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PRESENCE ET DEVENIR DES MICROPLASTIQUES DANS LE RESEAU D'ASSAINISSEMENT UNITAIRE - CAS DE L'AGGLOMERATION PARISIENNE

Rapport final
Thèse de doctorat de Minh Trang Nguyen
Décembre, 2023

- *Thèse réalisée au sein du laboratoire Leesu sous la direction de Rachid Dris, Johnny Gasperi et Bruno Tassin*

Occurrence and fate of microplastics in the sewage system and different pathways into the environment - case of Greater Paris area

by Minh Trang Nguyen

Thesis submitted in November 2023

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Preface

This PhD was a part of the LimnoPlast project, funded by the European Union's Horizon 2020 research and innovation programme under grant agreement No. 860720. The PhD was primarily conducted at the Water, Environment and Urban Systems Laboratory (LEESU) at École des Ponts ParisTech (ENPC) from September 2020 to November 2023. In addition, the PhD program included a three-month secondment at the Department of Biology, at Norwegian University of Science and Technology, Trondheim, Norway.

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DISSEMINATION AND COMMUNICATION

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Nguyen, M. T., Phuong, N. N., Dris, R., Gasperi, J., Gillet, T., Saad, M., Azimi, S. & Tassin, B. Microplastics in sewer sediments of the Parisian combined sewer network (Poster)

UNESCO – EU H2020 LimnoPlast conference 6-8 March 2023, Paris, France

Diving into freshwater microplastic pollution: Connecting water, environmental and social sciences

Nguyen, M. T., Dris, R., Gasperi, J & Tassin, B. Assessment of microplastics in wastewater treatment plant – occurrence, removal and fate (Oral)

NovaTech 2023 – Urban water 3-7 July 2023, Lyon, France

Nguyen, M. T., Dris. R., Phuong, N. N., Gasperi, J., Azimi, S., Rocher, V. & Tassin, B. Microplastic accumulation in sewer sediments and their pathways to the environment via combined sewer overflows (Poster)

Abstract

Plastics have become an integral part of modern human life. With a high-density population, metropolitan areas have become hotspots of plastic consumption and disposal. The abundance of microplastics (MPs) in urban wastewater reflects the plastic pollution issue in these areas. Since the contamination of humans and ecosystems with MPs are of great concern, understanding the inputs of MPs into the environment is crucial to support the implementation of mitigation measures. In this context, this PhD project focused on studying the occurrence and fate of MPs in the Parisian wastewater management system, aiming to investigate and evaluate various pathways through which MPs are released from urban areas into the surrounding environment.

Over the last decade, plastic research has primarily focused on the role of municipal wastewater treatment plants (WWTPs) in addressing MP-polluted wastewater. Existing water treatment technologies at these facilities have demonstrated high efficiencies in separating MPs from wastewater; however, WWTP effluents remain a significant input of MPs into the environment due to their large discharge volume. In addition, literature has highlighted the transfer of MPs into sewage sludge. This byproduct of water treatment serves as a potential source of MPs once disposed of into the environment. By investigating MPs in sludge at various treatment steps, this study found that current sludge treatment technologies were inefficient in completely removing MPs. There was no significant reduction in MP abundance observed after all treatment processes. Contamination levels remaining in the final treated sludge ranged from 8.6×10^4 to 4.5×10^5 particle/kg dry weight (dw) of MPs >25 µm, analyzed by µ-FTIR. Approximately 7 % of sludge-based MPs were returned back to the system via reject water from dewatering processes. Additionally, thermal treatment at high temperatures induced the fragmentation of plastic particles, leading to a reduction in their size. The findings in this study emphasize the potential incorporation and accumulation of MPs in agricultural soils via sludge application, resulting in soil contamination.

While MPs in WWTPs have been extensively studied over the last decade, little attention has been paid at their fate and occurrence during transport within the sewer network before reaching treatment facilities. To address this knowledge gap, this study investigated the potential

accumulation of MPs in sewer sediments, which serve as a stock of pollutants inside the sewer system. High concentrations of MPs, ranging from 5×10^3 to 178×10^3 particle/kg dw, were found in these sediments. This indicates the temporal storage of MPs in sewer sediments instead of arriving at WWTPs. This finding highlights the significant stock of MPs inside the sewer network and the associated risk of downstream transfer during wet weather events due to the resuspension of in-sewer sediments.

Combined sewer overflows (CSOs), one of the main untreated discharges from the combined sewage system, are expected to transfer a large number of MPs into receiving waters. However, research on this pathway is still limited. Therefore, a study to evaluate the quality of CSOs in terms of MP contamination and their potential to emit MPs into the environment was carried out. High MP levels were detected in CSOs during different storm events, ranging from 6.7×10^4 to 3.9×10^5 particle/m^3. At an annual scale, the number of MPs released with CSOs was equivalent to the massive load from treated wastewater, despite much lower discharge volumes. Thus, these findings confirm the significant role of CSOs as a land-based source of MPs into the surrounding environment during intense wet weather events.

In conclusion, this PhD project has provided data on MP contamination levels in various compartments of the wastewater management system, including the sewer network and the sludge-line treatment at WWTPs. It has also elucidated the contribution of various pathways for releasing MPs from urban areas into the environment, thereby underscoring the inadequacy of existing wastewater management systems in addressing plastic pollution.

Résumé français

Les plastiques font désormais partie intégrante de la vie humaine moderne. Avec une forte densité de population, les zones urbaines sont des points centraux de consommation puis d'élimination des plastiques. L'abondance des microplastiques (MP) dans les eaux usées traduit le problème de la pollution plastique dans ces zones. L'exposition des êtres humains et des écosystèmes aux MP étant très préoccupante, il est essentiel de comprendre les apports de MP dans l'environnement pour permettre le déploiement de mesures d'atténuation. Dans ce contexte, ce projet de doctorat s'est concentré sur l'étude de l'occurrence et du devenir des MP dans le système de gestion des eaux usées de la région parisienne, dans le but d'étudier et d'évaluer les différentes voies par lesquelles les MP des zones urbaines sont susceptibles de contaminer l'environnement.

Au cours de la dernière décennie, la recherche sur la contamination plastique s'est principalement concentrée sur le rôle des stations d'épuration (STEP) dans le traitement des eaux usées. Les technologies existantes de traitement des eaux dans ces installations ont démontré une grande efficacité dans l'abattement des MP mais les effluents restent un apport important de MP dans l'environnement. La littérature a mis en évidence le transfert de MP dans les boues d'épuration. Ce sous-produit du traitement de l'eau constitue une source potentielle de MP pour l'environnement, une partie des boues étant épandue sur des sols agricoles. En examinant les MP dans les boues à différentes étapes du traitement, cette étude a révélé que les technologies actuelles de traitement des boues ne permettaient pas de les éliminer complètement. Aucune réduction significative de l'abondance des MP n'a été observée après toutes les étapes du traitement. Les niveaux de contamination restant dans les boues traitées finales étaient compris entre $8,6\times10^4$ et $4,5\times10^5$ particules/kg de poids sec de MP >25 µm, analysés par µ-FTIR. Environ 7 % des MP des boues ont été renvoyés dans la file de traitement des eaux usées via les eaux de rejet provenant des processus de déshydratation. En outre, le traitement thermique à haute température a induit la fragmentation des particules de plastique, entraînant une réduction de leur taille. Les résultats de cette étude soulignent le potentiel d'incorporation et d'accumulation des MP dans les sols agricoles par l'épandage de boues, ce qui entraîne une contamination du sol.

Bien que les MP dans les stations d'épuration aient fait l'objet d'études approfondies au cours de la dernière décennie, peu d'attention a été accordée à leur devenir et à leur présence pendant leur transport des eaux usées dans le réseau d'égouts avant qu'elles n'atteignent les stations d'épuration. Pour combler cette lacune, cette étude a examiné l'accumulation potentielle de MP dans les dépôts en réseau d'assainissement, qui constituent des stocks de polluants à l'intérieur du réseau. Des concentrations élevées de MP, allant de 5×10^3 à 178×10^3 particules/kg de poids sec, ont été trouvées dans ces dépôts. Cette constatation met en évidence l'importance du stock de particules dans le réseau d'assainissement et le risque associé de transfert vers l'aval lors d'événements pluvieux en raison de la remise en suspension des dépôts.

Les débordements des réseaux unitaires via les déversoirs d'orage (DO), l'un des principaux rejets non traités du réseau, transfèrent un grand nombre de MP dans les eaux réceptrices. Toutefois, les recherches pour estimer ce flux sont encore limitées. C'est pourquoi une étude a été réalisée pour évaluer la contamination des eaux des réseaux unitaires par les MP et le rôle des DO dans leur transfert dans le milieu récepteur. Des niveaux élevés de MP ont été détectés dans les déversement au cours de différents épisodes de temps de pluie, allant de $6{,}7\times10^4$ à $3{,}9\times10^5$ particules/m^3. À l'échelle annuelle, le nombre de particules rejetées par les déversoirs d'orage serait équivalent à la charge massive des eaux usées traitées, malgré des volumes de rejet beaucoup plus faibles. Ces résultats ont donc confirmé le rôle important des déversoirs d'orage en tant que source terrestre de particules dans le milieu environnant lors d'événements pluvieux intenses.

En conclusion, ce travail de thèse a fourni des données sur les niveaux de contamination par les MP dans divers compartiments du système de gestion des eaux usées, y compris le réseau d'assainissement et le traitement des boues dans les stations d'épuration. Il a également permis de préciser la contribution des différentes voies de rejet des MP des zones urbaines dans l'environnement, soulignant ainsi l'inadaptation des systèmes de gestion des eaux usées existants dans la lutte contre la pollution plastique.

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Abbreviations

µ-FTIR	micro-Fourier Transform Infrared Spectroscopy
ABS	Acrylonitrile butadiene styrene
ASTM International	American Society for Testing and Materials
ATR	Attenuated total reflectance
BAF	Biologically active filter
CP	Cationic polyamine
CA	Cellulose acetate
COD	Chemical oxygen demand
CSOs	Combined sewer overflows
DDT	Dichlorodiphenyltrichloroethane
DF	Disc filter
DAF	Dissolved air flotation
DO	Dissolved oxygen
DM	Dynamic membrane
dw	Dry weight
EC	Electrocoagulation
EU	European Union
EPS	Expanded polystyrene
SIAAP	Greater Paris sanitation authority
GBS	Gross bed sediment
GDP	Gross domestic product
HDPE	High density polyethylene
HRT	Hydraulic retention time
iWWTP	Industrial wastewater treatment plant
LDPE	Low density polyethylene
MBR	Membrane bioreactor
MDF	Membrane disc filter
MPs	Microplastics
NPs	Nanoplastics
INSEE	National Institute for Statistical and Economic Studies

PA	Polyamide
PAC	Polyaluminum chloride
PAHs	Polycyclic aromatic hydrocarbons
PCBs	Polychlorinated biphenyls
PE	Polyethylene
PEq	Population equivalent
PEST	Polyester
PET	Polyethylene terephthalate
PFAS	Polyfluoroalkyl substances
POC	Particulate organic carbon
PP	Polypropylene
PS	Polystyrene
PU	Polyurethane
PVAC	Polyvinyl acetate
PVC	Polyvinyl chloride
RE	Removal efficiency
REACH	Registration, evaluation, authorization and restriction of chemicals
RSF	Rapid sand filtration
SAV	Seine Aval WWTP
SEC	Seine Centre WWTP
SEG	Seine Grésillons WWTP
SDS	Sodium dodecyl sulfate
SRT	Sludge retention time
US-EPA	United States Environmental Protection Agency
VC	Vinyl copolymer
WWTP	Wastewater treatment plant

General introduction

Plastics have become an integral part of our world as a novel material with a multitude of positive attributes, such as being lightweight, flexible, durable, waterproof and inexpensive. Consequently, they have supplanted conventional materials like metal, wood, glass and natural textile fibers, rendering them indispensable in various aspects of modern daily life. Human demand for plastic has continuously and exponentially grown, especially over the last two decades, culminating in a production of 400 million tons in 2022 (PlasticsEurope, 2023). Their applications cover nearly every sector, including packaging, construction, automotive, agriculture, medical care, personal hygiene, education, fashion and leisure. Furthermore, plastics are not only abundantly available, but also excessively utilized, given the dominance of single-use products, with up to 40 % of the annual production used for packaging only. The rapid growth of plastic waste is a consequence of massive production and consumption behavior. Existing solid waste management systems are inefficient in properly handling plastic waste, resulting in its leakage into the environment.

Owing to their durability and resistance, plastics tend to persist over extended periods (Webb et al., 2013; Fotopoulou & Karapanagioti, 2019). For example, the half-life of high density polyethylene (HDPE) pipes in the marine environment was estimated to be 1,200 years in Chamas et al. (2020). This leads to the accumulation of plastic waste in the environment, raising concerns about their potential impacts on both humans and ecosystems. Scientific efforts have therefore been dedicated to tackling this issue.

Once disposed of in the environment, plastic waste breaks down into smaller debris, which can be found in a wide range of sizes and shapes. Fragmentation also occurs in in-use plastic items, generating smaller plastic fragments and unintentionally releasing them into the environment. Great interest has been focused on microplastics (MPs), particles smaller than 5 mm, due to their specific size. This size not only renders them available to a wide variety of organisms but also provides a large surface area for the sorption of pollutants and the desorption of their own chemical additives. Moreover, capturing these particles using simple techniques is challenging.

Since their initial discovery in the ocean in the 1970s, MPs have now been detected worldwide across various environmental compartments, from marine to freshwater ecosystems, and even in the air (Capparelli et al., 2021; Roscher et al., 2021; Beaurepaire et al., 2022). In particular, MPs have been found abundantly in urban wastewater, reflecting the direct link to plastic products available in household environments. This phenomenon can also be attributed to other human activities within these densely populated areas, including recreation, transportation and industry. Consequently, local watercourses in proximity have become susceptible to plastic contamination due to inadequate urban wastewater treatment (Polanco et al., 2020; Fang et al., 2021; Werbowski et al., 2021). Therefore, it is important to quantify the presence of MPs within wastewater management systems and understand the role of discharges from these systems as sources of MPs into the surrounding environment. This knowledge will be valuable in the mitigation efforts to combat MP pollution.

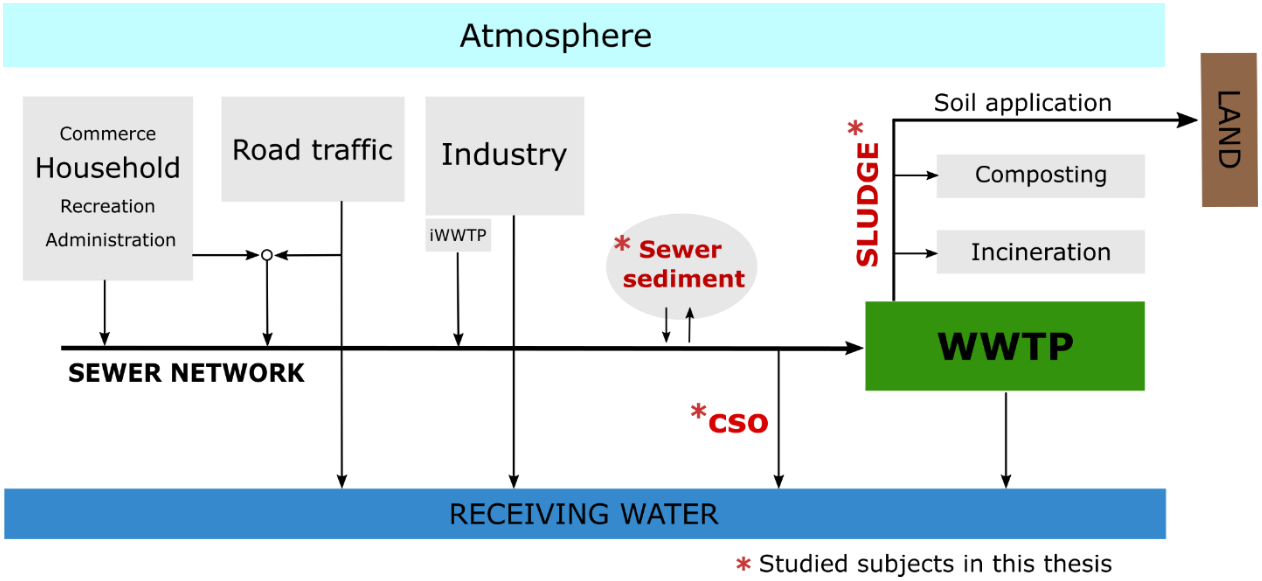

Figure 1: Principal role of the wastewater management system in handling MP fluxes in an urban environment. MPs generated from human living activities enter sewer systems along with wastewater. These particles are then transported through the sewer network to wastewater treatment plants (WWTPs). The main discharges from these treatment facilities into the environment include treated wastewater and treated sludge. Additionally, in case of combined sewer systems, untreated wastewater can be discharged before reaching WWTPs, known as combined sewer overflows (CSOs). The accumulation of sewer sediments inside the sewer network influences pollution levels in wastewater, particularly during wet weather events. The diagram also depicts other discharges of MP-contaminated wastewater into surrounding freshwater bodies (arrow for atmosphere)

The overall goal of this thesis was to elucidate the occurrence and fate of MPs in different compartments of the Parisian wastewater management system, as highlighted in Figure 1. Various potential pathways for discharging MPs from urban areas into the Seine River and the surrounding terrestrial environment were also evaluated with the obtained results. The scientific objectives of the thesis are described in more detail as follows.

Over the past decade, investigations of wastewater management systems concerning plastic contamination were carried out, with a primary focus on wastewater treatment plants (WWTPs). Research efforts have mainly aimed at understanding the occurrence and fate of MPs within these treatment facilities and evaluating the efficiency of existing water technologies for MP removal. The findings have documented high efficiency of WWTPs up to more than 90 % in separating MPs from the water phase (Lares et al., 2018; Akarsu et al., 2020; Koyuncuoğlu & Erden, 2023). Most plastic particles are removed during the screening and grit-grease removal processes, while remaining particles tend to be transferred into sewage sludge via primary and secondary sedimentation. Consequently, the disposal of MP-polluted sludge through soil application can serve as a pathway for releasing MPs into the environment (Figure 1). In contrast to water-line treatment, the existence and behavior of MPs through sludge-line treatment have not been clearly elucidated. Additionally, there is a lack of data on the impacts of different sludge treatment technologies on MPs (Hatinoğlu & Sanin, 2021). For this reason, the first aim of this study was to achieve a better understanding on MPs' occurrence throughout sludge-line treatment in WWTPs by investigating MPs in different sludge types, from raw sludge to final treated sludge. Estimated MP budget on an annual scale was carried out for different treatment steps, including centrifugation and anaerobic digestion. The results were expected to support the assessment of treatment technologies' impacts on MP particles, as well as evaluate the potential of treated sludge as a pathway for MPs entering into the environment.

Objective 1

Investigate the occurrence of MPs in sludge-line treatment at WWTPs in the Greater Paris area, from raw sludge to final treated sludge, using various treatment technologies

Assess the impacts of these treatment technologies on MPs

Evaluate the potential of treated sludge as a pathway for MPs entering the surrounding terrestrial environment

While research efforts have primarily focused on MPs within WWTPs, the occurrence and fate of these plastic particles before reaching treatment facilities have received limited attention. Previous works have reported changes in wastewater quality during its conveyance within the sewer system due to the sedimentation of particulate matter, forming wet bed deposits known as sewer sediments inside sewer pipes (Figure 1). These sediments can significantly contribute to pollution levels in wastewater when resuspended during wet weather events (Gromaire et al., 2001; Gasperi et al., 2010). Similarly, MPs may interact with mineral and organic particles present in wastewater during the transport, settling down and becoming trapped in sewer sediments. When these sediments erode, MPs can be released into water flow along with other pollutants. Therefore, the second goal of this study was to enhance the understanding of MP's transfer through sewer networks to WWTPs by monitoring their accumulation in sewer sediments. The findings are expected to support the assessment of in-sewer processes' contribution to the quality of combined sewer overflows (CSOs) concerning plastic pollution.

