especially for nanoparticles and smaller microplastics.

6.2 Implications for the Water Treatment Industry

Several implications were determined for the drinking water industry based on the above specific conclusions of the study and they include:

- The analysis of existing datasets derived from full-scale surveys and bench-scale studies can provide valuable insights into the diverse range of MNPs present in surface water sources, their behavior in water treatment systems, and their abundance in finished drinking water. This

analysis can increase awareness within the water industry regarding the prevalence of MNPs and also the importance of their removal in the overall water management process.

- The review of the existing findings can help DWTPs to consider MNP removal as part of their overall treatment objectives, such as turbidity removal and the treatment of other emerging contaminants. The limitations in the current treatment processes regarding MNPs removal may prompt the water industry to explore and implement advanced treatment technologies.
- The comprehensive evaluation of existing data and review of the potential toxicity associated with MNPs may influence regulatory agencies to prompt the water industry to monitor MNPs in source and finished drinking water, and potentially initiate establishing guidelines to ensure the protection of public health from the presence of MNPs in drinking water.
- The review of data highlights research gaps and areas requiring further investigation. It may inspire scientists and engineers to develop standardized techniques to characterize MNPs and conduct more studies on the behavior of MNPs during water treatment processes.
- The implementation of the proposed regulatory framework would enable water industries to revise their monitoring schedules, review their operational procedures, and strategically plan to include MNP removals when making necessary investments in new technologies and upgrades. This would involve capital investments and the need for experienced personnel, allowing the industry to proactively prepare for retrofitting treatment facilities in accordance with the regulatory requirements.

6.3 Limitations/Challenges

- MNPs are a diverse class of polymeric particles, consisting of a multitude of different particles with varying properties. This unique property of MNPs makes it challenging to categorize them and develop standardized methods for their detection and analysis. The complexity of MNPs requires a comprehensive understanding of their behavior and interactions to effectively address their presence, potential toxic impacts, and removal during water treatment processes.
- There is a limited number of research publications regarding MPs abundance in drinking water sources and their removal by different treatment processes. Though bench-scale studies provide some valuable insights, the translation of these findings to full-scale operation can be challenging. There is a need for more comprehensive full-scale surveys to assess the

performance of MP removal technologies in real-world conditions, considering the complexities and operational challenges of large-scale water treatment plants. The lack of a sufficient number of studies is posing a challenge in drawing robust conclusions.

- The absence of studies investigating the abundance and removal of NPs at the full-scale level and the presence of only a few studies looking at the bench-scale level hamper the understanding of the occurrence of NPs in drinking water sources and their removal pattern. The primary challenge lies in the small size of the NPs which makes it challenging to develop reliable methodologies to quantify NPs and differentiate them from naturally occurring particles. Current techniques have difficulties investigating particles within the nanometer scale, which may exhibit higher persistence during drinking water treatment processes.
- The existing published studies on MNPs have utilized various sampling and analysis methodologies. Differences in sample collection, preparation, sample volume, extraction techniques, and analytical methods can lead to discrepancies in the reported results. These methodological inconsistencies make the comparison and synthesizing of data very challenging. So, the absence of standardized protocols makes it difficult to draw definitive conclusions from studies and impedes the ability to establish consistent benchmarks for MNP removal in drinking water treatment processes.
- The presence of MNPs in source water exhibits a high degree of diversity regarding sizes, shapes, concentrations, and material properties. The existence of such different types or mixtures of MNPs makes it challenging to generalize the findings across the studies. The removal of MNPs appears to be independent of their varying properties, making it difficult to establish a definitive relationship between these factors.
- The published studies on MNPs encompass various geographic locations of the world, leading to differences in the properties of the water bodies due to varying surrounding conditions. The water characteristics, including the presence of substances such as NOM and divalent cations, can have an impact on the removal of MNPs. These substances have the potential to interact with MNPs and modify their behavior during water treatment processes.
- The MNPs employed in bench-scale studies are often not representative of the actual composition found in the source water. Besides, the use of deionized water in several bench-scale studies introduces variations in properties that differ from those of actual surface water.

These factors contribute to the limited applicability of findings from bench-scale studies to real-world scenarios.

- Insufficient description of the operating conditions and different treatment techniques including CFS and filtration processes in full-scale DWTP surveys, poses a limitation in understanding their performance. Besides, the reported conditions of these treatment processes vary among the published bench-scale studies. For example, the type and dosage of coagulant and coagulant aid, and treatment conditions can influence the efficacy of the CFS system, but the variations in these parameters across studies complicate the comparison of results. Similarly, there is insufficient reporting on the different specific conditions employed in filtration processes. Furthermore, membrane filtration has yet to be fully understood as a means of removing MNPs.
- The proposed regulatory framework is a preliminary approach based on an integrated long-term monitoring and research program. It would require significant investments of time and resources to turn it into reality.

6.4 Future Research/ Recommendations

Throughout this thesis, several areas were identified as requiring further investigation and future research should consider the following aspects:

- More research is warranted to elucidate the influence of different MNPs properties, including morphology, shape, size, surface characteristics, and associated pollutants, on their removal under various conditions. Additionally, a systematic study of their characteristics and water properties (e.g., pH, ions) is necessary to evaluate the efficiency and mechanism of MNPs removal by different conditions of CFS and filtration processes.
- Future research endeavors should prioritize the investigation of the behavior of smaller-sized fibrous or fragmental MNPs (which are commonly encountered in full-scale plants) within various treatment processes and under varying conditions to better reflect real-world scenarios.
- Exploring the fate and transport of NPs in DWTPs should be prioritized as detection and quantification techniques for NPs become more readily available. Also, efforts should be made to standardize and unify quantification methods for MPs, ensuring the development of reliable means of MPs quantification.

- Concurrent turbidity monitoring data from the DWTPs should be included in the reporting process to gauge the performance of CFS and filtration processes in the full-scale DWTPs. It will help to understand the overall effectiveness of the plant for turbidity removal and provide insights into the removal of MNPs under similar typical operational conditions.
- The proposed framework requires a feasibility study to assess its practicality and effectiveness. As it is currently a proposition, there is a substantial amount of work that needs to be done. If the framework is to be implemented, criteria and methods must be established and closely monitored. The process of implementation is expected to be lengthy and would involve various stages.
- The currently proposed framework does not account for the indirect health risks posed by adsorbed chemical contaminants on MNPs. Including this aspect in future work would enhance the overall comprehensiveness of the framework. Additionally, while the framework primarily focuses on surface water sources, incorporating groundwater sources in future iterations will broaden its scope.

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Appendix A

Supplementary Information for Chapter 3

The code used for generating 'Particle Graphing Tool'

```
import matplotlib.pyplot as plt
import pandas as pd
from matplotlib.lines import Line2D
import matplotlib as mpl
import streamlit as st
mpl.rcParams['figure.dpi'] = 400 #increasing the resolution of created
plots



# mpl.rcParams['font.family'] = ['serif']
# mpl.rcParams['font.serif'] = ['Times New Roman']
mpl.rcParams['font.size'] = 11 #setting the font size to 11 pt

class range_set():  #a specific class to represent a set of results with
ranges as individual enteries as opposed to points
    def __init__(self,dataframe): #initialization of class, the subset of
dataset is required for initiation
        self.data = dataframe #loading the sub dataframe as data
        self.DWTP = self.data['DWTPs'][0] #Identifying the Drinking Water
Treatment Plant (DWTP) that the subset represents
        self.Study = self.data['Study'][0] #Identifying the study
(publication) that the subset is extracted from

    def
plot(self,handle:list(),color,marker,offset,alpha,show_trend_line,trend_li
ne_alpha): #The rendering instructions for ploting the data, The plot is
similar to a scatter plot, however, each entery is consisiting of single Y
value and a rang of X values, as opposed to single X value
        x = [] #generating empty arrays (lists) for story x , y points,
the x value here is the average value of the range provided
        y = []
        for index, row in self.data.iterrows(): #For loop to iterate over
the subset enteries
            if row['Removal efficiency'] == 1: #plotting scenario when y
value is 1 (or 100%)
                plt.plot([row['Size range 1'],row['Size range
2']],[row['Removal efficiency']*100 + offset,row['Removal
efficiency']*100+ offset],color,marker='|',alpha=alpha) #plotting a
horizontal line at the removal efficiency of y with "|" marker on each end
to specify the range, the range limists are "size range 1" and "size range
2"
                x.append((row['Size range 2']+row['Size range
1'])/2)#adding the average of the size range as x of the scatter point
                y.append(row['Removal efficiency']*100+offset) #adding the
removal efficiency as y of the scatter point, the point has an offset to
avoid overlaying multiple lines at y = 1 or 100%
```

```
            else: #same process for other y values but without the offset
                plt.plot([row['Size range 1'], row['Size range 2']],
                         [row['Removal efficiency'] * 100, row['Removal
efficiency'] * 100], color, marker='|',alpha=alpha)
                x.append((row['Size range 2'] + row['Size range 1']) / 2)
                y.append(row['Removal efficiency'] * 100)
        if show_trend_line: #if enabled by the user, a dashed trendline
will connect the averages (scatter points) for each subset
            plt.plot(x,y,color=color,linestyle='--
',alpha=trend_line_alpha) #drawing the scatter line
        plt.scatter(x, y, marker=marker, color=color) #plotting the
averages (scatter points) for the subset

        handle.append(Line2D([0], [0], color=color, marker=marker,
linestyle='-', markerfacecolor=color,label=self.DWTP + ', ' +
self.Study))#generating legend handles based on the treatment plant and
study of the subsets


class data_set(): #class for loading and processing data sets
    def __init__(self,path):#class initializing
        self.data_set = pd.read_csv(path, header=0) #read dataset as a
dataframe from the csv
        self.DWTPs = self.data_set['DWTPs'].to_list() #Extract drinking
water treatment plant names from the dataset
        self.DWTPs = list(dict.fromkeys(self.DWTPs)) #Making the entries
in treatment plant names unique (remove all duplicates)
        self.sub_frames = [] #creating an empty list to store sub datasets
based on the treatment plant they originated from
        for item in self.DWTPs: #looping through the treatment plants to
divide the main dataset
            df_temp = self.data_set[self.data_set['DWTPs']==item]
#selecting sub_sets from the main dataset
            df_temp.reset_index(inplace=True) #reset the index on the
subset
            self.sub_frames.append(df_temp) #store the subset

        self.processed_sets = [range_set(s) for s in self.sub_frames]
#process the subsets using the "range_set" function defined earlier

    def plot_all(self,legend_style='outside'): #A class method to render
the plots, "legend_style" argument determines whether the legend is placed
inside or outside the plot's frame
        handles = [] #a list to store legend handles (entries)
        self.colors = (c for c in ['#e6194b', '#3cb44b', '#ffe119',
'#4363d8', '#f58231', '#911eb4', '#46f0f0', '#f032e6', '#bcf60c',
'#fabebe', '#008080', '#e6beff', '#9a6324', '#fffac8', '#800000',
'#aaffc3', '#808000', '#ffd8b1', '#000075', '#808080', '#ffffff',
'#000000','#e6194b', '#3cb44b', '#ffe119', '#4363d8', '#f58231',
'#911eb4', '#46f0f0', '#f032e6', '#bcf60c', '#fabebe', '#008080',
'#e6beff', '#9a6324', '#fffac8', '#800000', '#aaffc3', '#808000',
'#ffd8b1', '#000075', '#808080', '#ffffff', '#000000', '#fabebe',
```

```
'#008080', '#e6beff', '#9a6324', '#fffac8', '#800000', '#aaffc3',
'#808000', '#ffd8b1', '#000075', '#808080', '#ffffff',
'#000000','#e6194b', '#3cb44b', '#ffe119', '#4363d8', '#f58231',
'#911eb4', '#46f0f0', '#f032e6', '#bcf60c', '#fabebe', '#008080',
'#e6beff']) #generator object returning colors for the plot
        self.markers = (m for m in
['v','s','o','d','^','<','>','8','1','3','4','v','s','o','d','^','<','>','
8','1','3','4','v','s','o','d','^','<','>','8','1','3','4']) #a generator
object returning marker styles for scatters
        self.offset = (t for t in [-3.4,-3.3,-3.2,-3.1,-3,-2.9,-2.8,-2.7,-
2.5,-2,-1.5,-1,-0.5,0,0.5,1,1.5,2,2.5,2.7,2.8,2.9,3,3.1,3.2,3.2,3.3,3.4])
#a generator object returning values for 100% offset
        #self.offset = (t for t in [0,-0.5,+0.5,-1,1,-1.5,+1.5])
#depricated offset values
        self.num_keys1 = (k for k in range(1,100)) #generating unique keys
for interface components
        self.num_keys2 = (k for k in range(101,200))
        self.check_keys = (k for k in range(200,300))
        self.text_keys = (k for k in range(300,400))
        self.color_keys = (k for k in range(500, 600))
        self.num_keys3 = (k for k in range(600, 700))
        for set in self.processed_sets: #for loop to generate interface
for each processed set
            with st.expander(label=set.DWTP + ' ' + set.Study + ':
propertise'): #generating a dedicated expander for each sub set
                col1, col2 , col3 , col4 = st.columns(4) #making columns
for organization
                with col1: #refer to streamlit documentation for detailed
explanation of the objects below, the streamlit interface feed the desired
values for color, marker style, offset value and whether a trendline
should be drawn to the class methods above
                    color = st.color_picker(label='Color',
value=next(self.colors),key=next(self.color_keys))
                    trend = st.checkbox(label='Show trend-
line',value=True,key=next(self.check_keys))
                with col2:
                    marker = st.text_input(label='Marker
style',value=next(self.markers),key=next(self.text_keys))
                with col3:
                    offset = st.number_input(label='Offset at
100%',value=float(next(self.offset)),step=0.1,key=next(self.num_keys3))
                with col4:
                    alpha =
st.number_input(label='Transparency',value=1.0,min_value=0.2,max_value=1.0
,step=0.1,key=next(self.num_keys1))
                    t_alpha = st.number_input(label='Trend-line
Transparency',value=1.0,min_value=0.2,max_value=1.0,step=0.1,key=next(self
.num_keys2))


set.plot(handle=handles,color=color,marker=marker,offset=offset,show_trend
_line=trend,alpha=alpha,trend_line_alpha=t_alpha) #calling the "plot"
```

```
method of the class to render the plot using the inputs from the interface
(objects above) as arguments
        if legend_style == 'outside': #deciding where to locate the legend
using the logic flow below
            plt.legend(handles=handles,fontsize=11,loc='upper
center',bbox_to_anchor=[0.5,-0.15]) #bbox_to_anchor=[1.6,0.5]

        elif legend_style == 'inside':
            plt.legend(handles=handles,fontsize=9)
        else:
            pass

#%%
#initializing the main interface using Streamlit (refer to Streamlit
documentation, when in doubt :) )

st.set_page_config(layout='wide',page_title='Graphing tool')  #configuring
the page style
st.title('Graphing tool for particle removal efficiency data') #setting a
title for the app
st.caption('Developed by Sina Golchi for NSERC Chair in Water Treatment')
#a tiny caption

with st.sidebar: #rendering interface components in the sidebar
    csv = st.file_uploader(label='Upload CSV file',) #loading the csv file
containing the main datasets using the file loader


dft = data_set(csv) #process the CSV file using the data_set class

with st.sidebar: #getting some input from the user regarding the axis of
the plot
    st.markdown('''___''')
    st.caption('Axis options')
    col1, col2  = st.columns(2)
    with col1:
        xmin = st.number_input(label='x min',value=-5) #acquiring the
minimum of x axis
        ymin = st.number_input(label='y min', value=-1) #acquiring the
maximum of x axis
    with col2:
        xmax = st.number_input(label='x max',value=100) #acquiring the
minimum of y axis
        ymax = st.number_input(label='y max', value=110) #acquiring the
maximum of y axis
    xlog = st.checkbox(label='Logarithmic x axis') #whether the x axis
type to be set as logarithmic
    ylog = st.checkbox(label='Logarithmic y axis') #whether the y axis
type to be set as logarithmic

    st.markdown('''___''')
    st.caption('Legend option')
```

```
    L_type = st.selectbox(label='Legend
type',options=['outside','inside','no legend']) #acquiring the legend
style

# try:
dft.plot_all(legend_style=L_type) #calling the plot_all method to render
the main plot

if xlog:
    plt.xscale('log') #setting the x scale logarithmic if requested
if ylog:
    plt.yscale('log') #setting the y scale logarithmic if requested

plt.xlim(xmin, xmax) #setting the limits of the x axis
plt.ylim(ymin, ymax) #setting the limits of the x axis

plt.xlabel(r'Particle size range [$ \rm \mu m$]') #setting the x axis
label
plt.ylabel('Removal efficiency [%]') #setting the y axis label
plt.grid(axis='y',alpha=0.4) #setting up a grid
plt.gcf().set_size_inches(7,4) #setting the size of the graph

st.pyplot(plt.gcf()) #passing the matplotlib (plot) object to the
interface for final visualization
```

