UNIVERSITY OF CALIFORNIA,
IRVINE

Fate and Removal of Microplastics in Water Reclamation

DISSERTATION

submitted in partial satisfaction of the requirements

for the degree of

DOCTOR OF PHILOSOPHY

in Civil and Environmental Engineering

by

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Dissertation Committee:

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2022

DEDICATION

To

My parents, my friends, my advisor Diego Rosso, my colleagues and mentors,

whose love, support, and understanding have made this all possible.

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ACKNOWLEDGMENTS

First and foremost, I would like to express my deepest love and gratitude to my family. During all these years when we have been living in different parts of the world, my parents have been persistently providing encouragement, support and inspiration. I would have never had a chance to complete my degrees in the US without their selfless support.

I must express my sincere gratitude to my advisor and committee chair, Professor Diego Rosso, for his invaluable mentorship, support and tutelage during the course of my undergraduate research and PhD degree. He has enlightened me with an exemplary spirit of research, engineering, education and scholarship. It was a life-changing experience working with Prof. Rosso, and I look forward to future collaborations with him. I would also like to thank my committee members, Professor Adeyemi Adeleye and Russ Detwiler. They have enthusiastically and selflessly offered guidance and support, without which I would not have been able to defend my dissertation.

Many thanks are owed to all my friends, colleagues in Environmental Process Laboratory and UCI WEX Research Center for their friendship and help throughout these years, especially Dana Austin, Elinor Austin, Gina Habil. I must also thank my great mentors, Linda Tseng, Zeki Kayiran and Derya Dursun, who demonstrated the essence of academic as well as engineering industry experience.

Last but not least, I would like to acknowledge Department of Energy, Santa Margarita Water District, Brown and Caldwell, Water Environment Federation, and Hazen and Sawyer for research support.

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Engineers Without Borders of University of California, Irvine

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- Liu, L. and **Sun, Y**., Kleinmeyer, Z., Habil, G., Yang, Q., Zhao, L., Rosso, D., 2022. Microplastics separation using stainless steel mini-hydrocyclones fabricated with additive manufacturing. Science of The Total Environment 840, 156697. https://doi.org/10.1016/j.scitotenv.2022.156697 (Co-first authors)
- Adeleye, A.S., Xue, J., Zhao, Y., Taylor, A.A., Zenobio, J.E., **Sun, Y**., Han, Z., Salawu, O.A., Zhu, Y., 2022. Abundance, fate, and effects of pharmaceuticals and personal care products in aquatic environments. Journal of Hazardous Materials 424, 127284. https://doi.org/10.1016/j.jhazmat.2021.127284 (equal authorship distribution other than first author)
- Liu, L., Zhao, L., **Sun, Y.,** Gao, S., Jiang, M., Jiang, M., Rosso, D., 2021. Separation performance of hydrocyclones with medium rearrangement internals. J. Environ. Chem. Eng. https://doi.org/10.1016/j.jece.2021.105642
- **Sun, Y**., Garrido-Bserba, M., Molinos-Senante, M., Donikian, N.A., Poch, M., Rosso, D., 2020. Science of the Total Environment A composite indicator approach to assess the sustainability and resilience of wastewater management alternatives. Sci. Total Environ. 725, 138286. https://doi.org/10.1016/j.scitotenv.2020.138286

Conference Proceedings

- **Sun, Y**., Habil,.G, Liu, L., Rosso, D. "Microplastics separation using stainless steel mini-hydrocyclones fabricated with additive manufacturing" To be presented at the 2022 Association of Environmental Engineering and Science Professors (AEESP) Research and Education Conference, June 28-30, St. Louis, MO. USA

- **Sun, Y**., Habil,.G, Liu, L., Rosso, D. "A half year-long monitoring study of microplastics in a water resource recovery facility" To be presented at the 2022 Association of Environmental Engineering and Science Professors (AEESP) Research and Education Conference, June 28-30, St. Louis, MO. USA

- **Sun, Y**., Kleinmeyer, Z., Drake, D., Liu, L, Pasco, R., Johnson, R., Rosso, D." Monitoring of Microplastics in a Water Reclamation Process with an Improved Recovery Method". Presented at the 94th the Water Environment Federation's Technical Exhibition and Conference, October 16-20, 2021, Chicago, IL, USA

- **Sun, Y**., Enrique, Y., Drake, D., Kleinmeyer, Z., Donikian, N., Liu, L., Rosso, D. "Microplastics in wastewater treatment plants: Presence, abundance and removal using conventional and advanced technologies". Presented at the 260th American Chemical Society National Meeting, August 16-20, 2020, San Francisco, CA, USA

- **Sun, Y**., Han, Z., Xue, J., Zhu, Y., Zhao, Y., Zenobio, J., Adeleye, A. "Pharmaceuticals in wastewater treatment plants: Presence, abundance, and fate". Presented at the 258th American Chemical Society National Meeting, August 26-30, 2019, San Diego, CA, USA

- Han, Z., Zhu, Y., **Sun, Y**., Xue, J., Zhao, Y., Zenobio, J., Adeleye, A. "The use of conventional and emerging technologies for removing pharmaceuticals from wastewater". Presented at the 258th American Chemical Society National Meeting, August 26-30, 2019, San Diego, CA, USA

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ABSTRACT OF THE DISSERTATION

Fate and Removal of Microplastics in Water Reclamation

by

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Doctor of Philosophy in Civil and Environmental Engineering

University of California, Irvine, 2022

Professor Diego Rosso, Chair

Microplastics is an important class of contaminants of emerging concerns (CECs) and a rising environmental concern, as they have been ubiquitously detected in natural and engineered water systems. Water resource recovery facilities (WRRFs) have been found to be a sink as well as pathway of microplastics release to the environment. The target removal of microplastics from wastewater and reclaimed water needs to be built upon a comprehensive understanding of their occurrence and fate within WRRFs. To understand the seasonal occurrence and characteristics of microplastics in WRRFs, microplastics from wastewater influent, primary effluent, secondary effluent and tertiary effluent in a local WRRF were quantified and characterized over nine months with a set of validated methods for microplastics extraction. Higher abundance of microplastics were observed during winter season and laundry discharge was identified as a major contributor of microplastics in wastewater. Fine microplastics were abundantly found in the WRRF, especially in secondary and tertiary effluent, necessitating further investigation removing fine and nanoplastics. In addition, mini-hydrocyclone, a highly customizable and widely used hydraulic device, was tested as alternative for separating fine microplastics from water and wastewater, and achieved satisfactory separation efficiency. This confirmed their feasibility for pilot or full-scale applications.

Chapter 1 Introduction and objectives

1.1 Background information

Microplastic particles and fibers are an important class of contaminants of emerging concerns (CECs) and a rising environmental concern as the microplastics (MPs) input to the environment is increasing, and they are ubiquitously detected in natural and engineered water systems (Bui et al., 2020a; Gatidou, 2019; Ngo et al., 2019a). Microplastics are often defined as plastic fragments with size lower than 5 mm (Coffin, 2020; Sun et al., 2019a). More recently, plastics with size lower than 1 μm have been introduced as "nanoplastics" (Blair et al., 2017a) since they exhibit different physicochemical properties than microplastics (Gigault et al., 2018). However, such standards are not officially adopted. Based on sources, microplastics are classified into two categories: primary MPs are intentionally manufactured for particular applications, such as microbeads in personal care products (PCPs) (i.e. facewash, body scrub) and industrial scrubbers; secondary MPs result from the fragmentation of larger plastic particles (Prata, 2018; Sun et al., 2019a).

The recalcitrant chemical properties of MPs allow them to easily transport, accumulate, and persist in the environment (Campanale et al., 2020; Rodriguez-Mozaz et al., 2018). Studies on MPs' toxicity have shown that MPs can cause both acute and chronic toxic effects on living organisms (Besseling et al., 2015; Chae and An, 2017; de Sá et al., 2018; Judy et al., 2019; Mak et al., 2019; Yong et al., 2020). Short-term exposure of zebrafish to MPs has been found to cause disruption of oogenesis and metabolic activities, oxidative damages, local inflammation (Mak et al., 2019; Marana et al., 2022; Pei et al., 2022), whilst adverse effects of long-term exposure to MPs were observed more commonly, including reduced reproduction in earthworms and multiple

fish species (Chae et al., 2018; Pitt et al., 2018; Sobhani et al., 2021), increased mortality in *Daphnia Magna* and mussels (An et al., 2021; Cole et al., 2020), and reduced growth in mysid shrimp and silversides (Siddiqui et al., 2022). In addition, contaminants such as per- and polyfluoroalkyl substances (PFAs), heavy metals, persistent organic pollutants (POPs), or pathogens can be adsorbed onto MPs due to their large surface area and subsequently transported over a long distance by MPs, thus potentially reaching human bodies via the food chain (Gatidou, 2019; Godoy et al., 2019; Westphalen, 2018).

Over the last decade, numerous pathways of MP release to the environment and drinking water resources have been discovered, and amongst them are water resource recovery facilities (WRRF, also referred to as wastewater treatment plants; Kalčíková et al., 2017; Murphy et al., 2016; Xu et al., 2019). Although many studies have shown that WRRF could efficiently remove MPs from wastewater, MPs are still found abundant in WRRF effluent (Talvitie et al., 2015a; Xu et al., 2019a; Ziajahromi et al., 2017a). It is worth noting that existing treatment units in WRRF are not designed specifically to remove microplastics and their incidental and beneficial removal from wastewater is driven by physicochemical properties of MPs as well as operating parameters.

The rapidly increasing water demand along with extreme climate events such as floods, droughts and wildfires are imposing formidable challenges on water resource security and supply systems worldwide. To rise up to those challenges, a rising amount of effort from governments and water agencies has been devoted to water reclamation, including addressing the risks of MPs and other CECs. The monitoring of MPs' temporal occurrence, fate and transport within WRRFs with a set of reliable methods is an essential step towards the development of methods or strategies to remove MPs. The methods, technologies or strategies need to take practicality and feasibility

in to account as well as balance energy use with efficiency of removing microplastics from wastewater.

1.2 Objectives

The limited knowledge on the temporal occurrence and fate of MPs in WRRFs and technically feasible MP removal methods are the main drivers of this work. The objectives of this dissertation are:

1. To provide a comprehensive, systematic and critical review of literature of existing data (Chapter 2);
2. To provide a reliable, replicable set of methods to recover MPs from wastewater samples upon validation (Chapter 3);
3. To quantify the abundance of MPs in wastewater in different seasons, and understand the performance of different treatment process units in removing MPs (Chapter 4);
4. To develop and test potential method for MP removal (Chapter 5).

The outcome of this research imparts practical and actionable information to researchers and practitioners which will help improve the management of water reclamation in addressing MPs as well as lay the foundation for numerous future studies on MPs control and removal.

Chapter 2 Literature Review

2.1 Introduction

An increasing amount of effort to mitigate plastics and microplastics pollution worldwide has been driving research on MPs in water and wastewater systems. The abundance of MPs and the attempts to remove them at different stages of WRRFs have also been documented by several researchers. Such research is useful for informing the modifications and optimizations needed to enhance the removal of MPs from wastewater. Therefore, this meta-analysis synthesized current data to evaluate MPs removal and transport in WRRFs; investigate emerging technologies for MPs removal from wastewater; and discuss future directions for engineering practice, research, and regulation.

Heretofore, a number of reviews were published regarding MPs in wastewater, which focused on the occurrence and fate of MPs in WRRFs (Bui et al., 2020b; Habib et al., 2020; W. Liu et al., 2021; Ngo et al., 2019b; Sun et al., 2019a; Turan et al., 2021); sampling, pre-treatment, and analysis methods (Elkhatib and Oyanedel-Craver, 2020; Kang et al., 2020); MP removal technologies (Enfrin et al., 2019; Masiá et al., 2020; Rodríguez-Narvaez et al., 2021; Sun et al., 2019a); and MPs' impacts on sludge treatment (He et al., 2021; Azizi et al., 2021). As the number of studies on MPs in wastewater and biosolids is growing rapidly and MPs are gaining more attention on a local agency level, there is an urgent need for comprehensive and quantitative analysis of MPs in water and resources recovery. To support decision-making and strategizing among stakeholders, researchers and practitioners, we conducted a comprehensive and critical literature review in an attempt to identify and summarize 1) patterns in occurrence of MPs in WRRFs; 2) existing methods for sampling and sample processing and their frequency of use; 3) patterns and mechanisms in the removal of MPs at each treatment stage with various technologies

in traditional WRRFs; and 4) trends in the development of potential and emerging technologies to enhance MPs removal. Perspectives on future research and development on MPs in water and resource recovery are also discussed.

2.2 Methods

Various combinations of search keywords were used for searching on *Google Scholar*, *Science Direct* as well as *Web of Science*, including:

("microplastics" OR "micro-plastics") AND ("wastewater treatment plant" OR "WWTP" OR "water resource recovery facility" OR "WRRF" OR "sludge") AND ("occurrence" OR "transport" OR "fate" OR "removal")

A total number of 134 items were initially collected upon the search of keywords, including original research papers, systematic and critical reviews, standards, theses, book chapters and reports. Both qualitative and quantitative information regarding the following criteria were extracted from the 33 research papers that were written in English and contained relevant content: 1) sampling, sample processing and analytical methods used; 2) distribution of MP polymer types, shapes and colors; 3) abundance of overall MPs found in influent and/or effluent wastewater; 4) abundance of MPs at each treatment stage. Quantitative information was processed in MATLAB (Mathworks, 2021b) and sequentially used for synthesizing data tables and figures.

2.2.1 Calculation of overall abundance of microplastics and polymer types

In the calculation of the overall abundance of each polymer type in wastewater influent and effluent, studies that reported the distribution of each MP polymer type found but failed to report the overall MP abundance were eliminated from the final selection, as these papers' data

could not be used to calculate the relative abundance (i.e., MP L^{-1} for wastewater; MP g^{-1} dry weight for sludge) of each MP type. Conversely, papers that reported the overall abundance of MPs but failed to report the MP polymer type distribution (i.e., percentage) were also eliminated from the final selection. After initially considering 134 papers, the final selection was narrowed down to 22 papers. Of these papers, four included relevant data for wastewater and sludge, while two included relevant data for sludge only, and 20 included relevant data for wastewater only.

Data processing was performed by two research members, as this ensured that calculations could be verified and corrected if necessary. Polymer type distribution and overall MP concentration reported from each paper selected were used for the calculation of the relative abundance of each polymer type in MATLAB. Along with relative abundance, the mean relative abundance and standard deviation were computed. These latter two values were used to classify polymer types as "no abundance," "low abundance," "medium abundance," and "high abundance." Microplastics with a concentration of zero were classified as "no abundance"; MPs within a standard deviation below the mean relative abundance (i.e., between zero and the mean relative abundance) were classified as "low abundance"; MPs within a standard deviation above the mean were classified as "medium abundance"; and MPs higher than a standard deviation above the mean were classified as "high abundance". For studies that reported an influent value for a polymer type but not an effluent value—and vice versa—the missing value was considered unknown, as the origin or final destination, respectively, of these unknown values were undetermined. These unknown values were treated as Not A Number (NaN) values.

The computation of regional concentration of MPs in wastewater influent and effluent referenced 35 studies and data from 133 WRRFs around the globe. These include studies not incorporated in the relative abundance calculations (i.e., studies that reported only influent

abundance or only effluent abundance, or studies that did not specify distribution per polymer type). Outliers in the data were detected using the interquartile (IQR) method and removed.

2.2.2 Overall and stepwise removal efficiency of MPs in WRRFs

Out of the 35 research papers that reported concentration in wastewater influent and effluent, only 15 contained information on MPs' concentration at individual treatment steps. For the papers only presented figures, we utilized WebPlotDigitizer 4.5 (https://apps.automeris.io/wpd/) to extract desired concentration values. Overall, stepwise removal efficiency of MPs in relation to overall abundance and in relation to abundance at previous treatment step via each treatment step were computed by using Eqn.1, 2 and 3 respectively.

$$\%Overall = \frac{C_{inf} - C_{eff}}{C_{inf}} \times 100\% \quad \text{Equation (1)}$$

$$\%Stepwise_{overall} = \frac{C_i - C_{i+1}}{C_{inf}} \times 100\%,\ i \in N \quad \text{Equation (2)}$$

$$\%Stepwise_{previous} = \frac{C_i - C_{i+1}}{C_{i-1}} \times 100\%\ i \in N \quad \text{Equation (3)}$$

Where C_{inf} is the concentration of MPs in wastewater influent; C_{eff} is the concentration of MPs in wastewater effluent; $N = \{1,2,3, \ldots\}$ represents the number of treatment steps in a WRRF.

2.3 Sampling and analytical methods used for MPs in wastewater

The large variation of MP occurrence and characteristics has been observed, and could be partially related to the differences in MP sampling and separation methods (Gatidou, 2019; Sun et al., 2019a). The majority of existing studies used grab samples instead of composite samples (Fig. 2-1). In an intra-day MP variation study (Cao et al., 2020a), it was proved that MP occurrence in wastewater varied over time. Therefore, composite samples or multiple samples collected at the same time on every sampling day are subjected to less diurnal variation of MP concentration. However, it is still unclear whether MP occurrence is correlated to the variation of wastewater characteristics. During sample collection and subsequent procedures, most studies avoided using plastic containers as much as possible for wastewater and sludge samples and opted for metal and glass (Bayo et al., 2020b; Conley et al., 2019; Lares et al., 2018a; Long et al., 2019) (ex., aluminium shroud (Carr et al., 2016), or watch glass as covering (Lares et al., 2018a)). Nonetheless, some studies subsequently employed plastic containers to transport and store samples (Michielssen et al., 2016; Raju et al., 2020), while others explicitly stated that they cleaned and checked for contamination from the plastic containers being used (Blair et al., 2019a; Lares et al., 2018a). While sampling, volume reduction (Hidalgo-Ruz et al., 2012; Li et al., 2018) can be achieved by pre-treating samples *in situ* using sieves (Blair et al., 2019a; Carr et al., 2016; Gies et al., 2018; Lares et al., 2019a, 2018a; Liu et al., 2019; Long et al., 2019; Pittura et al., 2021) or surface filtering and skimming (Carr et al., 2016) before collecting and storing the samples. We noted that some studies provided details on how the sampling equipment was cleaned prior to sampling to avoid contamination or cross contamination (Carr et al., 2016; Long et al., 2019), but many studies did not explicitly mention such steps.

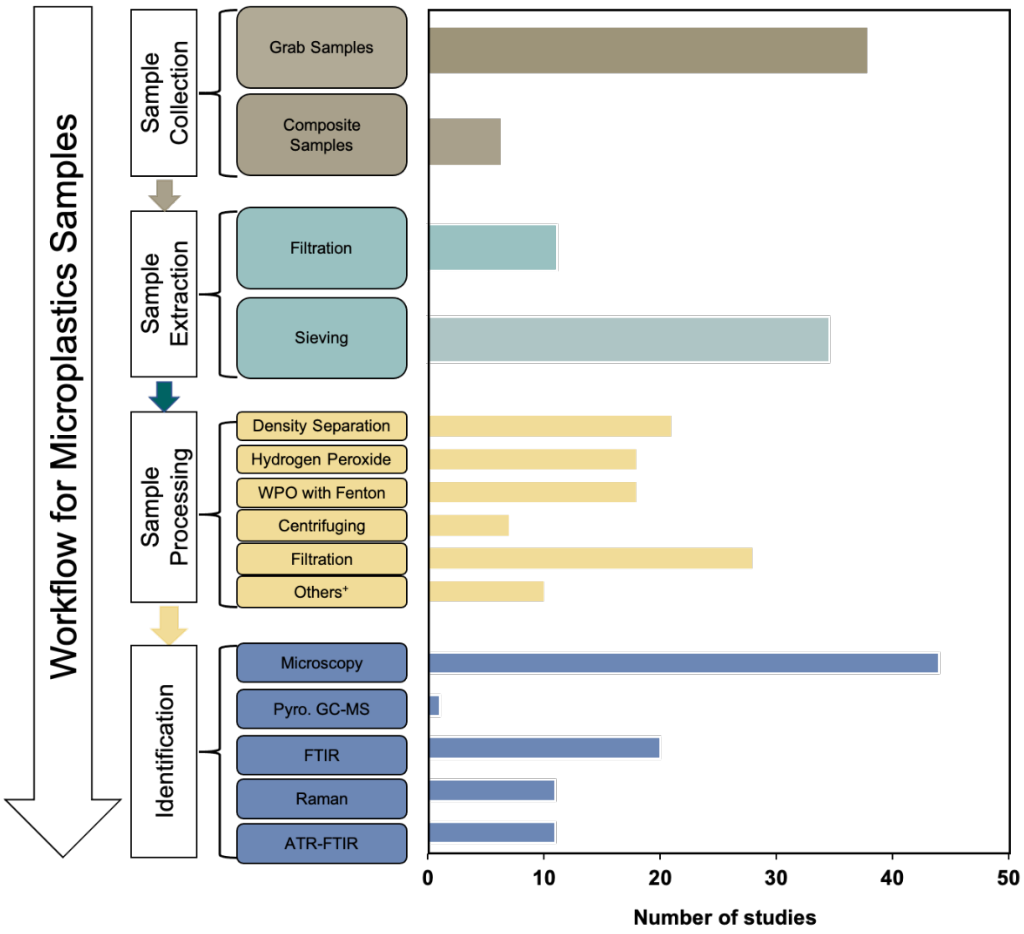


Figure 2 - 1 Sampling, sample processing and analytical methods used in existing studies (Others include enzymatic digestion, acidic or alkaline digestion, oil extraction, sonication, and sample suspension in organic solvent)

Microplastics are commonly separated by sieving and filtration (Fig. 2-2). Prior to the publishing of the first standard method (ASTM D8333-20) for the identification and quantification of MPs in water matrices of various solid concentration, sieves of various mesh sizes have been used in previous studies (Fig 2-2). Sieves with larger openings are more likely to miss small MPs, thus leading to underestimation of MP concentration and bias of size distrubution (Lares et al., 2018a; Simon et al., 2018a). Several studies adopted filtration devices or an automatic composite sample, since low MP concentration led to a need for a large volume of

wastewater sample (Dris et al., 2015; Naji et al., 2021a; Talvitie et al., 2017a, 2015b; Ziajahromi et al., 2017b). These devices may introduce currently unquantified bias or error, as pumps and samplers are associated with uncertainty.