Objective 2

Understand transfer of MPs to wastewater treatment plants by monitoring their accumulation in sewer sediments inside the Parisian sewer network

Assess indirectly the contribution of in-sewer process to the quality of CSOs concerning plastic pollution

Furthermore, a portion of wastewater is occasionally discharged into receiving waters without treatment for various reasons, including CSOs, which occur due to the surcharging of combined sewer systems (Figure 1). According to the literature, CSO discharges not only disrupt receiving water dynamics, reduce dissolved oxygen levels, but also introduce high loads of micropollutants into receiving waters (Gasperi et al., 2011; Passerat et al., 2011; Phillips et al., 2012; Launay et al., 2016). Regarding plastic pollution, CSOs are expected to be an important source of MPs released into the environment since they are composed of untreated wastewater and materials eroded from in-sewer deposits. However, research on the quality of CSOs remains limited, likely due to challenges in sample collection with the stochastic nature of these events. This results in a lack of data to evaluate the potentially major pathway of MP emissions via CSOs. Therefore, the third goal of this study was to investigate the emission of MPs along with CSOs into the environment during wet weather events. The obtained results will be used to evaluate the contribution of CSOs to plastic pollution in receiving water bodies compared to other point sources, mainly WWTP effluents.

Objective 3

Investigate the emission of MPs along with CSOs into the Seine River during intensive wet weather events

Evaluate the contribution role of CSOs to the level of plastic pollution in receiving water bodies compared to other point sources, mainly WWTP effluents, in the scale of Greater Paris area

In this context, this thesis presents the main findings of a three-year investigation on MP pollution in urban wastewater within the Paris megacity. The organization of this thesis is as follows:

Chapter 1 will first present the definition and related terms of plastic materials. Then, an overview of plastic research which has been dedicated to address plastic pollution is summarized and up-to-date knowledge of MPs in urban areas is provided. In detail, this section will introduce MP fluxes within an urban environment, thereby shedding light on the primary sources of MP emissions into the surrounding environment, as well as the role of wastewater management systems to address this emerging pollutant. Next, a comprehensive review of the most recent findings on MPs within the wastewater management system, especially on their occurrence and fate in water-line treatment at WWTPs is provided. As the effluent still contains MPs and acts as one of point source, research efforts have been dedicated to the development of innovative technologies designed to enhance the removal of MPs from wastewater, which is summarized in this chapter. Lastly, the remaining knowledge gaps of MP research in urban wastewater based on the literature review in this chapter, which inspired the present study, are mentioned.

Chapter 2 will introduce the selected urban environment of the study, which encompasses the Paris megacity, the Seine River, and the Parisian wastewater management system. The Paris megacity is as an ideal representative of a densely populated urban area. The Seine River serves as a local watercourse that endures anthropogenic pressures from human activities within its catchment area. The Parisian wastewater management system plays a crucial role in mitigating the impacts of urban wastewater on the receiving waters, in this case, the Seine River.

Chapter 3 is devoted on the study of MPs in sewage sludge within WWTPs. The main content of this chapter is extracted from the manuscript in preparation "*Microplastic contamination along different sludge-line treatments: case of Paris megacity*". Since MPs separated from water phase are transferred into sewage sludge, this byproduct of water treatment becomes a potential source of MPs into the environment upon disposal. The study therefore focused on the occurrence on MPs throughout sludge-line treatment in WWTPs. The findings provide a better understanding on the efficiency of existing sludge treatment technologies toward MP removal.

Also, the data help to assess the magnitude of MPs emitted into the environment via the disposal of treated sludge to agriculture land.

Chapter 4 presents the study on the occurrence and fate of MPs during transport with wastewater inside sewer network before reaching treatment facilities. This section is extracted from the manuscript under revision "*Microplastic accumulation in sewer sediments and its potential entering the environment via combined sewer overflows: a study case in Paris*". The findings indicate the accumulation of MPs in sewer sediments which can act as a major stock of MPs inside sewer network, thereby posing an associated risk of downstream transfer during wet weather events due to the resuspension of these in-sewer sediments. Data on MP content in sewer sediments also help to improve the indirect assessment of MPs entering the environment via CSOs.

Chapter 5 presents the investigation on the quality of CSOs regarding MP contamination. The results help to estimate MP flux discharged via CSOs to the environment during intensive wet weather events, thereby assessing the contribution role of CSOs to plastic pollution in receiving waters. The main content is extracted from the manuscript *"Combined sewer overflows – a neglected pathway of urban-based microplastics to the environment: Case study of the Parisian sewer network"*.

Chapter 6 will summarize and discuss the main research outcomes of the PhD project. Perspectives for future work to further contribute to research topic will be included. In addition, the limitations of the methodology applied in this study are also mentioned.

Chapter 1: Literature review

1.1. Plastic pollution and research needs

1.1.1. Plastics – definition and related terms

Plastic was originally used to describe a substance that could be molded and shaped. Nowadays, plastics refer to a wide range of synthetic or semi-synthetic materials having polymers as their main ingredient. Polymers are long chains of many repeating subunits, named ‘monomers’ (Crawford & Quinn, 2017). The invention of the first full-synthetic polymer in 1907, known as Bakelite, marked the beginning of the plastic age.

A system of symbols is regulated by American Society for Testing and Materials (ASTM International) for plastic identification (Figure 1-1), which aims to facilitate the process of identifying and separating plastics for recycling efforts. Plastics labelled from 1 to 6 represent commodity plastics, which are of the greatest commercial importance with a high rate of production. Plastics labelled as 7 refer to less commonly produced types.

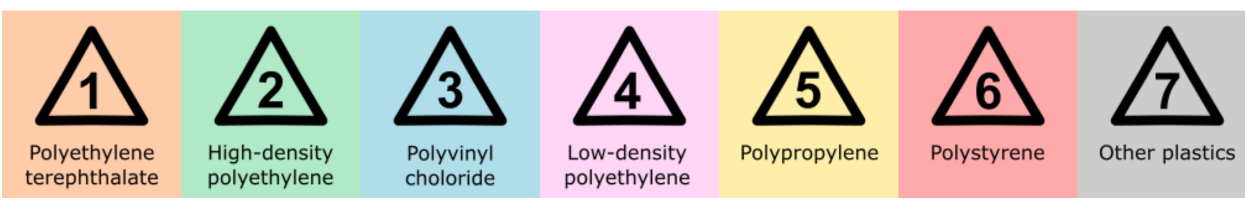

Figure 1-1: Identification code for different polymer types according to ASTM International

Plastics can generally be divided into two main groups: thermoplastics and thermosetting plastics. Thermoplastics can be melted and molded when heated up at a certain temperature, and they solidify upon cooling. This feature allows thermoplastics to be reshaped and recycled. In contrast, thermosetting plastics cannot be melted and reformed, making them unsuitable for recycling.

A polymer can consist of a single type of monomer, referred to as a ‘homopolymer’. For example, polyethylene (PE) is a homopolymer. A polymer can also be composed of two or more different types of monomers, known as a ‘copolymer’. For instance, acrylonitrile butadiene styrene (ABS) is a copolymer. Different types of polymers can be combined to obtain the advantageous properties of the original materials, which are categorized as polymer blends.

Biodegradable plastics are materials that can be broken down by biological organisms within a reasonable timescale, producing natural byproducts that are harmless to the surrounding environment such as water, carbon dioxide and biomass (Ammala et al., 2011; Andrady, 2015; Crawford & Quinn, 2017). This is distinct from bio-based plastics, which are derived from biomass resources. Bio-based polymers can be biodegradable or not, depending on their chemical structure.

The manufacturing process of plastics involves the use of various chemicals, known as additives, to enhance the material's properties or improve the production process itself. Common plastic additives are plasticizers, modifiers, colorants, flame retardants, and more. For certain uses, fillers such as talc are also added to polymers to increase the volume of plastics at low cost.

1.1.2. Plastics: from an outstanding material to an environmental issue

Plastics have become an integral part of modern life as a new material with many outstanding qualities. They are remarkable for being lightweight, versatile, durable, waterproof and inexpensive. These attributes have established plastics as an indispensable material in various aspects of daily life, contributing significantly to hygiene and overall comfort in human society. Plastic have replaced traditional materials such as metal, wood, and glass, finding applications in diverse sectors, including packaging, construction, automotive, agriculture, medical, education, leisure, and beyond. The global demand for plastics has consistently grown since the beginning of mass production in the 1950s. In particular, a surge in production volume has been observed in the last two decades, reaching over 400 million tons in 2022 (PlasticsEurope, 2023). China leads the world's top producers, accounting for one third of global plastic production, followed by countries from North American and Europe. Fossil-based plastics continue to dominate the plastic market, representing over 90 % of the world's production. Polypropylene (PP) and polyethylene are the most demanded polymers among diverse plastic materials.

Plastic products exhibit a wide range of service lifespans, which depend on their specific applications. In sectors like automotive, electronics and construction, plastics can endure several years or even decades. Conversely, plastic items used in agriculture, for example mulching films, have much shorter service lives, lasting only about one year (Jansen et al., 2019). In particular, plastic products used for food packaging and catering services, such as low-density prolyethylene (LDPE) bags, food wrap, plastic utensils, are often discarded after one-time use (Koelmans et al., 2014; Andrady, 2015; Chen et al., 2021). These items are commonly described as single-use plastics. PlasticsEurope (2023) reported that nearly 40 % of the global plastics were used for packaging only. Disposable mentality towards plastics and their overconsumption could be attributed to the affordability of this material. Consequently, massive production and excessive consumption habits of plastics have resulted in a rapid growth of plastic waste. Especially, urban areas have become hotspots of plastic waste generation due to their high density population. It is estimated that plastic waste accounts for 10-16 % of the total global municipal waste by weight (Barnes et al., 2009; Thompson et al., 2009; Muenmee et al., 2015).

The three primary options for managing plastic waste at the end of its life cycle are recycling, energy recovery, and landfill. Notable efforts have been made to enhance recycling practices among developed countries, with the Netherlands, Norway, Spain, and Germany taking the lead (PlasticsEurope, 2022). However, the rate of recycled plastics remains moderate, accounting for less than 10 % of total plastic waste in Europe alone. Moreover, most of the recycling is indeed downcycling. This occurs due to the deterioration in the properties of plastics during product lifetime and the impacts from the recycling processes, which leads to the recycled material not being qualified for the production of the same products (Simon, 2019). A significant quantity of plastic waste continues to be disposed of in landfills because it is a cheaper and more straightforward option when compared to recycling and incineration for energy recovery (H. Li et al., 2022). In many developing countries, plastic waste is dumped alongside other waste materials in open areas due to the low cost (Muenmee et al., 2015). Thus, the existing solid waste management systems, both in developed and developing nations, are generally inefficient in adequately handling plastic waste, resulting in the release of a significant fraction into the environment.

Because of their durable and resistant nature, plastics persist for extended periods under environmental conditions upon disposal. This leads to the accumulation of plastic waste in the environment. A prominent incident is the discovery of the Great Pacific garbage patch by Charles Moore in 1997. This has significantly raised awareness of both the scientific community and the general public about plastic pollution.

Plastic pollution has shown adverse impacts on the wildlife of both aquatic and terrestrial ecosystems. Gall and Thompson (2015) documented that more than 30,000 marine individuals, including turtles, mammals, and seabirds, got entangled in plastics debris. Most of these cases were caused by plastic nets and ropes that were either lost, abandoned, or discarded during fishing activities. Terrestrial animals also suffered from plastic pollution; they were often found with their heads stuck in plastic containers or their horns caught in plastic waste. Such entanglements typically result in physical injuries, restricted mobility, and even death in these animals (Derraik, 2002; Duncan et al., 2017). Additionally, plastic ingestion is documented among various species, causing serious issues such as choking, obstructed digestive tracts, and starvation Laist (1997) and Lavers et al. (2014). This situation has become increasingly common due to the widespread distribution and fragmentation of plastic waste in the environment.

1.1.3. Research on plastic pollution

As plastic waste has become ubiquitous in the environment, the unknown impacts of these debris on humans and other living organisms have concerned both scientific and public communities. Therefore, further research is requested to gain a better understanding of these related issues. Over the last few decades, scientific efforts have been dedicated to this purpose.

1.1.3.1. Definition and classification

Once disposed of in the environment, plastic waste undergoes weathering process influenced by various factors such as mechanical force, temperature, light and water (Crawford & Quinn, 2017). This makes large plastic debris become brittle and gradually break down into smaller fragments, which can vary widely in size and shape. In plastic research, these fragments can be divided into several size classes (i.e., macro-, meso-, micro- and nano-plastics), with the thresholds primarily determined by methodological limits (van Emmerik, 2021), as illustrated in Figure 1-2. Generally, plastic debris with the smallest dimension greater than 5 mm is classified as *macroplastics*, while those below this threshold are referred to as *microplastics* (Thompson et al., 2004; Arthur, C., J. Baker and H. Bamford, 2009). Similarly, the US-EPA defined *microplastics* as plastic particles ranging in size from 5 mm to 1 nm. These are the most commonly used definitions in the scientific community at the moment. Although the definition of *nanoplastics* is still a subject of debate, some publications define NP as plastic particles within the size range from 1 to 1000 nm, based on colloidal physics and chemistry (Gigault et al., 2018; González-Pleiter et al., 2019; Hartmann et al., 2019). In contrast, *nanoplastics* are plastic particles smaller than 1 nm according to the US-EPA.

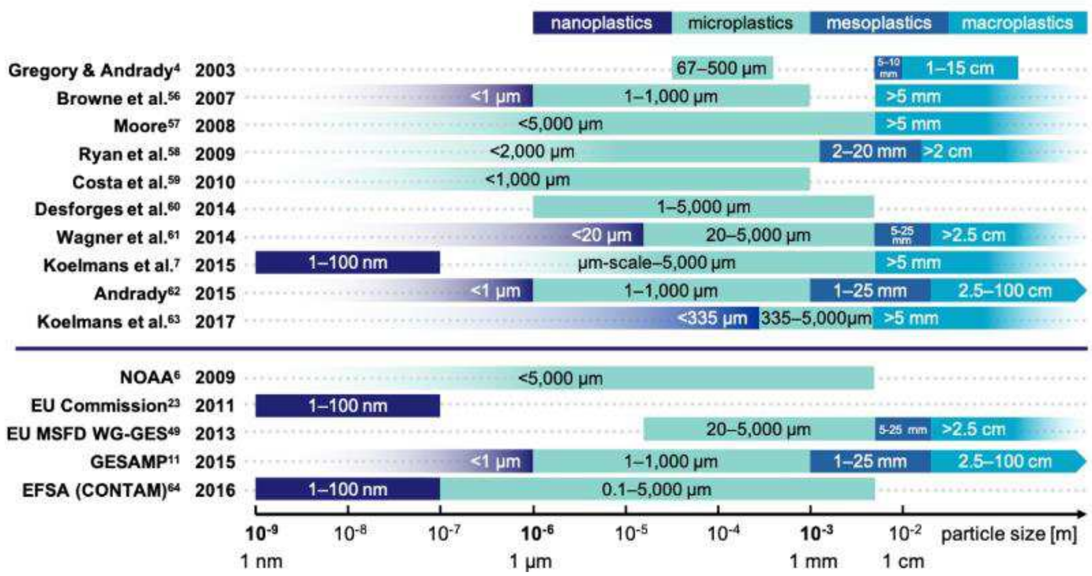

Figure 1-2: Various classifications of plastic debris according to size used in the literature and institutional reports (Hartmann et al., 2019)

1.1.3.2. Overview of research on microplastics

There has been a greater focus on MPs compared to macroplastics owing to their specific size. This size not only makes them available to a wide range of organisms, but also offers a large surface area for the sorption of pollutants on these particles. Meanwhile, it is challenging to capture MPs with simple techniques. The study of nanoplastics (NPs), which is a subclass of MPs, represents a new field of plastic research with advancements in identification technologies.

Since being first reported in the ocean in the 1970s, MPs have now been detected worldwide, from highly urbanized areas (Dris et al., 2018; Rao et al., 2020) to rural and remote locations (González-Pleiter et al., 2020; Materić et al., 2020). These particles were present in environmental samples taken from open seas and coastal areas (Bagaev et al., 2018; Capriotti et al., 2021), and from numerous freshwater systems, including rivers (Alam et al., 2019; Chauhan et al., 2021), streams (Dikareva & Simon, 2019; Montecinos et al., 2022), lakes (Eriksen et al., 2013; Wang et al., 2019), and reservoirs (Nocoń et al., 2020; Lin et al., 2021). They dispersed throughout the water column, from sediments to surface water (Rodrigues et al., 2018; Laermanns et al., 2021; Sekudewicz et al., 2021). MPs were also found to be present in the air (Dris et al., 2016; Beaurepaire et al., 2022). Especially, the abundance of MPs in wastewater was widely reported in the literature, showing the direct correlation between human activities and plastic pollution (F. Wang et al., 2020; Ziajahromi et al., 2021; Zhou et al., 2023).

In addition to detecting the distribution of MPs in the environment, plastic research has also paid attention to the toxicity of MPs and its potential impacts on human and ecological systems. MPs not only contain chemical additives from production process, but also carry various toxic (micro)pollutants from the surrounding media through absorption and adsorption (Endo & Koelmans, 2019; D. Zhou et al., 2022). These pollutants are polycyclic aromatic hydrocarbons (PAHs) (Fisner et al., 2017; Diepens & Koelmans, 2018), polychlorinated biphenyls (PCBs) (Velzeboer et al., 2014; Gauquie et al., 2015), dichlorodiphenyltrichloroethane (DDT) (Heskett et al., 2012), polyfluoroalkyl substances (PFAS), heavy metals, and pharmaceuticals (Wu et al., 2016; Ateia et al., 2020). Ecotoxicological studies have been conducted on a wide range of organisms, from aquatic to terrestrial environments, to understand the effects of MPs (Barboza et al., 2018; Yi et al., 2019; S. Zhang et al., 2019). For instance, adverse impacts on the ecophysicological functions of organisms have been documented after exposure to PS MPs, including metabolism and behavior (Cedervall et al., 2012; Mattsson et al., 2015), development and growth (Della Torre et al., 2014; Cui et al., 2017), survival and reproduction (Besseling et al., 2014; Sussarellu et al., 2016). The responses of tested organisms to MPs exposure mostly depended on particle characteristics (e.g., polymer type, size, morphology and surface alterations), while impacts from additive chemicals associated with the particles were rarely studied (Gomes et al., 2022). Future research aims to investigate potential direct and indirect plastic effects of MPs at the ecosystem level. Besides, since MPs were found in the air, in foods and drinks, human exposure to MPs via respiration and diet has been expected (Liebezeit & Liebezeit, 2013; Karami et al., 2017; Kirstein et al., 2020). Indeed, recent studies have detected plastic particles in human organs, such as intestine, lungs and liver, also in human blood and breast milk (Barceló et al., 2023). While no acute impact on human health has been reported, chronic impacts, which have been observed on animals in *in vivo* studies, are expected.

Meanwhile, the question of solutions for plastic pollution and the increasing abundance of MPs in the environment have been raised. Potential methods for addressing this issue have been summarized in the literature (Löhr et al., 2017; Eriksen et al., 2018; Prata et al., 2019), covering from production, application to disposal stage of plastic products (Wagner, 2022). For example, taxes and bans can be implemented to reduce plastic production and consumption, while incentives and subsidies can promote the replacement of greener materials which are compostable and biodegradable. Technological advancements should be harnessed to improve plastic waste management from recycling and recovery to disposal and cleanup efforts. Strategies for enhancing societal responsibility have also been mentioned, such as information campaigns, educational programs, and participation in cleanup activities. The realization of these solutions may be inefficient due to the complexity, diversity and uncertainty of plastic pollution (Wagner, 2022). The "*Key findings and recommendations*" from the Limnoplast project stated that the systemic solutions, which consider the whole plastic life cycle while taking into account technological, political and societal factors, are key to solving plastic pollution. A similar statement was made in Wagner (2022), which recommended to approach the plastics issue from a systems perspective, integrating the concept of 'circular economy'.

Overall, plastic is a new material that did not exist in nature before its invention. The increase in quantity of this material in the environment in micro-size without proper control poses a potential risk to human society and ecosystems. Therefore, it is necessary to minimize the release of plastics, especially MPs, into the environment. To achieve this, a comprehensive understanding of MP pathways in the environment is required before implementing further mitigation measures.

1.2. Microplastic research in urban areas

Plastic waste is directly linked to human activities. Urban areas, as a result, become hotspots of plastic waste generation due to their high density population. Consequently, MPs, as a representative of plastic pollution, are found abundantly in urban wastewater. This renders local surrounding watercourses susceptible to plastic contamination due to the inadequate treatment of urban wastewater. In this context, the occurrence and fate of MPs within urban areas have been investigated over the last decade to comprehend the contribution of these regions to plastic pollution levels in the environment.