Table A1: MPs abundance in the influent and effluent of the selected full-scale DWTPs, with overall removal efficiency of MPs by these DWTPs, irrespective of differences in MPs size ranges, shapes, polymer types, and any other properties in general

Reference	**MPs abundance in the influent of DWTPs (MPs/L)**	**MPs abundance in the effluent of DWTPs (MPs/L)**	**Overall removal efficiency by DWTPs (%)**
Jung et al., 2022	2.2 ± 1.3	0.02 ± 0.02	99.1
Wu et al., 2022	3738.36 ± 461.29	695.66 ± 39.32	81.18
Radityaningrum et al., 2021 (SDWTP-I)	26.8	12.3	54
Radityaningrum et al., 2021 (SDWTP-II)	35	8.5	76
Shen et al., 2021	2753* ± 161*	352* ± 15*	87.22*
Babel & Dork, 2021 (WTP1)	1180.5 ± 158	521 ± 61	55.9
Babel & Dork, 2021 (WTP2)	1463 ± 126	617 ± 147	57.8
Dalmau-Soler et al., 2021	0.96 ± 0.46	0.06 ± 0.04	93 ± 5
Sarkar et al., 2021	17.86 ± 2.66	2.75 ± 0.92	85
Kankanige & Babel, 2021 (Dry season)	1385 ± 192.82*	448.7 ± 106.38*	67.6
Kankanige & Babel, 2021 (Rainy season)	1796.6 ± 199.47*	769.4 ± 106.38*	57.2
Pivokonsky et al., 2020 (DWTP-Milence)	23 ± 2	14 ± 1	39.13 (40)
Pivokonsky et al., 2020 (DWTP-Plzeň)	1296 ± 35	151 ± 4	88.34 (88)
Wang et al., 2020	6614 ± 1132	930 ± 71	82.1–88.6 (85.35*)
Pivokonsky et al., 2018 (WTP1)	1473 ± 34	443 ± 10	70
Pivokonsky et al., 2018 (WTP2)	1812 ± 35	338 ± 76	81
Pivokonsky et al., 2018 (WTP3)	3605 ± 497	628 ± 28	83

**calculated from the survey*

Table A2: MPs abundance in the influent and effluent of the CFS unit of the selected full-scale DWTPs, with removal efficiency of MPs by those CFS units

(The values were reported irrespective of the differences in MPs size ranges, shapes, polymer types, and any other properties in general)

Reference	**MPs abundance in the influent of CFS unit (MPs/L)**	**MPs abundance in the effluent of CFS unit (MPs/L)**	**Removal by CFS (%)**
Jung et al., 2022	2.3±0.8	1.4±0.15	39.13*
Wu et al., 2022	7789.69	4487.87	42.5*
Radityaningrum et al., 2021 (SDWTP-I)	–	–	–
Radityaningrum et al., 2021 (SDWTP-II)	–	–	–
Shen et al., 2021	–	–	–
Babel & Dork, 2021 (WTP1)	1180.5	826	30.03*
Babel & Dork, 2021 (WTP2)	1463	987.5	32.5*
Dalmau-Soler et al., 2021	–	–	–
Sarkar et al., 2021	17.88	6.99	60.91*
Kankanige & Babel, 2021 (Dry season)	1298.5	823.2	36.6
Kankanige & Babel, 2021 (Rainy season)	1669.2	1222.9	26.7
Pivokonsky et al., 2020 (DWTP-Milence)	–	–	–
Pivokonsky et al., 2020 (DWTP-Plzeň)	–	–	62
Wang et al., 2020	6614	3472*	47.5*
Pivokonsky et al., 2018 (WTP1)	–	–	–
Pivokonsky et al., 2018 (WTP2)	–	–	–
Pivokonsky et al., 2018 (WTP3)	–	–	–

**calculated from the survey*

Table A3: MPs abundance in the influent and effluent of the filtration unit of selected full-scale DWTPs, with removal efficiency of MPs by those filtration units

(The values were reported irrespective of the differences in MPs size ranges, shapes, polymer types, and any other properties in general)

Reference	**MPs abundance in the influent of filtration unit (MPs/L)**	**MPs abundance in the effluent of filtration unit (MPs/L)**	**Removal by filtration (%)**
Jung et al., 2022	1.4±0.15	0.2±0.15	85.71*
Wu et al., 2022	1634.12	1361.57	16.68*
Radityaningrum et al., 2021 (SDWTP-I)	18.5	13.8	25.41*
Radityaningrum et al., 2021 (SDWTP-II)	–	–	–
Shen et al., 2021	–	–	–
Babel & Dork, 2021 (WTP1)	826	578	30.02*
Babel & Dork, 2021 (WTP2)	987.5	693	29.82*
Dalmau-Soler et al., 2021	–	–	–
Sarkar et al., 2021	–	–	–
Kankanige & Babel, 2021 (Dry season)	823.2	536.7	34.8
Kankanige & Babel, 2021 (Rainy season)	1222.9	844.6	30.9
Pivokonsky et al., 2020 (DWTP-Milence)	–	–	–
Pivokonsky et al., 2020 (DWTP-Plzeň)	–	–	20
Wang et al., 2020	3472*	2230*	36*
Pivokonsky et al., 2018 (WTP1)	–	–	–
Pivokonsky et al., 2018 (WTP2)	–	–	–
Pivokonsky et al., 2018 (WTP3)	–	–	–

**calculated from the survey*

Table A4: MPs abundance in the influent and effluent of the DWTPs, with their removal efficiency for every size group individually

(where a: 1-10 μm, b: 10-20 μm, c: 20-50 μm, d: 50-100 μm, e: 100-5000 μm)

Reference	Data type	Size range (μm)	Size category	MPs abundance in the influent of DWTPs (MPs/L)	MPs abundance in the effluent of DWTPs (MPs/L)	Removal efficiency by DWTPs (%)
Jung et al., 2022*	DWTP-A	10-20	b	0.02	0	100
		20-50	c	0.55	0	100
		50-100	d	1	0	100
		>100	e	0.8	0	100
	DWTP-B	10-20	b	0.01	0	100
		20-50	c	0.4	0	100
		50-100	d	1	0	100
		>100	e	0.72	0	100
	DWTP-C	10-20	b	0	0	100
		20-50	c	0.5	0	100
		50-100	d	0.86	0	100
		>100	e	0.92	0	100
Wu et al., 2022	Granular MPs	5-10	a	2760.14	379.24	86
		10-20	b	933.34	316.03	66
		20-100	c-d	41.8	0.31	99
		100-500	e	2.88	0.07	98
		500-1000	e	0.13	0.02	88
		1000-5000	e	0.1	0	100
	Fibrous MPs	100-500	e	0.61	0	100
		500-1000	e	0.76	0	100
		1000-5000	e	1.43	0	100
Radityaningrum et al., 2021	SDWTP-1	1-100	a-d	0.5	0.3	40
		101-350	e	4.5	1.3	71
		351-1000	e	13.5	8.5	37
		1000-5000	e	8.3	2.2	72
	SDWTP-2	1-100	a-d	1.5	0.7	53