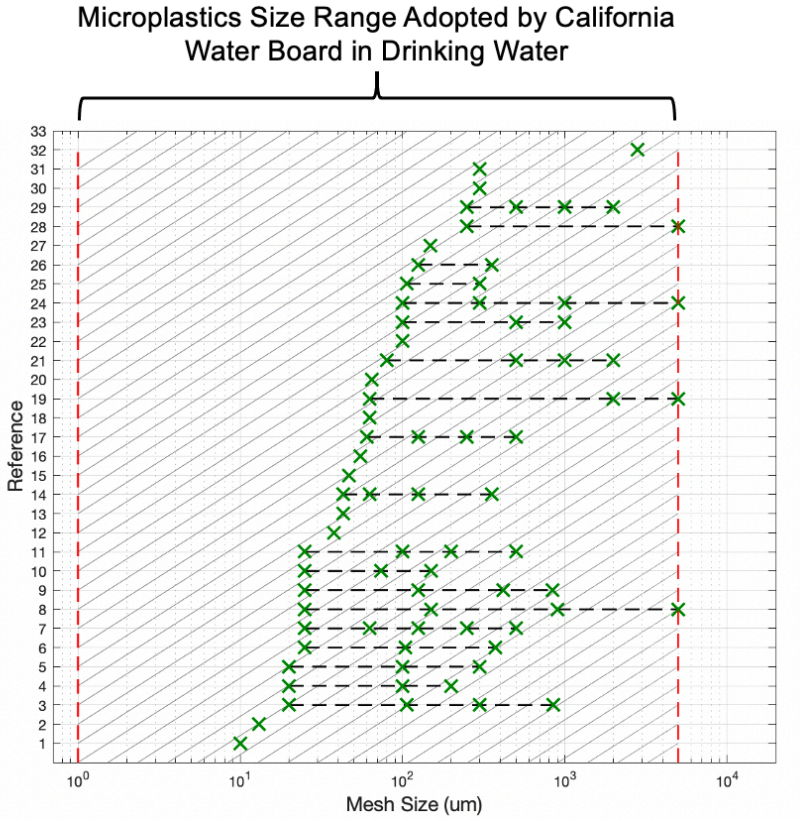


Figure 2 - 2 Mesh size used in previous studies to filter microplastics in wastewater (Reference listed in Table A1)

The sample extraction procedures usually consist of chemical and physical processes. The most frequently used methods are wet peroxide oxidation (WPO) and WPO with Fenton' reagent, followed by vacuum filtration. In some studies, the extraction procedures may process the sample according to its type or content (Carr et al., 2016; Gies et al., 2018; Liu et al., 2019; Long et al., 2019; Tang et al., 2020a; L. Zhang et al., 2021a). For instance, (Gies et al., 2018) processed the solid content and the liquid content differently. The settled solid content and the diluted sludge

solid content (stirred in distilled water and settled overnight under refrigeration) underwent WPO, while the liquid portion of the raw influent, primary effluent and secondary effluent underwent oil extraction protocol (liquid-liquid extraction of the MPs in canola oil) (Crichton et al., 2017). The minimal processing involved only filtration, as recommended by Lares et al. (2019a).

Sample extraction methods should avoid damaging MPs and be non-destructive for the count-based quantification. Previous digestion and extraction method comparison studies concluded that all previously used chemical digestion methods impose damages on MPs to some extent, changing characteristics such as surface morphology, surface area, and chemical composition (indicated by mass loss) (Lares et al., 2019b; Pfeiffer and Fischer, 2020; Tagg et al., 2017). These studies also commonly observed that polyamide (PA) is more vulnerable to chemical digestion, evidenced by higher mass loss after digestion. This means that the count of PA discovered in wastewater and sludge samples could be underestimated due to damage from reagents, creating a dilemma where neither damage on MPs from chemical digestion or high solid and organic residuals is desired.

The quantification and identification of MPs normally (recorded in 96% of existing studies; Fig. 2.1) involved a light or optical microscopy (e.g., (Conley et al., 2019; Leslie et al., 2017; Long et al., 2019; Naji et al., 2021b; Pittura et al., 2021; Raju et al., 2020) (Table S2) to differentiate the shape, colour, size, and structure of the particles to screen suspected microplastics. Then either all (Pittura et al., 2021), a subset (Bayo et al., 2020b; Blair et al., 2019a; Lares et al., 2019a; Liu et al., 2019; Long et al., 2019), or ambiguous and suspected particles (Carr et al., 2016; Gies et al., 2018; Vardar et al., 2021) were examined via Fourier Transformation Infrared (FTIR) spectroscopy (or μ-FTIR, equipped with attenuated total reflectance, ATR) (e.g., Cao et al., 2020b; Naji et al., 2021b; L. Zhang et al., 2021) or Raman spectroscopy/microscopy (or μ-Raman)

(e.g., (Lares et al., 2018a; Long et al., 2019; Vardar et al., 2021) to confirm or determine the material of the suspected MPs. Whilst spectroscopic methods can provide a count of MPs along with their polymer type, pyrolysis-Gas Chromatography-Mass Spectroscopy (Pyro GC-MS) can provide the mass of MPs and polymer, but is a destructive method.

The plethora of methods to extract microplastics from different matrices (i.e. wastewater, sludge, surface water, organisms, etc.) shows there is no single set of methods that is appropriate for all types of samples. From a water quality and utility management perspective, the immediate need is to monitor the abundance of microplastics. To meet this goal, sampling, handling, pre-treatment and analysis should be selected based on the level of detail of the information required (i.e., polymer type, morphology, concentration by count or mass) and aim to be rapid and reproducible at local facilities.

2.4 Occurrence of microplastics in wastewater

Microplastics are readily detected in WRRF influent, effluent, and sludge; there is significant variation in concentration, type (i.e., composition), abundance, shape, and size. To date, 21 studies have been conducted that provided data on relative polymer concentration in the influent and effluent, or sludge; and a total of 60 different polymers were identified. Concentration in wastewater influent varied within six orders of magnitude, ranging from 0.27 MP L^{-1} (Lv et al., 2019) to 18,285 MP L^{-1} (Simon et al., 2018a) and with an average of 229.1 ± 1,030.1 MP L^{-1}. Concentration in the effluent was generally lower due to MP removal during treatment, but still varied within four orders of magnitude across WWRFs: the lowest reported effluent concentration was 0.005 MP L^{-1} (Talvitie et al., 2017a), and the highest was 28.4 MP L^{-1} (Liu et al., 2019). Average effluent concentration was much lower than that of the influent, at 7.7 ± 9.6 MP L^{-1}. For sludge, meanwhile, concentration varied within three orders of magnitude, from 0.235 MP g^{-1} dry

weight (L. Zhang et al., 2021a) to 240.3 MP g^{-1} dry weight (Liu et al., 2019), with an average concentration of 84.1 ± 90.6 MP L^{-1}. The majority of these studies reported MP concentration within the tens and low hundreds range (Lares et al., 2018a; Uddin et al., 2020; Xu et al., 2019b) (Fig. S1), although extremely low and high concentrations were reported in some WRRFs beyond the 21 studies previously discussed. One study conducted in 10 Danish WRRFs reported an average influent concentration of 7,216 MP L^{-1} (Simon et al., 2018a). Furthermore, of the 21 studies, 19 provided data on MP abundance in the influent and effluent, while only 6 (Lares et al., 2018a; Liu et al., 2019; Magni et al., 2019; Tang et al., 2020b; Xu et al., 2020; L. Zhang et al., 2021a) detailed the abundance of specific MP types in sludge.

The occurrence and abundance of MPs in wastewater and sludge also differ substantially across WRRFs. Dozens of MP types have been detected in wastewater and sludge, and some of the most frequently reported are polypropylene (PP), polyester (PES), polyethylene (PE), polyamide/nylon (PA) and polyethylene terephthalate (PET) (Table 2-1). Highly abundant MPs include some of the most reported MPs, along with less frequently reported ones. The most abundant MPs are PP, PE, PET and polystyrene (PS). Polyester, PA and PET are commonly used as textile material, and thus more likely to enter WRRFs as textile microfibers from domestic washing machines (Almroth et al., 2018; Sillanpää and Sainio, 2017)

Table 2 - 1 Polymers detected in WRRFs with the frequency of papers referenced and relative abundance in the influent and effluent.

Polymer Abbreviation[a]	Density[b] (g cm^{-3})	No. Papers Referenced[c]	Influent Relative Abundance[d]	Effluent Relative Abundance[e]	Sludge Relative Abundance[f]
PP	0.83-0.92	11	****	+++	××
PES	1.24-2.3	10	***	++	××
PE	0.89-0.98	10	****	+++	××
PA	1.02-1.16	6	**	++	××
PET	0.96-1.45	6	****	++	××××
PS	1.04-1.1	5	****	+++	××
PVC	1.16-1.58	4	**	++	××
PE-PP	0.94	2	***	++	××
Rayon	1.7-1.8	2	****	++	××
PVA	1.19	2	**	++	×
PEUR	1.20	1	**	++	×
AS	1.05-1.08	1	**	++	×
PMMA	1.09-1.20	1	**	+	×
PAA	1.22	1	****	++	×
PC	1.2-1.22	1	*	++	×
Alkyd	1.24-2.01	1	**	++	×

[a] Polymers referenced in at least one paper: Polypropylene (PP), Polyester (PES), Polyethylene (PE), Polyamide (PA), Polyethylene Terephthalate (PET), Polystyrene (PS), Polyvinyl Chloride (PVC), Polyethylene and Polypropylene Copolymer (PE-PP), Polyvinyl Acetate (PVA), Polyether Urethane (PEUR), Acrylonitrile Styrene Copolymer (AS), Poly Methyl Methacrylate (PMMA), Poly Acrylate (PAA), Polycarbonate (PC). Due to limited space, please refer to SI for the complete table.
[b] References: (Duis and Coors, 2016; Kowalski et al., 2016; Li et al., 2018; Simon et al., 2018a; Sun et al., 2019a)
[c] References: (Bayo et al., 2020b, 2020a; Cao et al., 2020b; Gündoğdu et al., 2018; Lares et al., 2018a; Liu et al., 2019; Long et al., 2019; Lv et al., 2019; Magni et al., 2019; Murphy et al., 2016b; Raju et al., 2020; Simon et al., 2019a, 2018a; Talvitie et al., 2017a; Tang et al., 2020b; R. Wang et al., 2020; Xu et al., 2020, 2019b, 2018; Yang et al., 2019; L. Zhang et al., 2021a)
[d] * Refers to no abundance in the influent, ** refers to low abundance in the influent, *** refers to medium abundance in the influent, **** refers to high abundance in the influent (Refer to SI for MP polymer concentration calculations).
[e] + Refers to no abundance in the effluent, ++ refers to low abundance in the effluent, +++ refers to medium abundance in the effluent, ++++ refers to high abundance in the effluent (Refer to SI for MP polymer concentration calculations).
[f] × Refers to no abundance in sludge, ×× refers to low abundance in sludge, ××× refers to medium abundance in sludge, ×××× refers to high abundance in sludge (Refer to SI for MP polymer concentration calculations).

Large variations in the reported MP types and their abundance may be attributed to different sewer system and WRRF properties, such as population served, influent rate and treatment processes. Polymers frequently used in textile or manufacturing industries, such as PET, PA, PS and PE, are more abundant in industrial cities. Agricultural areas, meanwhile, may be associated with different MP types. Raju et al. (2020) reported that fruit stickers, despite not being previously addressed as a pollutant or a MP source, are frequently found as litter and may be a major source of polyurethane (PU). Additional studies on MP composition and abundance in multiple areas with similar commercial and economic motivations may provide insight into which polymer types are associated with certain industries. Furthermore, WRRFs associated with combined sewer systems were found to receive a higher concentration of MPs along with other small anthropogenic litter (SAL) such as tire fragments, road paint, and other plastic items that could discharge into WRRFs (Lee and Kim, 2018a; Michielssen et al., 2016). Storm drainage systems are direct pathways for land-based MPs into freshwaters (Liu et al., 2019; Vaughan et al., 2017). As previously discussed, combined sewer systems typically associated with higher MP concentrations, specifically MP fragments (Ó Briain et al., 2020), could potentially reduce the MP concentration directly released into the environment and direct it to WRRFs under normal weather conditions. Service population of WRRFs is another factor found to impact MP occurrence, yet contradicting conclusions on the correlation between MPs occurrence and served population were found in the literature. Mason et al. (Mason et al., 2016) investigated the MPs discharged from 17 WRRFs in the United States and concluded that facilities serving larger populations discharge more MPs. Zhang et al. (2021) found similar trends among four WWTPs in China. On the other hand, Mahon et al. (2017), Mintenig et al. (2017), and Long et al. (2019), who investigated MPs in multiple WRRFs in Ireland, Germany, and China, respectively, did not observe a significant

correlation between MPs occurrence in WRRFs and their served population. Instead, Long et al. (2019) suggested that served population size influences MP flux rather than concentration.

A diversity of information derived from studies conducted in different countries and regions—though mostly those of higher GDP (Fig. 2-3)—indicates that MP occurrence and abundance vary geographically. None of the papers included data in Antarctica, South America, or Africa. Of the 133 plants documented in the papers, nearly three-quarters were in Asia and only one was in Oceania. Europe had the highest influent abundance of MPs by far, at 2,708 ± 3,542 MP L^{-1}. The abundance in North America, Asia, and Oceania, were 138±10 MP L^{-1}, 71±75 MP L^{-1}, and 5±0 MP L^{-1}, respectively. Except for Oceania, the range of abundance between plants within each continent was extremely large. In Europe, the minimum reported abundance was 2.5 MP L^{-1} (Lares et al., 2018a) and the maximum abundance was 10,044 MP L^{-1} (Simon et al., 2018a). Forty-one papers reported average MP abundance in effluent wastewater within the same continents identified for influent abundance. Nineteen studies measuring effluent abundance were reported in Asia and 16 studies in Europe, while only 6 were in North America and 2 in Oceania. The highest to lowest average effluent MP abundance was Europe (17.41 ± 24.28 MP L^{-1}), Asia (2.96 ± 4.75 MP L^{-1}), Oceania (1.19 ± 1.20 MP L^{-1}), and North America (0.06 ± 0.005 MP L^{-1}). The percent removal, from highest to lowest, was North America (99.96%), Europe (99.36%), Asia (95.82%), and Oceania (77.20%). Microplastics is a global, rather than local, environmental challenge. As such, conducting research in WRRFs in different regions of similar conditions (e.g., climate, water consumption per capita, etc.) and in regions of different economic prosperity would be valuable.

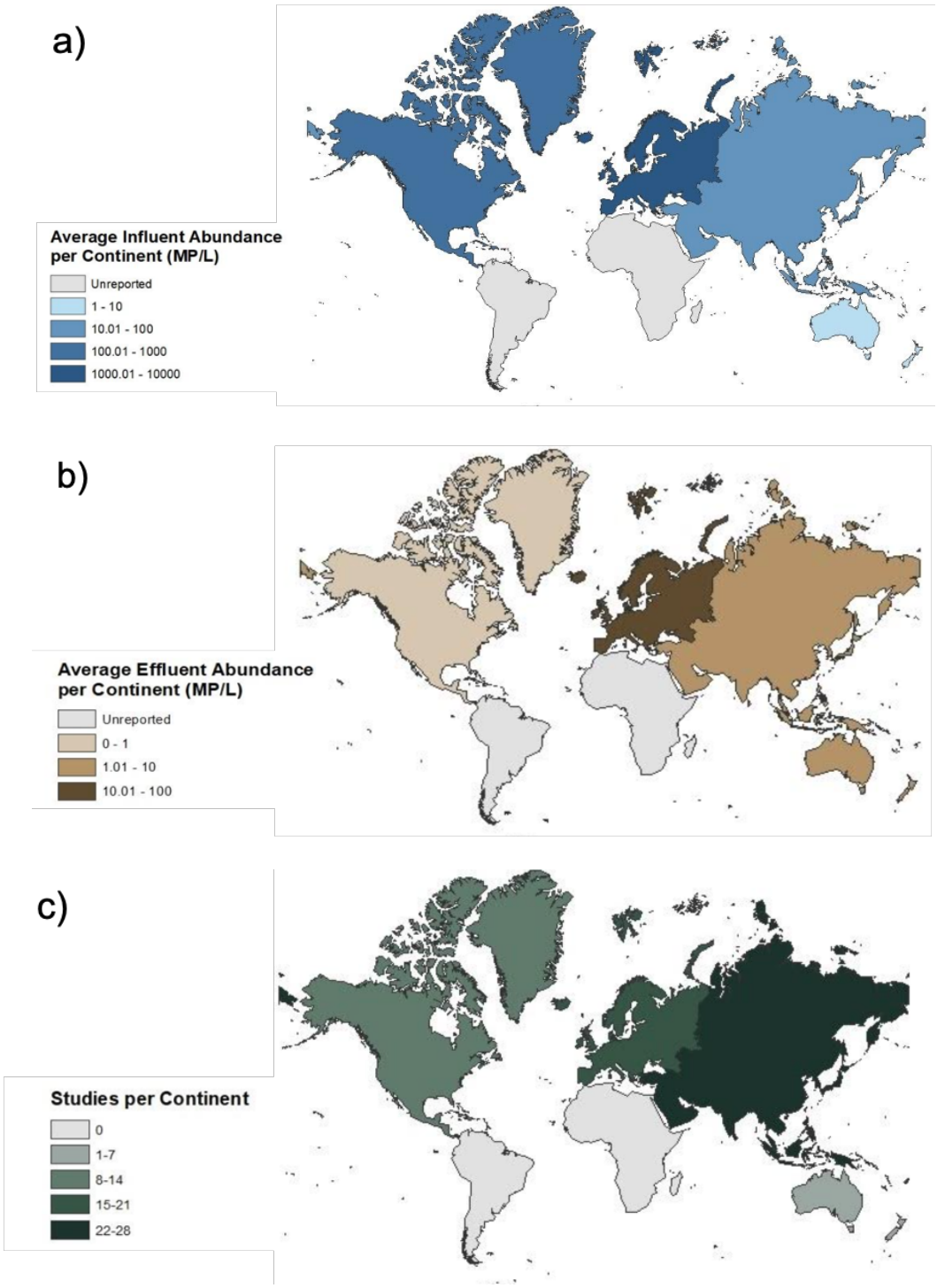


Figure 2 - 3 Mean concentration of MPs in wastewater a) influent and b) effluent, and c) number of studies conducted in different continents (Antarctica excluded as no studies were conducted there)

2.5 Removal of microplastics from wastewater

Existing studies on the occurrence and fate of MPs in WRRFs evidence that conventional wastewater treatment units are efficient in removing MPs from wastewater influent (Fig. 2-4). As shown in Fig.1, the majority of existing literature reports an overall removal of MPs of greater than 1-log (>90%). Regardless of the high removal efficiency, MPs ranging from 0.004 to 447 MPs L^{-1} (Fig. 2.4) were still detected in the effluent, corresponding to considerably high total discharge of MPs from WRRFs. Most WRRFs discharge millions of MPs daily, with the median value of total discharge being 7.04×10^{7} MP d^{-1}. Notably, although the average overall MP removal efficiency of tertiary facilities is 7% higher than that of secondary facilities (Fig. 2-4), the difference in MP removal between secondary and tertiary WRRFs is not statistically significant ($P > 0.05$), suggesting the necessity for additional treatment process(es) for MP removal in both secondary and tertiary facilities.

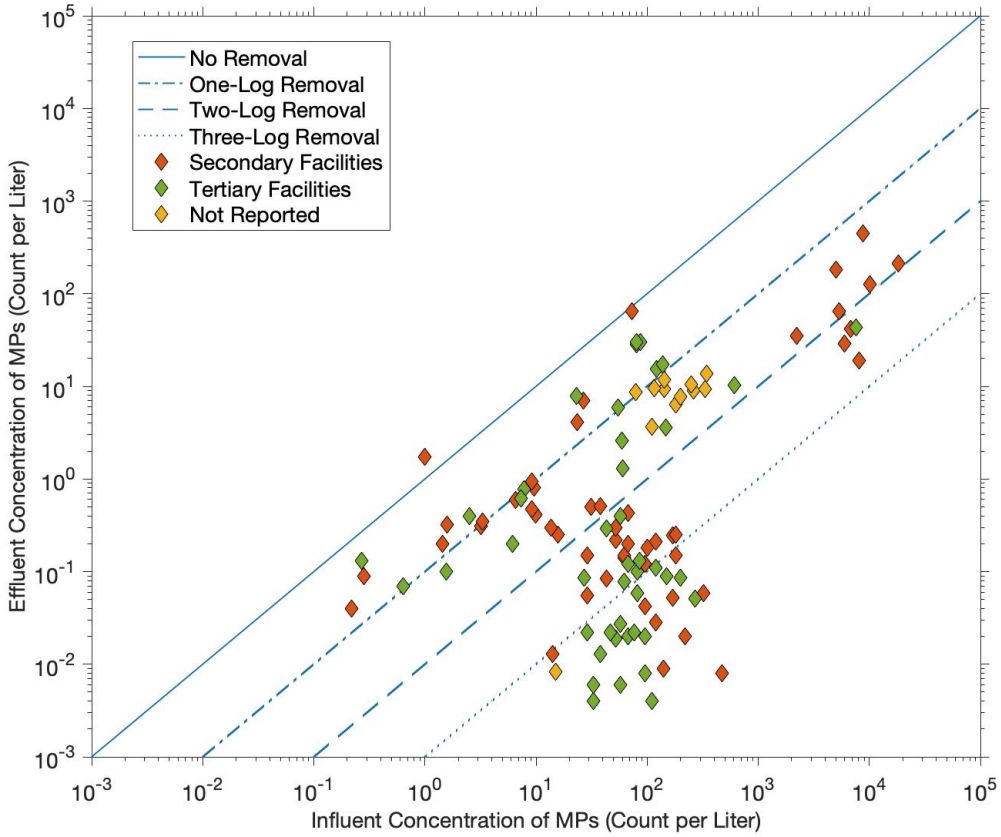


Figure 2 - 4 Microplastics removal after secondary and tertiary treatment at the same location, based on literature publications. The number of points collected in this aggregate plot is n = 110.

The removal MPs of different polymer types and sizes cross existing literature have also been examined in this study. The vast majority of the frequently detected polymers (e.g. PES, PET, PE) were effectively removed from wastewater stream in WRRFs (Fig. 2-5). It is worth noting that PET, PES, rayon and PA are widely used materials for synthetic clothing that predominantly appear as MP fibers in municipal wastewater (Lares et al., 2018a; Sun et al., 2019a; Ziajahromi et al., 2017b), while other MPs (e.g., PE, PVC, PS) are commonly detected as particles and fragments. This means that conventional wastewater treatment process units are typically not selective in removing any particular polymer or shape, but remove MPs as a group of contaminants instead.

Although high removal efficiencies of MPs have been commonly reported cross different studies, there has been a lack of uniform observation of each treatment step's performance in removing certain types or sizes of MPs due to differences in 1) treatment system configuration, 2) process units' operational conditions [e.g., hydraulic retention time (HRT), wastewater characteristics, mean cell retention time (MCRT)], and 3) sampling, sample processing and analytical methods. The mechanism of MP removal and the performance of each treatment step is discussed in the following sections.