1.2.1. Microplastic fluxes in an urban system: from inputs to receiving environments

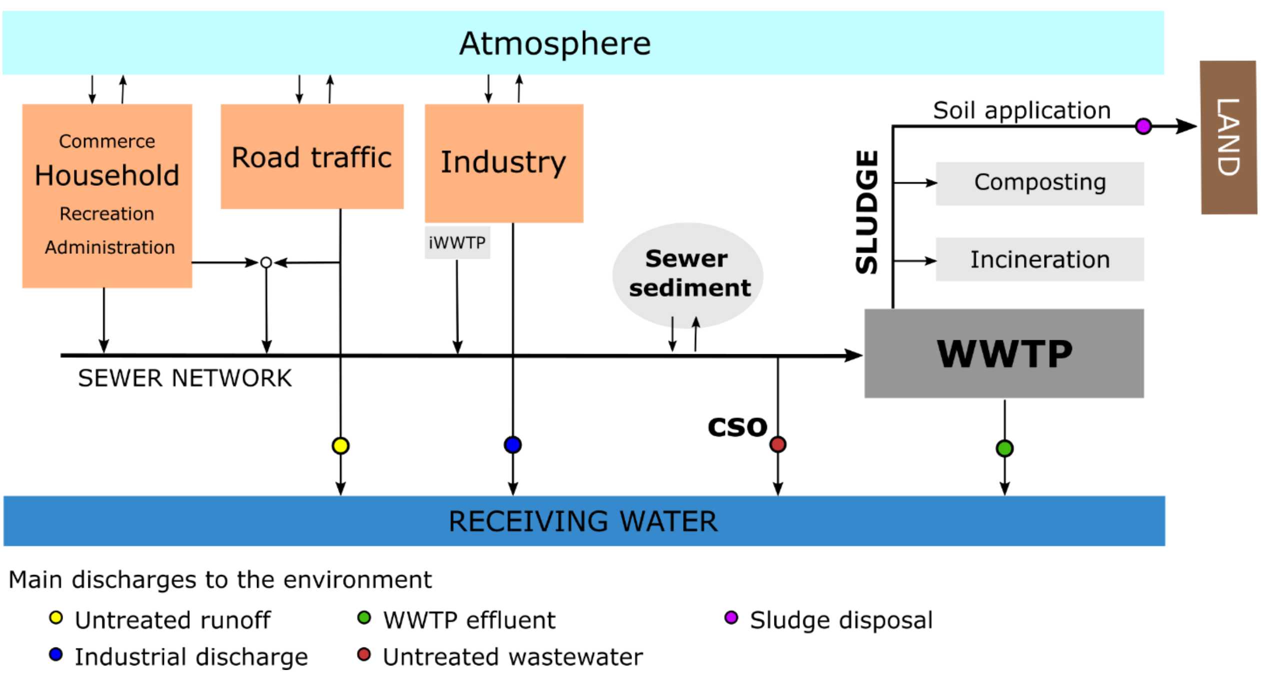

Figure 1-3: MP fluxes within a typical urban environment. The figure shows the path of MP flux from various sources into wastewater, and eventually, the natural environment. MPs primarily stem from human activities, including daily living, manufacturing, and commuting. Many of these particles find their way into

wastewater. Wastewater management systems, as a preventive measure, receive and treat wastewater, thereby protecting the surrounding environment from anthropogenic pressures. Land and nearby water bodies represent two primary environmental compartments that receive both treated and untreated discharges, either directly from sources or indirectly through wastewater management systems

Industrial activities have been identified as a primary source of MPs (Deng et al., 2020; Gkika et al., 2023), as recent studies have detected MPs in industrial wastewater with concentrations ranging from 183 to 443 particles/L (Franco et al., 2020; Chan et al., 2021; Van Do et al., 2022; Brown et al., 2023). The contamination level and the polymer composition of MPs found in industrial wastewater vary among different industries, reflecting the nature of manufacturing processes. For example, Chan et al. (2021) detected fiber-shaped MPs in the discharge of a textile wet-processing mill in China; F. Wang et al. (2020) reported a higher MP content in industrial wastewater from chemical factories compared to electroplating, especially for polystyrene (PS) due to its use as a raw material. Industrial wastewater is typically treated at industrial wastewater treatment plants (iWWTPs), serving as a component of human intervention. The efficiency of these plants can vary widely among different sites, ranging from 20 % (Van Do et al., 2022) up to 90 % (Franco et al., 2020). Other studies also documented the persistence of MPs in the effluent after treatment (Bitter & Lackner, 2020; F. Wang et al., 2020). In some cases, industrial wastewater enters sewer systems, eventually heading to municipal WWTPs for purification. Otherwise, it can be directly discharged into surrounding water bodies.

Human activities in residential areas and various services, including commercial, recreational, and administrative settlements, generate domestic wastewater. Since MPs are present in a wide range of products within these environments, they are found in this type of wastewater. For instance, microbeads can be detected following the application of personal care and cleaning products, where these particles are added as abrasive agents (Mason et al., 2016; Kalčíková et al., 2017). Due to environmental concerns related to plastic pollution, the use of intentionally-added MPs in wash-off products is now banned in several countries or restricted by several large cosmetic and pharmaceutical companies. The European Commission also released new regulations to restrict these particles under the EU chemical legislation REACH recently (European Commission, 2023). Nevertheless, MPs stemming from laundry activities is of great concern. Since textile production is dominated by synthetic fibers, they have been detected abundantly in domestic wastewater, especially from residential areas (Zhou et al., 2023). Polymers such as polyester (PEST), PE and polyamide (PA) are found to be dominant (Dris et al., 2015; Vollertsen & Hansen, 2017). Domestic wastewater is typically conveyed to municipal wastewater treatment plants for treatment.

Streets, roads and various other impervious surfaces are covered with dust and plastic particles originating from different human activities. Runoff water, stemming from precipitation events or street cleaning activities, can carry these particles into sewer systems. Plastic particles found in runoff can result from the erosion of car tires and other vehicle transportation parts, textile clothes, additives or coating materials, fragments of road marking paints and degradation of plastic items such as building materials and billboards, etc. (Cai et al., 2017; Dris et al., 2017; Vianello et al., 2019; Can-Güven, 2020). Tire-wear particles are likely the most significant contributor to MPs released into urban runoff (Kole et al., 2017; Järlskog et al., 2020). Besides, atmospheric deposition via precipitation, including rain and snowfall, also significantly transport MPs from the atmosphere into runoff. Klein and Fischer (2019) reported a correlation between the abundance of MPs in atmospheric deposition and storm events. Dris et al. (2016) estimated that 10^{10} particles fall from the atmosphere onto the Paris megacity each year through deposition process. Several studies have reported that stormwater runoff is highly polluted with MPs. For instance, Piñon-Colin et al. (2020), which investigated plastic pollution in stormwater runoff in Tijuana (Mexico), found hundreds of MPs per liter for various land uses. Similarly, Sun et al. (2023) documented high MP contamination levels, reaching up to 5,000 particles/L in urban surface runoff from residential catchments. MP-contaminated runoff can be either discharged directly to surrounding waters in case of separate sewer systems or conveyed along with wastewater to WWTPs in combined sewer systems.

Wastewater management systems play a crucial role in protecting the surrounding environment from pollutants stemming from human activities within urban areas. A wastewater management system comprises of two main components: the sewer network and the municipal wastewater treatment plants. The sewer network, consisting of underground pipes, pumping stations and other accessories, collects and conveys wastewater from different sources to WWTPs, where it is purified before being returned to the environment. Urban areas are typically equipped with either a combined sewer - stormwater system or a separate system. The combined system is commonly found in old town centers, where the sewer network transports a mixture of wastewater and storm water to treatment facilities. In contrast, the separate system is designed to convey sewage and stormwater independently. In this system, domestic wastewater heads to WWTPs, while runoff is discharged to surrounding waters with or without undergoing a basic treatment such as screening.

Thus, municipal WWTPs receives not only domestic wastewater, but also industrial wastewater in some instances, and runoff if the sewer system is combined. Talvitie et al. (2017b) found hundreds of MPs per liter in 24-hour composite raw wastewater samples taken at the influent of a WWTP in Finland. The variation in MP levels could be correlated with activities in households and commercial buildings throughout the day. Other studies also reported high MP levels in the influent of municipal WWTPs (F. Wang et al., 2020; Ben-David et al., 2021; Zhou et al., 2023). At WWTPs, pollutants in wastewater are removed during water-line treatment before treated wastewater is discharged as effluents. As a by-product from water treatment, sewage sludge undergoes different stages of sludge-line treatment before disposed of as biosolids. Since municipal WWTPs receive MPs from various sources, their discharges, including effluents and biosolids, are inputs of MPs to the environment.

Besides, it should be noted that not all wastewater collected by the sewer network reaches WWTPs. In combined sewers, while the volume and flow rate of domestic wastewater fluctuate during the day depending on the quantity of water use, stormwater can rapidly increase during wet weather occurrences. In case of intensive events, sewer network might become inadequate to deliver all the water to WWTPs. This leads to the overflow of excess water through gullies and manholes into the surrounding environment. Additionally, the capacity of WWTPs might be insufficient to handle the incoming inflow, resulting in the discharge of surplus water to the receiving water bodies to protect treatment systems. Combined sewer overflow is the term which refers to sewage spilled into the environment without treatment due to the surcharge of the combined system. Furthermore, technical issues or malfunctions of the sewer system can also result in the discharge of untreated wastewater into the surrounding recipients. These untreated discharges can also serve as potential sources of MPs into the environment.

During transport, particulate matter in wastewater can detach from the water phase and settle down, forming bed deposits inside sewer networks, named sewer sediments. This phenomenon often occurs during low-activity intervals such as nights and dry-weather periods or wherever water flow decelerates, such as abrupt changes in the shape or dimension of pipes, divergent or low slope sectors, etc. The accumulation of these in-sewer deposits reduces conveyance efficiency of the sewer network (Crabtree, 1989; Seco, 2014; Veliskova & Sokac, 2019), therefore, they require regular removal as a maintenance practice. The accumulated pollutants in in-sewer deposits can be released into the environment upon the disposal of sewer sediment waste if not properly managed, or they go back to water flow via the resuspension of these sediments during wet weather events. MPs may behave similarly to other pollutants in wastewater during transport through sewer network.

The surrounding environment receive different types of discharges from urban areas. Land and soil act as recipients for solid wastes from wastewater management systems, such as treated sludge and sewer sediments. The freshwater environment, on the other hand, receives various inputs, including WWTP effluent, untreated runoff, untreated wastewater, CSOs and more. Literature has reported the presence of MPs in these discharges (Ziajahromi et al., 2017; H. Chen et al., 2020; Treilles et al., 2021), making the receiving environment become susceptible to plastic pollution. Therefore, further research efforts focusing on the occurrence and fate of MPs in sewer management systems are required, in order to mitigate the amount of MPs release to the environment and address plastic pollution issue.

1.2.2. Microplastics in wastewater treatment plants

Investigations into sewer management systems regarding MP pollution have been conducted over the past decade. Since WWTPs are the primary components of sewer systems that employ various treatment technologies, research efforts have focused on these treatment facilities.

1.2.2.1. Efficiency of existing water treatment technologies towards microplastic removal

WWTP was initially designed to treat organic matter and nutrient pollution in domestic sewage. In parallel with the urbanization, WWTP has advanced in both capacity as well as technology to handle more pollutants entering the sewer systems, such as pharmaceuticals and chemical detergents, along with the combined flows from industrial activities in some circumstances. The emergence of MPs in wastewater now poses a challenge to existing WWTPs in ensuring the quality of effluent before discharging it to the environment. Upon arrival at WWTPs, wastewater undergoes a series of purification processes, as illustrated in Figure 1-4. Firstly, water passes through screenings to remove bulky objects such as bottles, cans and leaves. Water then undergoes grit-grease removal, where sand and heavy solids settle to the bottom of the basin, while fatty compounds rise to the surface with the support of air flotation and are later recovered through skimming. This stage aims to prevent damage to downstream mechanical equipment in the system and ensure a cleaner inflow for further treatment.

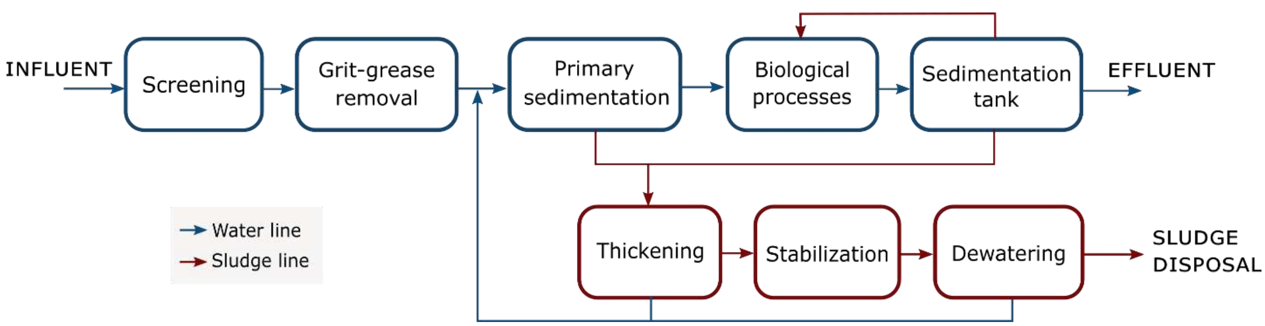

Figure 1-4: Diagram flow of a standard WWTP

After preliminary treatment, the water proceeds to primary sedimentation, where suspended matter is removed, forming primary sludge. The primary effluent then undergoes biological secondary treatment, where bacteria break down organic material and remove carbon, nitrogen and phosphates, which they use for their living activities. An air inflow is necessary for this step, allowing the production of bacterial biomass, which forms secondary sludge that is later separated from the water. Tertiary treatment is optional and serves to further remove nutrients and remaining impurities in the water. Depending on the design of the WWTPs, the hydraulic circulation may vary, including often recirculation of excess sludge and water from sludge-line treatment. Primary and secondary sludge are subject to sludge treatment processes, including thickening, stabilization and dewatering before disposal.

The detection of MPs in treated wastewater was reported in the last decade (Talvitie et al., 2015; Estahbanati & Fahrenfeld, 2016). This has spurred investigation to elucidate MP removal efficiency of existing technologies and the mechanism involved (Carr et al., 2016; Talvitie et al., 2017b; Ziajahromi et al., 2017; Bayo et al., 2020). Most findings showed that the full-scale of a conventional WWTP which combines physical, chemical and/or biological treatments can remove up to 96–99 % of MPs in raw wastewater (Gies et al., 2018; Lares et al., 2018; Blair et al., 2019); in which, 40–90 % of them can be retained during preliminary and primary treatment. Screenings does not provide considerable removal due to large mesh sizes. However, a part of MPs could get removed owing to their attachment to larger items. Since fibers are likely to get through screens because of their small diameter, screening works more effectively for plastic fragments (Ziajahromi et al., 2017). Grit and grease removal, as the subsequent process, is more efficient. The removal mechanism relies on the density of plastic particles where they are either settled or floated (Kurt et al., 2022). Particles with a density lower than wastewater like PE, expanded polystyrene (EPS) and PP tends to stay on water surface and be skimmed off with other floating materials (e.g., fat, grease and oil). PE microbeads were found dominant in grease waste in Murphy et al. (2016). The process is specifically improved with the help of flotation (Pramanik et al., 2021). In the meantime, plastic with density greater than wastewater might also settle and then get removed with sludge (Lares et al., 2018). Indeed, polyethylene terephthalate (PET, 1.38 g/cm^3) was found more abundant than PE and PP (0.90 - 0.94 g/cm^3) in primary sludge (Ziajahromi et al., 2021). Thus, at this stage of treatment, the physical characteristics of MPs (density, size and shape) play a crucial role in the removal efficiency (Reddy & Nair, 2022).

At secondary treatment, the removal is facilitated via adsorption and/or aggregation of MPs with solid flocs during biological processes, followed by sludge separation. In addition, biofouling which occurs with the presence of microorganisms may change the density of MPs, thereby increasing their settling ability during sedimentation (Kurt et al., 2022). In the literature, the highest removal efficiency of MPs was reported for the application of membrane bioreactor

(MBR) technology which combines biological processes and filtration (Table 1-1). However, it is important to note that solid flocs are often unstable and may release attached MPs back into the water phase (Carr et al., 2016). Furthermore, MPs can re-enter the wastewater treatment systems along with recycled activated sludge (Magni et al., 2019). According to research by Hidayaturrahman & Lee (2019), 77 % of the remaining particles in primary effluent can be further separated from the water phase during secondary treatment. Thus, although WWTPs are not specifically designed to address MPs, existing technologies are capable of removing the majority of them from wastewater.

Table 1-1: Performance of advanced technologies toward MP removal in WWTPs

Technology	Removal efficiency (RE)	References
Biologically active filter/ biofilter (BAF)	No significant impact on microlitter concentration	Talvitie et al. (2017b)
Disc filter (DF)	RE = 40-98.5 %	Talvitie et al. (2017a)
Rapid sand filtration (RSF)	RE = 97 %	
Dissolved air flotation (DAF)	RE = 95 %	
Membrane bioreactor (MBR)	RE = 99.9 % producing the final effluent with the lowest MP concentration	
Pilot-scale membrane bioreactor (consist of an aerobic and anaerobic a tank, and a submerged MBR unit with pore size 0.4 µm)	RE = 99.4 % higher than conventional activated sludge with RE = 98.3 %	Lares et al. (2018)
Ozone	RE = 89.9 %	Hidayaturrahman and Lee (2019)
Membrane disc filter (MDF)	RE = 79.4 %	
Rapid sand filtration	RE = 73.8 %	
Coagulation with Al-based coagulant	RE = 47.1 - 53.8 %	
Coagulation/flocculation (C/F) with ferric chloride ($FeCl_3$) polyaluminum chloride (PAC) cationic polyamine (CP)	RE of C/F + $FeCl_3$ = 99.4 % RE of C/F + PAC = 98.2 % RE of C/F + CP = 65 %	Rajala et al. (2020)
Sand filter	RE = 50 %	Magni et al. (2019)

To improve the quality of treated wastewater, many WWTPs have employed tertiary treatment or supplementary techniques. Table 1-1 provides an overview of the performance of several advanced techniques for removing MPs from secondary effluents. Membranes with small size cut-off showed the highest efficiency in MP removal. Ziajahromi et al. (2017) observed a reduction up to 90 % of MPs in primary effluent after reverse osmosis. However, this technique is impractical and cost-effective for a large scale treatment. Moreover, backwashing water from cleaning membranes contains MPs and needs to be treated properly. the combination of coagulation and flocculation with chemical aids is also well-known to remove colloidal particles remaining in wastewater after previous treatment steps. When coagulants such as iron (Fe) and aluminum (Al) salts are added, they neutralize the surface charges of suspended solids, disrupting their stability, which is maintained through electrostatic repulsion. As these particles approach each other due to Brownian motion and mechanical agitation, they form solid flocs through van der Waals forces, ultimately becoming part of the sludge blanket. This process effectively traps other suspended solid particles, including MPs. Rajala et al. (2020) assessed the performance of three commonly-used coagulants in enhancing MP removal during tertiary treatment. The results demonstrated that up to 99 % of spiked particles, specifically spherical PS particles with diameters of 1 µm and 6.3 µm, were separated from the water phase during the treatment, whereas settling alone could not remove them. Among the coagulants tested, ferric chloride and polyaluminum chloride were found to be more efficient than polyamine.

Besides, the RE of MPs in WWTPs is reported to vary for different size ranges. Magni et al. (2019) showed the decline in RE with decreasing size of the particles, e.g., 94 % for 5–0.05 mm; 77 % for 0.5–0.1 mm MPs and 65 % for 0.1–0.01 mm MPs. Similarly, the highest efficiency was obtained for larger-sized particles of 0.5–1.0 mm in Hu et al. (2022). Moreover, particles found in secondary effluent were in the range of 25–104 µm (Edo et al., 2019). Other studies also emphasized the abundance of small-sized particles remaining in treated wastewater (Mintenig et al., 2017; Talvitie et al., 2017b; Simon et al., 2018). In addition, during treatment processes in WWTPs, MPs have chance to break down into NPs due to mechanical actions (Enfrin et al., 2020; Pramanik et al., 2021), reducing the average size of remaining particles as well as the total removal efficiency.

1.2.2.2. Solutions to reduce microplastics in treated wastewater

Although a decent amount of MPs is removed from the water inlet with existing treatment processes, the outlet still contains small-sized particles, including those at the nanoscale. Given a large discharge volume, it acts as an important point source. Because of this, recent times have recorded the development of new technologies as well as the upgrade of traditional methods in order to reduce further MP contamination, especially in treated wastewater. These can be roughly categorized into two main groups: recovery methods, which try to capture plastic particle from wastewater, and degradation methods, which try to transform polymers into more easily-degradable products. Recovery methods consist of filtration, coagulation, phoretic interaction, magnetic and electrostatic separation, while degradation methods include biodegradation, electrochemical and photocatalytic degradation. A summary on these innovative technologies is provided in Table 1-2. More information can be found in the book chapter "*Microplastic and nanoplastic removal efficiency with current and innovative water technologies*" (*Clean Water*, n.d.).