		101-350	e	4.9	2.8	43
		351-1000	e	15.8	3	80
		1000-5000	e	12.8	2	84
Shen et al., 2021*	DWTP	1-10	a	2022.65	245.53	88
		10-50	b-c	333.33	62.89	81
		50-100	D	207.12	27.39	87
		>100	e	184.47	15.05	92
Babel & Dork, 2021	WTP-1	6.5-20	a-b	415.5	190	54
		20-53	c	386	169	56
		53-500	d-e	246	103	58
		>500	e	133	59	56
	WTP-2	6.5-20	a-b	630	259	59
		20-53	c	415	179	57
		53-500	d-e	289	123	57
		>500	e	129	56	57
Sarkar et al., 2021*	DWTP	<25	a-c	0.62	0	100
		25-50	c	3.16	0.21	93
		50-100	d	5.6	1.65	71
		>100	e	9.42	1.03	89
Kankanige & Babel, 2021	Dry season	6.5-53	a-c	506.5	202.5	60
		53-300	d-e	472.2	162.4	66
		300-500	e	244.4	51.7	79
		≥500	e	161.9	32.1	80
	Rainy season	6.5-53	a-c	609	323.5	47
		53-300	d-e	702.3	281.2	60
		300-500	e	312.6	105.7	66
		≥500	e	172.1	59	66
Pivokonsky et al., 2020*	DWTP-Milence: Fibers	≥5 to <10	a	-	-	-
		≥10 to <50	b-c	0.57	0.11	81
		≥50 to <100	d	1.53	0.32	79
		≥100	e	2.46	2.35	5
		≥1 to <5	a	11.63	8.6	26
		≥5 to <10	a	4.53	1.66	63

	DWTP-Milence: Fragments	≥10 to <50	b-c	1.72	0.74	57
		≥50 to <100	d	0.63	0	100
		≥100	e	0.34	0	100
	DWTP-Plzen: Fibers	≥5 to <10	a	5.38	0	100
		≥10 to <50	b-c	34.24	6.15	82
		≥50 to <100	d	58.84	2.31	96
		≥100	e	26.92	3.46	87
	DWTP-Plzen: Fragments	≥1 to <5	a	600	84	86
		≥5 to <10	a	356	44	88
		≥10 to <50	b-c	104	12	89
		≥50 to <100	d	88	0	100
		≥100	e	28	0	100
Wang et al., 2020	ADWTP	1-5	a	3760	793	79
		5-10	a	1520	136	91
		10-50	b-c	731	1	100
		50-100	d	379	0	100
		>100	e	224	0	100
Pivokonsky et al., 2018*	WTP-1	1-5	a	791	268	66
		5-10	a	468	133	72
		10-50	b-c	145	43	70
		50-100	d	78	0	100
		>100	e	-	-	-
	WTP-2	1-5	a	1070	104	90
		5-10	a	591	164	72
		10-50	b-c	156	69	56
		50-100	d	-	-	-
		>100	e	-	-	-
	WTP-3	1-5	a	1683	291	83
		5-10	a	1293	274	79
		10-50	b-c	390	61	85
		50-100	d	212	0	100
		>100	e	45	0	100

**calculated the abundance and removal efficiency from the provided data of the survey*

Table A5: MPs abundance in the influent and effluent of the CFS unit of DWTPs, with their removal efficiency for every size group individually

(where a: 1-10 μm, b: 10-20 μm, c: 20-50 μm, d: 50-100 μm, e: 100-5000 μm)

Reference	**Data type**	**Size range (μm)**	**Size category**	**MPs abundance in the influent of CFS unit (MPs/L)**	**MPs abundance in the effluent of CFS unit (MPs/L)**	**Removal efficiency by CFS (%)**
Jung et al., 2022*	DWTP-A	10-20	b	-	-	-
		20-50	c	0.25	0.08	68
		50-100	d	0.32	0	100
		>100	e	0.18	0	100
	DWTP-B	10-20	b	-	-	-
		20-50	c	0.5	0.1	80
		50-100	d	0.18	0	100
		>100	e	0.23	0	100
	DWTP-C	10-20	b	-	-	-
		20-50	c	0.43	0.12	72
		50-100	d	0.3	0.08	73
		>100	e	0.3	0	100
Wu et al., 2022	Granular MPs	5-10	a	4788.12	3233.76	32
		10-20	b	2972.34	1204.65	59
		20-100	c-d	-	-	-
		100-500	e	3.25	0.29	91
		500-1000	e	0.05	0.03	40
		1000-5000	e	-	-	-
	Fibrous MPs	100-500	e	0.43	0.15	65
		500-1000	e	-	-	-
		1000-5000	e	-	-	-
Babel & Dork, 2021	WTP-1	6.5-20	a-b	415.5	277.5	33
		20-53	c	386	259	33
		53-500	d-e	246	185	25
		>500	e	133	104.5	21
	WTP-2	6.5-20	a-b	630	395	37

		20-53	c	415	268	35
		53-500	d-e	289	220.5	24
		>500	e	129	104	19
Kankanige & Babel, 2021	Dry season	6.5-53	a-c	490.7	301.6	38.5
		53-300	d-e	468.4	315.4	32.7
		300-500	e	200.6	118.2	41.1
		≥500	e	138.8	88	36.6
	Rainy season	6.5-53	a-c	591.5	391.6	33.8
		53-300	d-e	651.4	484.4	25.6
		300-500	e	275.6	219.8	20.2
		≥500	e	150.7	127.1	15.7
Pivokonsky et al., 2020*	DWTP-Plzen: Fibers	≥5 to <10	a	5.38	2.31	57
		≥10 to <50	b-c	34.23	13.85	60
		≥50 to <100	d	58.85	22.31	62
		≥100	e	26.92	10.77	60
	DWTP-Plzen: Fragments	≥1 to <5	a	600	232	61
		≥5 to <10	a	356	132	63
		≥10 to <50	b-c	104	40	62
		≥50 to <100	d	88	32	64
		≥100	e	28	20	29
Wang et al., 2020*	ADWTP	1-5	a	3760.33	2650.33	30
		5-10	a	1519.67	623.67	59
		10-50	b-c	731	177	76
		50-100	d	379	14.33	96
		>100	e	224	6.67	97

**calculated the abundance and removal efficiency from the provided data of the survey*

Table A6: MPs abundance in the influent and effluent of the filtration unit of DWTPs, with their removal efficiency for every size group individually

(where a: 1-10 μm, b: 10-20 μm, c: 20-50 μm, d: 50-100 μm, e: 100-5000 μm)

Reference	Data type	Size range (μm)	Size category	MPs abundance in the influent of filtration unit (MPs/L)	MPs abundance in the effluent of filtration unit (MPs/L)	Removal efficiency by filtration (%)
Wu et al., 2022	Granular MPs	5-10	a	1135.04	842.26	26
		10-20	b	-	-	-
		20-100	c-d	24.04	1.66	93
		100-500	e	0.73	0.08	89
		500-1000	e	0.05	0.03	40
		1000-5000	e	0.1	0	100
	Fibrous MPs	100-500	e	0.11	0	100
		500-1000	e	0.9	0.02	78
		1000-5000	e	0.08	0.06	25
Radityaningrum et al., 2021	SDWTP-1	1-100	a-d	-	-	-
		101-350	e	5	3.5	30
		351-1000	e	8.5	5.5	35
		1000-5000	e	4	2.8	30
Babel & Dork, 2021	WTP-1	6.5-20	a-b	277.5	209.5	25
		20-53	c	259	192	26
		53-500	d-e	185	112.5	39
		>500	e	104.5	64	39
	WTP-2	6.5-20	a-b	395	293	26
		20-53	c	268	203	24
		53-500	d-e	220.5	136	38
		>500	e	104	61	41
Kankanige & Babel, 2021	Dry season	6.5-53	a-c	301.6	220.7	26.8
		53-300	d-e	315.4	225.6	28.5
		300-500	e	118.2	55.8	52.8
		≥500	e	88	34.6	60.7
		6.5-53	a-c	391.6	341.9	12.7

	Rainy season	53-300	d-e	484.4	327.2	32.5
		300-500	e	219.8	114.6	47.9
		≥500	e	127.1	60.9	52.1
Pivokonsky et al., 2020*	DWTP-Plzen: Fibers	≥5 to <10	a	-	-	-
		≥10 to <50	b-c	13.85	12.31	11
		≥50 to <100	d	22.31	5.38	76
		≥100	e	10.77	5.38	50
	DWTP-Plzen: Fragments	≥1 to <5	a	232	124	47
		≥5 to <10	a	132	64	52
		≥10 to <50	b-c	40	28	30
		≥50 to <100	d	32	0	100
		≥100	e	20	0	100
Wang et al., 2020*	ADWTP	1-5	a	2650.33	1896.33	28
		5-10	a	623.67	311.67	50
		10-50	b-c	177	22	88
		50-100	d	14.33	0	100
		>100	e	6.67	0	100

**calculated the abundance and removal efficiency from the provided data of the survey*

The Figures (A1-A6) with trend lines:

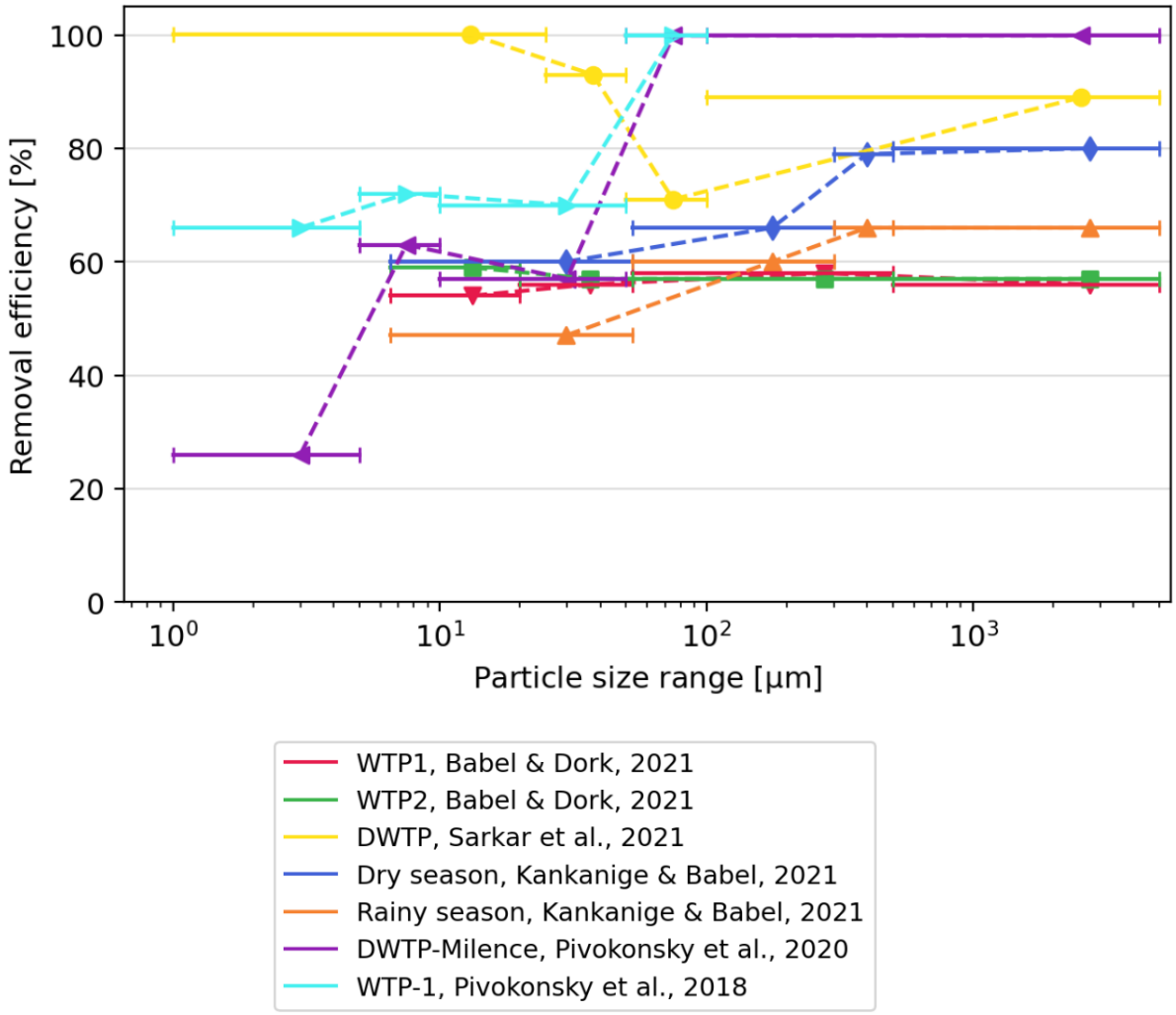


Figure A1: Overall removal efficiency of MPs for different size ranges by conventional DWTPs