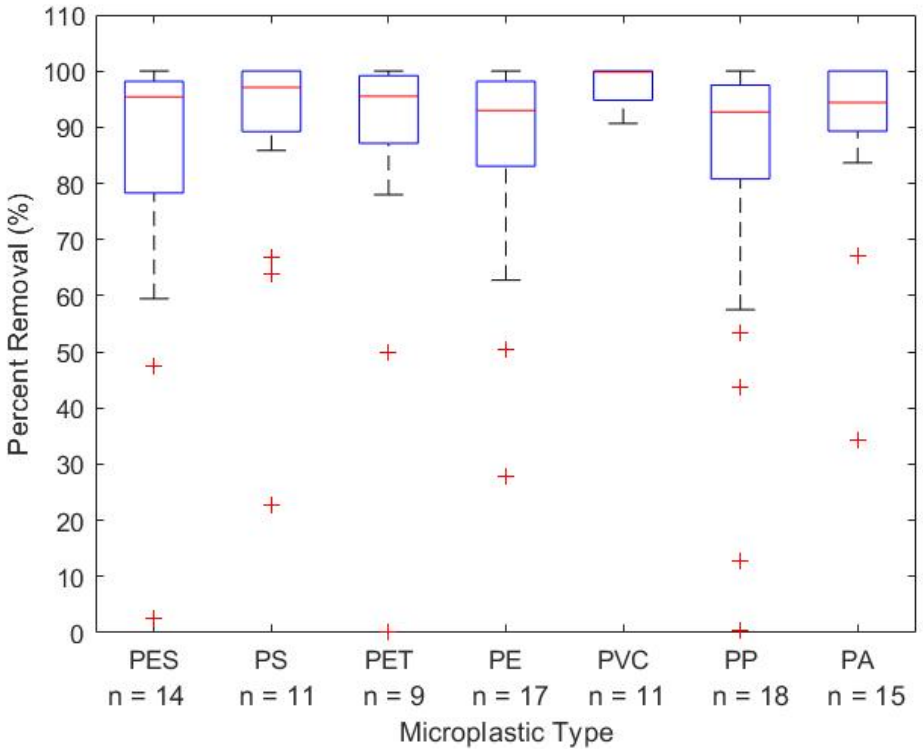


Figure 2 - 5 Average overall removal efficiency of each of the polymers from WRRFs (PES, polyester; PS polystyrene; PET, polyethylene terephthalate; PE, polyethylene; PVC, polyvinyl chloride; PP, polypropylene; PA, polyamide), and the number of studies each MP type

2.5.1 Primary treatment

Primary treatment is proven to be responsible for removing the majority of MPs, removing an average of approximately 60% of total MPs (Fig. 2-6) while being capable of removing up to 99%. During primary treatment, MPs denser than water and MPs trapped in solid flocs are removed via gravitational settling, whereas less dense MPs and MPs trapped in buoyant flocs are removed by skimming. The hydrophobic nature of MPs and the abundance of MP fibers promote the formation of MP aggregates and flocs, which favour the removal of MPs(Long et al., 2019). Although a substantial proportion of MPs can be removed from primary treatment, MPs can also become trapped in unstable flocs which do not settle efficiently, potentially leading to redistribution of MPs in the aqueous phase and allowing some to escape from the skimming and settling stages (Carr et al., 2016).

Many WRRFs around the globe are equipped with chemically-enhanced primary treatment (CEPT), where coagulants such as iron- or aluminum-based salts, and polymers are added to facilitate sedimentation. Microplastics are typically electrostatically charged, and coagulation with coagulants such as iron salts ($Fe_2(SO_4)_3.9H_2O$, $FeCl_3.6\ H_2O$) and aluminum salts ($KAl(SO_4)_2.12H_2O$, $AlCl_3.6H_2O$, $Al2(SO_4)_3.18H_2O$) can significantly improve the removal of MPs in sedimentation (Dey et al., 2021; Ruan et al., 2019). The type of coagulant plays an important role in the removal efficiency of particulates in CEPT, yet only a few studies addressed the effects of different coagulants and coagulation processes on MP removal. He et al. (2016) performed batch sedimentation tests of MPs with propylammonium chloride (PAC), propylammonium ferric chloride (PAFC) and poly ferric sulphate (PFS), observing optimal sedimentation of MPs along with preeminent removal of total suspended solids (TSS) and chemical oxygen demand (COD) with PAFC. (Ma et al., 2019) discovered that, in drinking water treatment, higher removal

efficiency of PE was induced by Al-based salt in comparison to Fe-based salt, as Al-based flocs had higher zeta potential at pH 7. Additionally, the use of polyacrylamide (PAM), a coagulation-enhancing reagent, improved the removal efficiency of PE by forming denser flocs. Since MPs are always negatively charged in water and Fe-based flocs are positively charged under acidic conditions, MPs could be easily adsorbed to Fe-based flocs (G. Zhou et al., 2021). The same study also pointed out that PE removal efficiency in the sedimentation process was affected by water characteristics, such as ionic strength, turbidity and natural organic matter (NOM). Given the variety of substances in wastewater, it is worthwhile to investigate the impacts of wastewater characteristics and the use of different coagulants on the removal efficiency of MPs in primary treatment.

2.5.2 Secondary treatment

Secondary treatment (also known as biological treatment) is designed to remove the majority of contaminants from wastewater via biological processes. The removal efficiency of MPs in secondary treatment varies significantly depending on the type of secondary treatment technology, and can reach up to 99% (Table A2). In this study, stepwise and proportional removal efficiency of secondary treatment were both computed using the concentration of MPs in the primary and secondary effluent in Table A2.

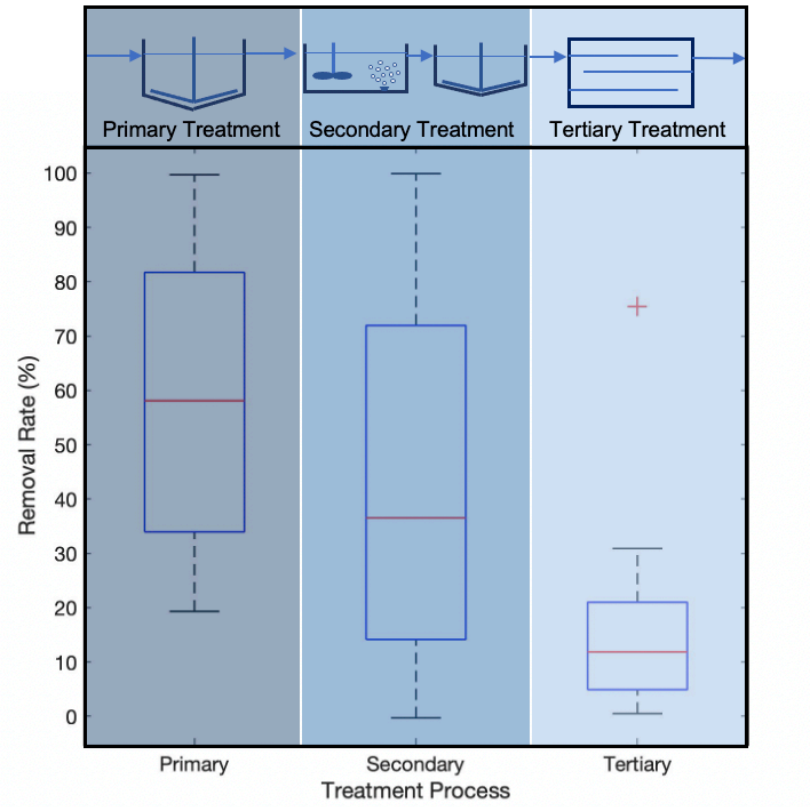


Figure 2 - 6 Removal rate of overall MPs from each treatment process ($n_{primary}$ = 12; $n_{secondary}$ = 17; $n_{tertiary}$ = 10; data from Table A2)

Activated sludge (AS) coupled with clarification tank [referred to as conventional activated sludge (CAS)] is the most commonly applied configuration of secondary treatment. Since MPs are non-biodegradable or slowly biodegradable (Blair et al., 2017b; Talvitie et al., 2017a), they are mainly removed via sedimentation during secondary clarification. During this stage, extracellular polymeric substances (EPS) secreted by microorganisms can potentially form hetero-aggregates with MPs (Enfrin et al., 2019; Zhang and Chen, 2020) which would sequentially be settled. In addition, the attachment of MPs to biomolecules on the cell surface by EPS increases the chances of contact between MPs and the crucial enzyme. However, such attachment may cause inhibitory effects on other crucial enzymatic activities, thus affecting the denitrification performance of the

secondary treatment (Zhang and Chen, 2020). Higher microorganisms commonly present in activated sludge, such as protozoa and rotifers, can potentially remove MPs by ingestion, as they commonly feed on suspended particulate matter (SPM) (Scherer et al., 2020). In general, CAS can further remove few MPs to >90% MPs from primary effluent (Table A2). It has long been recognized that operational conditions impact MP removal efficiency from activated sludge, but existing studies failed to associate various MP removal rates from AS with operational parameters (Lee and Kim, 2018a; Padervand et al., 2020; Vardar et al., 2021).

Other frequently applied secondary treatment technologies include membrane bioreactors (MBR), trickling filters (TF), and oxidation ditches (OD). Membrane bioreactors are increasingly being applied as a replacement for CAS due to its superior effluent quality (Blair et al., 2019a). Membrane bioreactor technology has been reported as an effective technology for removing MPs, as membrane filtration is more efficient at separating particulates (Lv et al., 2019; Michielssen et al., 2016; Talvitie et al., 2017c) (Table A2). It is worth noting that MPs can cause severe membrane fouling by clogging membrane pores or forming a filter cake, with the increased transmembrane pressure leading to higher energy demands and being likely to wear out the membrane faster (Li et al., 2020).

The removal of MPs using TF, OD, and other secondary treatment technologies has been insufficiently studied. Gies et al. (2018) and Lv et al. (2019) both found that TF coupled with secondary clarifier was able to remove more than 80% of MPs left from primary effluent. Trickling filter media has high specific surface area and porosity, and is covered with a layer of biofilm where microorganisms secrete EPS (*Wastewater Engineering: Treatment and Resource Recovery*, n.d.). Microplastics attached onto EPS then become fixed in the biofilm, which acts as an

attachable carrier for supporting biofilm growth, before MPs are sequentially left in the necrotic sludge or backwash (Zhang et al., 2020).

2.5.3 Tertiary treatment

Tertiary treatment is the final stage of wastewater treatment to polish water quality before discharge to the environment or to beneficial reuse for irrigation or recreational purposes. Tertiary treatment either uses physical-chemical processes (e.g., ozonation, chlorination, UV irradiation), physical separation [e.g., dissolved air flotation (DAF), rapid sand filtration (RSF)], or a combination of both to remove specific pollutants and pathogens.

In general, physical processes are more efficient in removing MPs than chemical processes. For instance, disc filters (DF) managed to remove 89.7% in a WRRF in California (Simon et al., 2019a), and a similar removal efficiency (89.5%) was found by (Talvitie et al., 2017a) in a Finish WRRF. Notably, the particle size for PES, different from other types of polymer, differed considerably before and after disc filtration, possibly ascribing to cake filtration and polyester fibers shedding from the disc filter cloth (Simon et al., 2019a). In contrast, the abundance of MPs changes marginally in chemical tertiary treatment units (<40%). Chlorination, ozonation, and UV irradiation can induce degradation of MPs (Enfrin et al., 2019; Lee et al., 2020). Chlorination conditions prevailing at WRRFs can lead to structural changes in some commonly detected polymers, such as PS and high-density polyethylene (HDPE) (Enfrin et al., 2019; Kelkar et al., 2019). UV irradiation is able to initiate degradation of polymers with a carbon-carbon backbone, resulting in chain scission (Gewert et al., 2015). In Kelkar et al. (2019), results from Raman spectroscopy revealed significant alterations in the composition of PP, PS and HDPE microplastics through prolonged exposure to UV sunlight. In the case of increased levels of suspended solids, which may contain MPs and nanoplastics, the tertiary stage reduces the ability for UV light to

reach potentially harmful microorganisms, thus reducing the operational efficiency of UV disinfection (Lindenauer and Darby, 1994). Given chemical processes' ability to degrade and fragmentate MPs, it is valuable to quantify MPs by mass in addition to by count, as measuring MPs by count can potentially lead to the false conclusion of an increase in the total quantity of MPs.

2.5.4 Retention of MPs in sludge

The majority of MP removal takes place in primary treatment through sedimentation, as previously discussed. Thus, primary sludge typically contains higher levels of MPs (by concentration) which are mainly composed of polymers with densities higher than water's density and bigger in size (Alavian Petroody et al., 2020; Leslie et al., 2017; Magni et al., 2019). In line with MPs found in wastewater, MP fibers are generally more prevalent in primary sludge due to an abundant discharge of MP fibers from laundry machines (Blair et al., 2019a; Gies et al., 2018; Lares et al., 2018a).

Microplastics entering secondary treatment are retained in secondary sludge by settling in the secondary clarifier through bioflocculation with EPS secreted by microorganisms, or by forming MBR sludge cake (Alavian Petroody et al., 2020; Feng et al., 2018; Lv et al., 2019). In general, MP concentration was found to be higher in MBR sludge in comparison to other types of secondary sludge (Lv et al., 2019; Michielssen et al., 2016; Talvitie et al., 2017a), which is in accordance with the higher MPs removal efficiency associated with MBR. Combined sludge (from primary and secondary treatment) contains approximately 88% of MPs from the influent (Table A2).

There are two major pathways for sludge disposal: 1) sludge undergoes thickening and dewatering and is directly disposed to the landfill; 2) thickened sludge is treated in aerobic or anaerobic digestion for stabilization and biosolids production. Aerobic digestion is an oxidation process in which the organic components are broken down by microorganisms, whereas anaerobic digestion is commonly used in large-scale WRRFs to reduce and stabilize organic content in sludge while harvesting biosolids and biogas. The fate of MPs in sludge treatment has important environmental implications due to land application of biosolids (Bayo et al., 2020a; Mahon et al., 2017). Whilst the impacts of sludge treatment processes on MP abundance and transformation have been reported in only a few studies, such as (Mahon et al., 2017) and (Petroody et al., 2021), the effects of MPs on treatment processes have been extensively studied (Pittura et al., 2021; Zhang et al., 2021). A high abundance of MPs in sludge has been found to cause adverse effects during sludge treatment, including reduced methane production, inhibited microbiological activities on anaerobic digestion, and increased greenhouse gas emission in aerobic digestion (Sun et al., 2020). For example, the concentrations of 20, 40, and 60 PVC particles g^{-1} TS decreased methane production by 9.4-24.2% (Wei et al., 2019b); PS with a quantity of 0.2 g L^{-1} or higher led to a decrease of 17.9-19.3% in methane production (Zhang et al., 2020). It needs to be noted that among the four stages of anaerobic sludge digestion (solubilization, hydrolysis, acidogenesis, methanogenesis) (*Wastewater Engineering: Treatment and Resource Recovery*, n.d.), methanogenesis is the most sensitive to the presence of MPs due to the susceptibility of methanogens' activities to the inhibitory environment caused by MPs (Mohammad Mirsoleimani Azizi et al., 2021; Wei et al., 2019a). Due to the impact from the abundance of MPs to the anaerobic process, biodegradable plastics that can degrade under environmental or industrial conditions need further investigation regarding their plausibility and practical use (Shah et al., 2008).

The presence of residual MPs in soil is evident when combining sludge and soil, which results in an increased MP count (Zubris and Richards, 2005). Biosolids are a major source of MPs in agricultural soil, depositing more than 10^2 times the MPs from plastic mulching or other plastic covers (Corradini et al., 2019). Soil with sludge deposits have on average of around 0.2-4 MPs per gram soil (Corradini et al., 2019; Nizzetto et al., 2016). Microplastic particles may also be able to flow due to soil porosity, causing the transport of MP particles to rivers and groundwater (Panno et al., 2019) and potentially impairing the value of biosolids as soil amendments or fertilizers.

2.6 Development of emerging methods for microplastics removal from wastewater

A considerable amount of MP residual could be released to the environment from WRRFs through effluent and sludge, necessitating the development of specific treatment processes aimed at MPs removal. In response to this need, numerous physical-chemical methods were invented or modified from conventional methods based on the properties of MPs and were tested at laboratory or pilot scale and have the potential to be implemented at plant scale in the future.

A number of studies focus on removing MPs from wastewater using purely physical or chemically enhanced physical separation methods mainly based on MPs' properties including different sizes, hydrophobicity, and densities (Herbort et al., 2018a; Perren et al., 2018a; Rhein et al., 2019). One big challenge to remove MPs from treated wastewater is the small sizes of the MP residuals (Lares et al., 2018a; Sun et al., 2019a, 2021). Introducing agglomeration with sol-gel or engineered nanoparticles (ENPs), or (electro-) coagulation could effectively increase MP aggregates' size through various mechanisms such as interparticle bridging, adhesion, or charge neutralization, thus making them easier to remove in the subsequent steps (Herbort et al., 2018a; Misra et al., 2020; Perren et al., 2018a; Rhein et al., 2019; Sorokhaibam and Ahmaruzzaman, 2014;

Zhang et al., 2021). Herbort et al. (2018) verified that sol-gel could be applied at an industrial scale by carrying out the test with process water and under changed climatic conditions regardless of the particle type and size. Perren et al. (2018), who investigated the electrocoagulation (EC) of MPs particles in synthetic wastewater, assessed the operating costs and specific mass removal rate (kg kWh^{-1}) at various current densities, showing the specific removal rate was higher at lower current density (11 A m^{-2}) thus providing practical implications of EC as an energy-efficient alternative for tertiary treatment.

Engineered nanoparticles (ENPs) are widely applied in fields ranging from medicine to environmental remediation (Ramakrishnan et al., 2021; Rhein et al., 2020). Magnetic seeded filtration (MSF), a two-step separation technology which induces agglomeration to target particles with magnetic seed nanoparticles and sequentially separates aggregates by magnetic forces, has been tested for MPs removal from water matrices and high efficiency (>95%) was observed from bench tests (Misra et al., 2020; Rhein et al., 2020, 2019). However, the current research on removing MPs using MSF leaves a number of fundamental questions unanswered regarding large-scale implementation such as regeneration of magnetic seed nanoparticles and optimization of the removal capacity of individual constituents to better apply to more representative wastewater operating conditions (Misra et al., 2020; Rhein et al., 2019).

Filtration has been proven to be effective at physically separating MPs over a wide range of sizes as previously discussed, i.e. MBR, UF, and RO, are highly efficient in retaining MPs whilst small MPs were still able to break through. One possible method to improve MPs removal rate is to alter properties of filtration media. Shen et al. (2021) used aluminosilicate filter media modified by cationic surfactant to separate MPs from solutions with various MP concentrations and achieved >96% removal of MPs through capturing, trapping and entanglement. For membrane

filtration, rapid clogging of the membranes is a major technical challenge. Beljanski et al. (2016) designed a gravitational filtration system to remove MPs from secondary effluent, featuring filters to retain MPs and filter backwashing. Although the gravity filtration system demonstrated high MPs retention and backwash efficiency (95-100%), the study was conducted with artificial MPs-wastewater solution and did not quantify the energy cost associated with the operation of the apparatus or backwash.

The recalcitrant and stable chemical nature of MPs makes their decomposition or conversion a time-inefficient process under normal temperature or pressure. Thus, the development of chemical removal methods is very limited at the moment, and studies on MP degradation were performed in pure water with a few exceptions. For instance, Zhou et al. (2021) designed adhesive polydopamine (PDA) Fe_3O_4 magnetic microrobots (MagRobots) loaded with enzymes that could degrade MPs, and verified their effectiveness in adhering to MPs and degrading MPs in wastewater. In addition, most chemical methods for MPs remediation do not directly eliminate MPs from wastewater, but mostly alter their properties to make them less recalcitrant to degradation. Tofa et al. (2019) grew ZnO nanorods for photocatalytic degradation enhancement, resulting in a 30% increase in the carbonyl index of the LDPE residuals. The photocatalytic oxidation of LDPE with excited heterogeneous ZnO photocatalyst led to the formation of low molecular weight functional groups of hydroperoxides, peroxides, carbonyl, and unsaturated groups on the LDPE surface, resulting in increased brittleness along with wrinkles, cracks, and cavities which later were found to be effectively degraded. This study leads to a potential way of using clean technology with reduced by-products (Tofa et al., 2019).

Notably, a few studies proved that MPs can be efficiently converted under high pressure and high temperature using supercritical water (Bai et al., 2020, 2019; Chen et al., 2019). This

shows that it could be a promising strategy to physically separate MPs from wastewater or treated wastewater, followed by a thermochemical step for decomposition. The technical readiness of emerging technologies for MPs' removal remains low at this moment, and they need to be tested at pilot and large scale for practicality and feasibility validation as well as balance energy use with efficiency of removing microplastics from wastewater.

2.7 Conclusion and future perspectives

Based on collected data and information, this paper critically discussed WRRFs' role and performance in retaining MPs and the methods for MP extraction from wastewater samples. The majority of MPs in wastewater influent can be effectively retained in sludge, with a small portion emitted in effluent. Primary treatment was found to be the most effective for removing MPs, with an overall removal rate of 66.6 ± 29.6 % of overall MPs entering the WRRF. Secondary and tertiary treatment performance is highly dependent on the type of treatment process applied.

The presence of MPs can inhibit microbial growth in secondary treatment, and tertiary treatment can induce MP degradation, leading to a production of smaller plastics which are more difficult to remove. Both of these phenomena highlight the need to remove more MPs during early-stage processes or via source control. Microplastic fibers are the most abundant shape identified, and PET, PP, and PE are the most common MP polymers detected in wastewater influent and effluent. Although WRRFs effectively removed all polymer types up to 99%, the median value of total daily discharge of MPs from WRRFs was 7×10^6 MP particles. High MP discharge from WRRFs creates a necessity for advanced treatment techniques to effectively remove MPs from the WRRF effluent and prevent their release into the environment.

Further consideration and research should be conducted regarding MPs seasonal occurrence and their abundance in sludge. Treated sludge is commonly used as fertilizer or soil amendment, which is an avenue for the MPs accumulated in the WRRF to enter the terrestrial environment and water bodies by either runoff or by flow through porous soil. The fate of MPs in sludge treatment remains unclear due to the lack of comprehensive studies on the fate of MPs in sludge treatment.

The most commonly used advanced treatment technologies, such as UF, RO and AOP, are energy-demanding, and MPs can expedite cake forming on filter-based technology. Other potential technologies for MP removal include mini-hydrocyclones (MHCs), magnetic seeded filtration (MSF), and eletrocoagulation (EC), but these technologies have only been tested on a lab-scale. Therefore, the efficiency, economic feasibility and implementation practicality of these technologies should be tested for MP removal in WRRFs in future studies.

The large variation in MP abundance and fate in wastewater observed in literature cross the globe is partially related to the differences in methods for MP sampling, extraction, and characterisation. Despite limiting the scope to this field, compared to the broader field of MP analysis in the environment, many ongoing activities were identified, with as many or more open questions still remaining. Whilst the characterization of MPs in water moves toward standardization for each sample type, the issue of rapid detection remains central to research and practice. In research, the rapid processing of samples would allow for more dynamic studies, which should address the effects of circadian and seasonal variations on the treatment process, as well as the occurrence of unusual process upsets. In engineering practice, a rapid test would provide the necessary signal for targeted process control of MP removal.

Source analysis and control are the logical steps to complement removal from the aggregate sewage that reaches treatment facilities. Systematic mapping of the nature and source of MPs should explain, for example, the role of MP fibers and their origin from residential and commercial laundering of synthetic fabrics. The comparative analysis of retrofitting multiple small sources vs. upgrading fewer centralised treatment facilities should inevitably include the engagement and acceptance of stakeholders and the analysis of overall environmental effects, as was the case, for example, of replacing incandescent bulbs with compact fluorescent or LED lights (Lim et al., 2013; Sandahl et al., 2006). Effective scientific communication on the topic of MPs to a wider audience beyond the engineering and scientific communities can potentially reduce MP influx in WRRFs.

Driven by toxicological concerns, government agencies in regions and countries where MPs have been widely studied have started to form policies for reducing or monitoring MPs in water matrices. In January 2020, Italy joined 12 other countries that have implemented a ban on the production of microbeads in rinse-off cosmetics (Pallone, 2015). In the United States, California is the first state in the United States to make an effort to address MPs in drinking water, with the SB-1422 California Safe Drinking Water Act proposing a definition of MPs in drinking water as a precedent for the detection and reporting of MPs in drinking water. Subsequentially, additional regulations are anticipated on the monitoring and removal of MPs from various matrices. Such regulations on monitoring and removal should be informed and refined based on risk assessment of microplastics in effluent, biosolids and reclaimed water in order to optimize economic and labour expenses in local agencies.