Table 1-2: Innovative technologies for removing MPs and NPs in water phase

Type of treatment		Technology	Main principle	Experiment	State of development	Pros (+)/ Cons (-)	Results	Reference
Recovery	Coagulation/flocculation	Separation using electrocoagulation (EC)	Hydroxide coagulants produced by EC can destabilize surface charges of suspended solids in aqueous media, enabling them to agglomerate and trap plastic particles	Application of aluminum-based EC to remove PS microbeads (300-355 µm) in wastewater analogue	Lab-scale	(+) High RE over a wide range of pH values (pH 3-10), independent of Cl– and HOCl species and current density (-) Chemical costs affect operation costs	RE of 89-100 % with the highest value observed at pH 7.5	Perren et al. (2018)
		Separation using sol-gel induced agglomeration	Bio-inspired pre-organized hybrid silica gel-based precursors once introduced into aquatic systems aid of plastic particles to agglomerate and get removed through filtration processes	Extraction of ultra-high molecular weight PE pellet (~100 µm) using different synthesized precursors	Lab-scale	(+) Agglomeration occurs independently of type, size, and amount of the trace substance concentration, and the external influences (pH, temperature, pressure)	Volume of agglomerate formed through sol-gel process is 666 times larger than that of the original particle	Herbort et al. (2018)
		Separation using plant-derived tannic acid	Particles are coated with chitosan and tannic acid to form phenolic surface. Then, metal ions (e.g., Fe3+) in aqueous media activate the coagulation of coated particles through metal-phenolic coordinate bonds	Removal of modified PS and PE beads (0.5–125 µm) through coagulation - filtration process	Lab-scale	(+) The method is highly reproducible and accurate. The efficiency is stable with different water conditions (e.g., pH, inorganic ions and natural organic material concentration)	RE of 96-99 % with pH 6-8, more efficient than conventional coagulation using Fe- and Al-salts	Park et al. (2021)
		Separation using composite metal calcium-aluminum	Particles are attached to flocculants and get removed during sedimentation	Removal of PS NPs (100 nm) through flocculation-	Lab-scale	NA	80 % of particles settled down with pH>7	Z. Chen et al. (2020a)

		(Ca/ Al) ions as flocculants		sedimentation process				
	Magnetic extraction	Separation using magnetic extraction	Plastic particles bound with hydrophobic Fe nanoparticles are recovered under magnetic effect	Removal of MPs in three different sizes, made of different polymer types in artificial seawater, freshwater analogue and sediment	Lab-scale	(+) The method is efficient for a wide size range, especially for MPs <20 µm (-) Magnetic extraction might cause fragmentation of MPs. RE depends on surface area to volume ratio of targeted particles and binding ability of nanoparticles. The method is more suitable for clean samples	92 % of PE and PS beads (10–20 µm) and 93 % of MPs >1 mm (PE, PET, PS, PU, PVC and PP) from seawater 84 % and 78 % of MPs 200 µm – 1 mm (PE, PS, PU, PVC and PP) from freshwater and sediments, respectively	Grbic et al. (2020)
		Separation using Magnetic Polyoxometalate-Supported Ionic Liquid Phases (magPOM-SILPs)	Magnetic nanoparticles coated with viscous POM-IL (magPOM-SILPs) attach to plastic particles, enabling them to be magnetically recovered	Removal of PS beads (1 and 10 µm) using magPOM-SILP	Lab-scale	(+) Using magPOM-SILPs can simultaneously remove multiple contaminants. The method can treat larger volumes of water than classical filtration	Over 90 % of PS beads are removed from spiked solutions	Misra et al. (2020)
		Separation using magnetic carbon nanotubes	Magnetic carbon nanotubes (M-CNTs) absorb on plastic particles, leading to their removal from aqueous media using permanent magnets	Removal of MPs (PE, PET and PA; 48 µm) using magPOM-SILP	Lab-scale	(+) M-CNTs can be recycled up to 4 times by thermal treatment. Removal efficiency is independent of COD, NH_4^+ and $PO4^{3-}$ in media, making this method applicable for wastewater treatment	100 % of added MPs were separated from the testing solution	Tang et al. (2021)

		Separation using core-shell supermagnetic iron oxide nanoparticles (SPIONs)	SPIONs adsorb to the plastic particle surfaces through attractive electrostatic and van der Waals interaction, then adhere them to larger aggregates that can later be magnetically collected	Removal of nano-size particles of three different polymers in different testing solutions (melamine resin, PS and PMMA)	Lab-scale	(+) SPIONs have moderate to non-toxic effects. Organic alkyl chain structures of SPION can attract both organic and inorganic particles	Efficient for NPs of different polymer types, different chemical structure and different water conditions	Sarcletti et al. (2021)
	Filtration	Separation using metal-organic framework-based foams	Plastic particles are captured by synthesized materials possessing interpenetrated pore structure	Application of a series of Zr-MOFs based foam materials for purifying simulated nanoparticle suspension (diameter ~260 nm) in water or seawater conditions	Lab-scale	(+) ZrMOF based foam materials can be recycled up to 10 cycles and produced in large-scale	RE up to 95 %	Y. Chen et al. (2020)
		Filtration using biochar	Plastic particles are immobilized by the microstructure of biochar	Removal of microplastic spheres (10 µm in diameter) by biochar	Lab-scale	(+) Low cost	RE > 95 % higher than using sand (60-80%)	Z. Wang et al. (2020)
			Sorption of NPs in solution on biochar generated from sugarcane bagasse-based	Removal of negatively charged polystyrene-based latex beads (<500 nm) by biochar synthesized at three different temperature	Lab-scale	(+) Fast and low cost (-) Efficiency depends on pH, organic matter and other competitive ions in solution	99 % of NPs removed with biochar pyrolyzed at 750 °C	Ganie et al. (2021)
		Biofiltration Filtration using biofilter	Plastic particles are retained by filter materials	Removal of MPs in the effluent of a WWTP using a biofilter made of stone wool, Filtralite	Pilot-scale implemented in WWTP	NA	Reduce 79-89 % MPs remaining in the effluent, in which remove 100 % MP	Liu et al. (2020)

				CLEAN HR 3-6 and granite gravel			>100 µm. Less polymer types are detected with no more PU, PS and acrylic	
		Dynamic membrane (DM) filtration	DM layer formed on the supporting mesh can further remove plastic particles during filtration process	Efficiency of DM system to remove particles ranging from 1.65 µm to 516 µm in synthetic wastewater	Lab-scale	(-) DM formation process was strongly affected by the concentration of micro-particles in the influent wastewater	The effluent turbidity <1 NTU after 20 min of filtration	L. Li et al. (2018)
		Filtration using a charged filter	Charged particles are trapped selectively with a novel 3D printed moving bed water filter	Filter was employed to treat polycarbonate contaminated synthetic water	Lab-scale	(+) low cost, energy-efficient; work for different water sources in different conditions	Separation of the NPs from water phase was enhanced	Gupta et al. (2021)
		Membrane filtration	NPs are retained using modified membranes	Removal of PS NPs (50, 100 and 500 nm) with three different modified membranes at low pressure	Lab-scale	(+) facile and scalable protocol; reduce energy consumption and remove bacteria	RE > 99 %	R. Wang et al. (2020)
	Phoretic interaction	Separation using acoustic focusing	Acoustophoretic forces act on particles with a positive or negative acoustic contrast factor, moving them toward the center or the walls of the microchannel for separation	Separation of PS MPs in suspension and MP fibers from the effluent of laundry machine using a bulk acoustic wave	Lab-scale	(-) This method may not be useful for relatively light and soft polymers such as low-density PE	BAW device was able to collect almost all MPs regardless of their shape	Akiyama et al. (2020)
		Separation using photocatalytic Au@Ni@TiO_2-based micromotors	Plastic particles are collected through phoretic interaction induced by photocatalytic activity	Removal of PS particles and MPs extracted from personal care products and open	Lab-scale	(+) The efficiency of light-driven micromotors is not limited to the certain materials and shapes	RE of 67-77 %	Wang et al. (2019)

			of synthesized micromotors	waters using photocatalytic TiO_2-based micromotors (Au@mag@TiO_2, mag = Ni, Fe)		(-) In spite of being coated with a thin Au layer, Ni might cause harm for the environment		
	Electrostatic separation (unpublished work)							
	Adsorption	Separation using synthesized material	NPs are removed from water phase via adsorption on CuNi carbon material (CuNi@C)	Removal of PS nanoplastics (~100 nm) using CuNi@C	Lab-scale	(+) CuNi@C can be reused at least 4 times (-) RE depends on pH of solution	RE = 98 % at concentration of CuNi@C 0.3 g/L Higher RE under acidic condition than alkaline; physical adsorption and monolayer coverage as the main mechanisms of the process	G. Zhou et al. (2022)
			Removal of NPs via adsorption on a new magnetic material	Removal of PS NPs from solution using Fe-modified fly ash material (NMA)	Lab-scale	(+) NMA can be synthesized with a simple method at low cost; they can be reused up to 4 times (-) the process depends on pH, interfering ions and ion strength, temperature	83.1 mg/g at room temperature	Zhao et al. (2022)
Degradation	Abiotic degradation	Degradation using anodic oxidation	Hydroxyls (*OH) generated by direct and indirect electrochemical process break the polymeric bonds and degrade plastic particles	Degrade PS microbeads (~26 µm) in synthetic suspension using electrooxidation process	Lab-scale	(+) Particles are not broken into smaller fragments, but transformed directly into the gaseous products, e.g., CO_2	High degradation efficiency of 89 %	Kiendrebeogo et al. (2021)

						(-) Energy cost was dominant in the value of total operating cost		
		Photocatalytic degradation using visible light	Reactive oxygen species such as hydroxyl (*OH) and superoxide (O_2^-) produced during photocatalysis process cause chain scission of polymer	Degrade PP spherical particles using visible light irradiation of zinc oxide nanorods (ZnO NRs) coated onto glass fibers substrates in a flow through system through photocatalytic reactor	Lab-scale	(+) This method is considered as environmental friendly solution via using sunlight as energy source and producing harmless by-products (-) This method requires long time reaction	Average volume particles reduces about 65 % after 456 h of exposure. By-products generated from the reaction have low toxicity effects on human and aquatic environment	Uheida et al. (2021)
		Photocatalytic degradation using visible light	Reactive oxygen species such as hydroxyl (*OH) and superoxide (O_2^-) produced during photocatalysis process cause chain scission of polymer	Degrade of PE plastics using the sunlight irradiation with polypyrrole/TiO_2 (PPy/TiO_2) nanocomposite as photocatalyst	Lab-scale	NA	Weight loss up to 54 % is reported after 240 h of exposure with the formation of cavities on PE plastic surface	Li et al. (2010)
	Biotic degradation	Enzymatic degradation	Using a number of microbial polyester hydrolases to degrade PET materials	Degrade PET films with a polyester hydrolase in an ultrafiltration membrane reactor to minimize the product inhibition of the enzyme	Lab-scale	(-) The limited activity of the polyester hydrolases requires long reaction times and their susceptibility against inhibition by intermediate hydrolysis products slows down degradation of PET materials	a weight loss of the PET films of only 6 % was achieved after a reaction time of 24 h	Barth et al. (2015)

NA: Information is not available

Plastic pollution challenges existing water treatment systems, especially with the abundance of MPs in wastewater. Efforts have been dedicated to improving the remediation of MPs in water systems. Different technologies, in which recovery methods accounted for a greater part compared to degradation methods, were studied. The application of intermediate products showed high efficiency in separating MP-NPs from the water phase. However, their fate after usage as well as their potential interaction with other co-existing pollutants remains unclear. Similarly, the impacts of by-products released from electrochemical and photocatalytic reactions on the environment need to be assessed. In addition, fragmentation of polymers during the process may increase small-sized particles, in turn worsening the issues. Bioremediation methods can be an effective and reasonable options, representing green solutions in dealing with plastic pollution. None of these innovative technologies perform as a single comprehensive solution. Instead, they were developed considering the integration with existing treatment facilities. For instance, coagulation techniques require a follow-up separation process, or magnetic extraction focuses on the small-sized fraction remaining in treated wastewater. All these technologies are either tested at a lab scale, or installed at a pilot scale in a WWTP. Thus, additional research is required to evaluate their efficiency and feasibility when working on a larger scale. Moreover, different criteria need to be considered when implementing the current systems with the new technologies, such as site location, land availability, existing plant design, influent load and cost (Conley et al., 2019). Furthermore, methods that are simple but effective in removing MPs (e.g., primary treatment) should be targeted (Reddy & Nair, 2022).

1.2.2.3. Research on microplastics in sewage sludge

Research on the occurrence and fate of MPs within WWTPs has determined a large part of these particles, once separated from the wastewater, end up in sewage sludge (Jiang et al., 2020). According to Carr et al. (2016), besides solids skimming in primary treatment, MPs were mainly transferred from wastewater to sludge through settling processes. In particular, the important role of the aeration tank in this transfer was highlighted in Hongprasith et al. (2020). Similarly, Talvitie et al. (2017b) reported that most of MPs arriving at WWTPs were eventually concentrated in sludge, with only 0.1 % remaining in the effluent. Other studies also documented that over 90 % of MPs are retained in sewage sludge (Zhang et al., 2020; Zhang & Chen, 2020). Since the final treated sludge is primarily managed through landfilling and soil application, an emission of MPs into the environment via sludge disposal is expected. Moreover, the presence of MPs in agricultural land, especially with higher levels in sludge-treated areas compared to non-treated one, has been reported (Corradini et al., 2019; Van Den Berg et al., 2020). These findings, therefore, underscored sludge as a source of MPs in soil environment. In this context, research efforts have been dedicated to quantifying the level of MPs in sewage sludge and understanding their fate through sludge-line treatment. These studies aim to contribute to the assessment of the WWTPs' overall efficiency toward MP removal and the potential emissions of MPs from these facilities into the environment.

Table 1-3 summarizes the concentrations of MPs in different sludge types from studies conducted worldwide, with values ranging from 8×10^2 to 2×10^6 particle/kg dry weight (dw). The majority of these studies reported average concentration levels in the range of 10^4 and 10^5 particle/kg dw. The substantial variation in the obtained results can be attributed to a multitude of factors, including socioeconomic status, population density, plastic consumption habits, weather conditions, and treatment technologies employed at the studied facilities. Additionally, the inconsistencies in the methodology applied for sample collection and analysis may contribute to these variations. Despite this, it is estimated that up to thousands of billions of MPs can be released into the environment annually via sludge disposal. According to Kedzierski et al. (2023), about 1.5 to 6.6 million tons of MPs would be present in soils on a global scale. The rapid accumulation of these particles in terrestrial ecosystems is expected due to the resistance nature of plastics.

Studies have also reported the composition and morphology of MPs found in sewage sludge, including size, shape and color. This data aims to provide insights into the sources and degradation status of these particles. Prevalent polymers detected in sewage sludge are PE, PP, PA, PEST (including PET) and PS (El Hayany et al., 2020; Alavian Petroody et al., 2021; Yuan et al., 2022), reflecting the origins of wastewater and the daily plastic consumption habits of society. For example, PP and PE are commonly found in personal care products, food packaging and shopping bag, while PET is associated with water bottles and beverage containers. PEST and PA are often found in synthetic textiles, and PS is used in food containers and insulation materials.

Most of studies have documented the predominance of MPs smaller than 500 µm (Mintenig et al., 2017; El Hayany et al., 2020; Ren et al., 2020), with particles lower than 300 µm accounting for the majority (Lee & Kim, 2018; Edo et al., 2019; Liu et al., 2019). According to the authors, small particles may more easily adhere to the surface of organic suspended solids in water, thereby becoming trapped by solid flocs in biological treatment processes. Meanwhile, these findings reflect the importance of screening and skimming during primary treatment to remove large-sized MPs, highlighting the potential of another source to the environment if primary treatment's waste is improperly handled (Ziajahromi et al., 2021). This also suggests the need to upgrade primary treatment technologies to enhance MPs removal (Pittura et al., 2020).

Among various shapes of MPs, fibers have been found dominant in many studies (Gies et al., 2018; Lares et al., 2018; X. Li et al., 2018; Edo et al., 2019). This phenomenon is commonly attributed to extensive production of synthetic textiles and laundry activities. Differently, some other reported the dominance of fragments in sewage sludge (Lee & Kim, 2018; Pittura et al., 2020; Ren et al., 2020; Yuan et al., 2022). These fragments can stem from industrial production processes and the breakdown of plastic products. According to El Hayany et al. (2022), MPs' shape may depend on the sources of MPs in wastewater, as well as lifestyle and consumption habit differences from country to country and region to region. While size and shape of sludge-based MPs are frequently reported, only a limited number of studies documented color of these particles.

As a byproduct of water treatment processes, sewage sludge typically undergoes a series of treatment processes, including thickening, stabilization and dewatering before disposal. While thickening and dewatering aim at diminishing the volume and weight of the sludge, stabilization aims to control odors and pathogen content in the sludge. Anaerobic digestion is one of the most commonly used technologies for sludge stabilization, which produces methane or biogas during the treatment, serving as an energy source for WWTP operations. Advanced treatment, like thermal conditioning/thermal hydrolysis and thermal drying, are sometimes applied to further remove pathogen and water content in the sludge. Thus, it can be seen that once MPs are transferred from water phase into sludge, they undergo various processing steps within the sludge-line treatment. In every treatment step, MPs within the sewage sludge can be altered. Therefore, the recent efforts have been dedicated to understanding the fate of MPs in various treatment processes. These investigations aim to provide insights into the impact of the treatments on MP particles and their efficiency in removing plastic particles from the matrix. Different mechanical techniques are employed during thickening and dewatering steps to remove water from sludge. Although no impacts of these techniques on MP particles have been reported, it has been documented that a part of MPs in sewage sludge can be released back into the water phase during these treatments (Talvitie et al., 2017b; Alavian Petroody et al., 2021; Salmi et al., 2021; Bretas Alvim et al., 2022). Owing to the distinct operating principles of each technique, varying concentrations of MPs, with different polymer compositions, can be found in reject water (X. Li et al., 2018). For example, centrifugation relies on the density difference between sludge and water, which may result in the release of low-density MPs back into the water. Thus, the circulation of MPs via reject water needs to be considered when evaluating removal efficiency of the technology. Concerning the stabilization step, the abundance of MPs in sewage sludge remained unchanged after anaerobic digestion, with little changes observed on particle surface (Li et al., 2022). This can be associated with the non-biodegradable nature of most plastics. Previous studies by Selke et al. (2015) and Gómez & Michel (2013) highlighted the resistance to degradation in conventional plastics (i.e., PP and PE), even when modified with additives to enhance biodegradability. Another biological process involved aerobic composting, which can be applied to sludge before its use in soil application. El Hayany et al. (2020), monitoring MPs behavior in lagooning sludge during the composting process, reported a decrease in particle size while the overall particle number remained relatively unchanged. In another study, more than 40 % of MPs from sewage sludge was reduced after hyper-thermophilic composting at temperatures ranging from 80 °C to 90 °C for 45 days (Z. Chen et al., 2020b). According to the authors, this reduction might result from the acceleration of hyper-thermophilic bacteria to MP biodegradation. However, it should be noted that the higher temperatures may lead to the fragmentation of MP particles, thereby causing them to fall below the size detection limit. In contrast, Edo et al. (2019) documented insignificant effects on MPs in sludge even after thermal processing at 300 °C. Li et al. (2022) also observed the slight decrease in MP concentration after thermal drying. However, Mahon et al. (2017) reported obvious cracking of PE MPs after thermal drying. In particular, Mahon et al. (2017) found that the abundance of MPs significantly increased after thermal hydrolysis treatment, while Li et al. (2022) observed deep cracks after the same treatment. This phenomenon is attributed to the combined effects of increased temperature and mechanical mixing, particularly elevated pressure (Mahon et al., 2017; Weithmann et al., 2018). In summary, research in this topic remains limited, and the findings are often contractive. Further research is therefore needed to gain a comprehensive understanding of the behavior of MPs in various sludge treatment processes.