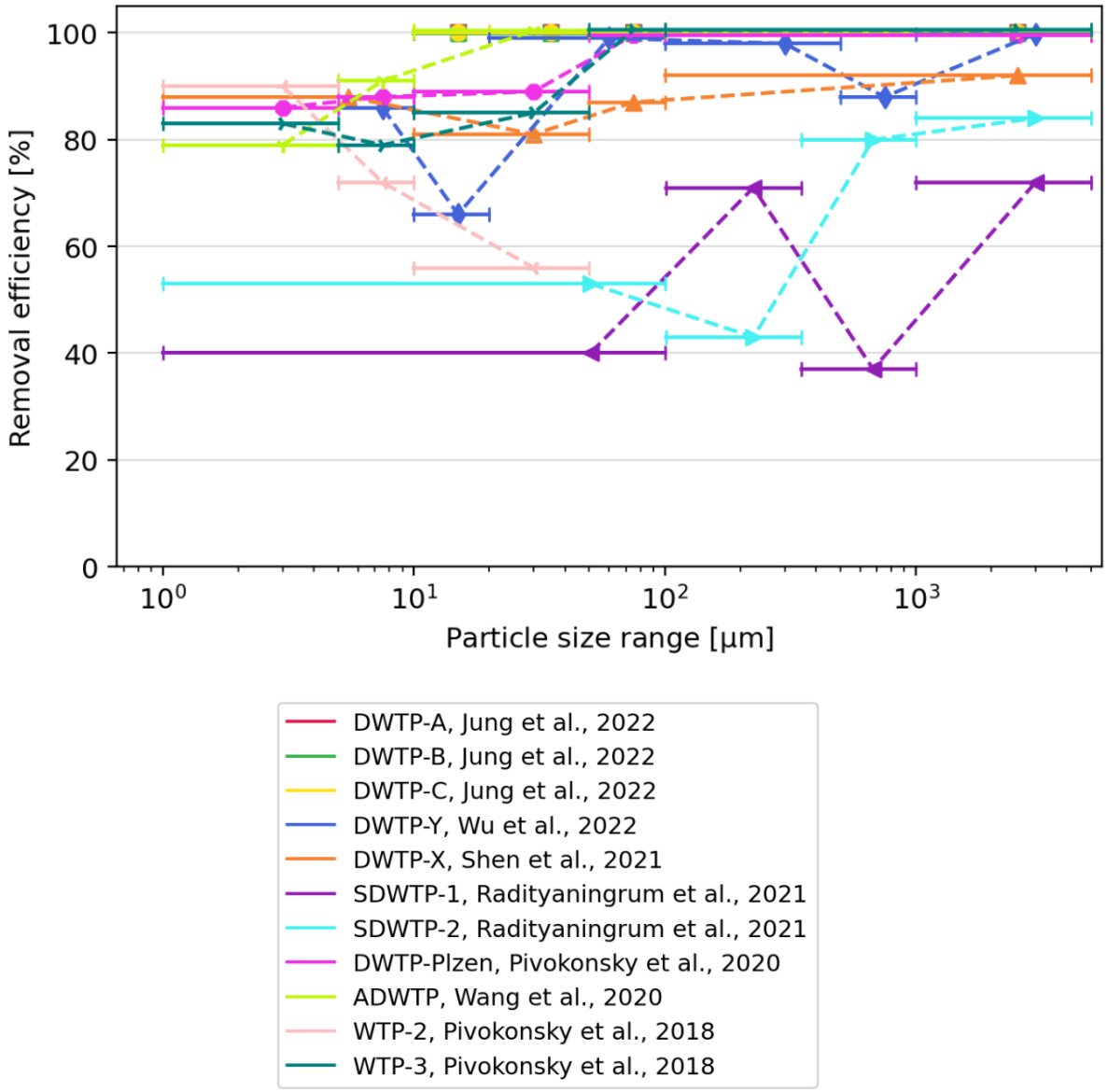


Figure A2: Overall removal efficiency of MPs for different size ranges by advanced DWTPs

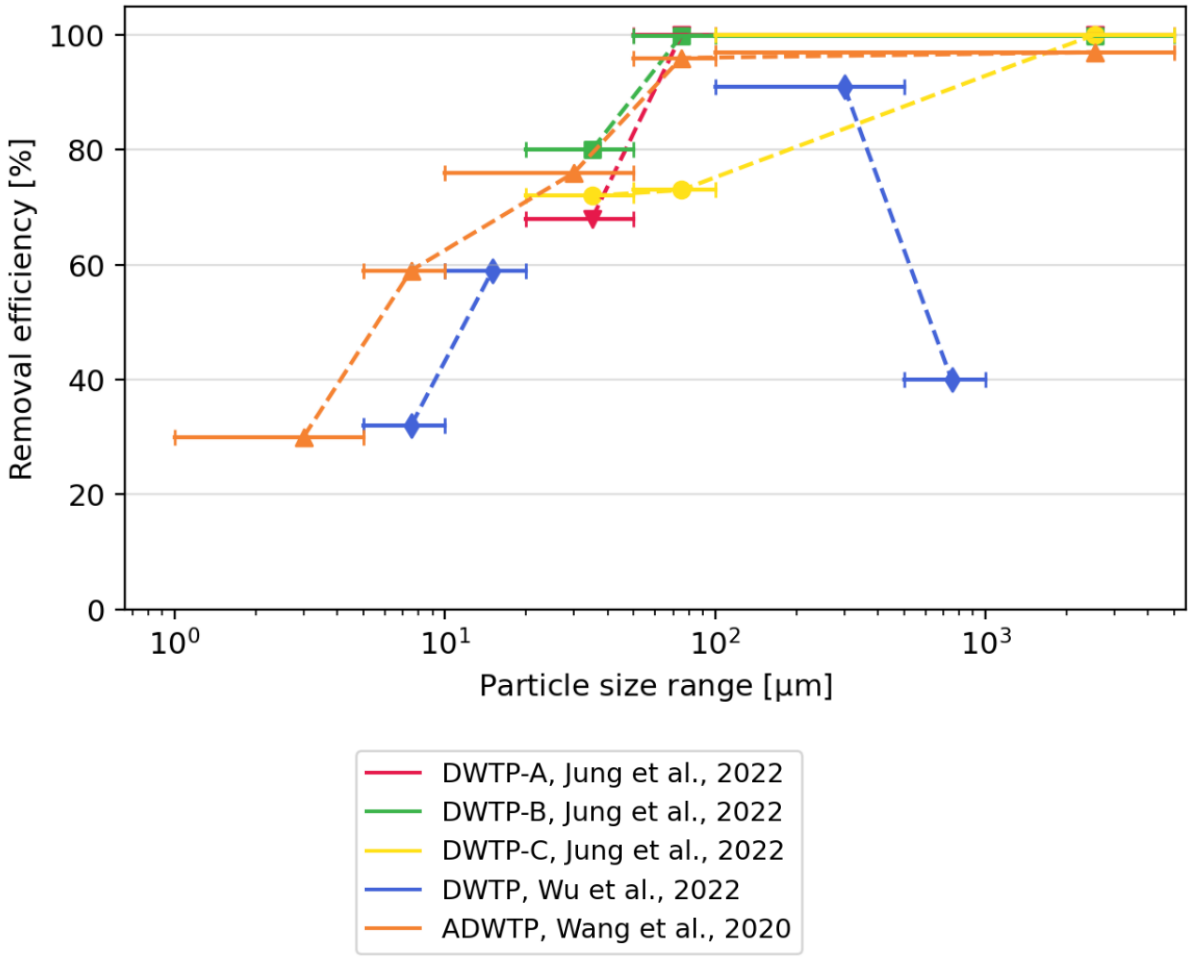


Figure A3: Removal efficiency of MPs for different size ranges by CFS unit of different DWTPs (upward trend)

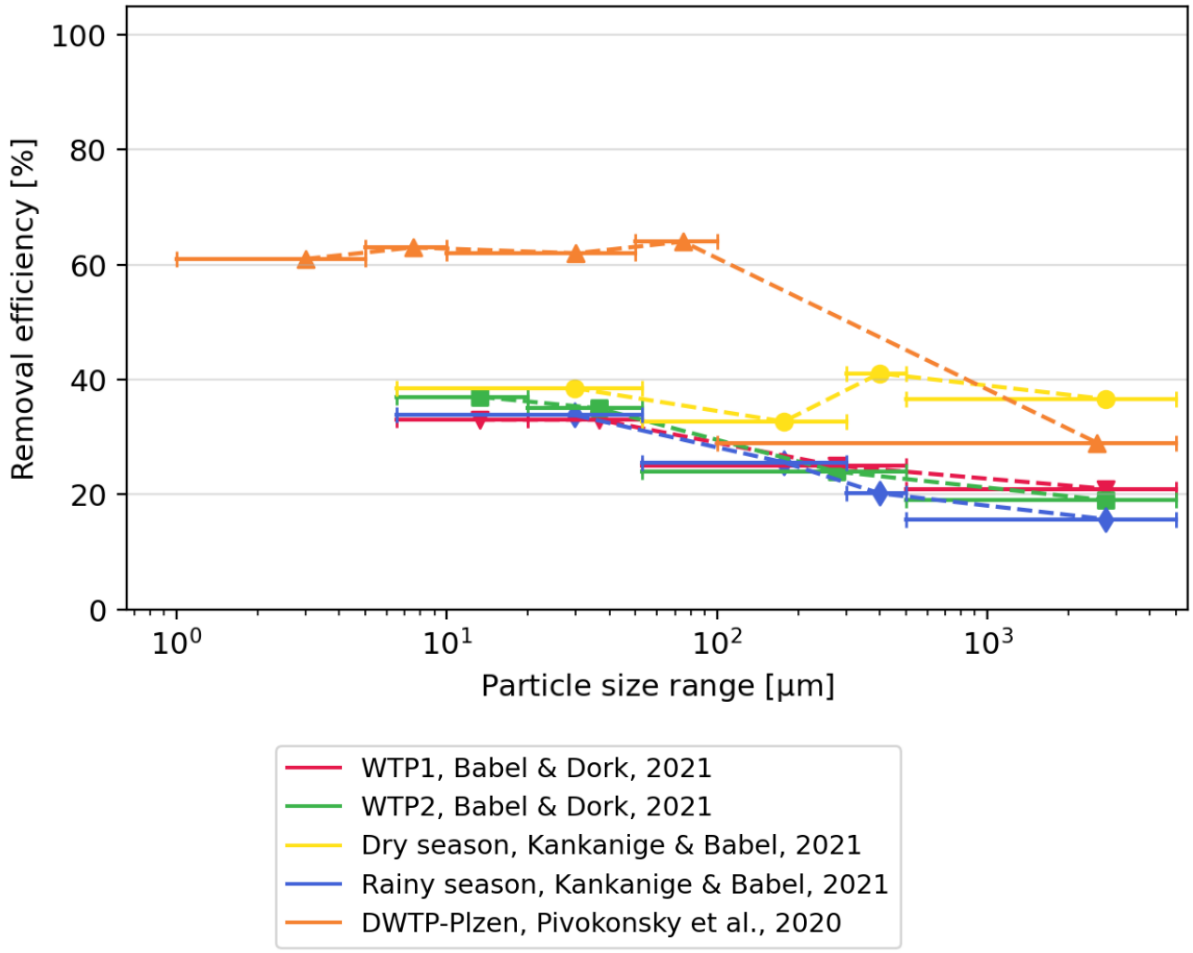


Figure A4: Removal efficiency of MPs for different size ranges by CFS unit of different DWTPs (downward trend)