Chapter 3 Method for Recovery MPs from Wastewater

3.1 Introduction

The investigation of MPs in water matrices is rapidly evolving around the globe. However, the separation of MPs, especially the fine ones, remains a major challenge. Chemical digestion to remove the mineral, biogenic organic substances is a crucial step for MP separation. The optimal chemical digestion method needs to minimize, if not completely eliminate, biogenic or natural organic material whilst preserve the integrity and original characteristics of the target MPs (Pfeiffer and Fischer, 2020) and produce a minimum amount of by-product.

As is discussed in Ch. 2, numerous chemical digestion methods have been previously used for recovering MPs from wastewater, but they had apparent disadvantages that prevented them from directly being adopted for this study. Existing methods evaluation studies proved that commonly used chemical digestion methods such as acidic and alkaline digestions, WPO, WPO with Fenton's reagent, could lead to damages on MPs to different extent (Hurley et al., 2018a; Lares et al., 2019a; Pfeiffer and Fischer, 2020; Tagg et al., 2017). In addition, Fenton's reagent could also lead to $Fe(OH)_3$ precipitation at certain pH and concentration (Fig. B1). Enzymatic digestion was an alternative with minimum damages to MPs, but was also associated with trade-offs like high costs and time inefficiency (Gies et al., 2018; Mintenig et al., 2017) which made it unsuitable for a long-term sampling like this study. To ensure the reliability, applicability and replicability of the digestion method applied later in the long-term monitoring of MPs, an objective evaluation of existing chemical digestion method on wastewater samples was conducted regarding three criteria: 1) mass recovery; 2) amount by-product; 3) changes in surface morphology. The evaluation encompassed acidic, alkaline digestion, WPO, and WPO with Fenton's reagent in different digestion environments.

3.2 Materials and Methods

3.2.1 Chemical digestion methods evaluated

The methods evaluated are summarized in Table 3-1. Method 1 to Method 4 were adopted in previous studies, and Method 5 was modified within the scope of this dissertation according to the formation of ferric hydroxide at different pH and Fe ion concentration. Method blanks were prepared for all methods listed as well as reference particles, and the results from method blanks were incorporated in the calculation of mass recovery of reference particles from each digestion method. It needs to be mentioned that the chemical digestions have been applied at different temperatures previously. For instance, Hidayaturrahman and Lee (2019) and Park et al. (2020) applied WPO at room temperature and at 60^{o}C respectively. However, according to findings reported in Pfeiffer and Fischer (2020), the differences in mass change at various temperature was insignificant.

For each trial, approximately 20 mg of each reference MP were weighted to 0.1 mg accuracy and transferred into pre-rinsed glass tubes, and 20 ml of respective solution was added to the test tubes with a pipet. The exposure time was 24 hours for all trials. All experiments were carried out in triplicates. After 24 hours, solids were separated from liquid by vacuum filtration with pre-weighed PC filter paper (0.8 μm, Millipore) and rinsed with Milli-Q water. The filter paper containing solids was then transferred to an aluminum pan covered with watch glass for drying at 40 ^{o}C. After drying, samples were weighed again to 0.1 mg accuracy. The mass recovery was then calculated based on the difference between the initial and final mass.

Table 3 - 1 Organic material digestion methods comparison

	Type of Reagent	Reagents	Reaction Environment
1	Oxidizer	30% H_2O_2 and 0.05 M Fe (II) solution at 1:1 v/v	Heated to 60 °C and stirred
2	Oxidizer	30% H_2O_2	60 °C
3	Acid	1 M HCl	Room temperature
4	Base	10% KOH	60 °C
5	Oxidizer	H_2O_2 (30%) with Fe at a ratio of 25:1 w/w and pH 3.5 buffer	Heated to 60°C and stirred

3.2.2 Origin and processing of reference MP particles

Four types of MP particles of different sizes and densities were used in this study (Table 3-2). These MPs have been abundantly found in wastewater (Sun et al., 2019b; Talvitie et al., 2017b; Xu et al., 2019c), freshwater (Akindele et al., 2019; Bharath K et al., 2021), seawater, and sediment (Ng and Obbard, 2006; Sun et al., 2018). Previous assessments of chemical digestion method such as Tagg et al., (2017), Hurley et al., (2018) and Pfeiffer and Fischer (2020) chose large, primary and pristine MPs, which are not representative of MPs in environmental matrices. In this evaluation, a mixture of large and fine MPs was selected. Furthermore, secondary PP particles were produced from 2-3 mm PP pellets (Fairfield™) in the laboratory with a kitchen grade blender to simulate the mechanical fragmentation of plastic pieces in garbage disposal. Ground PP were then sieved with stainless steel mesh of 25 µm, 150 µm and 900 µm. The surface morphology and chemical composition of secondary PP particles were examined using a scanning electron microscope (SEM; Megellan-X400) and Raman Spectroscopy (Renishaw®) respectively.

Table 3 - 2 Reference MP particles used in this study

Type of MP	Density (g cm^{-3})	Size Range (µm)	Source
Low-density Polyethylene (LDPE)	0.924	12-20	Primary
Polystyrene (PS)	0.96	900 - 1000	Primary
Polyamide (PA)	1.15	15-20	Secondary
Polypropylene (PP)	0.905	25 – 900	Primary

3.3 Results and discussion

3.3.1 Characteristics of secondary PP particles

The Raman Spectra were obtained for secondary PP of all three size ranges (Fig. 3-1). The differences in intensity between the original PP pellets and secondary PP was due to their different thickness, but the chemical nature of the secondary PP was unaltered. According to the SEM images of secondary PPs, the morphology and size were not uniform, as one would expect from actual secondary microplastics, thus the production of secondary PPs can be replicated.

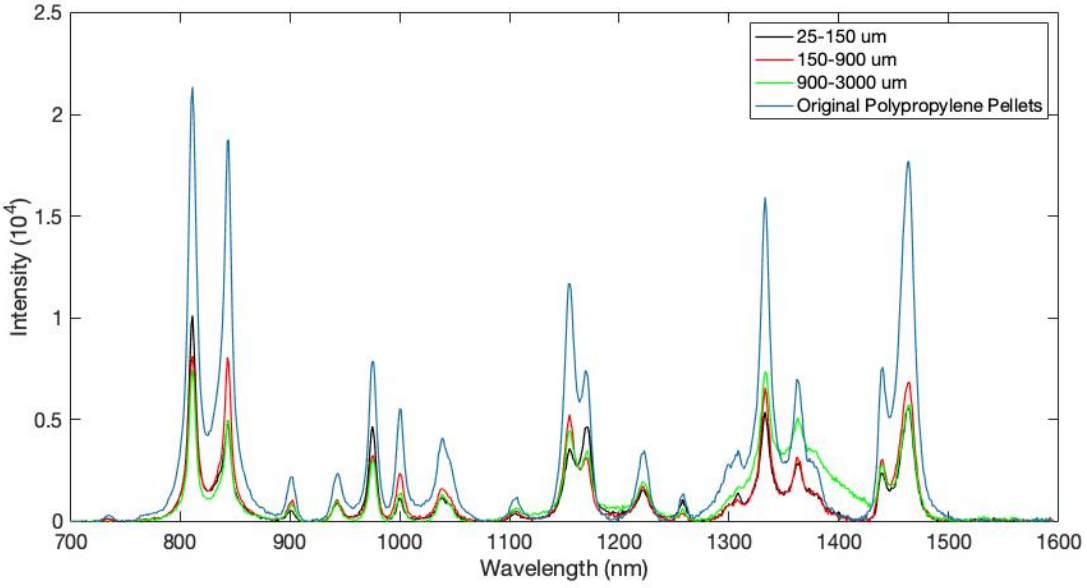


Figure 3 - 1 Raman Spectra of primary and secondary PP used in this study

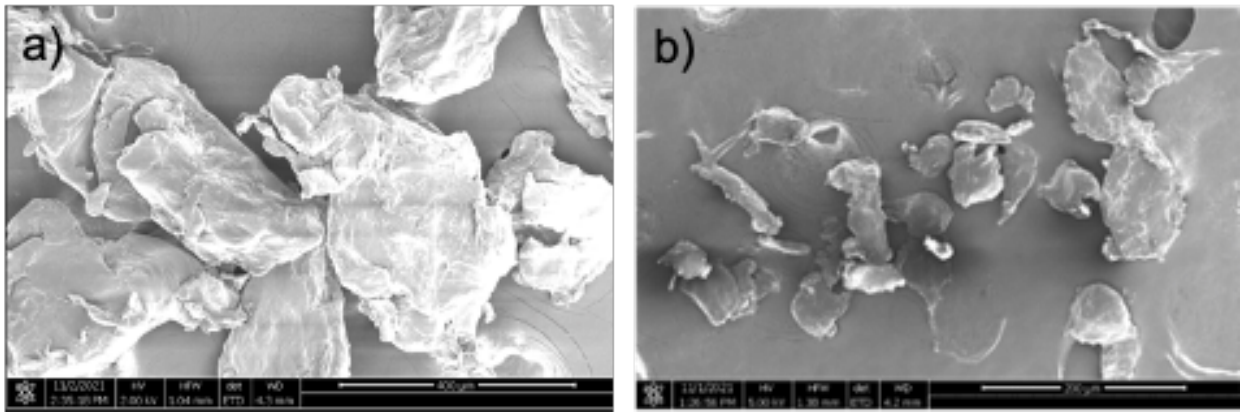


Figure 3 - 2 SEM images of a) secondary PP of 150-900µm; b) secondary PP of 25-150µm

3.3.2 Mass recovery and morphology of reference MPs

The results concerning the mass recovery of reference MPs from applied chemical digestion revealed the advantages of oxidizers over acid and base (Fig. 3-3). The average recovery efficiency was 88.5%, 82.7%, 83.1%, 92.6% and 92.8% for alkaline, acidic, WPO, WPO with Fenton's reagent and the modified digestion from this study respectively. The mass recovery from acidic digestion was approximately 10% lower than ones of WPO with Fenton's reagent. Acidic digestion had been reported to have a larger impact on plastic polymers as it could lead to partial degradation (Karami et al., 2017; Naidoo et al., 2017). The acidic digestion (HCl) also showed the greatest impacts on PA, which was in accordance with Pfeiffer and Fischer (2020). Figure 3-3 also demonstrated that different polymer types had different levels of resistance to chemical reagents. Polypropylene and Polyamide were associated with higher mass losses overall, which means in real sample processing, PA and PP are more likely to be underestimated, especially for mass-based quantification.

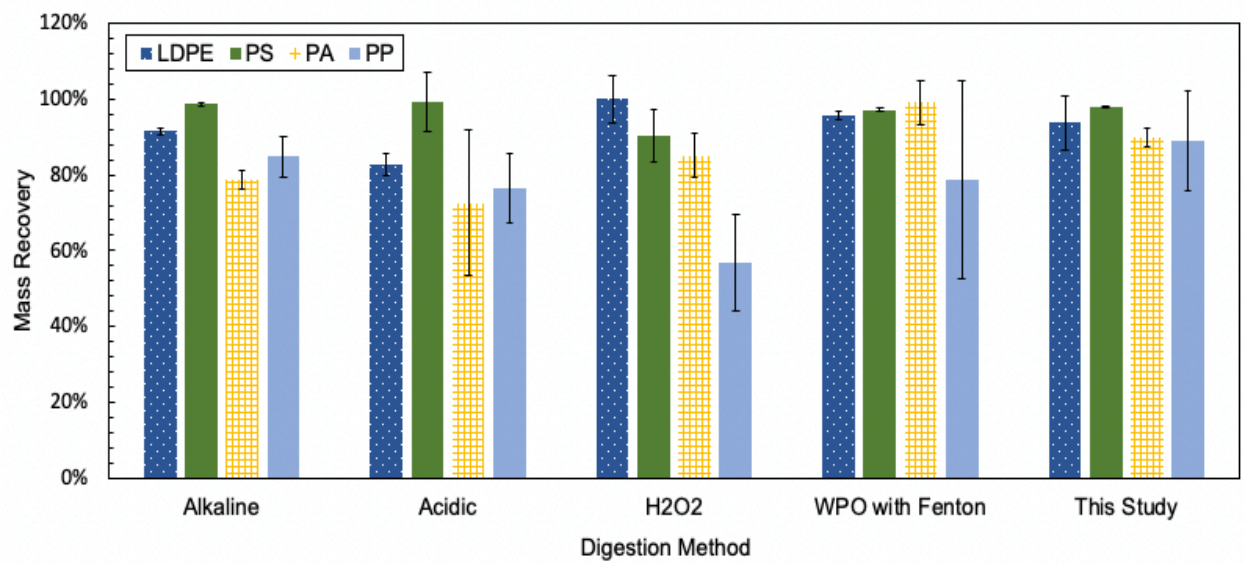


Figure 3 - 3 MPs' mass recovery from each chemical digestion method

Microplastic particles were examined under an SEM before and after digestion. The acidic digestion led to server damages on MP surfaces (Fig. 3-4), corresponding to a high mass loss whilst other digestion led to milder changes to MP surfaces. Notably, a layer of precipitates formed on the MPs' surface from Fenton's reagent as was indicated by the sharp edges on the rough MP surface (Fig.3-4). The orange-brown precipitates (Fig. B2) largely obstructed the visual observation of MPs under microscope. Although the majority of precipitate could be separated using density separation, it was still likely to have smaller MPs trapped in the iron precipitate and $Fe(OH)_3$ attach to the surfaces of MPs, changing its color and appearance in the characterization process. On the other hand, the sole application of H_2O_2 did not lead to any byproduct that would interfere with visual or spectral analysis, but led to a higher mass loss and was an inefficient digestion compared to the others (Pfeiffer and Fischer, 2020).

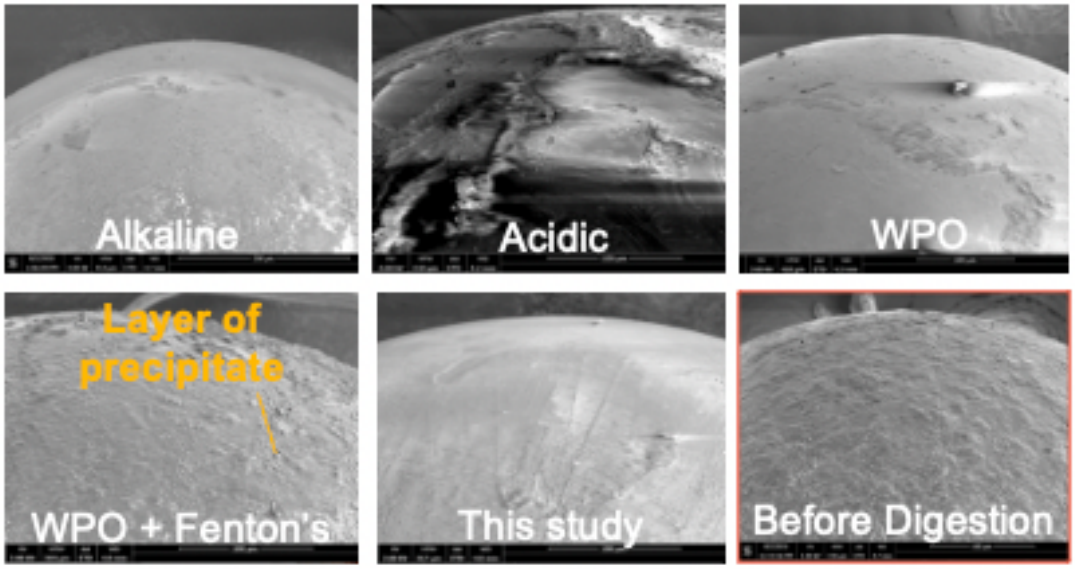


Figure 3 - 4 SEM images of PP surface before and after digestion with each method

3.4 Conclusion and limitations

Considering both byproducts and resistance of reference MPs to the chemical digestion, the outcome of this comparison reveal that the modified WPO proposed for the long-term sampling was achieved the most ideal results. It is worth noting that digestion efficiency was also an important criterion for chemical digestion methods, but it was excluded from this evaluation as the major concern was protecting target MPs for identification and characterization, and chemical methods selected for comparison was screened qualitatively for digestion efficiency. Destructive or inefficient methods were excluded prior to the experiments. However, it would still be value to further evaluate these methods for their efficiency with solids from actual wastewater sample.

Chapter 4 Microplastics' Occurrence and Fate in a Local WRRF

4.1 Introduction

As is shown in Ch. 2, a number of studies have been explored the occurrence, abundance, and fate of microplastics within WRRFs, by driven by toxicity and contaminants vector potential of MPs. However, most of the studies are short-term or one-time sampling. Cao et al. (2020) sampled multiples times during 24 hours to capture the diurnal trend, and Conley et al. (2019) sampled consistently for 3 months for intra-season variation of microplastics. To address this research gap, this study was launched to map the microplastics in wastewater over one year. The goal of this study is to understand the seasonality of microplastics in wastewater through, and their characteristics at different stages within a WRRF. Moreover, EPL team also investigated potential contribution of MPs from plastic compartments within an WRRF. Limitations and future work are also discussed in § 4.4.

4.2 Materials and Methods

4.2.1 Wastewater sampling

The samples were taken from Chiquita Water Reclamation Facility (CWRF) in Rancho Santa Margarita, California from August 2020 to April 2021, covering both dry and wet seasons. This facility serves 203,000 residents and has a treatment capacity of 10 MGD. The facility only receives municipal wastewater and is equipped with mechanical screens, grit chambers, chemically enhanced primary clarifiers, activated sludge, secondary clarifier, tertiary cloth filters and chlorination contact basins. The wastewater characteristics of the sampling months are displayed in Table C1.

The types of samples included primary influent (PI), primary effluent (PE), secondary effluent (SE) and tertiary effluent (TE) (Fig. 4-1). The sample volume was calibrated prior to the sampling campaign. Automatic composite samplers onsite were used for taking 24-hr composite samples, and all samples were taken on the last Thursday of each month. The sampling volume for primary influent and effluent was one gallon, and two gallons for secondary and tertiary effluent. The volume was calibrated prior to the start of sampling campaign.

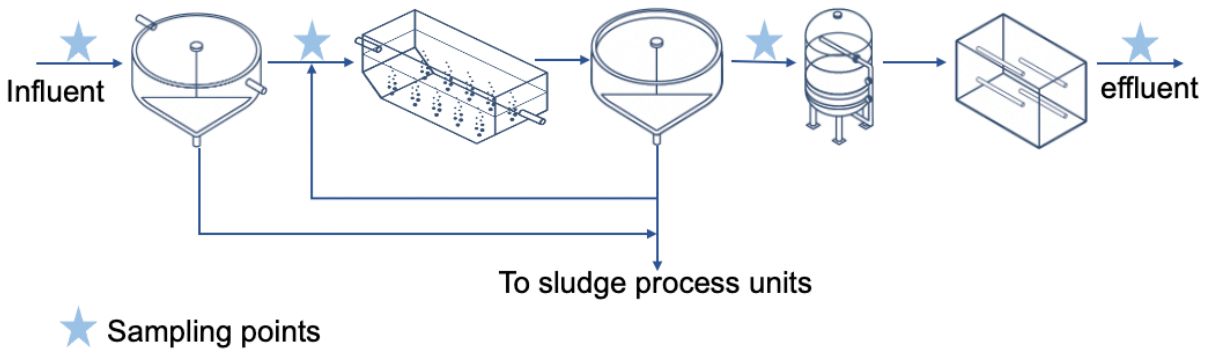


Figure 4 - 1 Process flow diagram of CWRF and sampling points

4.2.2 Sample processing

All samples were collected and transported in glass bottles pre-rinsed with Milli-Q water, and sieved in laboratory with stainless steel meshes with openings of 25 um, 150 um, 900 um, and 5 mm (Fig. 4-2). In order to capture the small microplastics, we vacuum filtered the filtrate from the 25-um sieve with 0.8 um PC filter membranes (Millipore). Solids retained on the meshes were then rinsed into Erlenmeyer flasks with Milli-Q water and dried at 60 C degrees.

The modified method described in Sec.3 was applied to eliminate the organic matrix from solid samples. Previously, this digestion method has been used in various studies with each of its own modifications (Dyachenko et al., 2017; Lares et al., 2018a; Mason et al., 2016; Vardar et al.,

2021). As aforementioned, these versions of WPO are associated with limitations such as large amount of precipitates, high acidity and reactivity. Therefore, our team made modifications to this digestion step based on the solubility of Ferrous precipitates in water at different pH (Fig. B1). In this step, 20 ml of 30% H_2O_2 was mixed with reduced concentration of Fenton's reagent (1 Fe: 25 H_2O_2 w/w), and the solution was kept stirred at 60 C degrees. The pH of the digestion was buffered to be approximately 3.5 as the optimal pH of WPO with Fenton's reagent is between 3-5 (Hurley et al., 2018b; Tagg et al., 2017). For samples with higher organic content, i.e. primary influent and primary effluent, additional 5 ml of H_2O_2 was added incrementally after the fifth hour of digestion. The amount of H_2O_2 needed was estimated based on the total suspended solids (TSS) of each type of sample.

Following chemical digestion, solids were separated using vacuum filtration with 0.8 um filter membranes (Millipore). To eliminate the interference of visual characterization and background noise in analytical instrument in a later step, we adopted the method in Zhao et al. (2017) and Fortin et al. (2019), in which solids retained on the membrane were resuspended in 7-10 ml of methanol through sonication. Because the membrane fragmented in sonication during the time intervals suggested by Fortin et al. (2019), a 30 seconds interval was used, and after 30 seconds of sonication in water bath, the filter membrane was retrieved from the suspension, rinsed with 1 ml of methanol, and observed for any remaining solids under an optimal microscope (Olympus, BH2). If any solids were seen, the above procedure was repeated. The methanol suspension was then evaporated to 5 ml and transferred to a counting chamber with glass pipets. The solids on counting chambers were then examined and characterized.

4.2.3 Microplastics identification and characterization

The counting chambers were directly examined under an optical microscope equipped with 10, 20, 40 and 60 times of objective lens (Olympus BH2). Suspected microplastics were characterized with Raman Spectroscopy (Renishaw). The operational parameters are summarized in Table 4.1. The color and shapes of suspected microplastics were documented, and the shapes included mainly fibers, fragments and particles.

Table 4 - 1 Operational parameters of Raman Spectroscopy

Wavelength	**Intensity**	**Aperture lens**	**time**	**Accumulation #**
532 nm or 785 nm	10%	50 X objective lens	10 (sec)	10

Polymer type was determined by comparing the query spectra to the Raman spectral library of MPs (Munno et al., 2020) using a data processing tool built in MATLAB. Data processing included baseline removal, wavenumber matching, data truncation and spectral linear kernel (SLK) (Khan and Madden, 2012) as spectral search algorithm. The complete code can be found in Appendix C.

4.2.4 Assessment of tertiary filter

A used tertiary cloth filter was obtained from CWRF. The cloth filter was pre-rinsed with Milli-Q water and cut into 2.5-inch by 2.5-inch squares (Fig. 4-2) for bench-scale filtration experiments. A small amount of fibers was directly retrieved from the cloth filter and its Raman spectrum was taken as a reference for fiber identification in filtrate. Two sets of cloth filters were tested for fibers, each of which were cleaned with a different procedure: The first set was cleaned with thorough rinse with Milli-Q water from all directions, and the second set was cleaned with

Milli-Q water rinse followed by 15-minute sonication. The purpose of comparing two cleaning procedure was to examine if the cleaning procedure with sonication would cause abnormal wearing of the cloth filter thus leading to higher counts of fibers. The cleaned filter cloths were then placed in between two flanges and the apparatus was set up vertically to ensure the flow of water was orthogonal to the filter surface. One liter of Milli-Q water was slowly poured through the filter and filtrate was vacuum filtered with 0.8 μm PC membranes. The membrane was examined under the optical microscope for any fibers shed by the cloth filters, and sequentially confirmed by Raman Spectroscopy.