Table 1-3: MP levels in sewage sludge in different countries. The data is shown in min-max value or average (± standard deviation) value

Location of WWTP	PEq (×10^3 inhabitant)	Sludge type	Concentration level (×10^3 particle/kg dw)	Size range (µm)	Reference
Spain	NA	Digested sludge	2033 ± 603	150-5000	Bretas Alvim et al. (2022)
		Dewatered sludge	1567 ± 199		
UK	1,580	Raw sludge	107.5	50-5000	Harley-Nyang et al. (2022)
		Thickened sludge	50.2		
		Digested sludge	180.7		
		Digested sludge (2nd)	286.5		
		Sludge cake	97.2		
		Pre-limed sludge	74.7		
		Limed sludge	37.7		
China	250	Dewatered sludge	44.4 - 750.0	25-5000	Wei et al. (2022)
China	NA	Raw sludge	22.96 - 51.41	25-5000	Yuan et al. (2022)
		Sludge cake	6.32 - 13.04		
Iran	105.8	Primary sludge	214 ± 16	37-5000	Alavian Petroody et al. (2021)
		Secondary sludge	206 ± 34		
		Thickened sludge	200 ± 13		
		Digested sludge	238 ± 31		
		Dewatered sludge	129 ± 17		
UK	38-320	Sludge cake	301 - 10,380	25-178	Horton et al. (2021)
Australia	234-700	Primary sludge	15.9 - 45.7	>25-5000	Ziajahromi et al. (2021)

		Secondary sludge	37.8 - 46.1		
		Dewatered sludge	48.5 - 56.5		
Morocco	NA	Raw sludge	40.5±11.9	100-5000	El Hayany et al. (2020)
		Dewatered sludge	36.0±9.7		
China	3,100	Returned activated sludge	36.3 ± 5.7	20-5000	Jiang et al. (2020)
		Sludge filter cake	46.3 ± 6.2		
Italy	80	Primary sludge	1.67	30-5000	Pittura et al. (2020)
		Secondary sludge	5.3		
		Dewatered sludge	4.74		
Australia	190	Excess activated sludge	7.91 ± 0.44[a]	>1.5	Raju et al. (2020)
China	100	Dewatered sludge	220	8-1000	Ren et al. (2020)
China	NA	Dewatered sludge	2.9 - 5.3 4.0±1.4	50-5000	Xu et al. (2020)
Spain	300	Raw sludge	133±59	25-5000	Edo et al. (2019)
		Treated sludge	101±19		
China	NA	Dewatered sludge	240.3±31.4	60-4200	Liu et al. (2019)
China	NA	Excess sludge (membrane tank)	1.6[a]	25-5000	Lv et al. (2019)
		Excess sludge (secondary setting tank)	0.7[a]		
Canada	1,300	Primary sludge	14.9±6.3	>1	Gies et al. (2018)
		Secondary sludge	4.4±2.9		

Finland	NA	Digested sludge	170.9±28.7	250-5000	Lares et al. (2018)
		Activated sludge	23.0±4.2		
		MBR sludge	27.3±4.7		
Korea	67.7	Sludge cake (AAO process)	14.9	106-5000	Lee & Kim (2018)
	235.7	Sludge cake (SBR process)	9.6		
	245.2	Sludge cake (media process)	13.1		
China	51.9-1,370	Dewatered sludge	1.6 - 56.4	37-5000	X. Li et al. (2018)
Denmark	NA	Digested sludge	169,000	20-500	Vollertsen & Hansen (2017)
Ireland	6.5-2,400	Anaerobic digested sludge			
		Treated sludge (thermal drying)	4.2 - 15.4	45-5000	Mahon et al. (2017)
		Treated sludge (lime stabilization)			
Germany	11-210	Dewatered sludge	1 - 24	<500	Mintenig et al. (2017)
Finland	800	Returned activated sludge	76.3±4.3	20-5000	Talvitie et al. (2017b)
		Treated sludge	186.7±26.0		
USA	NA	Treated sludge	1	45-400	Carr et al. (2016)
		Returned activated sludge	50[a]		
Scotland	650	Sludge cake	0.8	>65	Murphy et al. (2016)

PEq: population equivalent

[a] Unit: particle/L

NA: no data available

1.2.3. Knowledge gaps

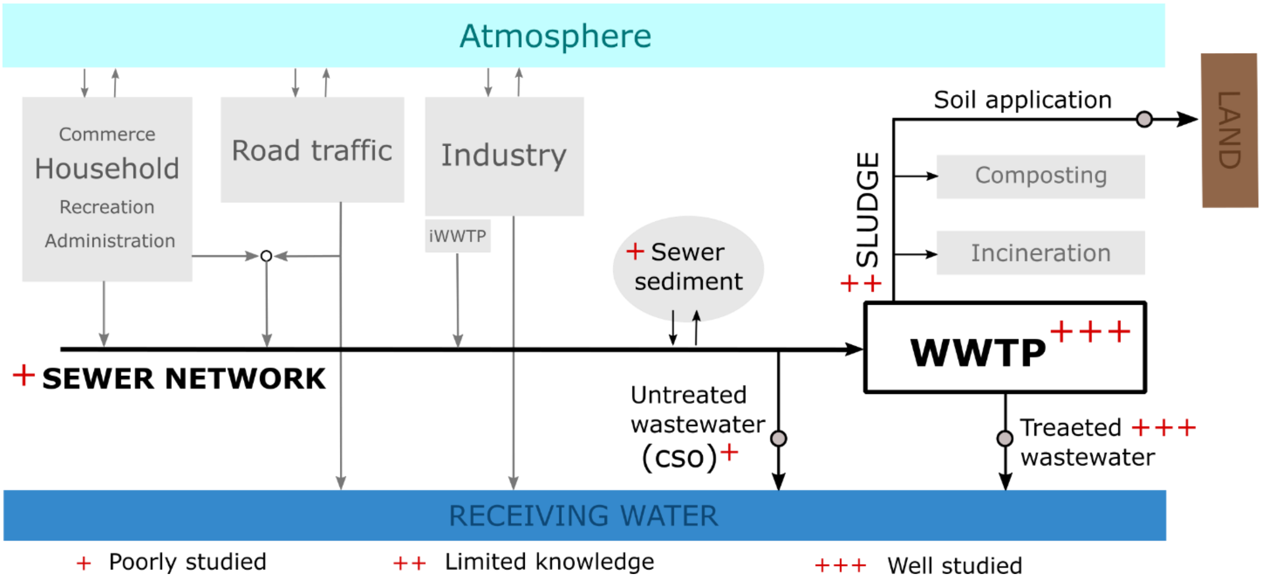

Figure 1-5: Scientific knowledge on the occurrence and fate of MPs in various compartments of the wastewater management system. The classification is based on the number of publications on the topic available on Google Scholar

Since MPs are transferred to sludge and remain in the system, the high RE reported in studies of MPs in WWTPs only demonstrates the efficiency of existing technologies in separating MPs from wastewater, but not the role of WWTPs in eliminating MPs from being released into the environment. RE is therefore a relative value based on author's judgements rather than representing an absolute truth. This can be attributed to the lack of a standardized methodology for assessing MP removal efficiency of WWTPs. To comprehensively understand the occurrence and fate of MP within WWTPs, research attention should be directed toward sludge management technologies. Initial studies on sludge-based MPs primarily focused on quantifying contamination level in sewage sludge (Gies et al., 2018; Lares et al., 2018; X. Li et al., 2018), while the effects of sludge treatment on MPs have been investigated more recently (Alavian Petroody et al., 2021; Harley-Nyang et al., 2022; Li et al., 2022). Despite this, research on MPs in the sludge–line treatment is still modest compared to the water treatment line, focusing on diverged topics. Especially, data on the fate of MPs during sludge treatment processes remain scarce. Therefore, further studies are required to gain a better understanding of MPs' occurrence and fate in sludge-line treatment at WWTPs.

Besides, MP occurrence and fate inside the sewer network before reaching treatment facilities has not been studied. Sewer sediments, which form as particulate matter separates from the water phase and settles down during sewage transport, have been proved as a source of pollutants contributing to pollution level in wet weather flows and CSO discharges into receiving water bodies. While travelling inside sewer network, MPs can become integrated with mineral and organic particles in wastewater, then settle down and get trapped in sewer sediments. When these sediments erode, MPs may be released into water flow alongside other pollutants. Therefore, it is imperative to investigate MPs within the sewer network, with particular focus on sewer sediments.

As one of the main untreated discharges which enters nearby water bodies, CSOs are expected to emit MPs from urban area to receiving water. Literature has documented an increase in MP pollution levels in various receiving waters due to CSO discharges. For example, Forrest et al. (2022) reported a sevenfold increase in MP level in watercourses downstream of a combined sewage outfall during one storm event in Ottawa, Ontario, Canada, compared to ambient conditions. Similarly, Rowley et al. (2020) established a strong relationship between MP contamination in the water column of the Thames River and CSO discharges from a nearby wastewater pumping station. While these findings illustrate the contribution role of CSOs to MP contamination level in freshwater environments, they are indirect or remain insufficient to estimate the quantity of MPs discharged via this pathway into the environment. To achieve a more accurate quantitative assessment, direct analysis on CSO samples is required. To the best of our knowledge, only a few studies have followed this approach: Dris (2016) collected water samples from a CSO outfall in Paris during three separate occurrences; while H. Chen et al. (2020) and Sun et al. (2023) analyzed wet weather flow samples collected from pumping station during CSO events. Consequently, there remains a knowledge gap regarding MP contamination in CSOs to date. Therefore, further investigations on the emission of MPs along with CSOs into the environment during wet weather events are essential.

The upcoming chapters will present an investigation focusing on three main objectives mentioned in the introduction. These findings aim to fill existing knowledge gaps in the study of MPs in wastewater management systems, thereby contributing to a comprehensive understanding of MP pathways in the environment.

Chapter 2: Study site

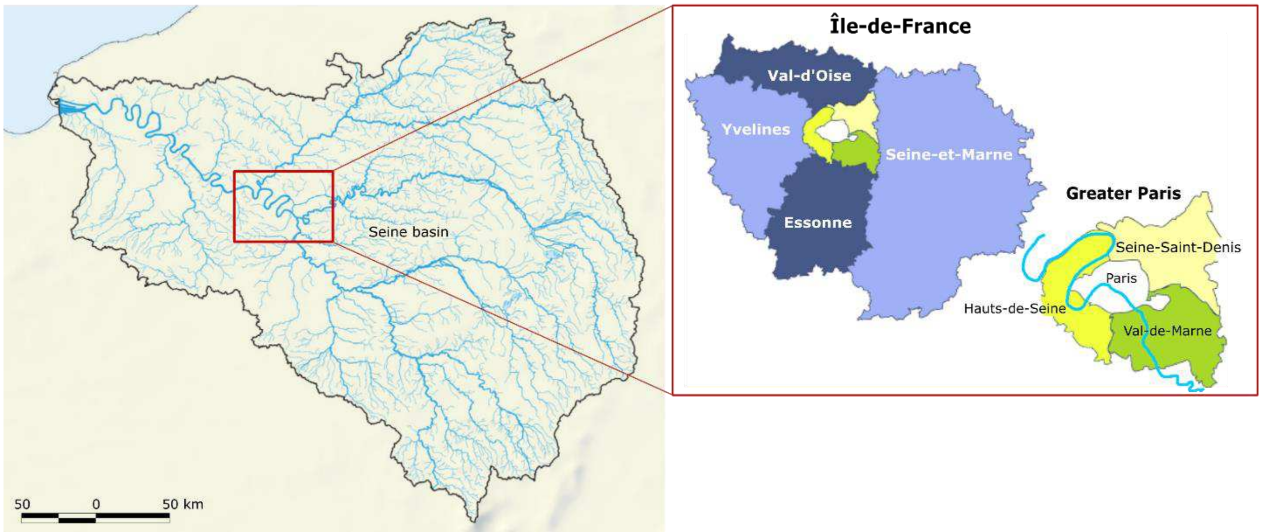

Figure 2-1: Île-de-France embedded in the Seine River catchment

2.1. Île-de-France – Paris megacity – Seine River

Île-de-France, also known as the Paris region, serves as the economic, political, and cultural center of France. This region covers an expansive surface area of 12,000 km², housing approximately 12 million people in 2022. It is divided into eight administrative departments organized into two circles: the inner circle, known as Greater Paris, includes Paris City (the capital of France), Hauts-de-Seine, Seine-Saint-Denis and Val-de-Marne; the outer circle includes Essonne, Seine-et-Marne, Val-d'Oise and Yvelines. Île-de-France has the highest per capita GDP (gross domestic product) among French regions, contributing to about 30 % of the total national GDP in 2019.

Paris megacity encompasses Greater Paris area and a part of the outer circle, covering a quarter of the Île-de-France's total area. This region accommodates about 10.7 million inhabitants from 412 municipalities, as reported by INSEE (National Institute for Statistical and Economic Studies). The Paris megacity is situated within the Seine catchment basin, with the river crossing the Paris City. The Seine River serves not only as one of the primary water sources for the region, but also as the recipient of various urban pollution discharges. The flow rate of the Seine is relatively modest compared to other rivers in France, with an average value of 310 m³/s (measured at Austerlitz bridge inside of Paris City). The Seine has a very low discharge dilution capacity, approximately 1.4 m^3/d/inhabitant, compared to 18 m^3/d/inhabitant for the Rhône in Lyon and 65 m^3/d/inhabitant for the Rhine in Strasbourg. This makes the river become more sensitive to point source pollution (Flipo et al., 2021b). During the low-flow periods in summer, the flow rate can decrease to approximately 80-100 m^3/s. Downstream of Paris, the WWTP effluent discharge, with an average flow of 25 m^3/s, can contribute up to 25-31 % of the downstream river flow. Thus, water management plays a pivotal role in shaping the discharge profile of the Seine River (Flipo et al., 2021a). Deterioration in the quality of the Seine's water was observed from the 1870s until the 1970s, attributed to urban development. Therefore, the Seine River represents a freshwater system profoundly affected by pollutants from urban areas within its catchment.

With its very high population density, human activities in the Paris megacity create numerous pressures on the environment, affecting not only on freshwater ecosystems, but also other environmental compartments, including soil and air. Therefore, this site is highly relevant for investigating the anthropogenic pressures on the surrounding environments, particularly plastic pollution.

2.2. Parisian wastewater management system - SIAAP

Sanitation in the Paris megacity is managed by a multi-stakeholder organization. The Greater Paris Sanitation Authority (SIAAP), established in 1970, serves as the public utility responsible for the transportation and treatment of wastewater in the Paris region. The drainage area covered by SIAAP spans approximately 1,800 km^2, encompassing the Greater Paris area and some surrounding suburbs (Figure 2-2). It serves 284 municipalities with a population of about 9 million inhabitants.

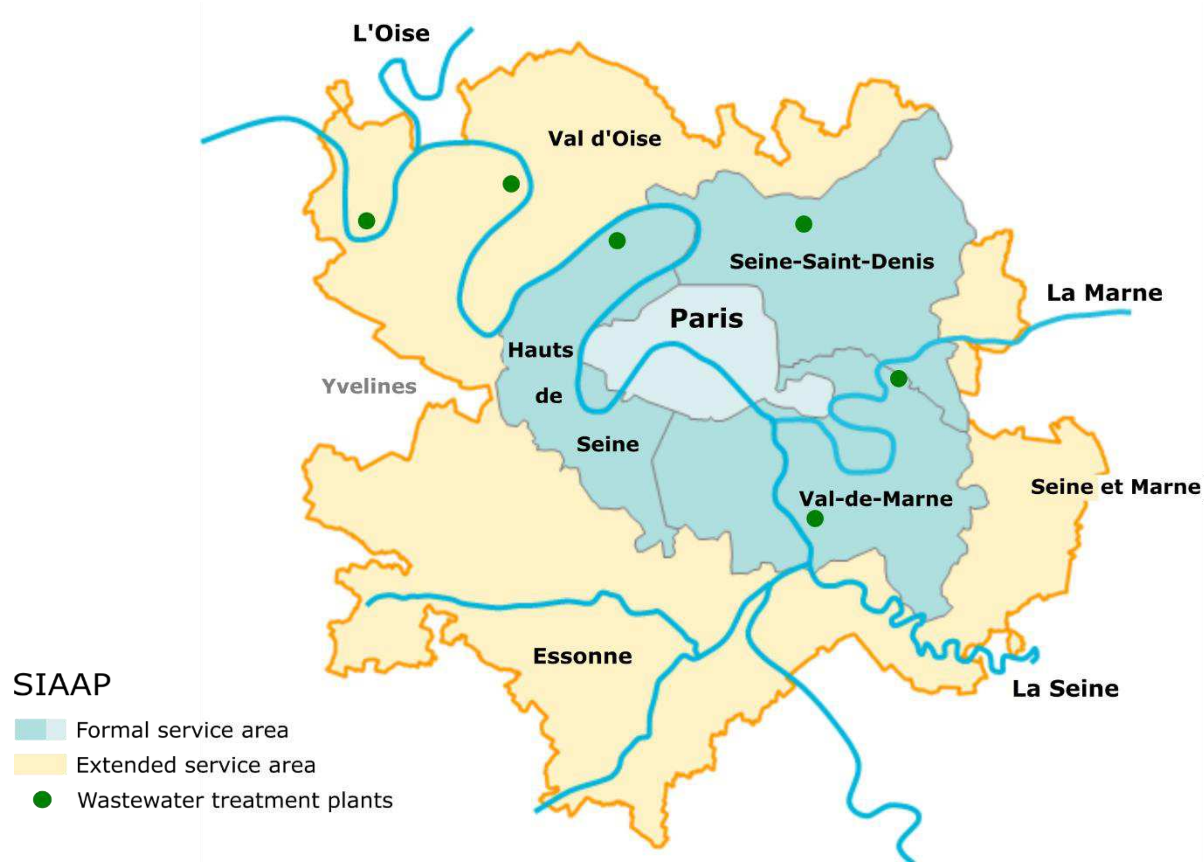

Figure 2-2: Wastewater service area operated by SIAAP within Paris megacity

SIAAP operates an extensive sewer system, which includes six WWTPs and approximately 400 km of main sewer networks. These primary networks are connected with over 15,000 km of municipal and intercommunal pipes. Additionally, the system incorporates basins and tunnels with a storage capacity of approximately 900,000 m³. In dry weather conditions, SIAAP treats about 2.5 million m³ of wastewater daily.

2.2.1. Sewer network

The Parisian wastewater management is organized into different levels, including collection, transport and treatment, with the participation of several operators. Domestic wastewater and stormwater are firstly collected within communities through municipal and intercommunal pipes. Subsequently, wastewater is conveyed through the department's network and then transferred to WWTPs via the main sewers, as illustrated in Figure 2-3. Parisian wastewater management comprises a combined sewer system in its central part and a separate system in the outskirts developed mostly after the Second World War. Paris City itself has a fully combined sewer system with a total line length of 2,100 km. The combined system transports a mixture of domestic wastewater and storm water to treatment facilities. In the separate system, domestic wastewater is directed to WWTPs, while runoff undergoes coarse screening and is discharged into surrounding waters. The quality of wastewater collection and the control of stormwater are determined at the municipal network level.

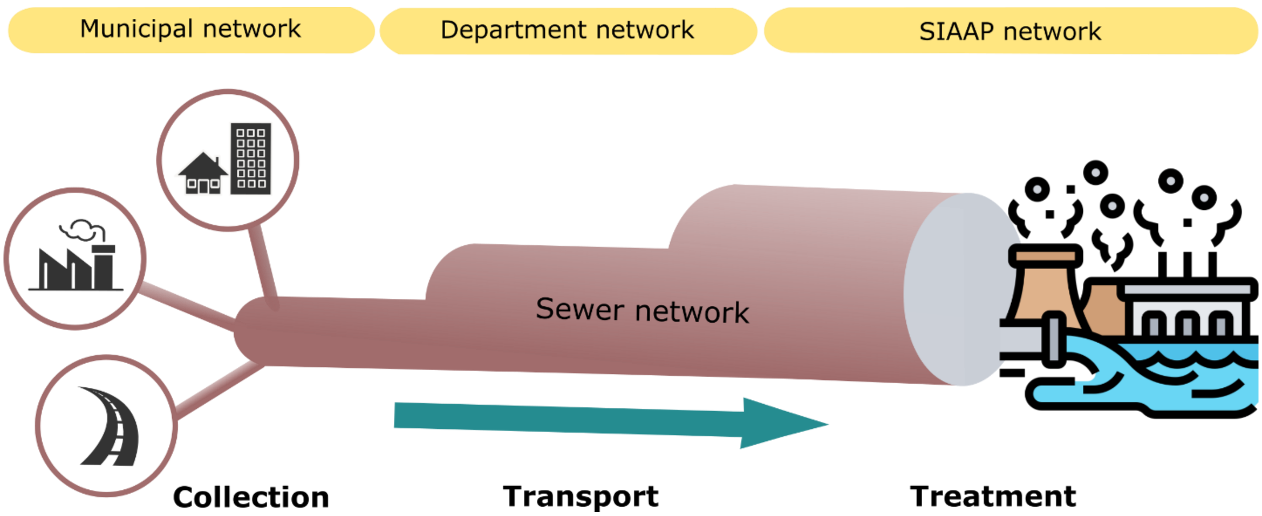

Figure 2-3: Flowchart of the wastewater transport within the Parisian sewage management system

The extensive network system of SIAAP has evolved over time in line with the growing urbanization of the region. It was built at varying depths, ranging from 3 m to 100 m below natural ground level, depending on the topography, with diameters that vary between 2.5 m and 6 m. The sewer network also consists of pumping stations and other accessories, supporting the transport of water flow. Because of the complexity of the separate sewer network, intentional and unintentional cross-connections of wastewater and stormwater sewers occur, known as misconnection. This issue leads to illicit discharges, which either contaminate runoff water by mixing it with sewage, or increase the hydraulic loading on the treatment system downstream. In addition, the main sewers transferring wastewater to WWTPs are oversized, allowing the daily water flow control and the transfer of capacity between WWTPs, a unique feature of the Parisian wastewater management system.