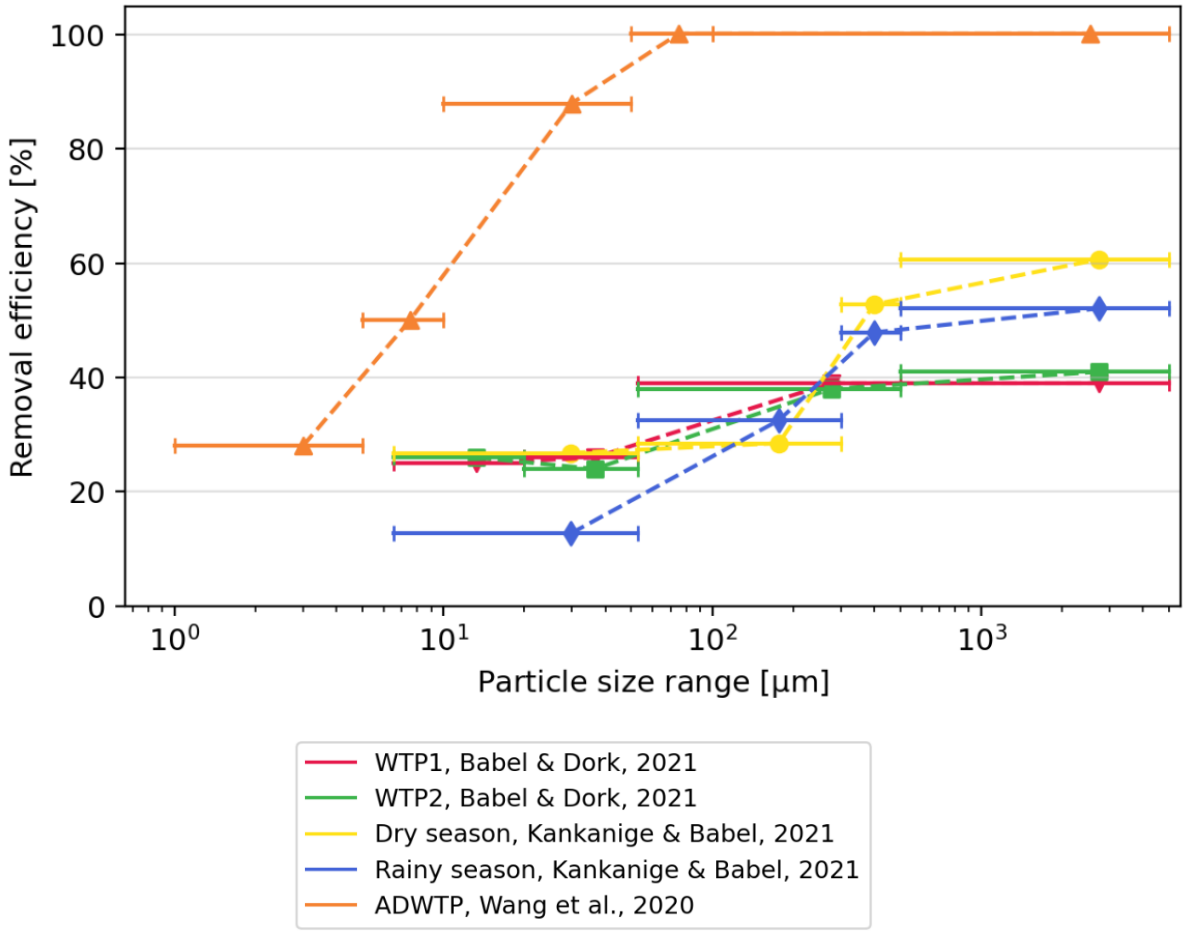


Figure A5: Removal efficiency (based on filter influent MP numbers) of MPs for different size ranges by filtration unit of different DWTPs (upward trend)

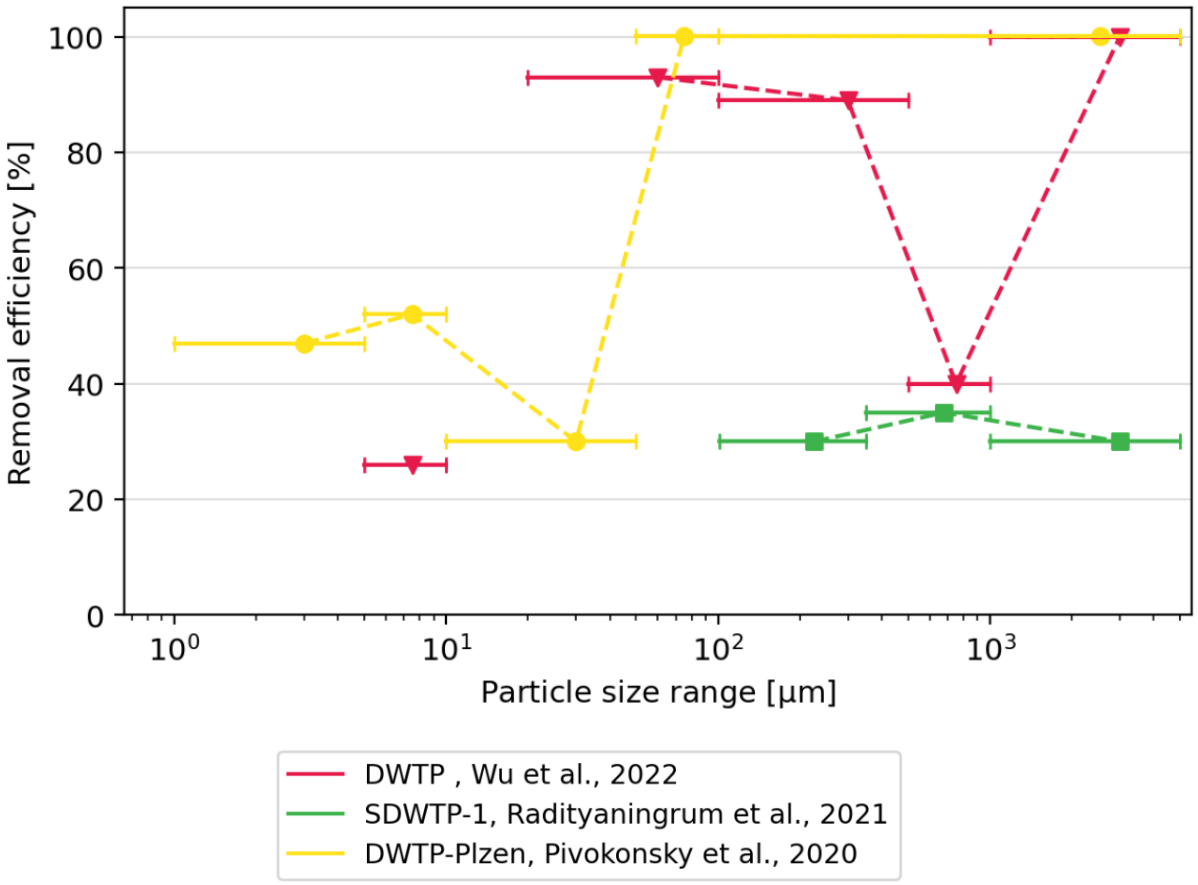


Figure A6: Removal efficiency (based on filter influent MP numbers) of MPs for different size ranges by filtration unit of different DWTPs (downward trend)

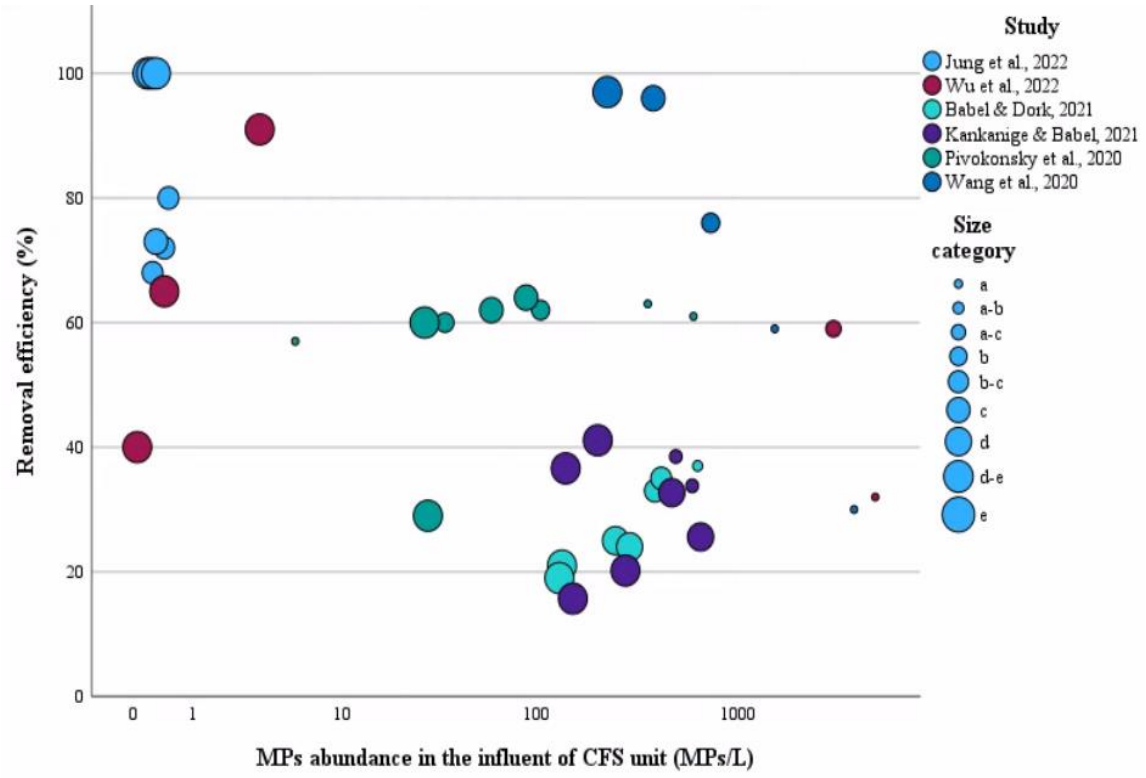


Figure A7: Relation between MPs removal efficiency by CFS process and their abundance in the influent of those CFS units of full-scale DWTPs for different size categories of MPs *(where a: 1-10 µm, b: 10-20 µm, c: 20-50 µm, d: 50-100 µm, e: 100-5000 µm)*