Figure 4 - 2 Cleaned and sonicated cloth tertiary filter

4.2.5 Quality and contamination control

Quality assurance and control (QA/QC) were applied to avoid contamination. Procedural blanks and positive control samples were used to identify potential contamination from sample processing and to account for MPs' loss during sample processing respectively. Procedural blanks

were processed once prior to the sampling campaign and once after. The positive control samples were composed of 20 mg reference MPs (5 mg LDPE, 5 mg PA, 5mg PVC and 5 mg PS) ranging from 12 μm to 900 μm in one liter of Milli-Q water.

In this study, the use of plasticware was minimized. Parafilm and plastic caps were replaced with aluminum foil or watch glass, and nitrile gloves, cotton clothing and bright blue lab coat (Normex®) were worn when the team was collecting or processing samples. In addition, all sampling equipment (i.e. glass storage bottles, stainless steel funnel, squeeze bottles), sieves and glassware were pre-rinsed with Milli-Q water before usage.

4.3 Results and discussion

4.3.1 Quality assurance and control

A total number of 2.8 ± 1.1 pieces of MPs per liter were identified from procedural blanks. The contamination could be traced to squeeze bottles and pipette tips because of their polymer types (LDPE and PP respectively) and colors. On the other hand, the mass recovery efficiency of the reference MPs was 89.2 ± 3.62 %, which was in the same range as reported in previous studies (Gies et al., 2018; Simon et al., 2018b). The loss of MPs was probably due to the chemical digestion and loose particles during transferring steps.

4.3.2 Abundance and removal microplastics

The abundances of MPs were expressed as count of MPs per liter of wastewater sample in this study. In general, MP abundances in wastewater influent were higher in the winter season (December to February), and the occurrence of MPs displays a negative correlation with temperature. The MP abundance of each month was statistically difference from one another

according the results of a One-way ANOVA test. There were a couple of studies that explored the seasonality of MPs in wastewater (Kim et al., 2022; Lares et al., 2018a; Roscher et al., 2022), but the findings differed substantially probably due to different living habits of the residents served by the WRRFs, sewage system type (combined vs. separated sewage system), type of wastewater received, etc. The average concentration of MPs in PI, PE, SE and TE were 119.9 ± 34.3, 38.7 ± 19.8, 35.3 ± 3.37 and 30.0 ± 5.66 MPs L^{-1} respectively. Noticeably, the concentration of MPs in primary effluent was increasing from summer to winter. The increase could be due to the lower sedimentation efficiency resulted from higher viscosity at lower temperatures. Moreover, higher concentrations of MPs had been observed in tertiary effluent than secondary effluent because of the fibers from tertiary cloth filters, which is evidenced by the higher percentage of PES fibers (§4.3.3) and results discussed in §4.3.4.

Approximately 73% of MPs was removed from the wastewater stream (Fig. 4-3). The majority of the removal took place in primary treatment, which was in agreement with the majority of the literature reviewed (§2.5). During primary treatment, dense MPs and MPs trapped in solid flocs are removed via gravitational settling; less dense MPs and the ones trapped in buoyant flocs could be removed by skimming. The removal of MPs from secondary and tertiary treatment was statistically insignificant. It is worth noting that MPs could undergo fragmentation in treatment process units (Bui et al., 2020a; W. Liu et al., 2021; Ngo et al., 2019a), and quantification of MPs by count, which was used in this study, could potentially lead to an overestimate of MPs and was unable to capture potential mass changes caused by prevalent plastics-degrading microbes in sewage and activated sludge (Wu et al., 2022). Thus, future studies are recommended to use both mass-based and count-based quantification when possible.

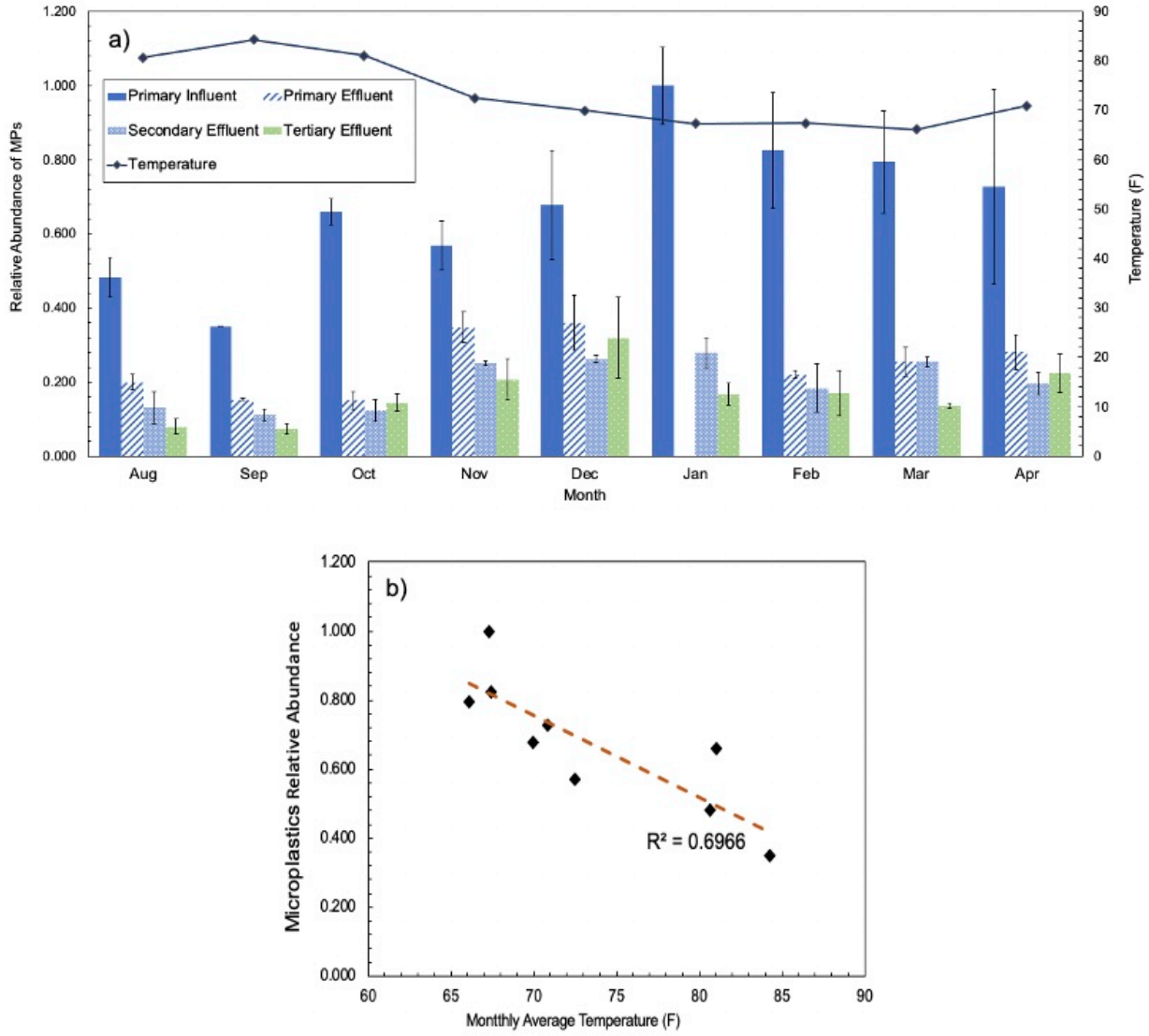


Figure 4 - 3 a) Relative abundance of MPs in different stage of the WRRF (* Primary effluent for January 2021 damaged; data presented as relative abundance as requested by funding agency); b) correlation plot of MPs in wastewater influent and monthly average temper

4.3.3 Size distribution, polymer type and morphology

The MPs collected in study was categorized into four size classes: <25 μm, 25-150 μm, 150-900 μm and 900-5000 μm. Microplastics smaller than 25 μm were the most prevalent

throughout the treatment stream and their abundance increased along the treatment processes (from 52% to 82%) (Fig. 4-4). The higher relative abundance of smaller MPs could be resulted from fragmentation of larger MPs or easier removal of larger MPs during treatment processes. The prevalence of smaller MPs, especially in tertiary effluent (Fig. 4.4), highlights the need to investigate the nature of those MPs and implement strategies to target their removal for water reclamation and reuse purposes. It should be noted that it is difficult to compare the differences in abundances of MPs of various size because a wide range of sieve sizes was used (Fig. 2-2).

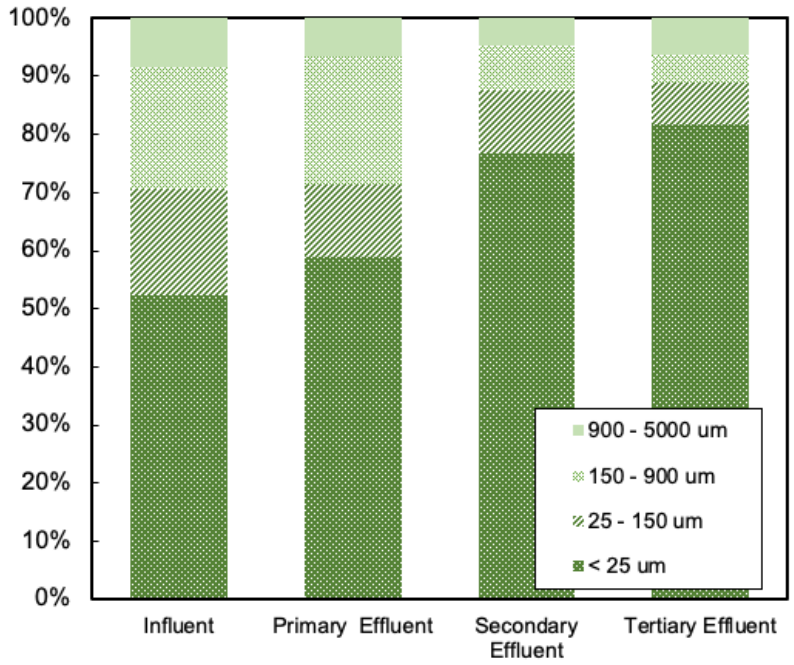


Figure 4 - 4 Average size distribution of MP in the four types of wastewater samples

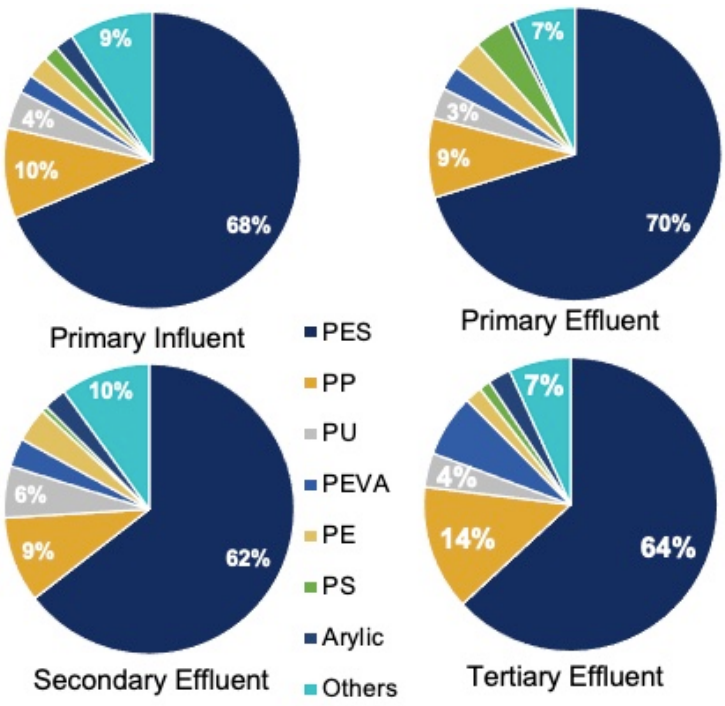


Figure 4 - 5 Average distribution of polymer types in the four types of wastewater samples

A total number of 16 types of polymer was identified from wastewater. A full list of polymer types can be found in Appendix C. Polyester was the most abundant type of polymer in all four wastewater samples, accounting for 68% of all MPs (Fig. 4-5), and all PES pieces detected were in shape of fibers. Similar findings were reported by Lares et al. (2018); Long et al., (2019) and Simon et al. (2018). These PES fibers were likely to originate from shedding of synthetic clothing. Polypropylene (PP) was the second most prevalent type of plastic, which was observed as fibers and fragments. Polypropylene is often used to make surgical/disposable masks. Their wide usage during the pandemic has been evidenced to prevalence of PP fibers in wastewater (Aragaw, 2020; Pikuda et al., 2022). It was also consistently observed that the distribution of polymer types only unsubstantially shifted from primary influent to tertiary effluent. This implies

that conventional processes in the WRRF were not selective in removing specific types of polymers, but only removed MPs on the whole.

4.3.4 Microplastic fibers from tertiary filter

A substantial number (23.6 ± 7.78 pieces per liter) of polyester fibers was detected in tertiary effluent. Used tertiary cloth filters were assessed as a potential source of PES fibers. In total, 18.8 ± 6.65 pieces of PES fibers per liter were found in filtrate from the cloth filter (cleaned with MilliQ-water only), which indicated that a large proportion of PES fibers in tertiary effluent could be traced to the cloth filter. The PES fiber concentration from the set cleaned with rinse and sonication was 157 ± 7.89 pieces per liter, showing sonication released more fibers than normal operational cleaning and was not suitable for cleaning the cloth filter. It should be noted that the cloth filers were used and at the end of their lifespan. Future experiments should be conducted with new cloth filters to confirm the results.

4.4 Conclusion and limitations

This study provided insights to seasonal variation of MPs in municipal wastewater through a nine-month sampling campaign. The results have shown a 73% overall removal of MPs ranging from smaller than 25 μm to 5000 μm in a WRRF located in southern California. The occurrence of MPs increased from summer to winter season, and was negatively correlated to temperature. Small size MPs were the most abundant, rising concerns of fine MPs and nanoplastics in treated wastewater. Fibers constituted the majority of MPs detected in this study, which indicated that laundry discharge was a primary source of MPs in municipal wastewater. Tertiary cloth filters were evaluated for their MP fiber shedding potential via bench-scale filtration test and 157 MP fibers L^{-1} was found in the filtrate.

Although this study covered summer and winter season, the nine-month sampling was a limitation as it was unable to capture a year-long variation of MPs or include more data points for a more comprehensive statistical analysis. Moreover, numerous pieces of treatment equipment in WRRFs is made of plastics, such as diffuser seats, conveyance pipes, membrane filters and the contribution of MPs from WRRFs needs to be more comprehensively and systematically evaluated in order to effectively remove MPs from wastewater.

Acknowledgement

The author would like to thank EPL team, UCI Laser Spectroscopy Lab for technical and instrumental support, Santa Margarita Water District for providing samples and on-site support.

Chapter 5 Mini-hydrocyclone for Microplastics Separation

5.1 Introduction

As discussed in preceding chapters, MPs are abundantly found in wastewater effluent, especially small ones, and mitigation of MPs in water reclamation can be achieved through direct removal or source control. This chapter presents mini-hydrocylone (MHC) as a novel method for MP separation, which could be utilized as a direct removal as well as source control device.

Hydrocyclones, a mature technology, have been applied to processing plastic waste and could be a potential candidate for MP separation (Fu et al., 2019; Gent et al., 2018; He et al., 2022; Malcolm Richard et al., 2011). Hydrocyclones have been utilized in a variety of industries such as petrochemical, food processing, pharmaceutical, bioprocesses, and wastewater treatment (Cilliers and Harrison, 2019, 1997; da Silva et al., 2020; L. Liu et al., 2021c; Yu and Fu, 2020), attributing to their large hydraulic load capacity, fast separation speed, cost effectiveness, low operation and maintenance requirements, consistency, and reliability (L. Liu et al., 2021a; Yang et al., 2011). Particle separation from the liquid phase is achieved under the three major forces acting on any given particle: centrifugal, buoyancy, and drag force (Shakeel Syed et al., 2017). A mini-hydrocyclone (MHC) has overall cylindrical geometry and configuration similar to a conventional hydrocyclone (CHC) (Fig. D1). However, MHCs are more efficient at separating finer particulates because they have a smaller nominal diameter resulting in a smaller cut size (Niazi et al., 2017; Yang et al., 2013). Numerous studies on MPs in water and wastewater indicated that smaller MPs ($< 100\ \mu m$) account for a considerable portion of total MPs and are difficult to remove (Jiang et al., 2020; Lares et al., 2018b; Mason et al., 2016; Z. Wang et al., 2020). Given MHC's ability to capture finer particles, they can be used to separate MPs from different water matrices.

Previously, various efforts (Chen et al., 2021; Cilliers and Harrison, 2019; He et al., 2022; Lv et al., 2018) have been made to employ MHCs for fine particles separation, including MPs, from water matrices. Lv et al. (2018) showed that the separation efficiency of fine particles from industrial wastewater using parallel MHCs reached 77.2% on average. Cillier and Harrison (2019) used MHCs to recover yeast flocs with median sizes between 20 and 200 μm, and achieved up to 85% yeast recovery in a single pass. He et al. (2022) used 3D printed MHCs to separate polymethylmethacrylate (PMMA) particles in the size range 5-20 μm with recovery rate up to 95%. Notably, the particles in these studies are all denser than water ($>10^3$ g cm^{-3}). Microplastics in the environment have densities both higher and lower than water's density, and the treatment of MPs less dense than water still needs to be investigated. Moreover, it is also crucial to assess MHC's MPs separation performance under environmental conditions.

The goal of this study is to investigate the performance of MHCs in separating fine MP particles of density higher and lower dense than water's, i.e. polyamide (PA) and low-density polyethylene (LDPE). To this end, three MHCs with different sizes and configurations are designed and manufactured with metal 3D printing. Mini-hydrocyclones' separation efficiency is evaluated at different operational parameters, including split ratio, flow rate, concentration, and configuration (with single MHCs and MHCs in series). To assess the MHCs' performance under environmental conditions, separation tests of single MHCs are also carried out with MPs weathered by UV and in synthetic wastewater effluent. Future directions and limitations are also discussed. This is the first study where MHCs are designed and used for separation of MPs of different densities in water and an environmental matrix, providing a basis for future operation and application of MHCs in MPs treatment.

5.2 Materials and Methods

5.2.1 Design and manufacturing of MHCs for MP separation

Microplastics in water matrices have densities ranging from 0.92 to 2.30 g·cm^{-3} (Sun et al., 2019b). The optimal geometric parameters for the MHC change depend on the physical parameters of the MPs (primarily density) (L. Liu et al., 2021b; Martínez et al., 2008). Two types of MHCs with the same main diameter (D_c = 10 mm for both) and different other geometric parameters were designed in EPL to separate MPs with densities both higher and lower than water's density, and manufactured by 3D printing with stainless steel (Fig. 5.1). Their geometric parameters are listed in Table 5-1. For the convenience of description herein, they are referred as MHC_H (separating the high-density MPs) and MHC_L (separating the low-density MPs).

Table 5 - 1 Geometric parameters for MHC_H and MHC_L

MHC types	D_o / D_c	D_u / D_c	L_c / D_c	L_v / D_c	L_d / D_c	a / D_c; b / D_c	θ
MHC_H1 and MHC_H2	0.2	0.18	0.52	0.38	3	0.15; 0.3	7°
MHC_L	0.2	0.25	1.06	0.16	9.63	0.18; 0.32	5.8°

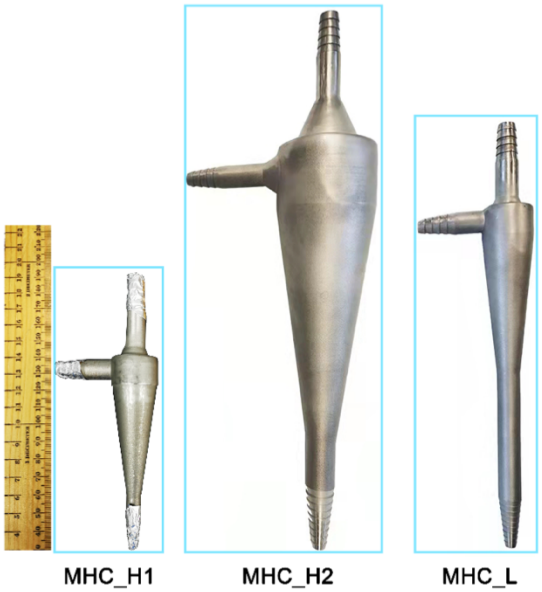


Figure 5 - 1 The prototypes of 3D printed stainless steel MHCs

5.2.2 Microplastic particles, synthetic stormwater and wastewater effluent

Microplastics selected for testing in this study were polyamide (Nylon; GoodFellow) and low-density polyethylene (LDPE) particles (Shiyansanzhou Tech., Inc.), which have been frequently detected in wastewater systems. The size range and densities can be found in Table 3-2, and their SEM images are included in Appendix D (Fig. D2).

Microplastics abundantly found in the environment are often weathered and oxidized by environmental conditions such as UV exposure, acidity, mechanical abrasion (Alimi et al., 2018; Bui et al., 2020a; Hanif et al., 2022). Therefore, LDPE and PA particles were exposed to UV in a solar simulator (40 mW cm^{-2}; RAYONET) for 15 hours and used for separation test in ultra-purified water, synthetic stormwater and wastewater. The LDPE and PA were examined with Raman microscopy before and after UV exposure, and no change in chemical composition was observed. The Raman spectra are included in the SI (Fig. D3).

Microplastics in storm runoff can be a major contributor to MPs in the influent of WRRFs connected to combined sewers (Lee and Kim, 2018b; Picó and Barceló, 2019; Shruti et al., 2021). Thus, the MHCs' separation performance was also evaluated with UV-weathered synthetic stormwater. The synthetic stormwater composition (Table 5-2) was determined based on Weisbrod et al. (1999) and Dunphy et al. (2007), and constituent concentrations were verified to be within typical concentration ranges of California highway runoff (Kayhanian et al., 2019). Furthermore, previous chapters convey the need to further separate MPs from treated wastewater. Therefore, MHCs' separation efficiency was also tested in synthetic wastewater effluent (Table 5-3). The synthetic wastewater composition was adopted from Xu et al. (2014), Ma et al. (2020) and McVeigh and Weatherley (1999) to simulate secondary wastewater effluent.