2.2.1.1. Sewer sediments inside the sewer network

The Parisian sewer system experiences the formation and accumulation of sewer deposits inside sewer pipes. This occurs when particulate matter in wastewater separates from the water phase during transport and settles to form bed deposits. It typically happens during dry-weather periods when a flow with high suspended solids concentrations passes through the system. In-sewer deposits also form when water flow decelerates, such as in areas with abrupt changes in the shape or dimension of sewer pipes, divergent or low slope sectors and particularly in the oversized sewer pipes arriving at WWTPs. Thus, the accumulation of in-sewer sediments is temporal and spatial dependent, directly linked to water velocity inside the sewer network (Seco, 2014).

Sewer sediments are a complex and highly heterogeneous aggregate of particulate matter present in wastewater. They can be categorized into three primary types: gross bed sediment (GBS), organic layer and biofilm, based on the nature of their constituent materials (Rocher et al. 2004), as shown in Figure 2-4. While GBS has a high mineral content, the other types contain mainly organic matter. Sewer sediments have been reported as a storage of pollutants, with high concentration levels, inside the sewer network (Ashley & Crabtree, 1992; Crabtree, 1989).

Figure 2-4: Types of sewer sediments inside the sewer network (Rocher et al., 2004)

The accumulation of in-sewer deposits inside sewer pipes reduces the system's conveyance capacity, alters the hydraulic flow regime and leads to several issues, such as surcharging, flooding and premature activation of overflows, and even blockages (Crabtree 1989; Seco 2014; Veliskova & Sokac 2019). Moreover, during wet weather events, the acceleration of flows can resuspend in-sewer deposits, releasing trapped pollutants into the water flow (Gasperi et al., 2010). A large fraction of total suspended solids, metals, chemicals and micropollutants in wet weather flows can be attributed to the erosion of sewer sediments (Chebbo et al., 1995; Ashley et al., 2004; Gasperi et al., 2010). Thus, the presence of these sediments not only complicates the operation of sewer systems, but also has adverse impacts on pollution levels in wet weather flows and the water bodies that receive these discharges. Therefore, it is required to regularly remove sewer sediments inside the sewer network as a part of maintenance practices.

To prevent the excessive buildup of sewer sediments that can impede wastewater transport, sand chambers, acting as sediment traps, were installed throughout the Parisian sewer system. About 100 sand chambers were distributed across the network in Paris City (Rocher et al., 2004). These chambers typically have the same width but are deeper than the sewer. This induces abrupt decrease in the flowrate of wastewater when passing through, thereby allowing particles to detach from water phase and settle down, as depicted in Figure 2-5. Sand chambers are regularly cleaned as part of the system maintenance. The extracted sediments are then sent to specialized treatment centers. In total, 3,000 tons of sewer sediments in wet weight are removed from sand chambers and the network each year (personal communication).

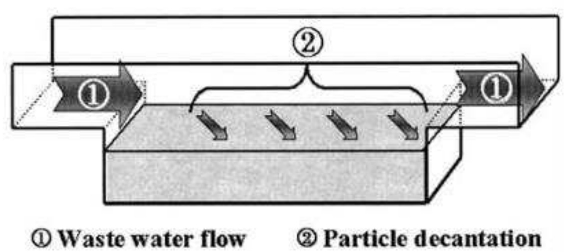

Figure 2-5: Design of sand chamber installed inside the sewer network (Rocher et al., 2004)

In this study, MP analysis was conducted using sewer sediments from sand chambers, as they exhibit similar characteristics to those found throughout the network, as reported in Rocher et al. (2004).

2.2.1.2. Overloading of the system leading to CSOs

During intense rain events, when a substantial volume of runoff enters the wastewater management system, the sewer network may become overwhelmed and unable to deliver all the water to WWTPs. This results in the overflow of excess water through storm spillways into the surrounding environment. Additionally, the capacity of WWTPs may be inadequate to manage the incoming inflow, leading to the discharge of the surplus to protect the treatment systems. This phenomenon is commonly referred to as CSOs, denoting the discharge of sewage into the environment without proper treatment due to the overloading of the combined system.

The impacts of CSOs on receiving waters have been well documented in the literature. For example, Passerat et al. (2011) reported a rapid increase in the flowrate of the Seine River during a storm event, rising from 157 m^3/s up to 367 m^3/s. This demonstrates how a high-volume discharge in a short-time frame can significantly affect the flow of receiving waters. Additionally, turbulence levels rise during these events, leading to increased turbidity and reduced photosynthesis activity of phytoplankton (Matzinger et al., 2012; Riechel et al., 2016). One of the main effects observed in receiving waters during and after CSO events is a deficit in dissolved oxygen (DO). This phenomenon results from the mixing with low-DO wet weather flows and the degradation of organic matter emitted with it (Riechel et al., 2016). Passerat et al. (2011) documented a high load of solid matter, reaching up to 830 mg/L in the discharge into the Seine River during the first 30 minutes of the event before gradually decreasing to 110 mg/L afterward. In addition, CSO discharges carry significant loads of micropollutants into waterbodies (Musolff et al., 2009). For instance, Launay et al. (2016) and Gasperi et al. (2011) detected a variety of organic and hazardous substances in CSO samples, including pharmaceuticals and personal care products, urban biocides, industrial chemicals, flame retardants, plasticizers and polycyclic aromatic hydrocarbons (PAHs), and more. According to Phillips et al. (2012), concentrations of some micropollutants in CSO discharges could be up to 10 times higher than in treated wastewater. In particular, for substances with removal efficiencies in WWTPs >90 %, CSO discharges can contribute 40−90 % of their annual load. Moreover, the accumulation of metals in the recipient's sediment after CSO events can affect its aquatic ecosystems, including inhibiting reproduction in some sensitive macroinvertebrate species (Schertzinger et al., 2018). The sanitary quality of receiving waters can also be impaired by CSOs. Passerat et al. (2011) found that fecal indicator bacteria in CSO discharges to the Seine River can be as high as in raw wastewater. Thus, CSO events cause chemical, physical and biological impacts, leading to the deterioration of the ecological health of receiving waters. These impacts may become more severe during low-flow periods due to limited dilution factors (Montserrat et al., 2013).

SIAAP has a stormwater storage capacity of 955,000 m^3, and when combined with department networks, the total storage capacity reaches 2.5 million m^3. This capacity comprises both underground and open-air storage tanks, as well as reservoirs tunnels. Despite this substantial capacity, approximate 20 to 40 CSO discharges occur in the Paris megacity each year, leading to the discharge of approximately 21 million m^3 into the Seine River. In this study, MP analysis was conducted using water samples collected from two major CSO outfalls in the Paris megacity, namely La Briche and Clichy.

2.2.2. Wastewater treatment plants

The sewage arriving at SIAAP's WWTPs is composed of various sources, including wastewater, stormwater/runoff, infiltration water, and non-potable water used for street cleaning and flushing the sewer network. Sewage composition is illustrated in Figure 2-6, with data from the year 2017. The fluctuations in wastewater and permanent infiltration water over the course of a year are correlated with groundwater levels.

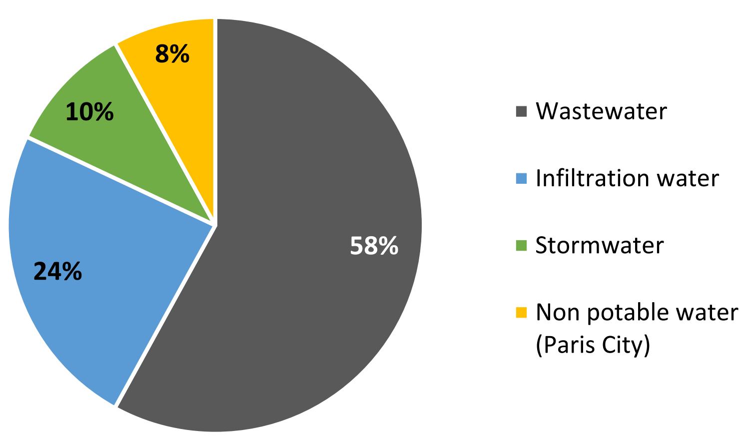

Figure 2-6: Different inputs of water flow entering the Parisian sewer management system

SIAAP operates six WWTPs that utilize advanced technologies in the field of water treatment. Their goal is to meet new regulations and society expectations, especially aiming to reach the 'good status' for receiving waters as stated in the European Water Framework Directive (2000/60/EC of October 23rd, 2000). These WWTPs use either conventional processes as activated sludge and extended aeration, or compact biofilters and membrane bioreactors, which allow a short hydraulic retention time of about 3 hours. For the treatment of sewage sludge, a wide range of dewatering technologies are employed. Figure 2-7 illustrates the wastewater management systems operated by SIAAP in Paris megacity. Seine Aval WWTP (SAV) is the largest plant, treating up 40 % of the wastewater generated in the service area of SIAAP, which amounts to 1.5 million m^3/day.

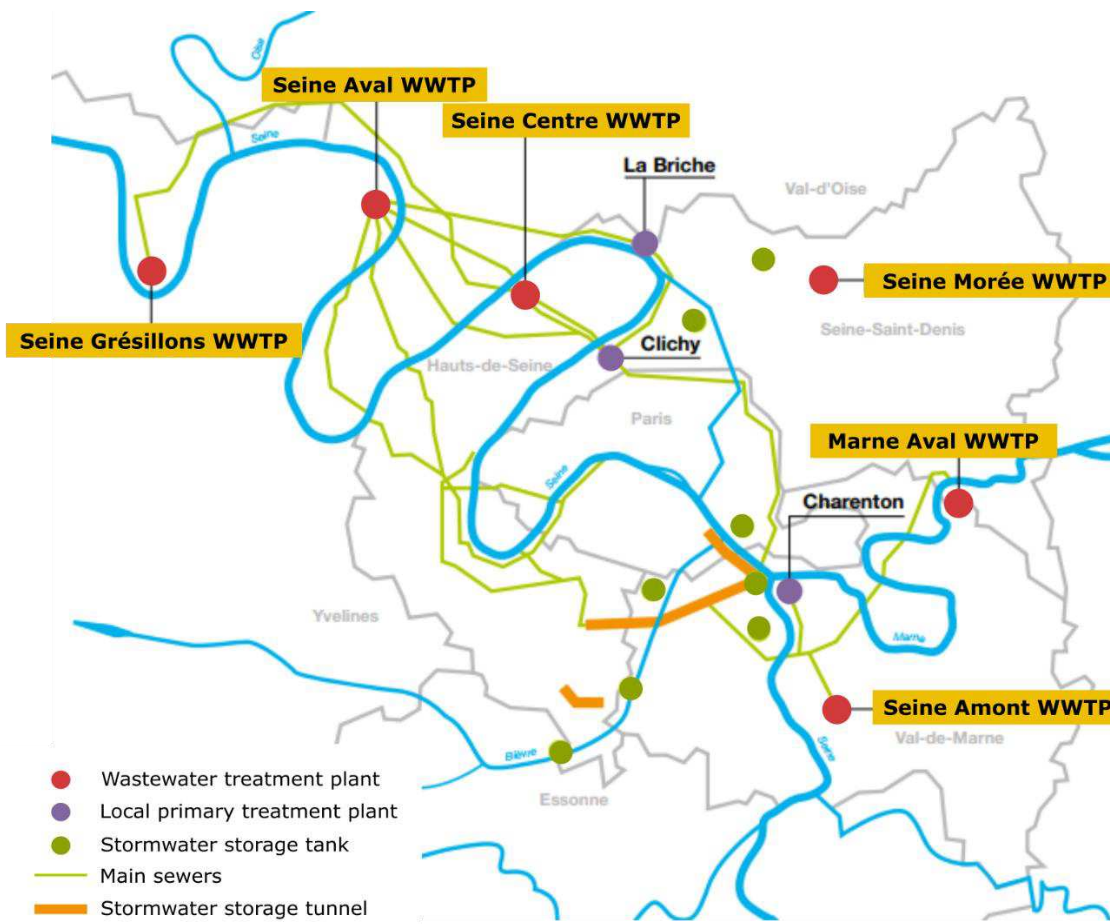

Figure 2-7: The Parisian wastewater management system operated by SIAAP

Most of these WWTPs discharge their effluents into the Seine River, amounting to approximately 2.4 million m^3 per day during dry weather conditions. The Marne Aval WWTP discharges into the Marne River, just upstream of the confluence with the Seine River.

Different sludge treatment technologies are employed at SIAAP's WWTPs. While sludge in the Seine Centre WWTP (SEC) and the Marne Aval WWTP is entirely incinerated, sludge from other plants is used for composting, soil application, cement production, pyrolysis or incineration. In the case of SAV, entire sludge is used for agriculture as a substitute for fertilizers and amendments. Most of the sludge production (up to 85 %) is spread directly in 13 departments, and a portion is composted beforehand.

Three out of the six WWTPs that cover a large panel of technological solutions were selected for MP analysis on sewage sludge in this study, namely SAV, SEC and Seine Grésillons (SEG). They are located in the downstream of Paris City, with SEC and SEG receiving the same wastewater. The specific characteristics with these three plants are provided as follows.

Table 2-1: The characteristics of the three studied WWTPs

	Seine Aval	Seine Centre	Seine Grésillons
Year of commissioning	1940	1998	2007
WWTP capacity (m^3/day)	1,500,000	240,000[a] 404,000[b]	300,000[a] 315,000[b]
PEq (inhabitants)	5 million	1 million	1.2 million
HRT	~8 h	~3 h	~3 h
Water treatment	Air stripping/Screening Grit-grease removal Primary sedimentation Activated sludge Secondary sedimentation Physicochemical treatment Biofiltration (nitrification + denitrification)	Screening Grit-grease removal Physicochemical treatment Biofiltration (carbon + nitrification + denitrification)	Screening Grit-grease removal Physicochemical treatment Biofiltration (nitrification + denitrification)

Sludge treatment	Thickening Anaerobic digestion Thermal conditioning Press filtration	Thickening (flotation) Dewatering Incineration	Thickening Anaerobic digestion Dewatering Thermal drying
Operation conditions	$SRT_{digestion}$ = 20 days $T_{digestion}$ = 37 °C $SRT_{thermal}$ = 45 minutes $T_{thermal}$ = 200 °C $P_{thermal}$ = 20 bars	$SRT_{thermal}$ >2 s $T_{thermal}$ = 850 °C	$SRT_{digestion}$ = 10-15 days $T_{digestion}$ = 55 °C $T_{thermal}$ = 140 °C
Sludge production (ton/year in dry mass)	69,000	21,000[c]	13,800
Sludge disposal	85 % soil application 15 % composted before soil application	N.A	89 % composted

HRT – Hydraulic retention time; SRT – Sludge retention time; PEq – Population equivalent; T – Temperature; P – Pressure; N.A – no information available

a: dry-weather mode; b: wet-weather mode; c: mass before incineration

Modified from Mailler et al. (2017) with an updated information from SIAAP

Chapter 3

Microplastic contamination along different sludge-line treatments: case of Paris megacity

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Abstract

Microplastics (MPs) are abundantly present in urban wastewater, reflecting the plastic consumption habits of modern lifestyle. Literature has reported the efficiency of municipal wastewater treatment plants (WWTPs) in separating MPs from wastewater. However, the transfer of MPs from water phase into sewage sludge turns this treatment byproduct into a plausible primary source of MPs upon disposal into the environment. Hence, attaining a comprehensive grasp of MP occurrence and fate throughout sludge-line treatment becomes crucial. For this purpose, an investigation into MPs in raw sludge, digested sludge, dewatered sludge and final treated sludge was conducted at several WWTPs within the Paris agglomeration. The results exhibited a substantial MP contamination in all sludge types, with concentrations ranging from 86.5×10^3 up to 493.3×10^3 particle/kg dry weight (dw) for the size range 25-500 µm. There was no reduction in MP abundance throughout the treatment process. Additionally, approximately 7 % of sludge-based MPs were returned back to the system through reject water stemming from centrifugation, as determined by the budget balance analysis. This indicates the inefficiency of current sludge treatment toward MP removal. Besides, the impact of treatment technologies on the size distribution of MPs was observed, especially after dewatering with centrifugation and thermal dying at a high temperature up to 200 °C for 45 minutes. Given that soil application is one of the prevalent strategies for sludge disposal, the presence of MPs in

treated sludge implies their potential incorporation and accumulation in agricultural soil. This also demonstrates a significant emission of MPs from urban wastewater into the terrestrial environment and highlights the importance of sludge management practices in addressing this issue.

Highlight

- Sludge treatment in WWTPs is inefficient for addressing microplastic contamination
- Microplastic levels remaining in final treated sludge ranged from 8.6×10^4 to 4.5×10^5 particle/kg dw
- Microplastic concentrations in sludge increased after centrifugation
- An internal circulation of microplastics within WWTPs via reject water, approximately 7% of total particles in raw sludge
- Thermal treatment induced fragmentation of plastic particles

3.1. Introduction

The modern world is witnessing an increase in the abundance of MPs - an emerging pollutant stemming from human activities. Urban areas have become hotspots of MP pollution due to high density population. Urban wastewater, which is highly polluted with MPs, reflects this issue. Local surrounding watercourses thereby become susceptible to plastic contamination due to the inadequate treatment of urban wastewater. According to Sato et al. (2013), upper-middle-income and high-income countries, on average, treat 38 % to 70 % of their generated wastewater, while it was only 8 % in low-income nations.

Wastewater management plays a crucial role in mitigating the anthropogenic pressures exerted by urban areas on their surrounding water bodies. Sewer systems convey wastewater to WWTPs, where it undergoes purification processes before being discharged to the environment. With the emergence of MPs in wastewater, scientific efforts have been dedicated to understanding their occurrence and fate in WWTPs over the last decade, mainly throughout water treatment line (Carr et al., 2016; Talvitie et al., 2017b; Ziajahromi et al., 2017; Bayo et al., 2020). Although the high MP removal efficiencies from water phase were reported, WWTP effluents remain a significant transport way of MPs from urban areas into the environment due to their large discharge volume (Ziajahromi et al., 2017; Magni et al., 2019). MPs are transferred from wastewater into sewage sludge. This poses a risk of MP emission into the terrestrial environment via sludge disposal. Therefore, knowledge of the existence and behavior of MPs in sludge needs to be elucidated.

Research on MPs in the sludge management of WWTPs has been conducted to a limited extent compared to the water treatment line. While initial studies primarily focused on identifying the magnitude of the problem (Gies et al., 2018; Lares et al., 2018; X. Li et al., 2018), recent publications also investigated the effects of sludge treatment on MPs (Alavian Petroody et al., 2021; Harley-Nyang et al., 2022; Li et al., 2022). Despite this, data on the fate of MPs during sludge treatment processes remain scarce. The obtained results on sludge-based MP contamination levels have shown wide variations among different studies (Hatinoğlu & Sanin, 2021). This can be attributed to multiple factors that impact the occurrence of MPs in sewage sludge. First, composition and quantity of generated wastewater, which determine the sludge-based MP content, differ among study sites. The results from the differences in population density, land-use, economic development level, consumption habit and waste management practices in each cities and regions (El Hayany et al., 2022). Additionally, apart from domestic wastewater, industrial wastewater is also treated in municipal WWTPs in some cases (Z. Long et al., 2021; Zhou et al., 2023). Depending on wastewater management systems, stormwater can be conveyed along with wastewater to treatment facilities in case of combined sewer systems. This leads to the contribution of runoff and resuspension of sewer sediment to an increased pollution in wastewater during intensive wet weather events, including MPs. Moreover, various configurations are operated at each WWTP, with different technologies applied depending on its capacity and serving area, technological choices, ground area and financial investment. Therefore, it is of interest to study MP contamination in different types of sludge throughout the treatment line at several facilities using the same methodology. This is particularly important given the absence of a standardized methodology, which hampers data comparison between different studies and the extrapolation of data from small-scale studies to larger ones.