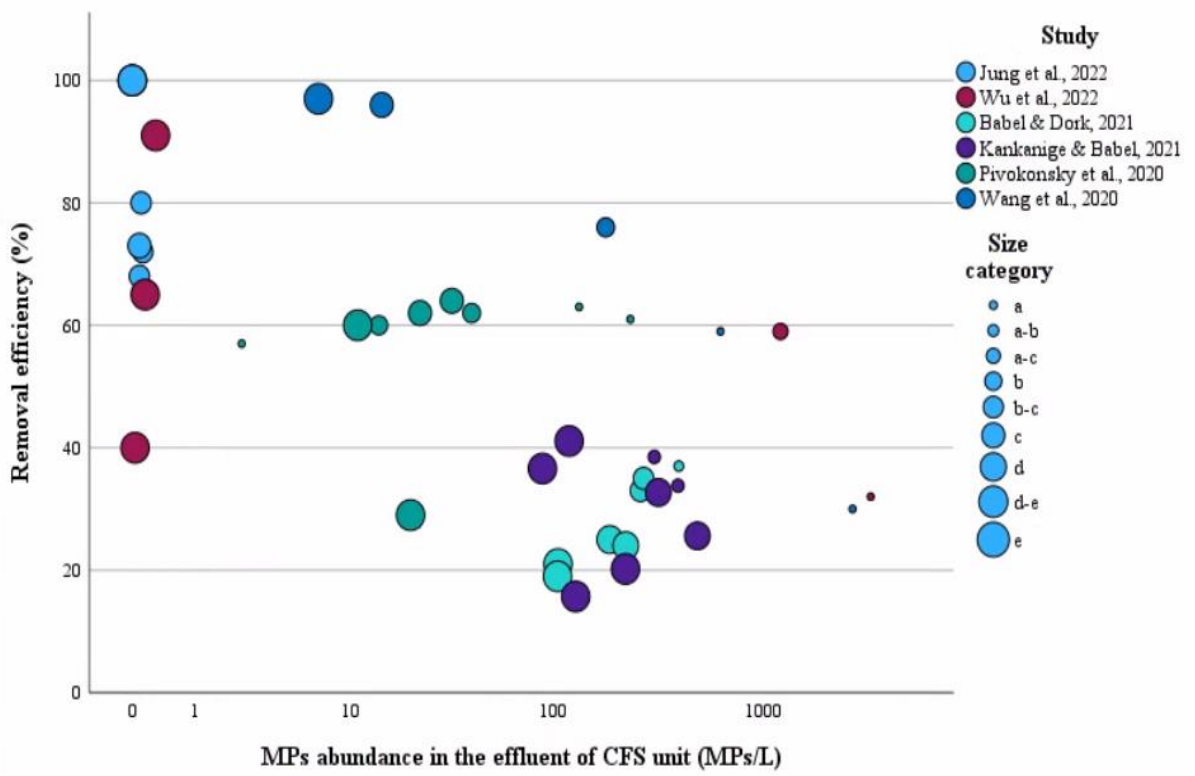


Figure A8: Relation between MPs removal efficiency by CFS process and their abundance in the effluent of those CFS units of full-scale DWTPs for different size categories of MPs *(where a: 1-10 µm, b: 10-20 µm, c: 20-50 µm, d: 50-100 µm, e: 100-5000 µm)*

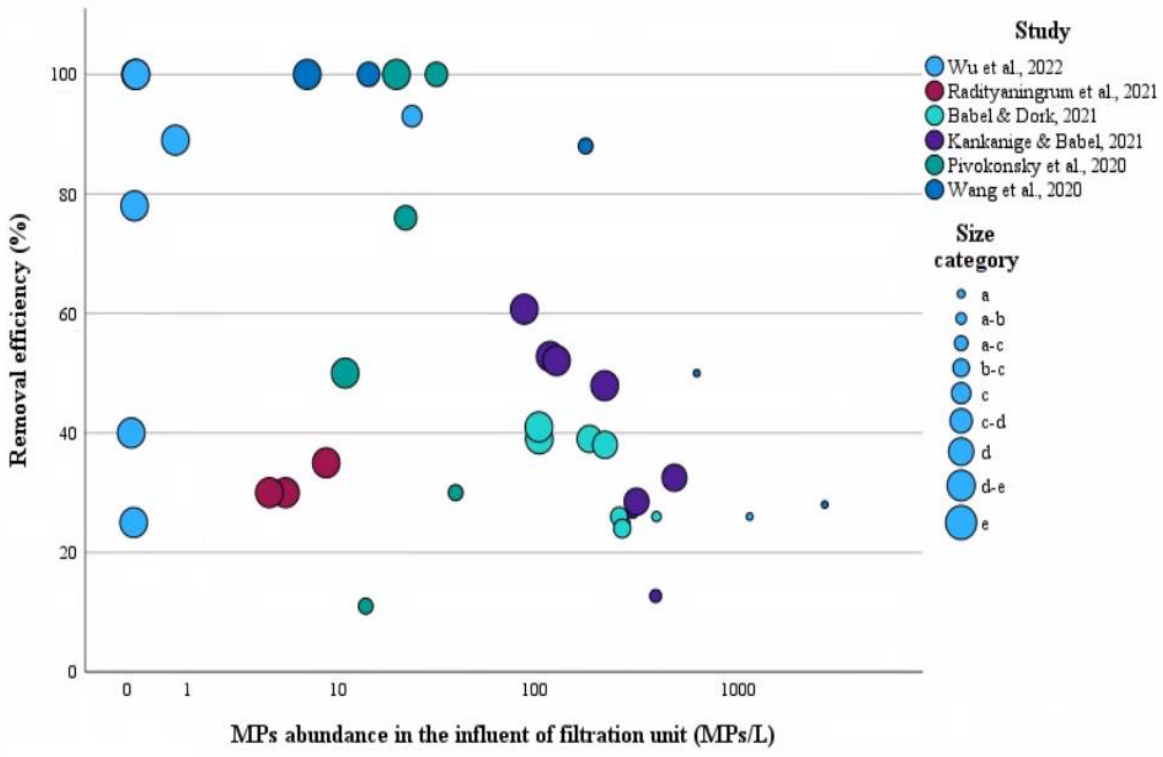


Figure A9: Relation between MPs removal efficiency (based on filter influent MP numbers) by filtration processes and their abundance in the influent of those filtration units of full-scale DWTPs for different size categories of MPs

(where a: 1-10 µm, b: 10-20 µm, c: 20-50 µm, d: 50-100 µm, e: 100-5000 µm)

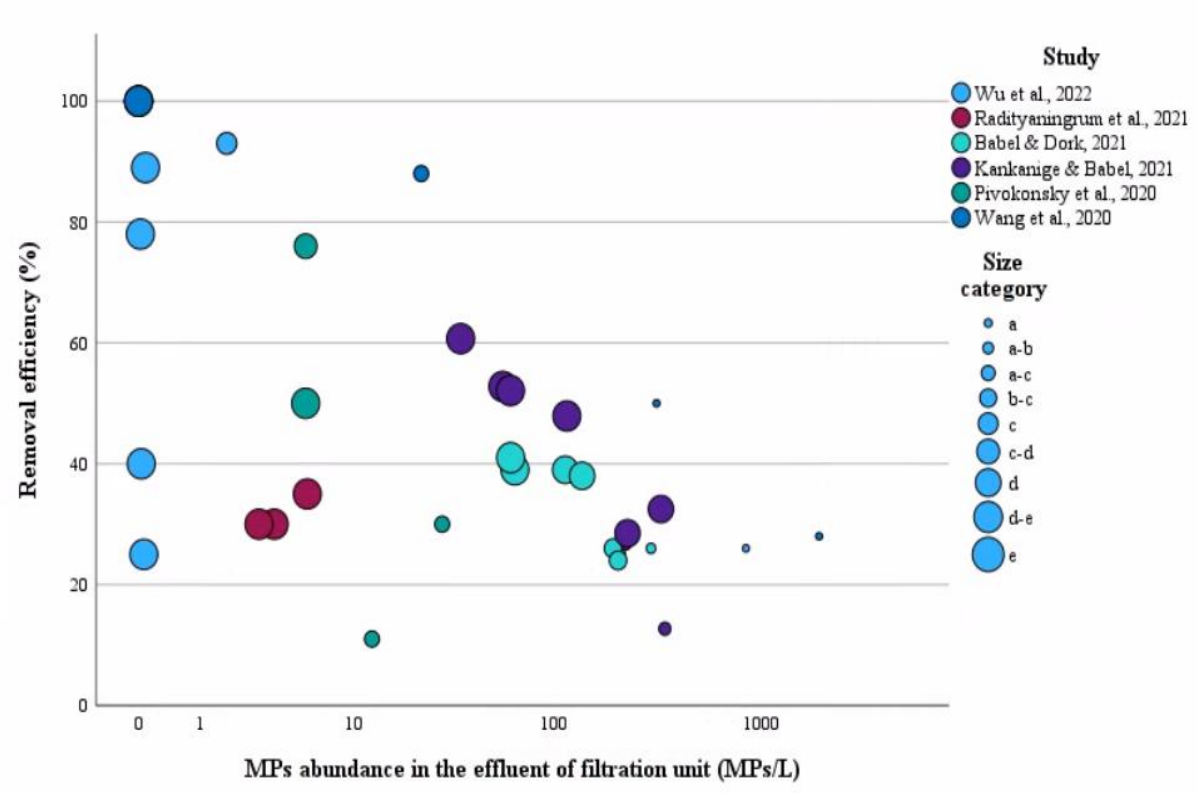


Figure A10: Relation between MPs removal efficiency (based on filter influent MP numbers) by filtration process and their abundance in the effluent of those filtration units of full-scale DWTPs for different size categories of MPs

(where a: 1-10 µm, b: 10-20 µm, c: 20-50 µm, d: 50-100 µm, e: 100-5000 µm)

Appendix B

Supplementary Information for Chapter 4

Table B1: Removal efficiency of different-sized MPs by CFS using optimized dosages of Al-based coagulant in synthetic water irrespective of MP polymer types, shapes, and other properties [Dataset for Figure 4.1 (a)]

No	Study	MPs Size (µm)	Removal efficiency (%)	Coagulation condition
1	Zhang et al. (2022)	1	89.3	PACl 400 mg/L
		1	98.5	PACl 400 mg/L & PAM 20 mg/L
2	Zhang et al. (2021)*	1-100	33	PACl 50 mg/L
		100-400	42	
		400-500	99	
		1-100	89	PACl 50 mg/L & PAM 50 mg/L
		100-400	81	
		400-500	100	
		1-100	68	PACl 50 mg/L & SA 50 mg/L
		100-400	69	
		400-500	100	
		1-100	75	PACl 50 mg/L & ASA 50 mg/L
		100-400	67	
		400-500	96	
3	Shao et al. (2020)	250-300	15.8	PACl 30 mg/L
4	Shahi et al. (2020)*	10-30	52	Alum 30 mg/L
		30-50	100	
		50-70	100	
		70-100	100	
5	Xia et al. (2020)	1-5	75	$AlCl_3$ 250 mg/L
11	Bayarkhuu & Byun (2022)	1	10	PACl 0.5 mg/L
		10	60	PACl 9 mg/L
12	Zhou et al. (2021)*	1-500	75	PACl 60 mg/L (PS)
		500-1000	99	
		1000-5000	97	
		1-500	30	PACl 90 mg/L (PE)
		500-1000	7	
		1000-5000	4	
13	Ma, Xue, Hu, et al. (2019)*	1-500	40	$AlCl_3$ 135 mg/L
		500-1000	24	
		1000-2000	16	
		2000-5000	9	
14	Na et al. (2021)*	10	41	$AlCl_3$ 10 mg/L
		20	90	
		45	94	
		90	95	

**calculated the removal efficiency from the provided data of the study*

Table B2: Removal efficiency of different-sized MPs by CFS using optimized dosages of Al-based coagulant in surface water irrespective of MP polymer types, shapes, and other properties [Dataset for Figure 4.1 (b)]