Table 5 - 2 Characteristics of the synthetic stormwater used in these experiments

Synthetic Stormwater Constituents	**Value**	**Chemicals used**
pH	6.8	NaOH
Dissolved Organic Compound	15 mg L^{-1}	Humic Acid
Na^{+}	16.5 mg L^{-1}	NaOH, NaCl, Na_3PO_4, $NaNO_3$
Cl^{-}	14.5 mg L^{-1}	NaCl
Cu^{2+}	0.08 mg L^{-1}	$CuSO_4$
SO_4^{2-}	0.12 mg L^{-1}	$CuSO_4$
Hardness	12.2 mg L^{-1}	$CaCO_3$
Total Phosphorus	0.6 mg L^{-1}	Na_3PO_4
NO_3^{-}	2 mg L^{-1}	$NaNO_3$, $Pb(NO_3)_2$
Pb^{2+}	0.08 mg L^{-1}	$Pb(NO_3)_2$

Table 5 - 3 Characteristics of the synthetic wastewater used in these experiments

Synthetic Stormwater Constituents	Value	Chemicals used
pH	7.1	
Na^+	49.3 mg L^{-1}	Na_2HPO_4, $NaNO_3$, $NaHCO_3$
Cl^-	77.4 mg L^{-1}	$CaCl_2$, KCl
Ca^{2+}	6.81 mg L^{-1}	$CaCl_2$
SO_4^{2-}	27.1 mg L^{-1}	$MgSO_4$
Mg^{2+}	9.03 mg L^{-1}	$CaCO_3$
K^+	16.8 mg L^{-1}	KCl
Total Phosphorus	5.89 mg L^{-1}	Na_2HPO_4
NO_3^-	53.3 mg L^{-1}	$NaNO_3$
NH_4^+-N	25.2 mg L^{-1}	NH_4Cl

5.2.3 Hydraulic circuit and optimization of operating parameters

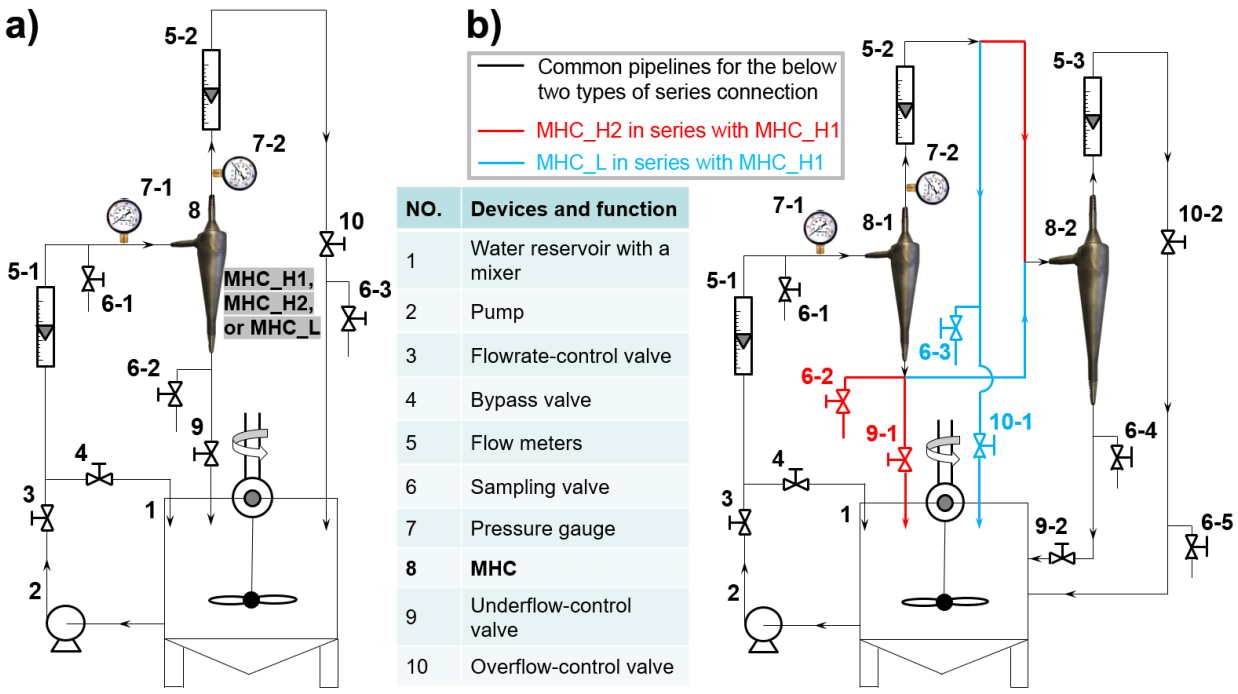
Two hydraulic circuits were built for the separation efficiency test (Figs. 5-2 a and b). MHCs' overflow and underflow were channeled back into the reservoir to maintain the consistency of particle concentration. The flowmeters, pressure gauges, and control valves were installed for the adjustment of operating parameters such as split ratio and feed flow rate. Using a hydraulic circuit instead of one directional flow experimentation significantly reduced experimental waste.

The first hydraulic circuit (Fig.5-2 a) was used for the single-stage MHC experiments to study the separation performance of a single type of MP particles in an MHC. The specific experimental scheme is shown in Table 3. The optimization of operating parameters of MHC_H1 & MHC_L and the comparison between MHC_H1 & MHC_H2 were be obtained through single-stage MHC experiments. For high-density particles (the PA particles, in this study), most of them in the reservoir after going through MHC_H1 or MHC_H2 flowed through the underflow valve (Valve 9) while the purified water flowed out from underflow outlet. Inversely, when using

MHC_L most of the particles (the LDPE particles, in this study) were removed through overflow outlet as MHC_L targets removal of low-density particles, and the purified water was discharged from underflow outlet. After the hydraulic circuit became stable as indicated by constant readings on the pressure gage and flow meter, samples (taken in triplicates) were taken from the sampling valves.

The second hydraulic circuit (Fig. 5-2b) was for the two-stage MHC series which includes MHC_H2 in series with MHC_H1. The goal here was to further enhance the separation efficiency of a type of high-density MP in the mixed liquid) which was conducted with three different feed pressures Pi (0.043, 0.134 and 0.193 MPa). The split ratios for both MHCs were 35%, and the feed particle concentration was 25 mg l^{-1}. In this experiment, the PA-water mixture entered the MHC_H2 for primary separation and its overflow passed through MHC_H1 for a secondary separation to further reduce the concentration of particles. Another two-stage MHC experiments (MHC_L in series with MHC_H1) was carried out under two different feed concentrations Ci (25 mg L^{-1} and 50 mg L^{-1}) to separate the high- and low-density MPs in the mixed liquid simultaneously. The feed pressure here was 0.245 MPa and the split ratios for both MHC were 35%. In this experiment Nylon-LDPE-water mixture first entered MHC_L for LDPE separation, and MHC_L's underflow stream was fed to MHC_H1 for PA separation. Both hydraulic circuits were maintained at ambient temperature.

Throughout the experiments, operational parameters include the feed flow rate Q_i, split ratio R_f and feed pressure P_i. The split ratio for MHC_H1 and MHC_H2 means the ratio of flow rate of underflow outlet (Q_u) to feed flow rate (Q_i), while the split ratio for MHC_L is the ratio of flow rate from overflow outlet (Q_o) to Q_i.



NO.	Devices and function
1	Water reservoir with a mixer
2	Pump
3	Flowrate-control valve
4	Bypass valve
5	Flow meters
6	Sampling valve
7	Pressure gauge
8	**MHC**
9	Underflow-control valve
10	Overflow-control valve

Figure 5 - 2 Layouts of the experimental processes: a) Single-stage MHC experiment; b) Two-stages MHC experiments.

Table 5 - 4 Single-stage MHC experimental parameters

MHC types	C_i *1	Q_i *2	R_{fH1} or R_{fL}
MHC_H1	5, 10, 25, 50, 100	0.068, 0.091, 0.114, 0.136, 0.159	R_{fH1}:15%, 25%, 35%, 40%, 45%
MHC_H2	/	0.227, 0.302, 0.363, 0.420, 0.454	/
MHC_L	/	0.068, 0.091, 0.114, 0.136, 0.159	R_{fL}: 15%, 25%, 35%, 45%
Remarks	Q_i=0.159, R_{fH1} = 30%	C_i = 25; R_f = 35%	C_i = 25; Q_{iH1} = 0.159, Q_{iL} = 0.136

*1the units of C_i are mg L^{-1}; *2the units of Q_i are m^3h^{-1}.

5.2.4 Calculation of separation efficiency

The total mass separation efficiency (E_t, hereafter referred to as total efficiency) and grade separation efficiency (E_g, referred to as grade efficiency) were all used to describe the separation efficiency of MPs. E_t refers to the ratio of dispersion phase mass from underflow outlet (for MHC_H1 and MHC_H2) or overflow outlet (MHC_L) to feed. The E_t for MHC_H1, MHC_H2 and MHC_L are shown in Eq. 5.1 and 5.2, respectively, and E_t for MHC_H2 in series with MHC_H1 and MHC_L in series with MHC_H1 are presented in Eq. 5.3 and 5.4, respectively.

$$E_t = \frac{M_u}{M_i} = \frac{Q_u C_u}{Q_i C_i} = \frac{Q_i C_i - Q_o C_o}{Q_i C_i} = 1 - \frac{Q_o C_o}{Q_i C_i} = 1 - (1 - R_{fH}) \frac{C_o}{C_i} \tag{5.1}$$

$$E_t = \frac{M_o}{M_i} = 1 - (1 - R_{fL}) \frac{C_u}{C_i} \tag{5.2}$$

$$E_t = \frac{M_{uH2} + M_{uH1}}{M_i} = 1 - \{1 - [R_{fH2} + (1 - R_{fH2}) \cdot R_{fH1}]\} \cdot \frac{C_{oH1}}{C_i} \tag{5.3}$$

$$E_t = \frac{M_{oL} + M_{uH1}}{M_i} = 1 - \{1 - [R_{fL} + (1 - R_{fL}) \cdot R_{fH1}]\} \cdot \frac{C_{oH1}}{C_i} \tag{5.4}$$

Where M_o, M_u and M_i represent the particle mass from overflow outlet, underflow outlet, and feed respectively; C_o, C_u, and C_i refer to the particle concentration from overflow outlet, underflow outlet, and feed respectively; R_f means the split ratio. The subscripts H1, H2, L indicate the parameter under the corresponding MHC type, such as C_{oH1} means the particle concentration from MHC_H1's overflow outlet, M_{uH2} is the particle mass from MHC_H2's underflow outlet. R_{fH} is a general term for the split ratio of high-density MHCs.

The total efficiency at which particles with size d_{pi} in the feed are separated is called the grade efficiency $E_g(d_{pi})$. E_g shows the relationship between the particle size and the corresponding grade efficiency with a curve (noted as grade efficiency curve). In this curve, the cut size (noted

as d_{50}) means the particle size when the E_g equal 50%, which is one of the important metrics of MHC separation performance.

5.2.5 Sample processing methods

The total efficiency was obtained by measuring the concentration of MP particles in each type of sample. The feed, underflow, and overflow samples were collected in glass graduated cylinders with their volumes recorded. The samples were vacuum filtered using glass fiber filter papers (1.5 μm, Hach) and the filter papers were dried in an aluminium weighing pan at 75 ^{o}C for 4 hours. The concentration of MPs was then calculated by using the volume of sample filtered and the mass of MPs. Triplicates were taken for all samples and the standard deviations were presented as error bars.

To obtain the grade efficiency, the feed, overflow, and underflow samples needed to be processed immediately after collection to minimize sedimentation, flotation, and aggregation. The collected samples were sonicated for 20 minutes in an Ultrasonic bath (Fisher Scientific) to form homogenous suspension before going to the particle analyzer. Once sonicated, the sample was then pumped through a laser particle analyzer for the size distribution of the MPs in each sample. The acquired size distributions were used to calculate the grade efficiency as described in §2.4. To ensure that the next samples read properly, the particle analyzer was backwashed with DI water before the next suspension was read.

5.2.6 Contamination Control

To avoid MP particle contamination, the use of plasticware was minimized and replaced with metal or glassware. After each trial with MHCs, the hydraulic circuit and reservoir were cleaned with soap and rinsed with Milli-Q water. The MHCs were thoroughly cleaned, air dried

after each trial, and stored with their inlets and outlets sealed with aluminum foil to prevent rusting. To identify potential contamination from laboratory space (Milli-Q water left open to the air on the bench for 8 hours) and experiment setup (the hydraulic circuit), blanks were taken for each set of experiment and summarized in Appendix D. Although several pieces of MPs were identified in each blank sample, their masses were unmeasurable. Notably, the majority of the MP contamination from the lab environment were fibers shed from Nomex lab coats, easily identifiable by their color and composition.

5.3 Results and discussion

5.3.1 Optimal operational conditions for microplastics' separation

The grade efficiency E_g under different feed particle concentrations (C_i) was evaluated by taking MHC_H1 as example. For the particles larger than 20 μm, the grade efficiency was consistently over 80% for all tested C_i. Microplastic particles could form aggregates during the spiral movement and the MHC_H1 demonstrated high efficiency in separation of those aggregates (50-100 μm).

Among the feed particle concentration values (C_i) at which the separation efficiency was evaluated**,** C_i = 25 mg L^{-1} showed the highest efficiency in the particle size range of 20-30 μm, while in the range of 30-100 mm, C_i = 50 mg L^{-1} was consistently the highest (>95%). The grade efficiency for smaller particles 2-30 μm was significantly lower at C_i of 5 mg L^{-1} and 10 mg L^{-1}. At lower *C*i, the chance of collision among particles decreased during the spiral motion given the difficulty of formation of larger-sized aggregates. Additionally, the magnitude of the fluid drag

force acting on particles enlarged as particle size decreased and the direction of drag force become more random, causing uncertain motions of particles (Zhang et al., 2017).

Feed flow rate (Q_i) determines the fluid tangential velocity in the MHC which directly affects separation efficiency (Wang et al., 2016). The separation efficiencies (E_t and E_g) under different Q_i can be seen in Fig. 5-3. Both grade and total separation efficiency grew higher as the feed flow rate varied from 0.068 to 0.159 $m^3 \cdot h^{-1}$ because a higher feed flow rate led to higher tangential velocity of the mixed phase, resulting in an increasing of centrifugal force on particles. However, the higher centrifugal force could also cause a reduction of the aggregation effect between particles and potentially breakup aggregates due to the stronger shearing effect. Therefore, the increase in total efficiency became smaller as flow rate was further increased for MHC_H1 and the separation efficiency decreased for MHC_L (Fig.5-3). Meanwhile, the separation efficiency of the finest particles (2-10 μm) was consistently lower (37%-61%) in comparison to larger ones for both MHC configurations, which was in accordance with previous studies. Because of their smaller masses, fine particles were subjected to smaller centrifugal force compared to larger particles or aggregates and associated with random movements under the enlarged fluid drag force (Zhang et al., 2017). The separation efficiency could be potentially improved by chemical enhancement in future studies.

A one-way ANOVA test was performed to check whether the total efficiencies of different flow rates as well as for each type of MPs were statistically different. Although the standard deviation at some flow rates overlapped, the total efficiencies were found statistically different and the total efficiency of MHC_L (for LDPE) was significantly lower than MHC_H1 (for Nylon) (Fig. 5-3 b and d), because the density differential between water and LDPE was smaller. By comparison,

the optimum feed flow rates were of MHC_H1 and MHC_L are 0.159 m^3h^{-1} (0.245 MPa) and 0.136 m^3h^{-1} (0.172 MPa), respectively.

The split ratio (R_f) was an important operational parameter that directly affected the separation efficiency of particles. Figure 5-4 demonstrates that the grade efficiency (E_g) increased as the split ratio (R_f) was varied from 15% to 35% and the obvious fishhook effect took place at the split ratio of 15% for both MHCs. The low split ratio decreased the velocity of mixed fluid around the underflow outlet, resulting in a lower turbulence intensity here under which the fine particles (2 - 20 μm) were not easily washed out from the wake flow of coarse particles. Therefore, more fine particles were separated with the coarse particles at a smaller split ratio, thus improving the separation efficiency of these fine particles.

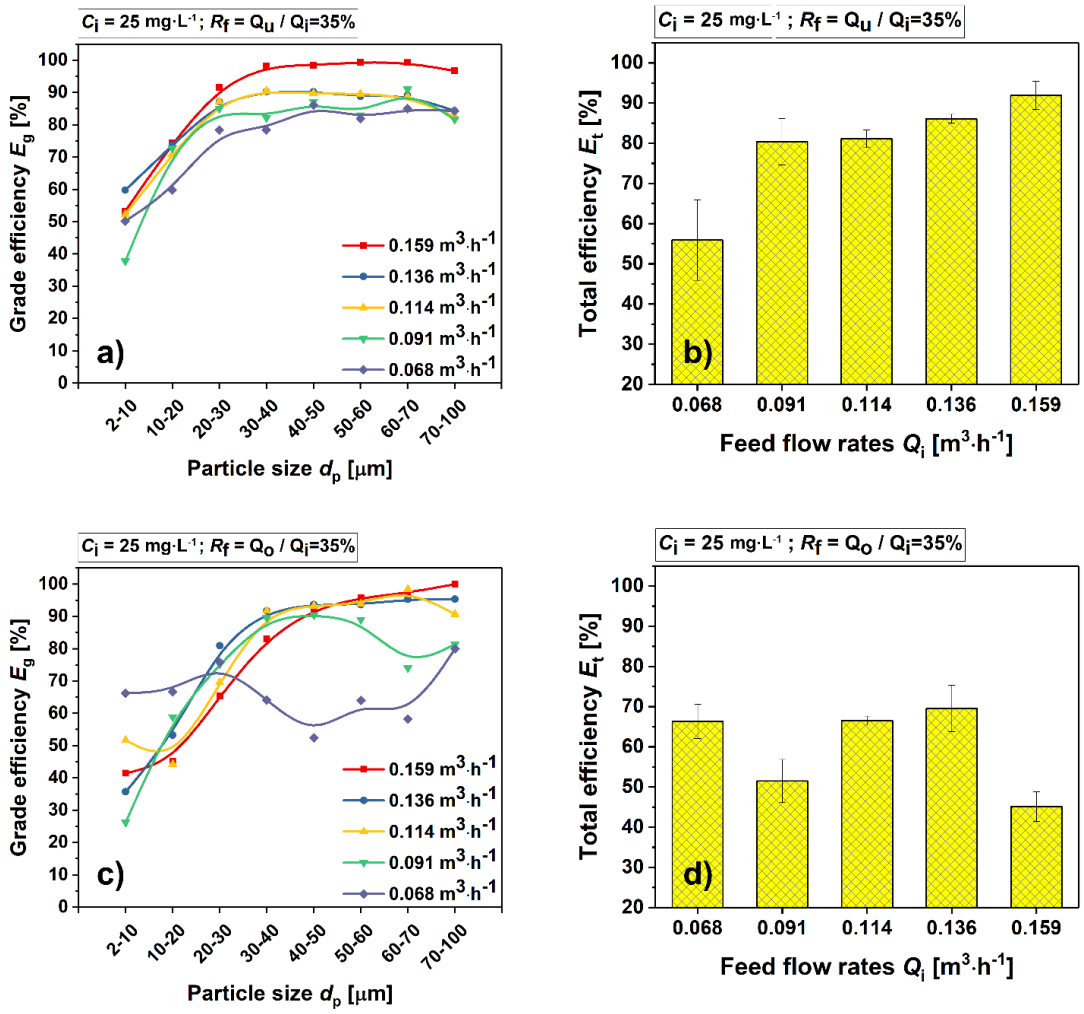


Figure 5 - 3 Grade and total efficiencies under different feed flow rates. a) Grade efficiency of MHC_H1; b) Total efficiency of MHC_H1; c) Grade efficiency of MHC_L; d) Total efficiency of MHC_L.

As the E_g (d_{pi}) shows comparisons of particle at different size range, it can be found R_f at 35% had the most numbers of highest-efficiency points for both MHCs (Fig. 5-4), which was also expressed in the E_t comparisons in Fig. D4 in Appendix D. Consequently, the optimal separation efficiencies were obtained at the split ratio of 35%. Exampled by MHC_H1, when the split ratio was set to 15%, it means that only 15% of the mixed fluid was discharged from underflow outlet. As a result, a part of particles that were originally moving near the underflow were not able to be

discharged from underflow outlet, and instead escaped from the overflow outlet. This adverse effect was improved as split ratio R_f increases, resulting in a better separation efficiency. However, the efficiency decreased when the split ratio was larger than 35% because the larger amount of water also flowed through underflow outlet although the number of particles in the underflow increased, causing a reduction of particle concentration at underflow outlet.

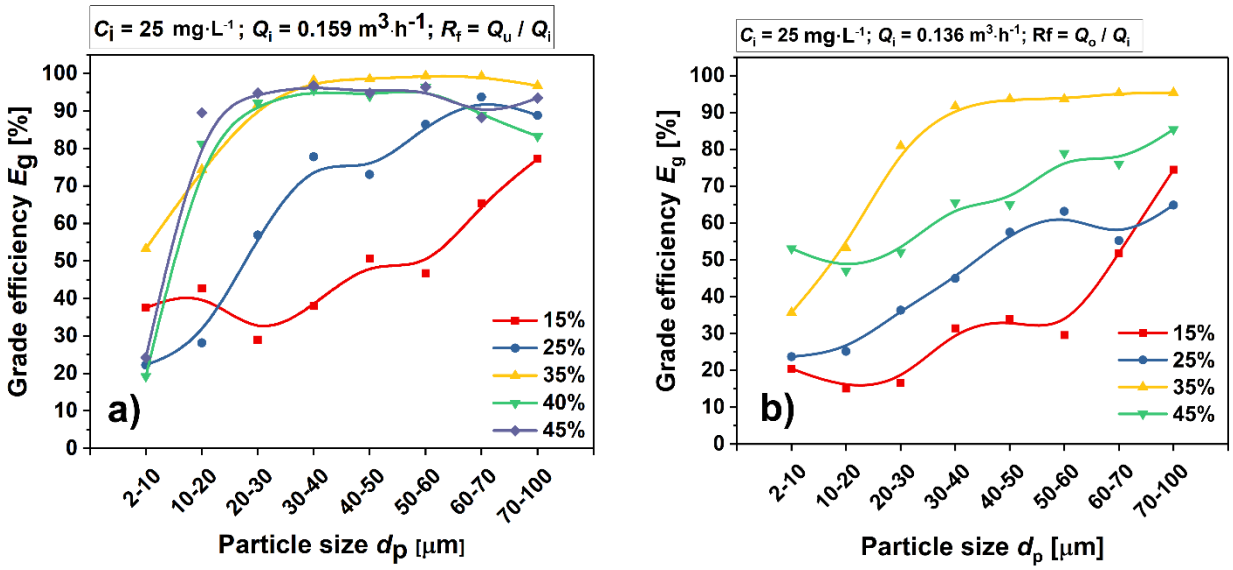


Figure 5 - 4 Grade efficiency curves under different split ratios (in percent). a) For MHC_H1; b) For MHC_L.

5.3.2 MHCs arranged in series

To further improve the separation efficiency of the same kind of MPs and to study the separation efficiency when two kinds of MPs (with density higher and lower than water) were present in the mixture, the separation experiments of MHC_H2 in series with MHC_H1 and MHC_L in series with MHC_H1 were carried out (Fig. 5-2 b). The total split ratios (R_{H2_1} and R_{L_H1}) of the two series connections were calculated in Eq. 5.5 and 5.6, respectively. R_{fH1}, R_{fH2} and

R_L in Eq. 5.5 and 5.6 were set to 35% which were determined or selected in these single MHCs' experiment previously. Herein, the reduced total separation efficiency (E_{rt}) and grade separation efficiency (E_{rg}) were used for comparisons to eliminate the effect of split ratio on separation efficiency. The calculations of E_{rt} are shown in Eq. 5.7, and the calculation of E_{rg} is similar to E_g in "5.2.4 Calculation of separation efficiency".

$$R_{H2\_1} = R_{fH2} + (1 - R_{fH2}) \cdot R_{fH1} = 0.35 + 0.65 \cdot 0.35 = 0.5775 \tag{5.5}$$

$$R_{L\_H1} = R_{fL} + (1 - R_{fL}) \cdot R_{fH1} = 0.35 + 0.65 \cdot 0.35 = 0.5775 \tag{5.6}$$

$$E_{rt} = \begin{cases} (E_t - R_{H2\_1})/(1 - R_{H2\_1}) & \text{(MHC\_H2 in series with MHC\_HI)} \\ (E_t - R_{L\_H1})/(1 - R_{L\_H1}) & \text{(MHC\_L in series with MHC\_HI)} \end{cases} \tag{5.7}$$

Where R_{H2_1} means the total split ratio of the two-stage MHCs separation system when MHC_H2 connected in series with MHC_H1; R_{L_H1} refers to the total split ratio when MHC_L connected in series with MHC_H1; Eq. 2.4 offered the mean of R_{fH1}, R_{fH2} and R_{fL}.