Being aware of the latest findings on MPs in sludge and recognizing the existing knowledge gap, the study was designed to investigate the occurrence and fate of MPs throughout sludge-line treatment in multiple WWTPs within Greater Paris area in France. With the obtained results, the study aimed to (i) assess the efficiency of sludge treatment in WWTP toward MP contamination, (ii) elucidate impacts of different technologies on MPs, and (iii) evaluate the contribution of treated sludge to the emission of MPs into the terrestrial environment.

3.2. Materials and methods

3.2.1. Study site

Paris megacity is the most populated area in France with about 10 million residents. Three out of six WWTPs serving the region were selected for this study, namely SAV, SEC and SEG. They are located in the downstream of Paris City and work under the supervision of SIAAP. The effluent from these WWTPs ends up into the Seine River. The characteristics of each WWTP are provided in Table 2-1 in Chapter 2.

SEC receives about 240,000 m^3 of wastewater per day. Raw sludge from water treatment line undergoes centrifugation for volume reduction, producing about 21,000 tons of dewatered sludge in dry mass per year. Dewatered sludge is then incinerated at 850 °C, releasing bottom ash and smoke, which is treated specifically to avoid odor problem.

SAV treats 1,500,000 m^3 of wastewater per day. Produced sludge first undergoes centrifugation and flotation for water removal before entering mesophilic anaerobic digestion at 37 °C for 20 days. About 40 % of organic matter in sludge is transformed into biogas, while pathogens and parasites are eliminated during this treatment. Digested sludge then undergoes thickening, thermal conditioning (200 °C, 20 bar for 45 minutes) and press filtration. Final treated sludge (or sludge cake) reaches an average dryness of 50 %. A small part of sludge production (about 15-20 %) stemming from the clariflocculation unit is dewatered with centrifugation, achieving an average dryness of 20 %. The plant produces about 69,000 tons of sludge in dry mass each year. Most sludge cake is applied directly in agricultural land of 13 departments as a substitute for fertilizers, while a small part is composted before soil application.

SEG treats 300,000 m^3 of wastewater per day. After thickening step, sludge undergoes a thermophilic digestion (55 °C for 10-15 days) where about 36 % of organic matter is reduced. Digested sludge passes through centrifugation and then thermal treatment at 140 °C. The plant produced about 38 tons of sludge in dry mass per day, equivalent to about 13,000 tons per year. Most of final treated sludge is composted before being applied in agriculture.

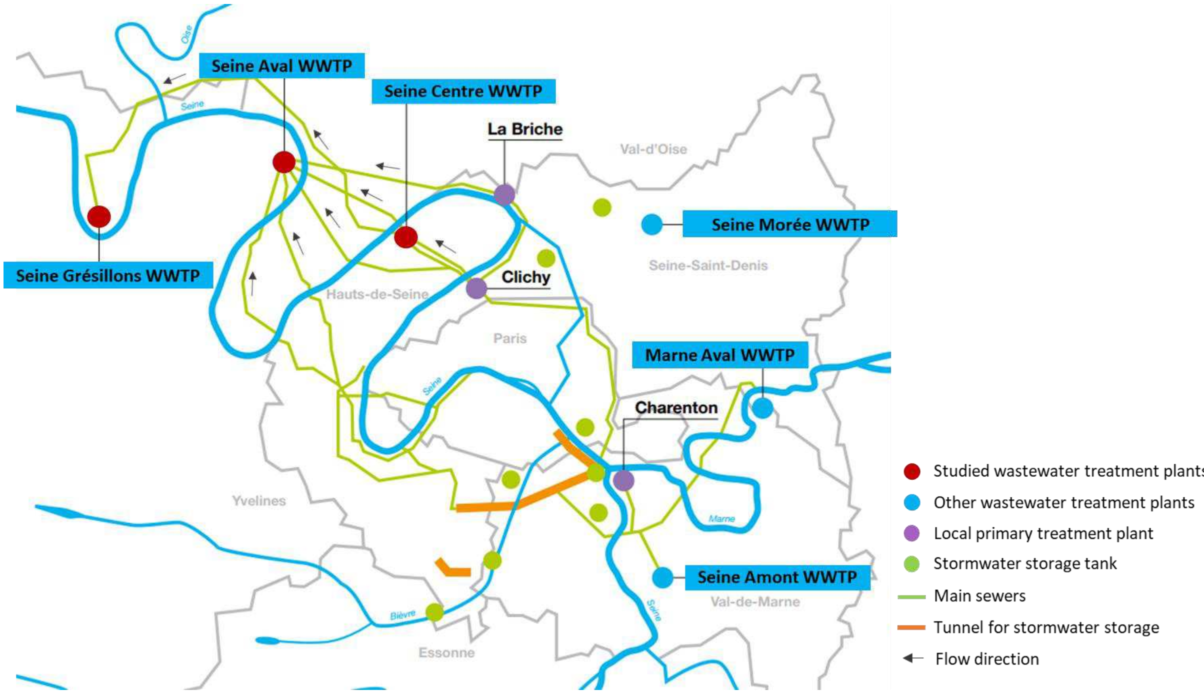

Figure 3-1: Seine Aval, Seine Centre and Seine Grésillons operated by SIAAP

3.2.2. Sample collection

Sampling was carried out from July 2021 to March 2022 at three WWTPs: SAV, SEC and SEG. Different types of sludge were collected, including raw sludge (6 samples), digested sludge (5 samples), dewatered sludge (6 samples) and sludge cake/treated sludge (4 samples). Raw sludge was the mixture of primary and secondary sludge from water-line treatment before undergoing any sludge treatment. Digested sludge was monitored after the digestion process completed. Dewatered sludge was the residue that came out of centrifugation units, and sludge cake/treated sludge was collected at the end of the sludge treatment. Reject water samples were also sampled from dewatering/thickening step at SEC (3 samples) and SAV (2 samples). Due to technical constraints, sludge samples were all punctually collected and stored in glass jars of 250 mL. Glass bottles of 2 L were used for reject water. All samples were kept in the refrigerator at 5 °C before analysis. The sludge-line treatment scheme of three studied WWTPs with sampling points are shown in Figure A-1.

The characteristics of each sludge type were included in Table A-1. Dry matter content (DM in %, in which 1% = 10 g/L) and volatile matter (VM in %DM) were shown in mean ± standard deviation values. In addition, reject water was measured for total suspended solids (TSS in mg/L).

3.2.3. Sample processing

3.2.3.1. For sewage sludge

Sewage sludge is a complex environmental matrix since it comprises microbial biomass and extracellular polymeric substances such as polysaccharides, glycoproteins, nucleic acids, lipids and humic acids (Zhong et al., 2017). Besides, high cellulose contents stemming from toilet papers can be found in sewage sludge because it is hardly degraded during biological processes (activated sludge and anaerobic digestion) (Simon et al., 2018; Wielinski et al., 2018; Philipp et al., 2022).

After consulting different procedures used in previous studies (Löder et al., 2017; Hurley et al., 2018; Al-Azzawi et al., 2020), a pretreatment protocol was developed to isolate MPs from sewage sludge matrices, combining chemical oxidation and enzymatic treatment (Figure 3-2). This protocol was first tailored to dewatered sludge and then applied for the other sludge types. Freeze-drying was carried out for raw sludge and digested sludge which contained high amount of water content, while dry matter content was determined particularly for other sludge types. This step allowed the introduction of a same amount of subsample to the same treatment protocol regardless of sludge type.

0.5 g dw sludge was placed in a 500 mL glass beaker. The beaker was filled with 100 mL and then stirred at 250 rpm to soften sample. A metal spoon was used to enhance the disintegration of sludge cake and treated sludge due to their hardness nature. The beaker was heated up to 30 °C, and then 15 mL of $FeSO_4.7H_2O$ 0.1 M, pH = 0.8 (acidified with concentrated H_2SO_4) and 10 mL of H_2O_2 50 % were added into the beaker to start the reaction. Heating was removed and temperature was monitored and kept under 45 °C. An ice-water bath was placed close by in case the solution overheated. 10 mL of H_2O_2 50 % was added gradually after 15-30 minutes in order to enhance the reaction. This step aimed to remove easily degradable OM present in the sample as much as possible. The reaction was monitored for 3 hours and then the beaker was let stand overnight. The reaction continued gently up to 24 hours afterwards. Next, sample was treated with 5 mL of sodium dodecyl sulfate (SDS) 10 % at 40-45 °C, 250 rpm for 24 hours. This step aims to alter the protein structure of organic matter in the sample matrix, thereby enhancing the effectiveness of the following enzymatic treatment. However, SDS needed to be removed throughout after treatment to avoid impact on the enzymatic treatment as well as the identification with FTIR later on. Sample was then treated with 10 mL TRIS buffer pH=10 and 10 mL lipase enzyme to remove lipid compounds in sample matrix. 10 mL TRIS buffer pH=9 and 10 mL protease were applied in the next step to break down protein. 15 mL of acetate buffer pH=4.5 and 15 mL of cellulase were then used to decompose cellulose and polysaccharides. Buffers were added to maintain an optimum pH condition for each specific enzyme. The sample was kept in a water bath at 35 °C at 50 rpm for 24 hours during enzymatic treatment steps. Fenton reaction was repeated to remove the remaining impurities in the sample. At the end of each treatment, sample was concentrated on 10 µm metallic filters using a metallic filtration unit. More than one filter was used for each sample if needed. The filtration funnel was rinsed thoroughly to avoid sample loss during transfer. Before going to the next step, sample on metallic filters was placed in the same beaker with 100 mL Milli-Q and resuspended in an ultrasonic bath for one minute.

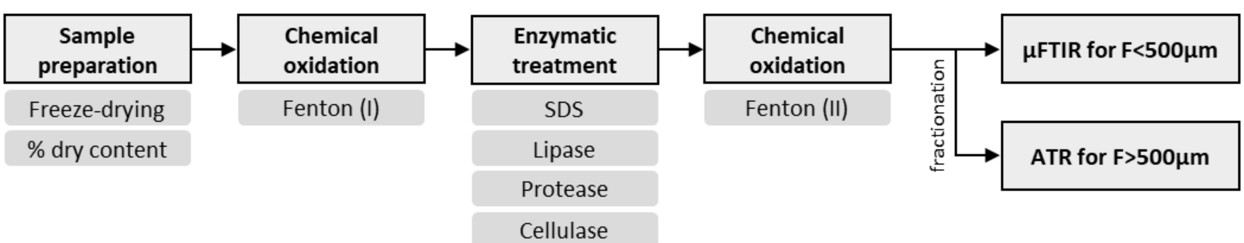

Figure 3-2: Schematic of the pretreatment protocol to extract MPs from sewage sludge matrices

3.2.3.2. For reject water

250 mL of reject water was placed in a 2 L glass beaker. A small volume of H_2O_2 30 % was added each time into the beaker to start and maintain oxidation treatment. The solution was stirred at a high speed of 1000 rpm without heating. H_2O_2 was added until no more foaming was observed. A total of 20-30 mL H_2O_2 30 % was used. The reaction was monitored for 3 hours and let to stand overnight. The sample was concentrated on 10 µm metallic filter at the end of the treatment.

Next, sample was resuspended in 100 mL of Milli-Q before 5 mL of SDS 10 % was added. The treatment was kept for 24 hours at 40-45 °C and 250 rpm. 10 mL TRIS buffer pH=9 and 10 mL protease were added in the next step. The reaction was kept in a water bath at 35 °C at 50 rpm for 24 hours.

After chemical oxidation treatment, treated sample of sewage sludge and reject water was fractionated using a 500µm stainless steel mesh. While fraction larger than 500µm (F>500µm) was kept on the mesh in a Petri dish for further analysis, the smaller fraction (F<500µm) was concentrated on a 10 µm metallic filter.

3.2.4. Sample analysis

3.2.4.1. Size fraction F>500 µm

The 500 µm mesh was inspected under a stereomicroscope (Leica M125C, 8-100x magnification). Suspected plastic particles were photographed and documented for shape and size using Histolab software (version 11.5.1). These particles were then analyzed for chemical composition using a FTIR spectroscopy. The measurement was performed on a Nicolet iS5 spectrometer (Thermo Scientific) coupled to an iD7 ATR–Diamond accessory. The wavenumber is set for 4000 – 400 cm^{-1}, 16 scans were carried out with a spectral resolution of 4 cm^{-1}. The software OMNIC (Thermo Fischer Scientific) was used to compare obtained spectra to reference database with a score out of 100 returned as the goodness of match. A particle is considered to be successfully identified when the match is higher than 70.

3.2.4.2. Size fraction <500 µm

Sample on the 10 µm metallic filter was resuspended in an ultrasonic bath and deposited on Anodisc filters (Ø25 mm, pore size of 0.2 µm, Whatman®). The analysis was then carried out with an automated µ-FTIR imaging in transmission mode (Nicolet iN10 MX, Thermo Scientific, 25×25 µm pixel resolution). The setting included the wavenumber range of 4000 – 400 cm^{-1} and 16 scans with a spectral resolution of 4 cm^{-1}. Data processing was carried out later with the siMPle software (version 1.1.β, developed by Aalborg University, Denmark and Alfred Wegener Institute, Germany) to identify the polymer composition of the particles.

Transmission mode is preferred when analyzing MP samples with FTIR spectroscopy. This action aims to avoid complex refraction which may produce uninterpretable spectra when particles with irregular shapes are irradiated under reflection mode (Harrison et al., 2012). However, particles larger than 500 µm are unsuitable for FTIR measurement in transmission mode because of their thickness. Therefore, two different methods were needed for two different fractions.

3.2.5. Blank and quality control

Cotton lab coats and nitrile gloves were worn to minimize sample contamination during lab processing. Samples were handled under a clean bench with laminar flow and covered with aluminum foils whenever possible. Glass apparatus were burned at 525 °C for 2 hours and metal ware were rinsed thoroughly with ultrapure water before use. All solutions in direct contact with sample (i.e., enzymes and buffers) were filtered through GF/D (pore size 2.7 µm). In addition, five procedural blanks were carried out to monitor potential contamination. PE, PP and polyvinyl chloride (PVC) particles were found in blanks, ranging from 0 to 9 particles per sample with the size less than 500 µm. The results showed a low contamination level during laboratory work and they were not used for data correction.

3.2.6. Data analysis

The Shapiro-Wilk test was conducted to test the normality of the particle size distribution. Due to the abnormal distribution of the dataset, non-parametric statistics including the Kruskal-Wallis test and then the post-hoc Dunn test were applied to examine the difference between median size of MPs in different sludge types of each WWTP. The level of significance set is $\alpha=0.05$. All the results of the statistical tests are provided in Annex I. The graphs and figures were prepared using R version 4.0.0 and Inkscape version 1.1.

3.2.7. Estimated annual budget at the technological scale

The total number of MPs in different types of sludge on an annual scale was calculated by multiplying the total dry weight of sludge (ton/year) by the mean number concentration of MPs (particle/g dw) found in this study. In the case of reject water, the annual total MP counts was determined by multiplying the total volumetric flow (m^3/year) by the obtained MP number concentration (particles/L). This approach allows the establishment of the MP budget at various treatment processes whenever applicable. Table A-2 provides the equations for calculating the total MP particles at each treatment step.

3.3. Results

3.3.1. Size fraction F>500 µm

Table 3-1: Contamination level of large-sized MPs in different sludge types and returned water

	Min - max	Median	Unit
Raw sludge (n=6)	1.86 – 7.95	3.67	particle/g dw
Digested sludge (n=5)	1.94 – 7.31	4.05	
Dewatered sludge (n=6)	<2.14 – 2.59	1.05	
Treated sludge (n=4)	<2.23 – 5.74	1.02	
Reject water (n=5)	<4 – 40	-	particle/L

MP particles larger than 500 µm (MPs>500 µm) were present in all sludge types regardless treatment plant, with a total of 31 particles detected. While 100 % of raw sludge and digested sludge samples contained MPs>500 µm particles, it was 50 % of samples for dewatered and treated sludge. For reject water, only two samples from SEC contained MPs>500 µm, with a total of 13 particles. The number concentration ranges for MPs>500 µm in all samples are shown in Table 3-1. The most common polymers found were PE, PET, PS, poly(11-bromoundecyl methacrylate) (PBMA) and ethylene/propylene copolymer.

Most of MPs>500 µm were fragments. The images of some particles are shown in Figure 3-3. Fibers were also observed in samples; however, they are mostly entangled on the mesh. This made the sorting out of individual fibers for chemical identification infeasible. In addition, fibers have a very thin and elongated shape, which results in a weak signal during ATR measurements due to the limited contact area. As a consequence, only a few fibers were identified, accounting for less than 10 % of the total particles. Detected MPs>500 µm were transparent or in color such as green, red, black, white and yellow.

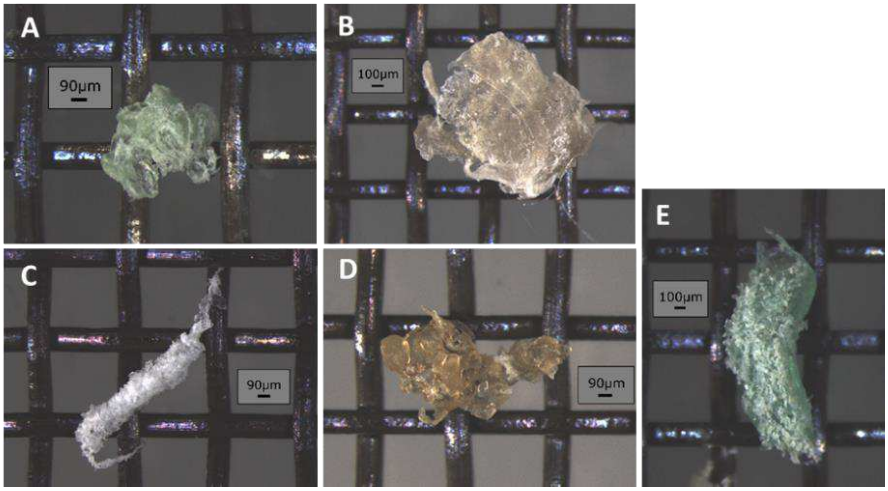

Figure 3-3: Exemplars of MPs>500 µm detected with ATR-FTIR. The black bar shows the scale of the image. A: PE in raw sludge; B: PE in digested sludge; C: PE in dewatered sludge; D: copolymer in treated sludge; E: PE in returned water

3.3.2. Size fraction F<500 µm

3.3.2.1. Concentration level and annual budget

Table 3-2: MP contamination in different sludge types (particle/g dw) and reject water (particle/L)

Type	Raw sludge	Digested sludge	Dewatered sludge	Sludge cake/Treated sludge	Reject water
Unit	particle/g	particle/g	particle/g	particle/g	particle/L
Seine Centre (SEC)	170.9	-	327.5	-	268
	197.1	-	404.7	-	724
	-	-	460.9	-	1120
Seine Aval (SAV)	211.0	259.4	-	86.5	52
	223.1	360.6	-	316.0	68
	488.7	-	-	450.2	-
Seine Grésillons (SEG)	341.4	239.1	231.8	298.0	-
	-	270.6	350.6	-	-
	-	277.0	493.3	-	-

Table 3-2 summarizes MP number concentrations found in sludge and reject water samples in this study. High levels of MPs were detected in all sludge types collected from three studied plants, ranging from 86 to 488 particle/g dw. The estimated annual budget of MPs in raw sludge indicated that approximately 10^{12}-10^{13} particles/year were transferred from water-line to sludge-line treatment within WWTPs (Table 3-3). MPs were found in reject water from dewatering process in SAV and SEC, with average concentrations of 60 and 704 particle/L, respectively. In the case of SEC, the annual MP budget in reject water amounted to approximately 7 % of that found in raw sludge before centrifugation.

Dewatered sludge exhibited slightly higher levels of MP contamination compared to other sludge types, with average concentrations of 397 particle/g dw at SEC and 358 particle/g dw at SEG. The MP contamination levels in the final treated sludge ranged from 86 to 450 particle/g dw for SAV and nearly 300 particle/g dw for SEG, corresponding to an annual budget of approximately 1.5×10^{12}–8×10^{12} particle/year.