No	Study	MPs Size (µm)	Removal efficiency (%)	Coagulation condition
7	C. Li et al. (2021)	100	98.9	$Al_2(SO_4)_3.18H_2O$ - 3.4 mg Al/L
8	Xue et al. (2021)*	3	86	Alum 30 mg/L (R)
		6	77	
		25	78	
		45	64	
		90	45	
		3	97	Alum 30 mg/L (L)
		6	86	
		25	91	
		45	89	
		90	64	
9	Lapointe et al. (2020)	90	99	Alum 2.73 mg Al/L & PAM 0.3 mg/L (PES)
		140	84	Alum 2.73 mg Al/L & PAM 0.3 mg/L (PS)
		15	89	Alum 2.73 mg Al/L & PAM 0.3 mg/L (PE)
		140	82	
10	Zhang et al. (2020)	10-20	1	$Al_2(SO_4)_3$ 20 mg/L & PolyDADMAC 0.5 mg/L
		45-53	13.6	
		106-125	2.1	
11	Bayarkhuu & Byun (2022)	1	80	PACl 15 mg/L
		10	90	
14	Na et al. (2021)*	10	33	$AlCl_3$ 10 mg/L
		20	80	
		45	89	
		90	90	

**calculated the removal efficiency from the provided data of the study*

Table B3: Removal efficiency of different-sized MPs by CFS using optimized dosages of Fe-based coagulant in both synthetic and surface water irrespective of MP polymer types, shapes, and other properties

[Dataset for Figure 4.2]

No	Study	MPs Size (µm)	Removal efficiency (%)	Coagulation condition
12	Zhou et al. (2021)*	1-500	63	DI water – $FeCl_3$ 60 mg/L (PS)
		500-1000	99	
		1000-5000	96	
		1-500	16	DI water – $FeCl_3$ 90 mg/L (PE)
		500-1000	7	
		1000-5000	6	
13	Ma, Xue, Hu, et al. (2019)*	1-500	18	DI water – $FeCl_3$ 277.8 mg/L
		500-1000	12	
		1000-2000	9	
		2000-5000	8	
14	Na et al. (2021)	10	34	DI water – $FeCl_3$ 10 mg/L
		20	77	
		45	88	
		90	90	
		10	37	River water – $FeCl_3$ 10 mg/L
		20	71	
		45	89	
		90	84	
15	Ma, Xue, Ding, et al. (2019)*	1-500	14	DI water – $FeCl_3$ 277.8 mg/L
		500-1000	9	
		1000-2000	5	
		2000-5000	5	

**calculated the removal efficiency from the provided data of the study*

Table B4: Removal efficiency of different-sized NPs by CFS using optimized dosages of coagulants in synthetic water irrespective of NP polymer types, shapes, and other properties

[Dataset for Figure 4.3]

No	Study	NPs Size (µm)	Removal efficiency (%)	Coagulation condition
1	Zhang et al. (2022)	0.05	85.6	PACl 400 mg/L
		0.1	86.3	
		0.5	91	
		0.05	98.5	PACl 400 mg/L & PAM 20 mg/L
		0.1	98.5	
		0.5	98.5	
11	Bayarkhuu & Byun (2022)	0.1	78.3	PACl 3 mg/L
		0.1	71.3	IL(2) 6 mg/L
		0.1	75.3	PACl 3 mg/L & IL(2) 6 mg/L
		0.1	78.9	PACl 15 mg/L
		0.1	72.2	IL(2) 12 mg/L
		0.1	87.7	PACl 15 mg/L & IL(2) mg/L
18	Lu et al. (2021)	0.53	1	Al_{13} 0.27 mg/L (Pristine)
		0.22	88	Al_{13} 0.27 mg/L (Weathered)
19	Gong et al. (2022)	0.05	96.6	$AlCl_3$ 10 mg/L
		0.05	95.8	$FeCl_3$ 10 mg/L
		0.05	98	PACl 5 mg/L
20	Hofman-Caris et al. (2022)	0.05	82.5	$FeCl_3$ 12/18 mg/L
		0.2	26.5	
21	Peydayesh et al. (2021)	0.5	98.2	lysozyme amyloid fibrils 9.4-20 mg/L

**calculated the removal efficiency from the provided data of the study*

The Figures (B1-B4) with trend lines:

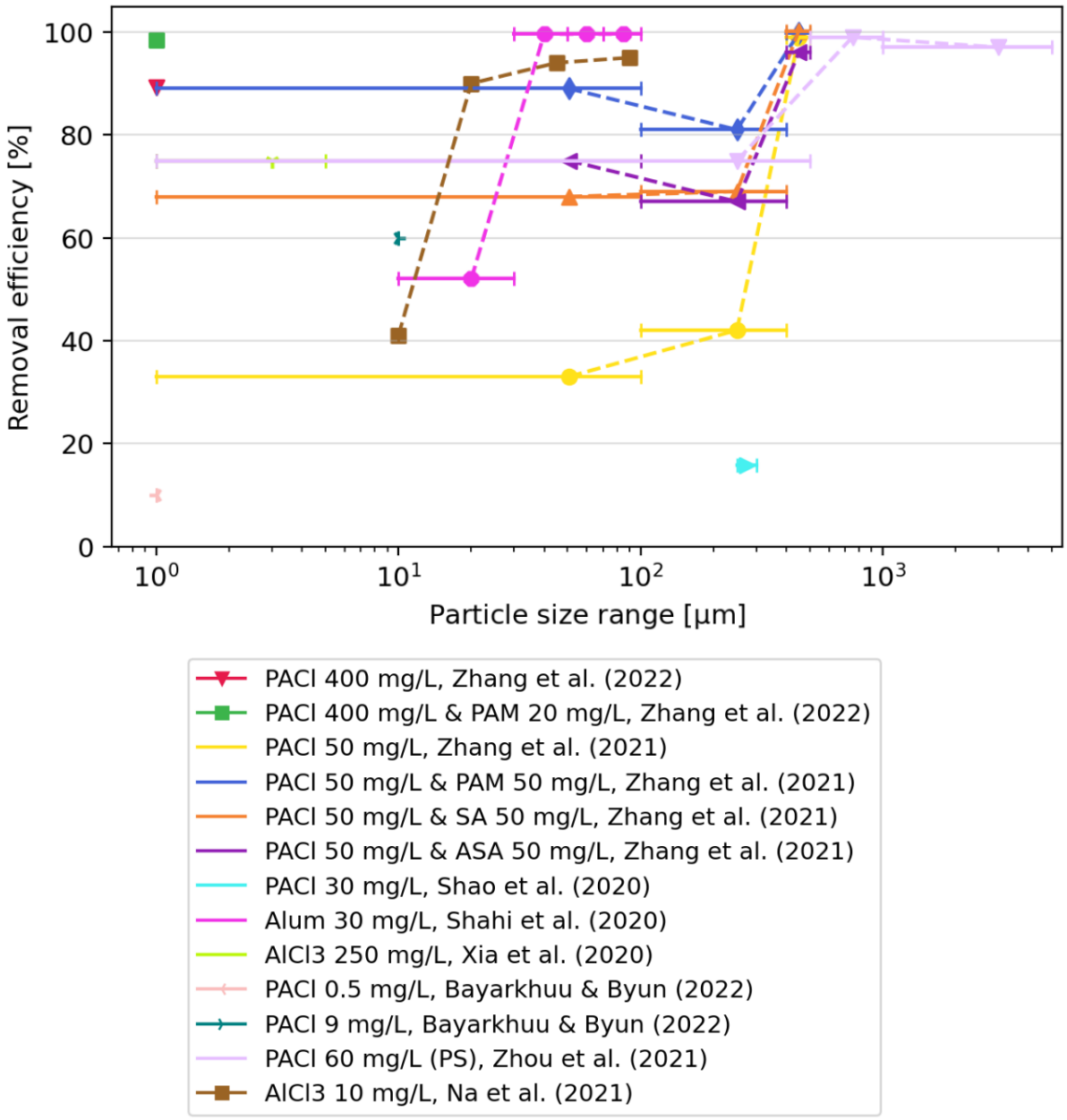


Figure B1: Removal efficiency of different-sized MPs by CFS using optimized dosages of Al-based coagulant in synthetic water irrespective of MPs polymer types, shape, and other properties *(PACl-poly-aluminum chloride, PAM-polyacrylamide, SA-sodium alginate, ASA-activated silicic acid, PS-polystyrene)*

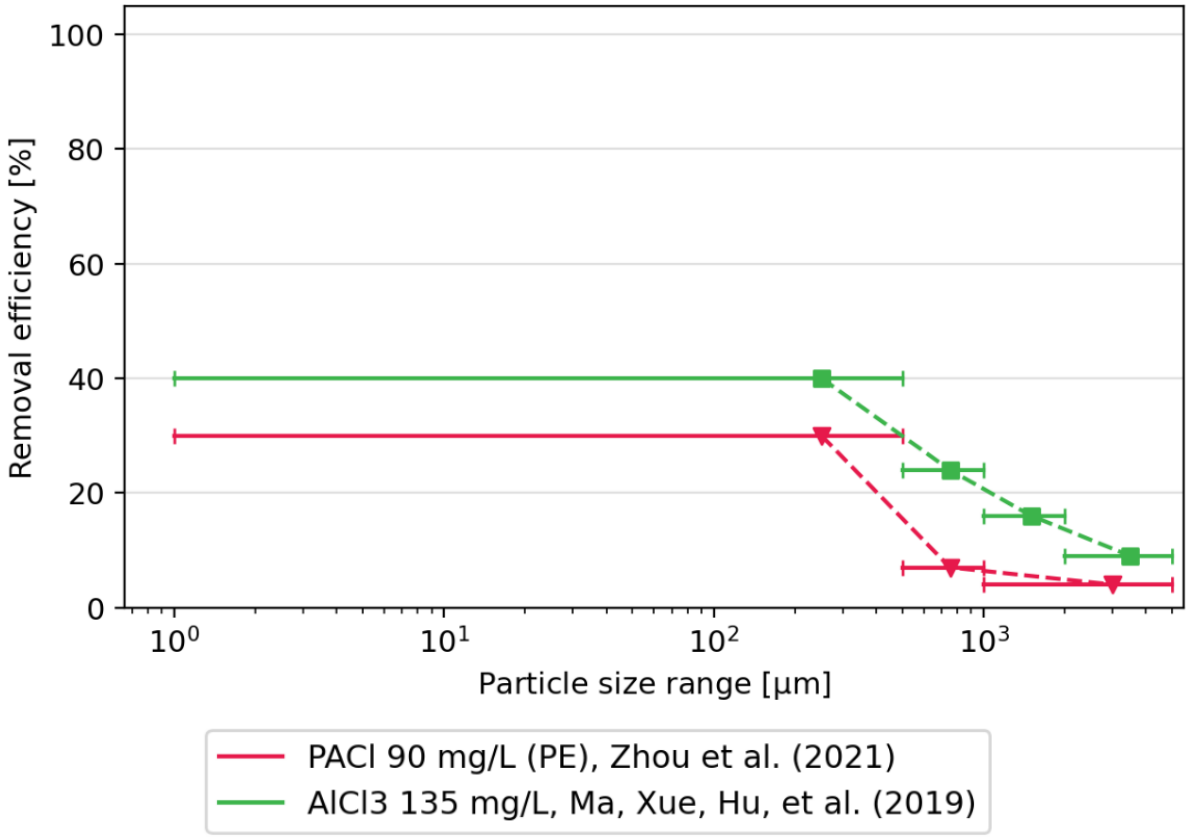


Figure B2: Removal efficiency of different-sized MPs by CFS using optimized dosages of Al-based coagulant in synthetic water irrespective of MPs polymer types, shape, and other properties *(PACl-poly-aluminum chloride, PE-polyethylene)*