MHC_H2 and MHC_H1 were arranged in series for the test of the enhancement of a secondary MHC for the particles with the same density. The proportion and concentration distribution of particles with different sizes from the feed and outlet flow were the most direct expression of the particle's whereabouts and Fig. 5-5 provides the relevant information when feed pressure was 0.134 MPa. Figs. 5-5 a-f present the proportion (Figs. 5-5 a, c, and e) and concentration distributions (Figs. 5-5 b, d, and f) of particles of different sizes, and the feed and overflow outlet of MHC_H1, MHC_H2 and two MHCs in series. From Figs. 5-5 a), c) and e), it can be found that most of the particles from feed liquid have a particle size of 2-30 μm (the number exceeds 96%). Among them, particle size at 2-10 μm, 10-20 μm and 20-30 μm accounted for 21.1%, 63.8%, and 12.0% (average values of Figs. 5-5 a, c, e), respectively. The proportion of

particles between 10-100 μm from feed was higher than that of overflow in all three cases, while particles between 2-10 μm were the opposite. In other words, compared with the particles from the feed, the reduction of particles in the range of 10-100 μm in the overflow outlet was higher than that in the range of 2-10 μm (the sum of the proportions of particles in all ranges is equal to 100%), which confirmed that the separation efficiency of 2-10 μm particles was lower.

Figs. 5-5 b, d, and f present the particle concentration in the overflow of MHC_H1 and MHC_H2, and the total overflow of MHC_H1 and MHC_H2 in series system. For 2-10 μm particles, the ratio of the overflow particle concentration to feed particle concentration of MHC_H1 (73.4%) was significantly lower than MHC_H2 (91.5%) and two MHCs in series (89.9%) according to Eq. 5.1, which also means MHC_H1 had higher separation efficiency for 2-10 μm particles than the other two cases (single MHC_H2 and MHC_H2 in series with MHC_H1). A MHC_H1 higher than two MHCs in series can be explained as following: the fluid in the second-stage MHC (MHC_H1) in the system in-series was unable to reach sufficient tangential velocity since only 65% of the fluid flowed into the second-stage MHC (MHC_H1) from the overflow outlet of the first-stage MHC (MHC_H2). Therefore, the MP particles (2-10μm) in MHC_H1 (flow rate from the overflow of MHC_H2 is 0.103 $m^3 \cdot h^{-1}$) in the series were unable to get enough centrifugal force to move to the wall area, resulting in lower E_{rg} than in the single staged MHC_H1 (feed flow rate was 0.114 $m^3 \cdot h^{-1}$ when the feed pressure was 0.134 MPa).

Compared with a single MHC_H2 and MHC_H1, arranging MHC_H1 and MHC_H2 in series respectively improved E_{rt} from 67.72% and 70.9% to 76.01% when feed pressure P_i was equal to 0.134 MPa, and improved from 65.96% and 78.6% to 85.93% for $P_i = 0.193$ MPa. These results actively demonstrate that the second-stage MHC (MHC_H1) of two MHCs in series was

capable of separating the particles which escaped the first-stage MHC (MHC_H2), especially for the fine particles that were difficult to separate in MHC_H2. The change trend of solid recovery (the fraction of dry solids in the feed that reports to the concentrated underflow) in Fig. 5-6 b) is similar to E_{rt}. However, the trend of concentration ratio is different: two MHC in series exhibited lower concentration ration than a single MHC, because the total split ratio of two MHC in series was larger than a single MHC.

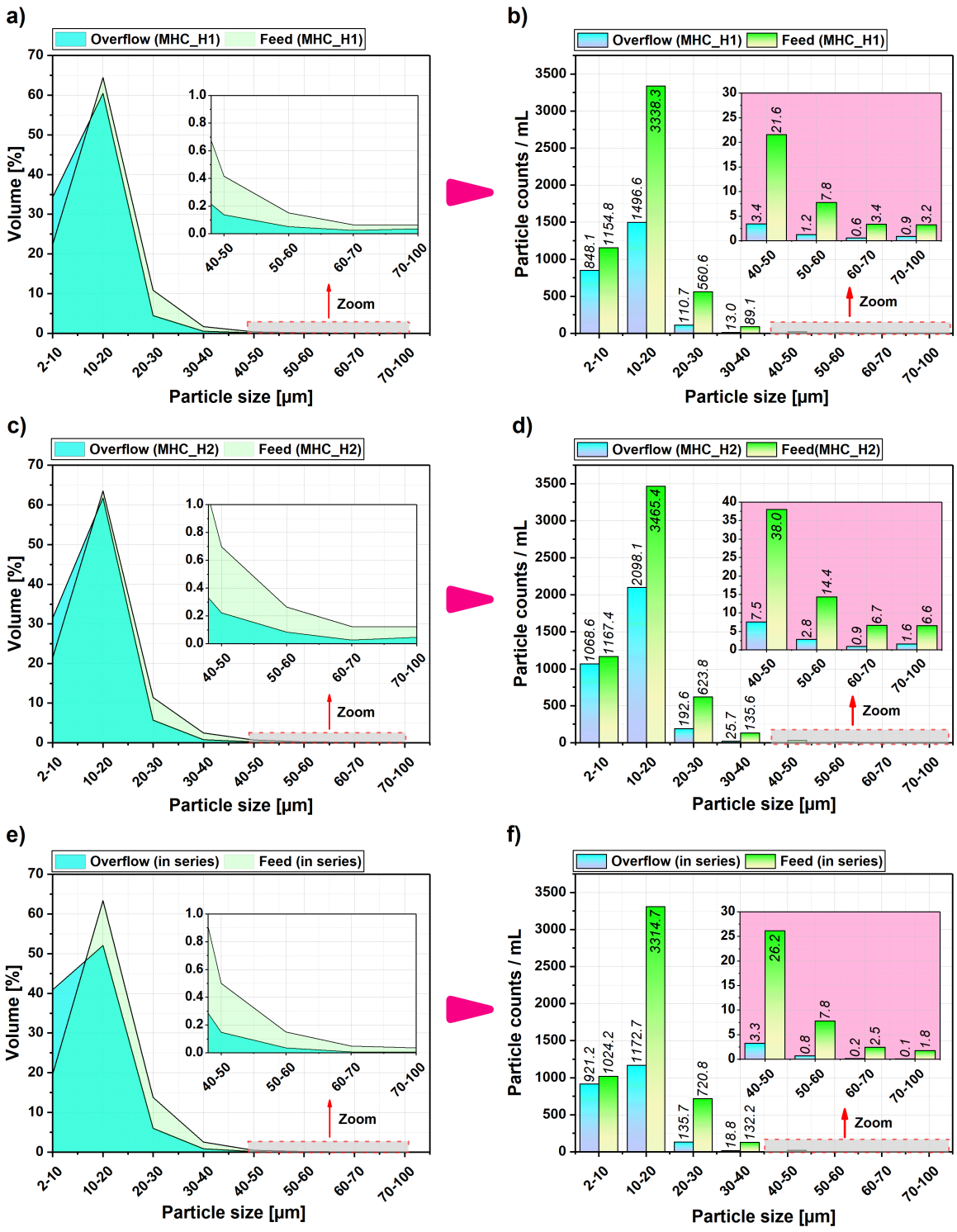


Figure 5 - 5 The proportion and concentration distribution of particles with different sizes from the overflow outlet and feed. a), c) and e) represent the proportion; b), d) and f) refer to the concentration distribution.

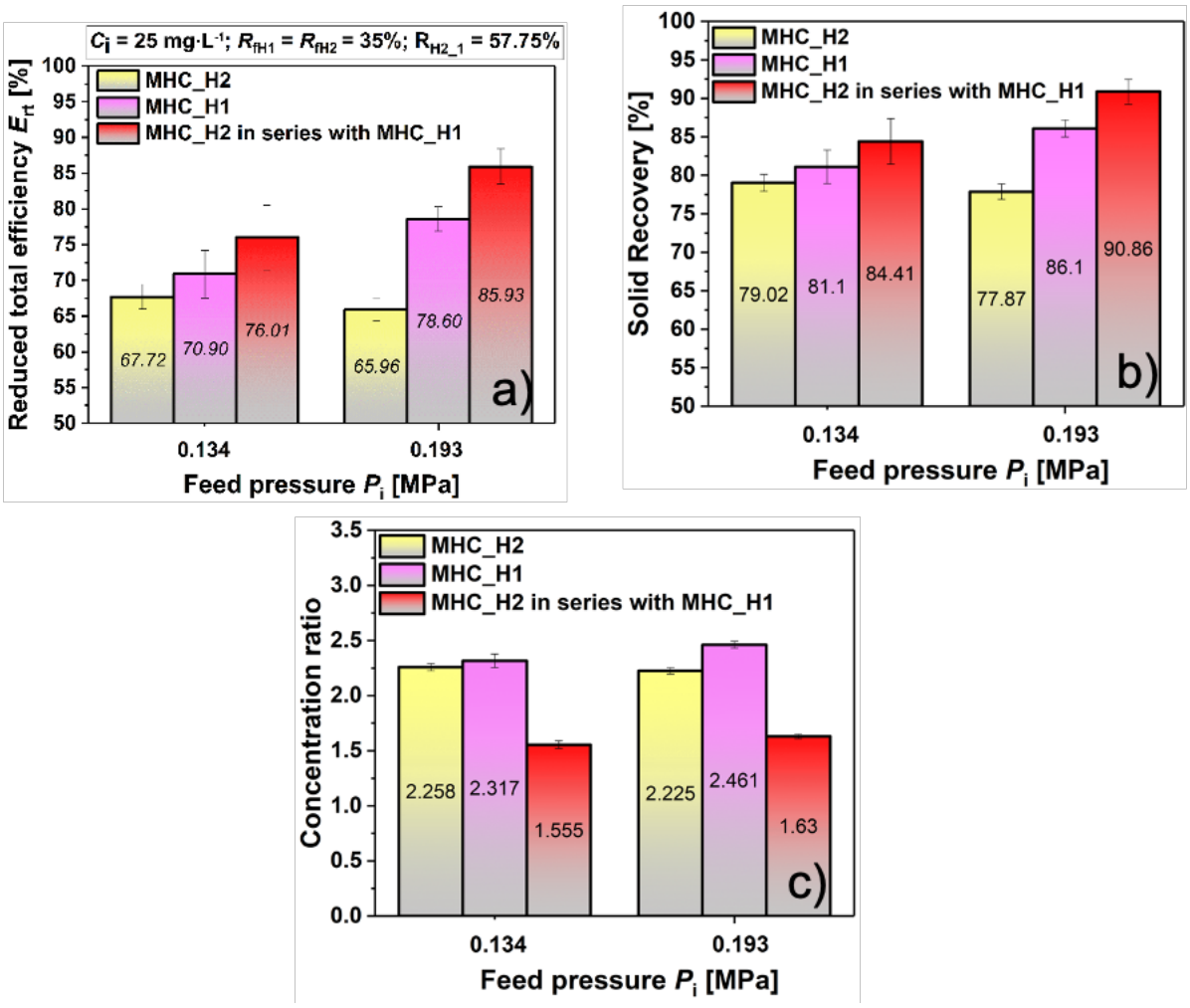


Figure 5 - 6 Separation performances of MHC_H1 and MHC_H2 and the two connected in series. a) The reduced total efficiency; b) Solid recovery; c) Concentration ratio

In real world applications, wastewater contains MPs of density that is both higher and lower than water density. Given this variability, the research was carried out by connecting MHC_L and MHC_H1 in series. Figure 5-7 exhibits the reduced grade efficiency E_{rg} when the feed liquid included both LDPE and Nylon particles, and the reduced grade efficiency curves of single MHC_L and MHC_H1 were also given for comparison. E_{rg} of a single MHC_L or MHC_H1 was expectedly higher than in the system in series for both 25 and 50 ppm. As most of the LDPE in the first-stage MHC (MHC_L) were spiraling upward (upward movement to separate from overflow

outlet), while most of the Nylon within MHC_L were spiraling downward since Nylon's density was higher than water's density. This combination of movement caused an increase in the chance of collision between LDPE and Nylon particles, which magnified the velocity loss of particles during the movement. LDPE particles, which could be separated from the overflow outlet smoothly, likely changed their direction of motion after colliding with Nylon particles, making LDPE to be discharged from the underflow outlet along with B. The same situation also took place in the second-stage MHC (MHC_H1). Moreover, the E_{rg} curve changing trend of in series system for both 25 and 50 ppm was similar to that of a single MHCs for particles in the range 2-40 μm, while the expected fluctuations in efficiency appeared for particles in the 40-100 μm range.

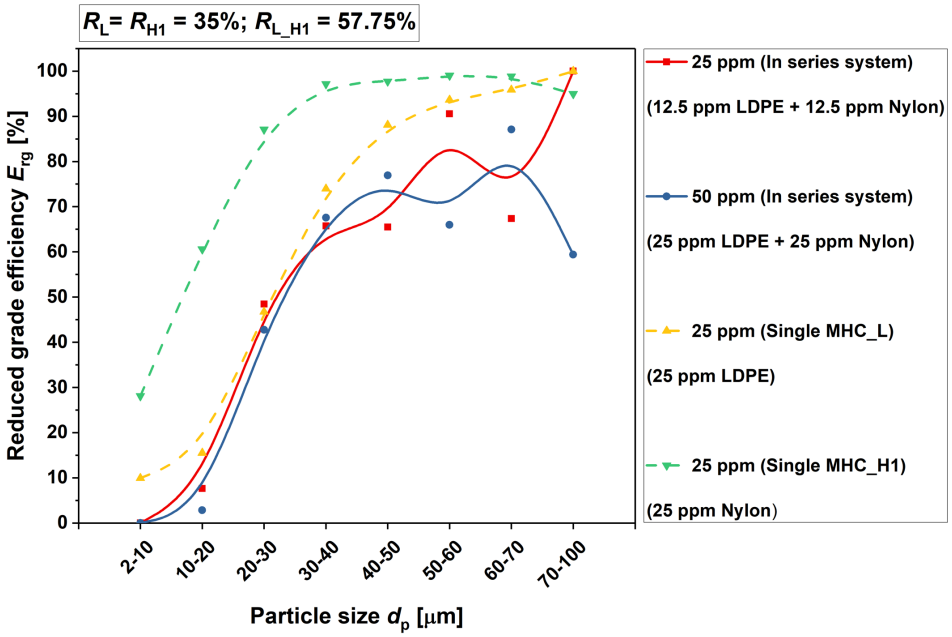


Figure 5 - 7 The reduced grade efficiency of MHC_L and MHC_H1 and the two connected in series

5.3.3 MPs separation in environmental matrices

The UV-exposed MP particles were used for separation tests in ultra-purified water as well as synthetic stormwater, at the optimal operational conditions determined previously. The Raman Spectra (Fig. D2) showed that there were no substantial changes in the virgin MPs from the 15-hour UV exposure, and no fragmentation or breakage of MPs were observed by visual observation with microscopy. The separation efficiency of UV-exposed LDPE and PA particles in ultra-purified water was not statistically different from the one of previous experiments. On the other hand, the separation efficiency of PA in synthetic stormwater and wastewater effluent reached 98.1% and 93.6%, which was 7.7% and 3.2% higher than the one in ultra-purified water respectively. The separation efficiency of LDPE improved by 3.8% in synthetic stormwater compared to ultra-purified water. However, only the difference in PA particles separation efficiency was statistically significant ($p = 0.038$). The high ionic strength in synthetic stormwater compressed the double layers and reduced the zeta potential of MPs (Li et al., 2019). Thus, the collision and aggregation of MPs were more likely to occur (Wu et al., 2007; Zhang et al., 2021) in synthetic stormwater, and MHCs were more efficient in removing larger particles/aggregates, as seen in previous sections. The separation efficiency in MHCs was primarily governed by density differentials. The improved separation efficiency of PA particles in stormwater and wastewater effluent showed the MHCs' suitability for separation of other types of abundant MPs in stormwater with similar and higher densities, such as tire wear particles (Baensch-Baltruschat et al., 2020; Kayhanian et al., 2012). However, this experiment did not account for the impact on MHCs' separation performance of other solids that could be present in real stormwater or wastewater effluent, such as sediments, microorganisms or EPS. Future studies should assess MHCs' separation of MPs under the interference of other solids.

In practical applications, multiple MHCs need to be installed in parallel to ensure that each MHC can obtain a suitable feed flow rate and that each single MHC maintained high separation efficiency (Chen et al., 2013; Huang et al., 2013; Lv et al., 2018). The number of MHCs can be determined from site-specific flow rate. In addition, when the mixed liquid contains both fine and coarse MP particles, a parallel system of conventional hydrocyclone can be used first (due to the larger feed flow rate of conventional hydrocyclones, fewer conventional hydrocyclones are required) to remove coarse MP particles, and then, the MHC parallel system could be used to process the remaining small-sized MP particles that are difficult to remove by conventional hydrocyclone, which can help minimize the head loss caused by MHCs.

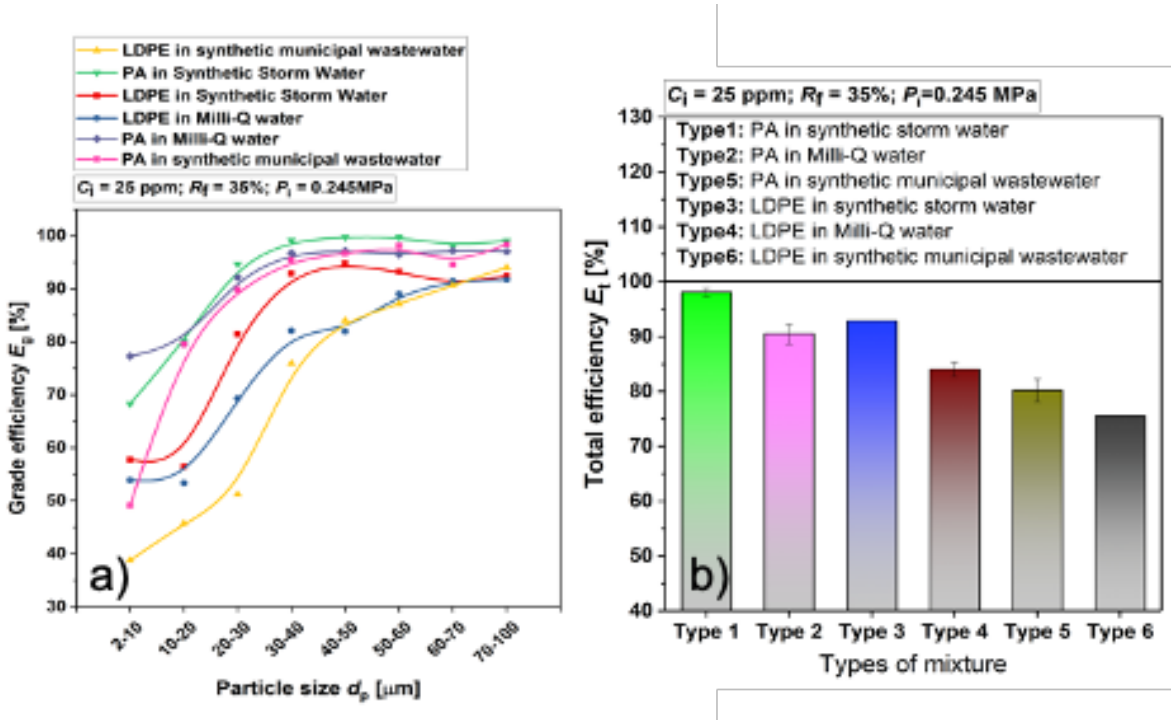


Figure 5 - 8 a) Total mass separation efficiency of UV exposed MPs in MilliQ water, synthetic stormwater and wastewater; b) Grade separation efficiency of UV exposed MPs in MilliQ water, synthetic stormwater and wastewater

5.4 Conclusions and Limitations

The effective removal from wastewater of MPs with various densities is an urgent matter for water reclamation. However, the strategy and methods are scarce. This study presented MHCs as a novel method to separate MPs from water matrices. Mini-hydrocyclones were manufactured by 3D printing stainless steel, and MP separation performance was evaluated for LDPE and PA at various operating conditions and setup. The findings from the work can be summarized, as follows:

- MHCs can effectively separate MPs denser and less dense than water under their optimal operating conditions, but the separation of MPs denser than water was better than the MPs less dense than water. The separation of less dense MPs could be enhanced by improving on the MHCs' geometric parameters.
- MHCs in series increased the separation efficiency of the same kind of microplastics and also verified the relatively high separation efficiency when multiple types of particles (of varying densities) exist in the mixed liquid simultaneously. However, this separation efficiency of MHCs in series was lower than the efficiency of a single MHC.
- The improved separation efficiency of MPs in synthetic stormwater indicated that the presence of dissolved solids in environmental water bodies can promote the removal of MPs using MHCs.

Although MHCs were shown to be applicable to the treatment of MPs, especially denser MPs, future experiments need to be conducted with different MP morphologies, like scales, fibers, rods, and in more diverse types of environmental matrices to validate the broader application of MHCs for MPs removal. Moreover, the volumetric recovery was governed by the split ratio, which can also be further optimized through design parameters.

Chapter 6 Summary

The growing concerns of microplastics pollution in water resources and rising needs of water reclamation require better understanding of microplastics' temporal variation, and urge development of microplastics removal technologies. This thesis provided critical information that can be used to enhance the understanding WRRFs' role as barrier and pathway of microplastics to reclaimed water and aquatic environment, and presented mini-hydrocylcones as a potential method for microplastics' separation from wastewater.

The major findings are summarized as follows:

1. The occurrence and abundance of microplastics in wastewater differed geographically and was dependent on the type of wastewater as well as characteristics of WRRFs;
2. A number of chemical digestion methods had been applied to remove natural and biogenic material from wastewater samples, and they can lead to damages on target microplastics to different extent. Using a reduced level of Fenton' reagent while keeping the pH in an optimal range with a buffer led to minimum amount of damages to target microplastics as indicated by mass loss and surface morphology.
3. Microplastics were more abundantly found in wastewater during winter season. However, seasonal variation of microplastics in wastewater could be facility specific.
4. Primary treatment removed the majority of microplastics in influent and the removal during secondary and tertiary treatment was insignificant.
5. The most commonly detected polymers in primary influent, primary effluent, secondary effluent and tertiary effluent were polyester and polypropylene, the majority of which were in the form of fibers. This means laundry discharge is the main contributor of microplastics to municipal wastewater. Fine microplastics (<25 μm) were the most prevalent throughout

the WRRF, and relatively more abundant in secondary and tertiary effluent as larger microplastics were removed during earlier stages.

6. A substantial amount of PES fibers was generated from the filtration process, illustrating that tertiary cloth filters could contribute to microplastics' occurrence in tertiary effluent.
7. Mini-hydrocyclones were designed and 3D printed with stainless steel. The optimal separation efficiency of microplastics in pure water was lower than the ones in synthetic stormwater and wastewater, which confirmed its suitability real-world application.

In conclusion, this thesis provided a comprehensive framework and understanding of the occurrence and target removal of microplastics in wastewater treatment process, which set a baseline for future research on the further evaluation of microplastics' mass balance in WRRFs and development and implementation of removal technologies.