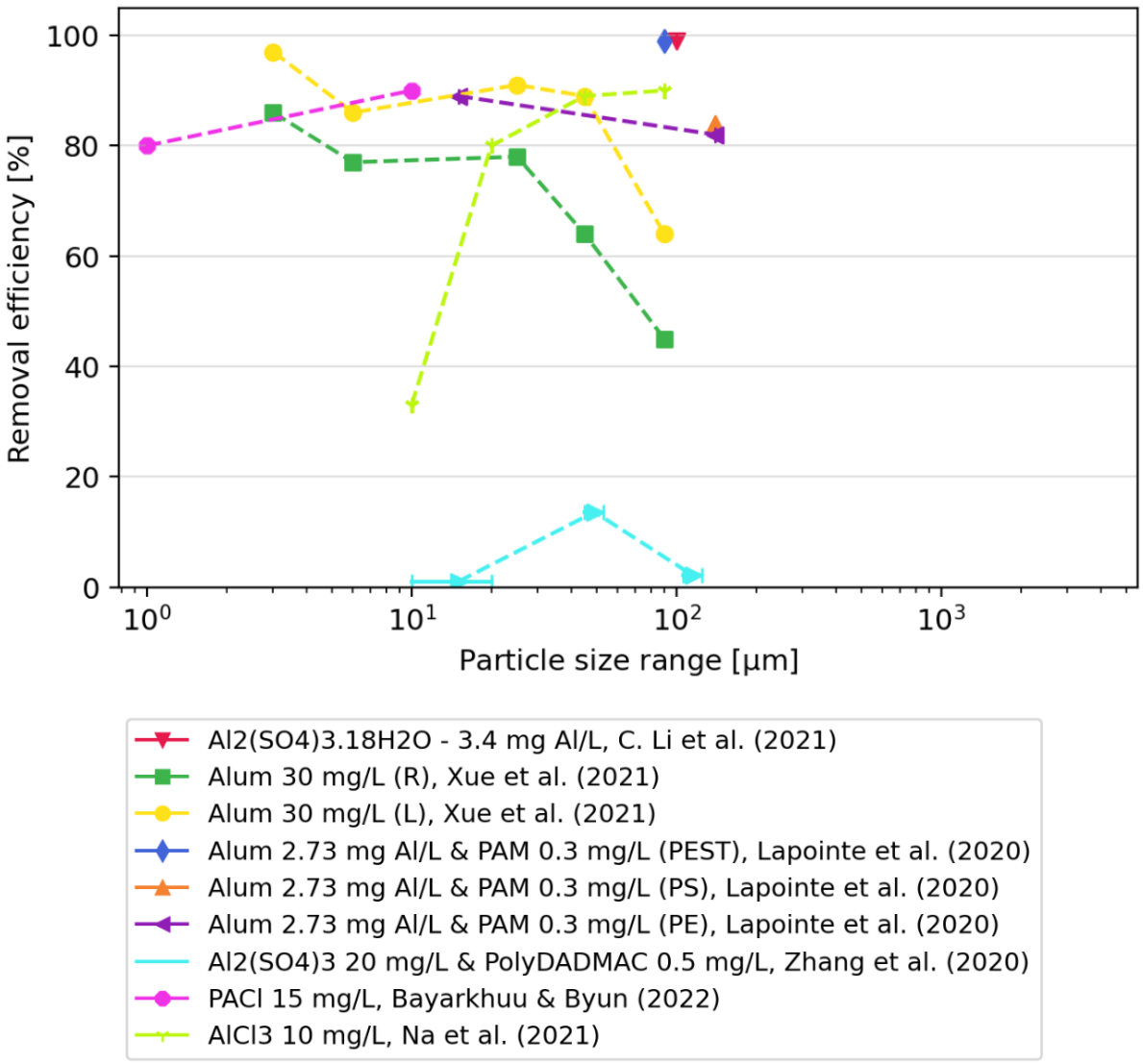


Figure B3: Removal efficiency of different-sized MPs by CFS using optimized dosages of Al-based coagulant in surface water irrespective of MPs polymer types, shape, and other properties *(PACl-poly-aluminum chloride, PAM-polyacrylamide, PolyDADMAC- Diallyldimethylammonium Chloride, PE-polyethylene, PS-polystyrene, PES-polyester)*

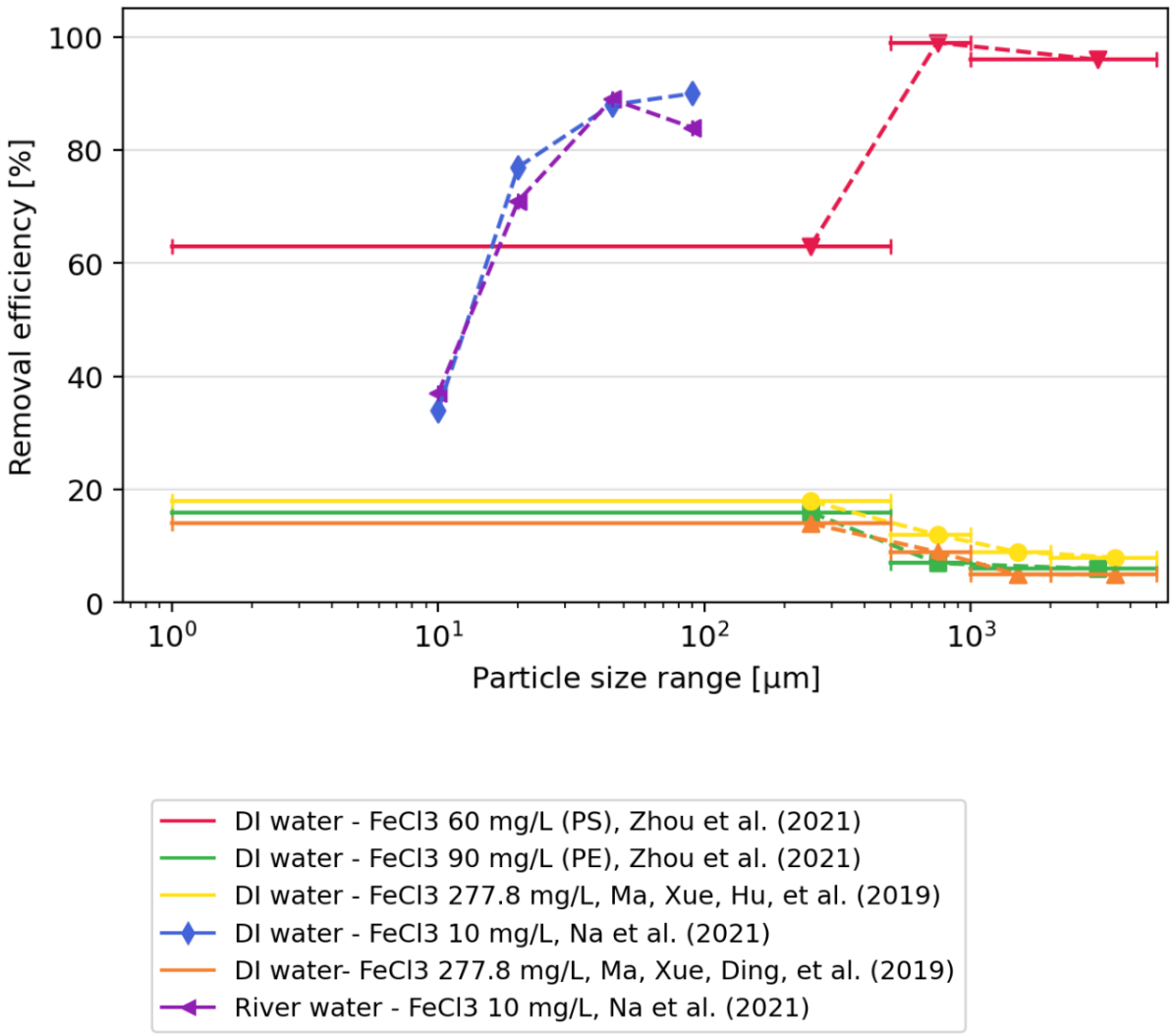


Figure B4: Removal efficiency of different-sized MPs by CFS using optimized dosages of Fe-based coagulant in both synthetic and surface water irrespective of MPs polymer types, shape, and other properties

(DI-deionized, PE-polyethylene, PS-polystyrene)

Appendix C

Supplementary Information for Chapter 5

Table C1: Global actions for the regulation of MPs in the environment

(Singh et al., 2022)

Country	Regulation	Product category	Definition of microplastics
Canada (in force)	Microbeads in Toiletries Regulations (Canada Gazette, Part II: Vol. 151, No. 12, 2017)	Toiletries, meaning any personal hair, skin, teeth, or mouth care products for cleansing or hygiene, including exfoliants	Microbead: plastic microbeads that are ≤5 mm in size, any plastic particle, including different forms such as solid, hollow, amorphous and solubilized
France (in force)	Decree prohibiting the placing on the market of rinse-off cosmetic products for exfoliation or cleansing that contain solid plastic particles (Amendment CD1857, 2019)	Rinse-off cosmetic products for exfoliation or cleansing	Solid plastic particles, with the exception of particles of natural origin not liable to persist in, or release active chemical or biological ingredients into the environment or to affect animal food chains
Ireland (in force)	Microbeads (Prohibition) Act, 2019 (S.I. 52 of 2019)	Outlawed the sale, manufacture, import, and export of products containing microplastics	Solid particle which is water insoluble and size ranging from 5mm-1 nm
New Zealand (in force)	Waste Minimization (Microbeads) Regulations, 2017 (Section 23(1)(b), 2017/291)	Wash-down cosmetic products, cleaning products	Microbead: a water-insoluble plastic particle that is less than 5 mm at its widest point
Sweden (in force)	Draft regulation prohibiting the placing on the market of rinse-off cosmetics that contain solid plastic particles which have been added for exfoliating, cleaning, or polishing purposes (Ordinance (1998:944), 4 – 4b)	Rinse-off cosmetics products	Solid particles of plastic which are 5 mm or less in size in any dimension and which are insoluble in water
United Kingdom (England, Wales, Scotland)	The Environmental Protection (Microbeads) Regulations, 2017/18 (2017 No. 1312)	Rinse-off personal care products	Microbead: any water-insoluble solid plastic particle of less than or equal to 5 mm in any dimension
United States (in force)	Microbead-free Waters Act, 2015 (Public Law 114-114,	Rinse-off cosmetics products	Microbead: any solid plastic particle that is less than 5 mm in size and is

	114th Congress, Dec. 28, 2015)		intended to be used to exfoliate or cleanse the human body or any part thereof.
Belgium (notified)	Draft Sector Agreement to support the replacement of microplastics in consumer products (Notification Number 2017/0465/ B' (2 October 2017)	–	–
China (notified)	'Opinions on further strengthening the clean-up of plastic pollution' which laid out the plan to ban the manufacture of daily chemical products containing plastic microbeads	Daily chemical products	–
India (notified in 2017, implementation since 2020)	Classification for cosmetic raw materials and adjuncts, Part 2: List of raw materials generally not recognized as safe for use in cosmetics (BIS IS 4707-2:2017)	Cosmetic products	Non-biodegradable polymeric microbeads
Italy (notified)	Draft technical regulation banning the marketing of non-biodegradable and non-compostable cotton buds and exfoliating rinse-off cosmetic products or detergents containing microplastics (Notification No. 2018/258/1)	Exfoliating rinse-off cosmetics products and detergents	Water insoluble solid plastic particles of 5 mm or less, referring to definition in Commission Decision EU 2017/1217 of 23 June 2017
Japan (notified in 2018)	Enacted a bill aimed at reducing the use of microplastics, contained in some cosmetics and other products	Cosmetic and other products	–
South Korea (notified in 2021)	Draft regulation on Safety Standards etc. of Cosmetics	Cleansing products, dental cleansing products	Microbead: less than or equal to 5 mm in size