Chapter 7 Future Perspective

Based on the outcome and limitations of this and previous study, future research on microplastics in wastewater should be centered around investigation of microplastics in sludge and biosolids as well as their co-removal with other contaminants, and improving technical readiness of removal technologies.

As presented in Ch. 2 and 4, the majority of microplastics in wastewater influent was retained during primary treatment, implying that a large proportion of microplastics were retained in sludge. A few studies *(inter alia,* Petroody et al., 2021; Pittura et al., 2021; Takdastan et al., 2021) investigated the abundance of microplastics in sludge, yet no studies to this date have conducted comprehensive sampling of each solid treatment process unit to close the mass balance. Therefore, the future focus is recommended to be shifted to microplastics in sludge and biosolids for implications in land application.

In addition, the increasingly strict requirements on wastewater effluent quality for reclamation purposes also entail the need for target removal of other CECs, such as pharmaceuticals and Per- and Polyfluoroalkyl substances (PFAs). Given microplastics' contaminant vector potential, future research should systematically investigate the interaction between microplastics and other CECs, and sequentially their co-removal from wastewater and sludge.

As microplastic pollution is rapidly gaining attention worldwide, researchers around globe are developing physical, chemical, biological technologies or strategies to mitigate microplastics from wastewater, and most proposed technologies were tested with pure water and at bench-scales as was seen in Ch.2. New technologies need to be tested at pilot and large scale, as well as in more

complex matrices for practicality, feasibility and infrastructural integrability validation as well as balance energy use with efficiency of removing microplastics from wastewater.

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Appendix A Supplementary Materials for Literature Review

Table A1. Mesh sizes used for MPs extraction in literature

Reference	Mesh Size (um)
(Bayo et al., 2020b)	100
(Blair et al., 2019b)	2800
(Cao et al., 2020a)	100 500 1000
(Conley et al., 2019)	43
(Edo et al., 2020)	25 104 375
(Gies et al., 2018)	63
(Gündoğdu et al., 2018)	55
(Jiang et al., 2020)	38
(Lares et al., 2018a)	250 5000
(Lee and Kim, 2018a)	106 300
(Leslie et al., 2017)	300
(Liu et al., 2019)	47
(Long et al., 2019)	43 63 125 355
(Lv et al., 2019)	25 62.5 125 250 500
(Magni et al., 2019)	63 2000 5000
(Magnusson and Norén, n.d.)	300
(Mason et al., 2016)	125 355
(Michielssen et al., 2016)	20 106 300 850

(Murphy et al., 2016b)	65
(Simon et al., 2018a)	80 500 1000 2000
(Simon et al., 2019b)	10
(Takdastan et al., 2021)	25 125 420 840
(Talvitie et al., 2015b)	20 100 200
(Talvitie et al., 2017a)	20 100 300
(Tang et al., 2020b)	149
(Uddin et al., 2020)	250 500 1000 2000
(F. Wang et al., 2020)	13
(Xu et al., 2019c)	25 74 150
(L. Zhang et al., 2021b)	25 100 200 502
(Ziajahromi et al., 2017b)	60 125 250 500

Table A2. Removal efficiency of microplastics at each treatment step

Relative 1st %	1st Removal %	Secondary Treatment process	2nd Removal %	Relative 2nd %	Tertiary Treatment Process	3rd Removal %	Relative 3rd %	Effluent Conc. (MP/L)	Overall Removal %	Reference
99	99.7	MBR	0.70	33				0.4	99	(Lares et al., 2018a)
		AS	-0.30	-66.7				1.0	98	
78.3	79.6	AS	20.1	92.6				0.25	98.41	(Murphy et al., 2016b)
91.6	93.1	TF	6.75	80.8				0.5	98.4	(Gies et al., 2018)
19	19.3	AS	71.3	88				0.31	90.3	(Bayo et al., 2020b)
		MBR	99.9							(Talvitie et al., 2017a)
60.7	61.7	AS	31.1	79.2	Nitrification tank and filter	4.90	60	0.2	96.7	(Blair et al., 2019b)
26.1	26.5	OD	73.9	> 99	UV	0.50	> 0	0.13	51.9	(Lv et al., 2019)
		MBR	57.2	96.9						
40.7	41.4	AS	16.6	28.5	Chlorination	7.7	16.7	28.4	64.5	(Liu et al., 2019)
		TF	99.9							(Michielssen et al., 2016)
60.8	61.8	AS	23.7	60.5	Chlorination	4.7	30.6	5.9	89.2	
82.5	83.8	AS	1.0	5.8	Chlorination	12.1	73.5	2.6	95.6	
50.2	51.0	AS	36.5	73.4	Bio Filter	11.6	87.3	10.2	98.3	(Talvitie et al., 2015b)
				68.2	Chlorination	30.1				(Ziajahromi et al., 2017b)
					Disc Filtration	89.7				(Simon et al., 2019b)
16	25.0	AS	61.0	48	UV disinfection	21	14	30		(Jiang et al., 2020)
49	54.5	AS	71.1	40	Chlorination	31	5	0.84	90.87	(Takdastan et al., 2021)

Appendix B Method Validation Supplementary Material

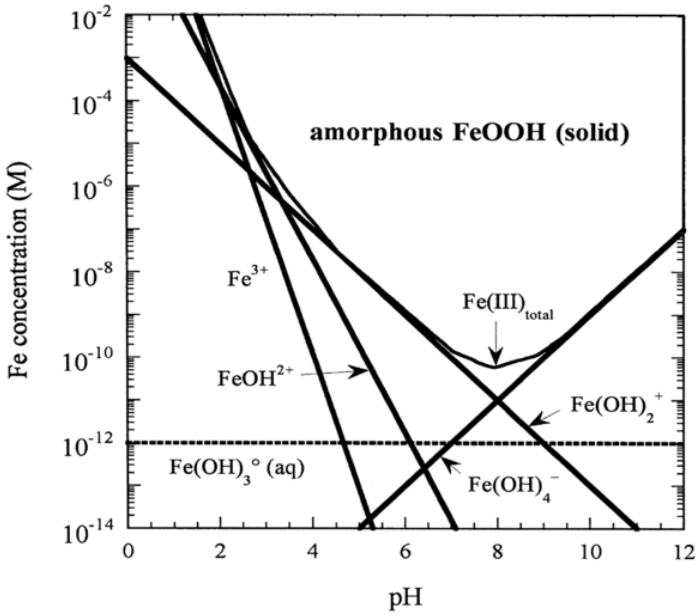

Figure B1. Ferric hydroxide formation in relation to pH and Fe concentration

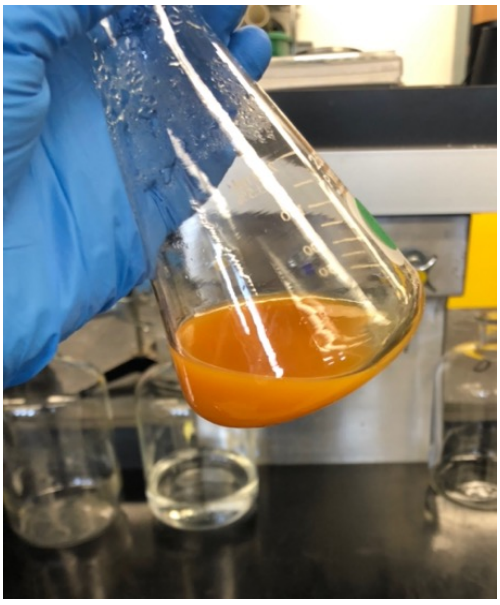

Figure B2. Ferric hydroxide formation during digestion

Appendix C Supplementary Material for MP Monitoring

1. MATLAB code of data processing tool

```
% Anne Sun
% Spectral Search Tool


close all; clear all;clc

%% Query Spectra
file=fopen(query spectra file.txt');

for i = 1:length(file)
placeholder(i,:) = textscan(file(i), '%f %f','headerline',1);
end
UnknownMP=flip(cell2mat(placeholder));
KnownMPnormalize=NaN(300, 4000);
    NormalizeUnknownMP=NaN(300, 4000);
    k=NaN(300,4000);

%% Comparison with library
cd '/Users/annesun/OneDrive/Microplastic Project/Raman Code and standard
MPs/Library'
z = dir('*.txt');

% load all data in the library folder, change to your local folder
% % rows:numbers of data points; coluum: wave and intensity; layer: # of
% % files)
% % lengths of different MP spectrum are different, created 4000 spots to
% % accomodate for the extended scans
% save z.mat
% load('z.mat')
datastorage = zeros(4000,2,length(z));
expression = '\s';

for i = 1:length(z)

    % Load the name of MPs and name of the files
    filename = {z(i).name};
    splitStr(i) = regexp(filename,expression,'split');
    MPname(i) = {splitStr{1,i}{1,1}};

    % Load data in to datastorage matrix
    data = load(filename{1,1});
    datastorage (1:length(data),:,i) = data; % first column wavelength
    % second column intensity

    [plugins,k(i,1:length(plugins)),SLK(i)]= Sorter(data,UnknownMP);
    KnownMPnormalize(i,1:length(plugins))=plugins(1,:);
    NormalizeUnknownMP(i,1:length(plugins))=plugins(2,:);
end

cd '/Users/annesun/OneDrive/Microplastic Project/Raman Code and standard MPs'
```

```
%% sorting in order of highest similairty to lowest
[sorting,order_SLK]=(sort(SLK));
order_SLK=flip(order_SLK);
disp('Percent similarity to the stock MP')
A = flip(sorting);
disp(A(1:4))
disp('the order of the most similair MP type by name')
B= MPname(order_SLK);
disp(B(1:4))

%% shows the top three matches in graphs

figure(1)
%SLK subplots
subplot(4,1,1)
plot(k(order_SLK(1),:),NormalizeUnknownMP(order_SLK(1),:),'red')
title('SLK')
xlabel('Wavelength (nm)')
ylabel('Intensity')
hold on
plot(k(order_SLK(1),:),KnownMPnormalize(order_SLK(1),:),'black')
legend('Unknown
MP',convertCharsToStrings(MPname(order_SLK(1))),'location','northwest')
subplot(4,1,2)
plot(k(order_SLK(2),:),NormalizeUnknownMP(order_SLK(2),:),'red')
xlabel('Wavelength (nm)')
ylabel('Intensity')
hold on
plot(k(order_SLK(2),:),KnownMPnormalize(order_SLK(2),:),'black')
legend('Unknown
MP',convertCharsToStrings(MPname(order_SLK(2))),'location','northwest')
subplot(4,1,3)
plot(k(order_SLK(3),:),NormalizeUnknownMP(order_SLK(3),:),'red')
xlabel('Wavelength (nm)')
ylabel('Intensity')
hold on
plot(k(order_SLK(3),:),KnownMPnormalize(order_SLK(3),:),'black')
legend('Unknown
MP',convertCharsToStrings(MPname(order_SLK(3))),'location','northwest')
subplot(4,1,4)
plot(k(order_SLK(4),:),NormalizeUnknownMP(order_SLK(4),:),'red')
xlabel('Wavelength (nm)')
ylabel('Intensity')
hold on
plot(k(order_SLK(4),:),KnownMPnormalize(order_SLK(4),:),'black')
legend('Unknown
MP',convertCharsToStrings(MPname(order_SLK(4))),'location','northwest')



%% function for interpretting, setting the same size, and Interpolation of
the data

function [vnorm,iq,SLK]=Sorter(data,UnknownMP)%change [] to [Mod_Euc,Base]
for mod Euclidean or change to [vnorm] for Euclidean
```

```
bottom=floor(data(1,1))+1; top=floor(data(end,1)); %finds max and min
wavelength
fclose('all');

BottomForMP=floor(UnknownMP(1,1))+1; TopForMP=floor(UnknownMP(end,1)); %finds
max and min wavelength

if bottom>BottomForMP %creates the order for the wavelength form highest
starting wavelength to lowest ending wavelength
    if top>TopForMP
    iq= (bottom:TopForMP);
    else
        iq=(bottom:top);
    end
else
    if top>TopForMP
    iq=(BottomForMP:TopForMP);
    else
        iq=(BottomForMP:top);
    end
end
values2= interp1(UnknownMP(:,1),UnknownMP(:,2),iq);values=
interp1(data(:,1),data(:,2),iq); %interperulates at integers of the same
wavelength which is i
for z=1:length(values)
    vnorm(1,z)= (values(z)-min(values))/(max(values)-min(values));
    vnorm(2,z)= (values2(z)-min(values2))/(max(values2)-
min(values2)); %normalization equation

end


%% Simple Kernel method
[Base, Y_new] = baseline(vnorm(2,:)');


w = 70; % window size

for j = (1+w:length(vnorm)-w) %solves for the simalairty at each section
    summation=0;s=0;

    for p = (j-w:j+w) %for the second part of the SLK equation the summation
part
        s =((vnorm(1,j)-vnorm(1,p))*(vnorm(2,j)-vnorm(2,p)));
        summation=s+summation;
    end

SLK(j-w)=vnorm(1,j)*vnorm(2,j)+summation;

end
SLK=sum(SLK)/length(SLK);

end

%% Baseline Functions
function [Base, Corrected_Spectrum]=baseline(Spectrum)
```

```
%Input
%-------
%Spectrum: vector of size (N*1)

%Output
%-------
%Base: Identified Baseline vector of size (N*1)
%Corrected_Spectrum: Corrected Spectrum vector of size (N*1)

l=length(Spectrum);

lp=ceil(0.5*l);

initial_Spectrum=[ones(lp,1)*Spectrum(1) ; Spectrum ;
ones(lp,1)*Spectrum(l)];

l2=length(initial_Spectrum);

S=initial_Spectrum;

n=1;

flag1=0;

while flag1==0

n=n+2;

i=(n-1)/2;

[Baseline, stripping]=peak_stripping(S,n);

A(i)= trapz(S-Baseline);

Stripped_Spectrum{i}=Baseline;

S=Baseline;

if i>3
    if A(i-1)<A(i-2) && A(i-1)<A(i)
        i_min=i-1;
        flag1=1;
    end
end

end

Base=Stripped_Spectrum{i_min};

Corrected_Spectrum=initial_Spectrum-Base;
Corrected_Spectrum=Corrected_Spectrum(lp+1:lp+l);

Base=Base(lp+1:lp+l);
```

```
end

function [Baseline, stripping]=peak_stripping(Spectrum,Window)
stripping=0;

y=sgolayfilt(Spectrum,0,Window);

n=length(Spectrum);

Baseline=zeros(n,1);

for i=1:1:n
   if Spectrum(i)>y(i)
       stripping=1;
       Baseline(i)=y(i);
   else
       Baseline(i)=Spectrum(i);
   end

end
end
```

Table C.1 Characteristics of wastewater samples (Provided by SMWD)

	Average Inf TSS (mg L^{-1})	**Average Inf BOD (mg L^{-1})**	**Primary Eff TSS (mg L^{-1})**	**Primary Eff BOD (mg L^{-1})**	**Secondary Eff TSS (mg L^{-1})**	**Secondary Eff BOD (mg L^{-1})**	**Tertiary Eff TSS (mg L^{-1})**	**Tertiary Eff BOD (mg L^{-1})**
August	465	280	100	150	7.98	39	1.73	6.7
September	416	260	112	143	7.95	37	1.27	7.0
October	456	319	123	149	8.81	59	1.02	6.6
November	599	362	123	147	8.75	22	0.99	5.9
December	566	343	142	176	8.26	14	1.04	5.8
January	539	340	124	175	8.03	14	0.93	5.4
February	530	312	121	154	18.6	20	0.89	6.0
March	470	316	128	168	9.32	30	0.97	5.1
April	370	303	135	168	8.60	42	1.64	4.1

Appendix D Supplementary Material for Mini-Hydrocyclone Study

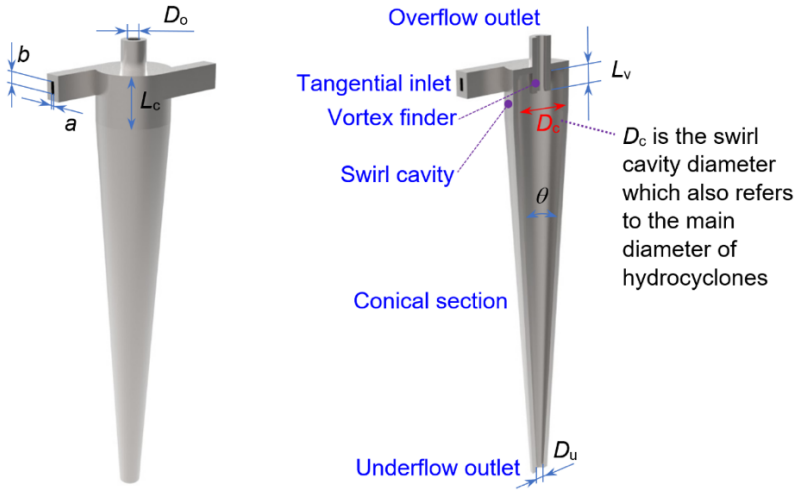

Figure D1. Schematic diagram of a typical mini-hydrocyclone (MHC)

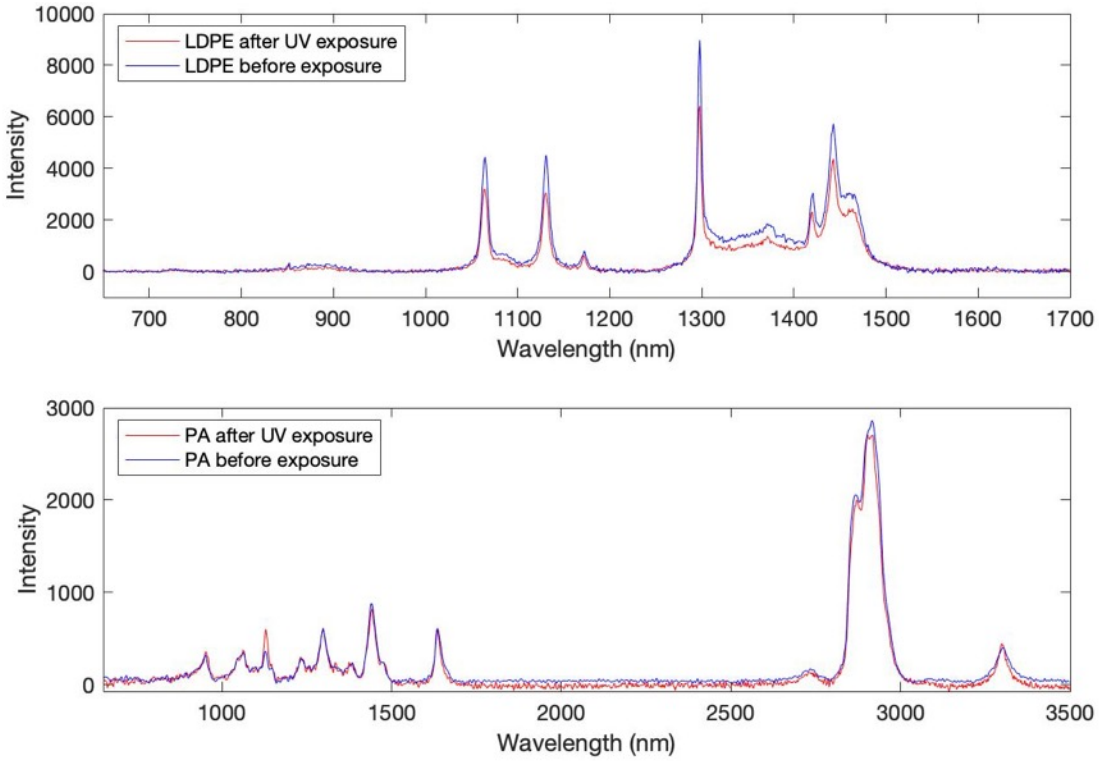

Figure D2. Raman Spectra of LDPE and PA before and after UV exposure

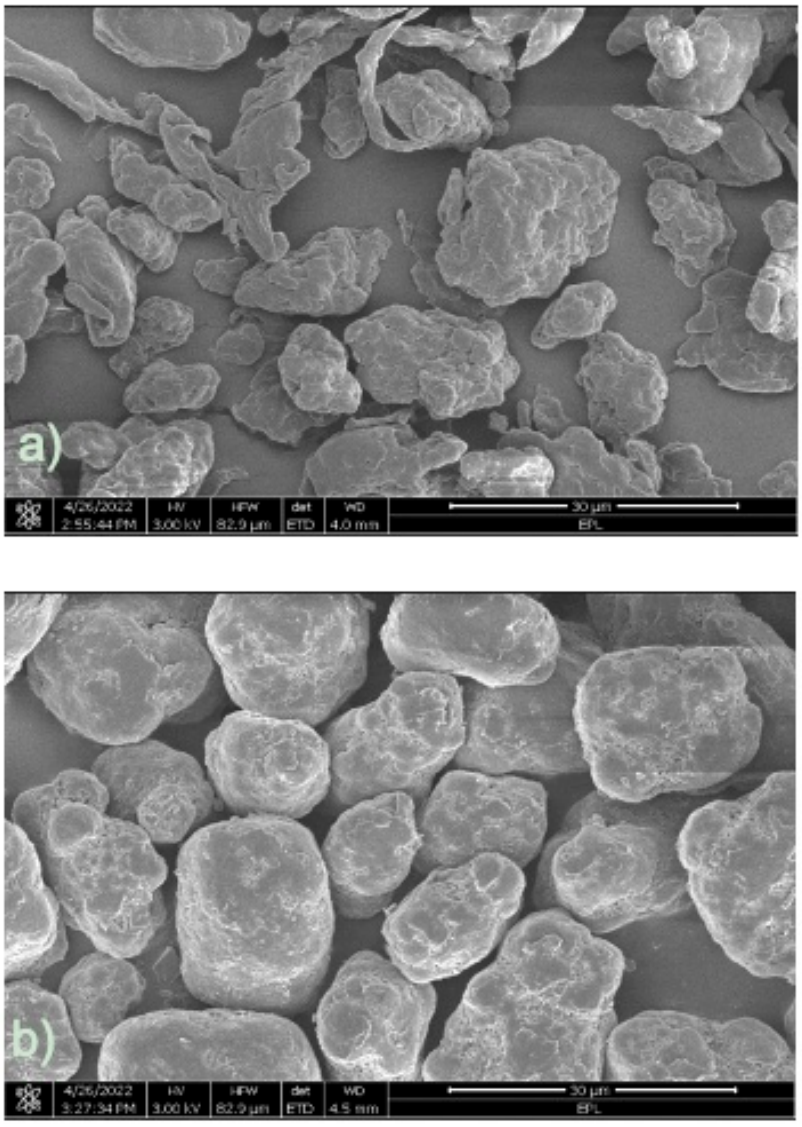

Figure D3. a) SEM image of LDPE; b) SEM image of PA

Table D1. Contamination Control Samples

Type of samples	**MP Contamination (# of MPs/ L)**
MilliQ water	1.33 ± 0.58
Lab space	0.262 ± 0.093 (MP/cm^2)
Feed with MilliQ water (Procedural blank)	4.4 ± 3.9
Underflow with MilliQ water (Procedural blank)	7.33 ± 3.06
Overflow with MilliQ water (Procedural blank)	5.33 ± 2.31
Feed with synthetic storm water (Procedural blank)	6.67 ± 5.03
Underflow with synthetic storm water (Procedural blank)	15.33 ± 9.87
Overflow with synthetic storm water (Procedural blank)	4.67 ± 1.15