Table 3-3: Estimated total number of MPs in different sludge types and reject water on an annual scale based on the data provided by SIAAP for the year 2021 (unit: particle/year)

Type	Raw sludge	Digested sludge	Dewatered sludge	Sludge cake/Treated sludge	Reject water
Seine Centre (SEC)	3.5×10^{12}	-	6.4×10^{12}	-	1.1×10^{11}
	4.0×10^{12}	-	7.9×10^{12}	-	3.0×10^{11}
	-	-	9.0×10^{12}	-	4.6×10^{11}
Seine Aval (SAV)	2.5×10^{13}	2.2×10^{13}	-	1.5×10^{12}	-
	2.7×10^{13}	3.1×10^{13}	-	5.6×10^{12}	-
	5.9×10^{13}	-	-	8.0×10^{12}	-
Seine Gresillons (SEG)	7.0×10^{12}	2.7×10^{12}	-	2.7×10^{12}	-
	-	3.0×10^{12}	-	-	-
	-	3.1×10^{12}	-	-	-

3.3.2.2. Particle size distribution

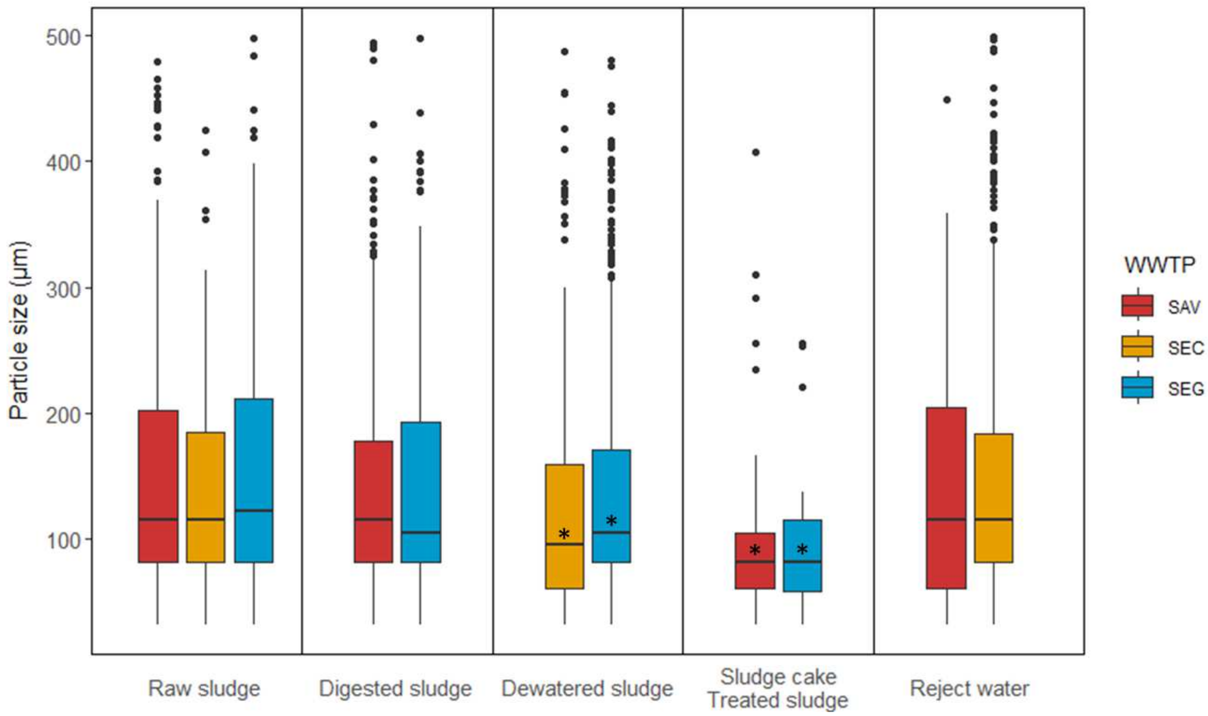

*Figure 3-4: Size distribution of MPs (25-500 μm) in different sludge types and reject water from three studied WWTPs. The box (IQR=Q3-Q1) presents the 50 % of the central size dataset. The line inside the box (Q2) presents the median particle size. The upper whisker shows the data range up to Q3+1.5*IQR. The lower whisker shows the data range down to Q1-1.5*IQR. In which, Q1-First quartile, Q2-Second quartile, Q3-Third quartile and IQR-Interquartile range. Outliers of the dataset are located outside the whiskers and shown in black dots. The asterisks show a significant change in median size of MPs in one sludge type compared to particles in raw sludge from the same plant with p-value <0.05.*

Most of MPs detected in sewage sludge are smaller than 200 μm. The median size particle ranged from 80 μm in sludge cake/treated sludge up to 120 μm in raw sludge and reject water. Particles found in raw sludge of three plants have a similar size distribution with no significant difference found among their median particle size.

The size of MPs after anaerobic digestion changed insignificantly in both SEG and SAV, while the significant reduction in size particle was observed for dewatered sludge in SEC and SEG. Similar, MPs in sludge cake/ treated sludge were significantly smaller than particles in other sludge types (Figure A-3, Figure A-4).

MPs in reject water from SAV and SEC have similar median size to particles in corresponding raw sludge, ranging from 115 to 122 µm. The size distribution of MPs in SAV reject water varied in a larger range compared to the case of SEC.

3.3.2.3. Polymer composition

13 different polymer types were detected in sewage sludge samples, in which, four main polymers (PE, PP, PEST and PS) accounted for more than 80 % up to 94 % regardless of sludge type and treatment plant. Polymer diversity remained stable after anaerobic digestion and dewatering. It decreased remarkably after thermal treatment, from 13 down to 7 for SAV and from 13 down to 5 for SEG. The polymer composition of MPs in different sludge types from three studied plants is summarized in Table A-3 in Annex I.

3.4. Discussion

3.4.1. Microplastic contamination in sewage sludge

The magnitude of MP levels in sewage sludge found in this study was similar to the data reported in the literature, most of which fell within the range of 10^4 – 10^5 particles/g dw, as illustrated in Table 1-3. However, the present study documented relatively higher concentrations than most other publications, regardless of sludge type (Pittura et al., 2020; Ziajahromi et al., 2021; Harley-Nyang et al., 2022; Yuan et al., 2022).

High MP levels found in raw sludge highlighted the substantial transfer of MPs from wastewater into sludge during water-line treatment processes. SEC and SEG receive wastewater from the same area, while SAV serves a larger catchment (Figure 3-1). However, the quality of MPs found in raw sludge from three WWTPs was similar regarding polymer composition and particle size. This suggests the homogeneity of wastewater throughout the Parisian sewer system. Thus, the findings from this study provide insights into the polymers present in wastewater in Paris megacity. The variations in MP level in raw sludge from different plants, which were also reported in previous studies, may show the influence of different factors such as influent load and water treatment technologies. Seasonal factors may also result in the variation of MP contamination level when samples were taken from different time points. However, it is important to consider potential biases resulting from the limitations of the current methodology for sludge-based MP analysis.

The levels of MPs remaining in treated sludge in this study were consistent with the findings of Talvitie et al. (2017b) and Horton et al. (2021), but higher than that observed in sludge cake or limed sludge in Lee & Kim (2018), Jiang et al. (2020) and Harley-Nyang et al. (2022). This emphasizes an important pathway of MP emission from WWTPs into the environment via sludge disposal. For example, in case of SAV, approximately 62,000 tons of sludge cake in dry mass are directly spread across 13 different departments per year, resulting in about 5.4×10^{12} – 2.8×10^{13} MPs being emitted into the environment from this plant.

MPs>500 µm were present infrequently in sludge samples, regardless of sludge type. The similarities were observed for particles in the size range 250-500 µm. This shows the efficiency of screening and grit-grease removal steps in separating large-sized particles from the water phase. As scum generated from these treatments is not included in the sludge and disposed of separately, another pathway of MPs originating from WWTPs should be considered (Alavian Petroody et al., 2021; Monira et al., 2023). The treatment of such waste materials differs from country to country. For example, in Australia, Zajahromi et al. (2021) reported that about 70 % MPs removed from wastewater during screening and grit-grease removal are transported to landfill. Since a small number of particles >500 µm were detected in this study, an increase in sample volume is recommended to obtain more robust data on this infrequent fraction. Similarly, most of reject water samples had less than one particle detected in 250 mL in this study, equivalent to a concentration <4 particle/L. This also indicates the need for analyzing a larger sample volume in future studies.

3.4.2. Impacts of treatment processes on microplastic particles

3.4.2.1. Microplastics after anaerobic digestion

Negligible changes in MP number concentration as well as particle size was observed after anaerobic treatment at SAV in this study. Similar, Li et al. (2022) reported that the MP content in sludge remained unchanged after anaerobic digestion, with little alterations observed on the particle surfaces. This can be attributed to the non-biodegradable nature of plastics, particularly conventional polymers. Previous studies by Selke et al. (2015) and Gomez Gómez & Michel (2013) emphasized the resistance to degradation in conventional plastics such as PP and PE, even when modified with additives for enhanced biodegradability. In addition to microbial degradation activities, the medium-temperature operation of anaerobic digestion at SAV appears to have minimal impact on MP concentrations in sewage sludge.

Compared to SAV, a slight decrease in MP contamination level was observed in SEG after sludge undergoes thermophilic digestion at 55 °C. Increase temperature above 50 °C may have impacts on plastic particles (Koelmans et al., 2019). However, due to the limited number of raw sludge samples in SEG, no statement can be drawn.

3.4.2.2. Microplastics in reject water

The detection of MPs in reject water from centrifugation at SEC indicates the release of MPs from sewage sludge back into the liquid phase. This phenomenon can be attributed to the breakdown of the extracellular polymeric structure of sludge flocs, resulting from biological decomposition during anaerobic digestion and/or the centrifugal force applied during dewatering. This breakdown leads to the separation of attached particles from the sludge and their recirculation back to the water-line treatment. Several studies have also reported high concentrations of MPs in centrifuge reject (Talvitie et al., 2017b; Nakao et al., 2021; Bretas Alvim et al., 2022). The variations in MPs levels among these studies could result from the difference in influent loads, technology choices in water and sludge treatment systems, as well as operational conditions of each facility.

Reject water in SAV also contained MPs but with lower concentrations. It can be attributed to the difference in operating principles of the employed technologies (X. Li et al., 2018). While reject water in SEC stemming from centrifugation, reject water in SAV stemming from mostly gravitational/static thickening which depends a lot on particle density. It may explain why only PP and PE – low-density polymers – were detected in reject water in SAV. Thus, mechanical water removal in sludge management results in the detachment of MPs from the sludge and their release into the liquid phase. Other studies also had the same findings, for instance, Alavian

Petroody et al. (2021) detected MPs in the supernatant after flotation/dynamic thickening. Similarly, Salmi et al. (2021) and Nakao et al. (2021) documented the presence of MPs in the effluent following gravimetric thickening (Table A-4).

Depending on each WWTP, the water stemming from these sludge treatment processes is returned back to the system either with or without treatment. In case of no treatment, this creates an internal circular pathway of MPs within WWTPs. The findings in this study show that approximately 7 % of MPs in raw sludge were transferred to reject water after centrifugation. Salmi et al. (2021) estimated that MP levels in reject water can be up to three times higher than the influent load in raw wastewater. All in all, this partly indicates the insufficiency of current technologies towards MPs removal in WWTPs. The recirculation and accumulation potential of MPs inside the system may have adverse impacts on the plant performance. Some effects of plastics particles on methane and hydrogen production from sludge have been reported (Hatinoğlu & Sanin, 2021). Thus, reject water requires proper management to reduce MP load imposed on the treatment step where it is returned. Besides, targeting this internal recirculate flow may be a new approach in improving the removal efficiency of WWTPs in addressing plastic pollution (Salmi et al., 2021).

3.4.2.3. Microplastics in dewatered sludge after centrifugation

An increase in the number concentration of MPs was observed in dewatered sludge after centrifugation at SEC. The budget established for this treatment step also showed an increase up to 200 % of the total particle counts on an annual scale. This phenomenon can be attributed to the significant reduction in the particle size of MPs after the treatment. Similarly, an increasing in number concentration after centrifugation, along with a decrease in particle size, was also observed in SEG. These findings suggested that centrifugation may have an impact on the fragmentation of plastic particles in sewage sludge. Besides, the reasonable levels of MPs detected in the inlet and outlets of the centrifugation unit at SEC indicate the reliability and validity of the methodology employed in this study.

3.4.2.4. Microplastics after thermal treatment

The MP concentration found in the final treated sludge in SAV and SEG was lower compared to the previous treatment, but a reduction in particle size was documented. These results suggest the fragmentation of plastic particles during the final treatment, causing them to fall below the size detection limit. Harley-Nyang et al. (2022) also reported the lowest average size range in the end product of sludge-line treatment. Additionally, polymer diversity has decreased after the final treatment, suggesting the impact of thermal treatment on the polymer composition in sludge. The spectra of PE found in sludge cake/treated sludge showed a worse match to the reference spectra (Figure A-5), thereby solidifying this hypothesis.

3.4.3. Plastic contamination in soil related to sludge disposal

According to Decree No. 97-1133 of the French government relating to the spreading of sludge from water treatment on agricultural soil, a maximum of 3 tons of dry matter of treated sludge can be applied per ha over a period of ten years. Based on the MP levels found in the final treated sludge in this study, the number of MPs, ranging from 2.6×10^{8} - 1.4×10^{9} particle/ha/year, can be applied to agricultural soil. Compared to MP inputs from atmospheric deposition, estimated at about 4.6×10^{7} – 2.8×10^{8} particle/ha/year (Beaurepaire et al., in preparation), sludge disposal appears to contribute a higher amount of MPs to plastic contamination in the terrestrial environment.

It should be noted that plastics are known for their resistance, which allows them to endure in the environment for extended periods. Furthermore, according to Schell et al. (2022), there is limited movement of MPs from agricultural soils to other environmental compartments. The study revealed a low infiltration capacity of MPs into deeper soil layers or underground water, and negligible impact from surface runoff in transporting these particles to nearby watercourses. Therefore, increasing MP levels in terrestrial ecosystems, leading to soil contamination, is expected due to the accumulation of these particles in agricultural land via sludge disposal.

3.5. Conclusion

In this study we investigated MP contamination levels at different sludge treatment stages at three WWTPs in Greater Paris area. Obtained results indicated the efficiency of existing treatment technologies in completely removing MPs in sewage sludge. MP concentrations in the range of 8.6×10^4 to 4.5×10^5 particle/kg remained in treated sludge, leading to the release of up to 10^{13} particles into agricultural soil per year solely from SAV. A certain amount of MPs was internal circulated inside the system with reject water from dewatering step. While anaerobic digestion had insignificant impacts on plastic particles, thermal drying at high temperature induced the reduction in the size particle distribution. This work demonstrates a significant emission of MPs from urban wastewater into the terrestrial environment via sludge disposal and highlights the importance of sludge management practices in addressing this issue.

Chapter 4

Microplastic accumulation in sewer sediments and its potential entering the environment via combined sewer overflows: a study case in Paris

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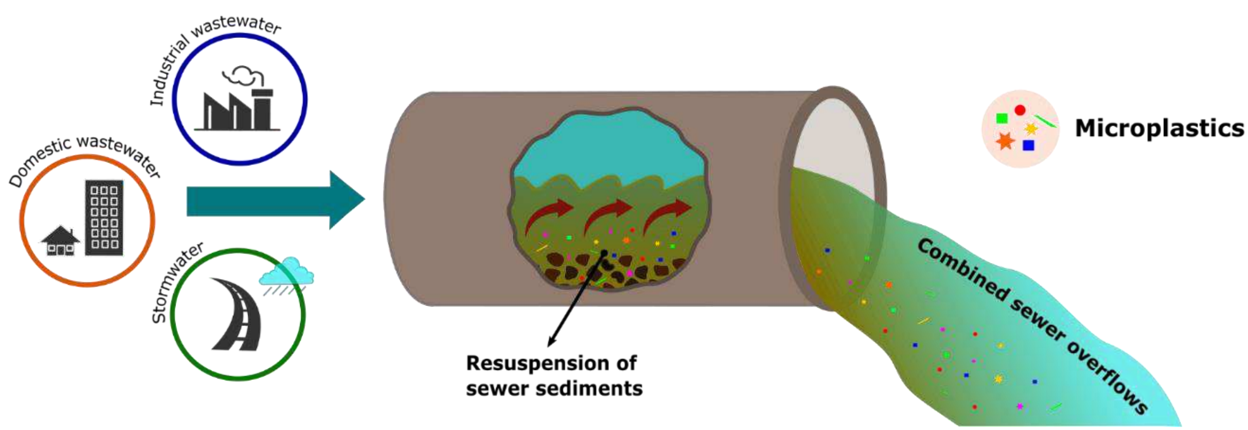

Abstract

During wet weather events, combined sewer overflows (CSOs) transfer large amount of particulate matter and associated pollutants into surrounding water bodies, thereby deteriorating the recipients' ecological health. Resuspension of sewer sediments during these events contribute significantly to pollution level of these discharges. However, how much this in-sewer process contributes to CSOs' quality regarding microplastic (MP) pollution is little known. Therefore, an investigation on sewer deposits inside the Parisian combined sewer network was carried out. The study found high MP concentrations stored in this matrix, ranging from 5×10^3 to 178×10^3 particle/kg dry weight (dw). Polymer composition is similar to what found in raw wastewater, containing a high proportion of polyethylene and polypropylene. Thus, the results indicated the persistence of MPs in sewer network during transport during dry weather periods to treatment facilities. Once the resuspension of sewer deposits happens, MPs can be released into water flow and get discharged along with CSOs. This highlights another potential pathway of MPs into freshwater environment.

Highlights

- Initial investigation on MP occurrence and fate in sewer networks
- High concentration of MPs up to 178×10^3 particle/kg dw stored in sewer sediments
- Polyethylene and polypropylene were the most common polymer types

Keywords: combined sewer overflows, combined sewers, microplastics, plastic pollution, sewer sediment, wet weather flows

4.1. Introduction

Sewer sediments are bed deposits found inside sewer networks, formed by particulate matter which detaches from water phase and settles down during transport of sewage. This phenomenon often occurs during low-activity intervals and dry-weather periods when flow rate and turbulence levels are moderate. Sewer sediments also mount wherever water flow decelerates, for instance, abrupt changes in shape or dimension of pipes, divergent or low slope sectors, etc. Thus, the accumulation of sewer sediments are temporal and spatial dependent, directly linked to water velocity inside sewer network (Seco, 2014).

Sewer sediment is moist, heterogeneous and complex in composition. However, they can be subdivided into gross bed sediment (GBS), organic layer and biofilm based on the nature of materials (Rocher et al., 2004). While GBS has a high mineral content, the other types contain mainly organic matter. The main components in inorganic fraction of sewer deposits are particles swept from different surfaces of impervious areas by runoff water, surrounded soil coming along with groundwater infiltration and rusted metal of the sewer, etc. While, organic fraction mostly stems from sanitary wastewater which contains high load of suspended solids (Ashley & Crabtree, 1992; Ashley et al., 2004; Arthur et al., 2015). During wet weather events, sewage inside combined sewer systems accelerates, accompanied by an increasing turbulence level, which leads to the transport of sewer sediments in different modes (Butler et al., 2003). Some tend to roll, slide or leap as bed-load inside sewer network (e.g., grit and gross solids), some get resuspended and carried in suspension (e.g., organic solids, silt and fine sand). Thus, sewer sediments' materials (including pollutants) can be released and added into wet weather flows during these occurrences, known as erosion/resuspension of sewer sediments.

During intensive events, wet weather flows can be discharged into the environments without any treatment. These discharges are coined as combined sewer overflows (CSOs) in case of combined sewer systems. Previous studies highlighted CSO pollution and its adverse impacts on the health of the receiving water bodies (Hvitved-Jacobsen, 1982; Phillips et al., 2012; Rechenburg et al., 2006), as well as indicated remarkable contribution of sewer sediments to quality of CSOs (Chebbo et al., 1995; Gromaire et al., 2001; Ashley et al., 2004; Gasperi et al., 2010). The accumulation of these in-sewer deposits also causes more frequent CSOs due to reduction in conveyance efficiency of the sewer network (Crabtree, 1989; Seco, 2014; Veliskova & Sokac, 2019). Therefore, sewer sediments can act as a sink-source of pollutants that contribute to rising pollution level in wet weather flows and receiving water bodies.

Microplastics (MPs) are found abundant in wastewater as a consequence of using and overusing plastics in modern daily life. While the evolution of MPs in wastewater treatment plants (WWTPs) have been well studied in the last decade (Liu et al., 2021; Z. Xu et al., 2021; Krishnan et al., 2023), it was not the case for their occurrence and fate before reaching these treatment facilities. During transport inside sewer network, MPs may integrate with mineral and organic particles in wastewater, settle down and get trapped in sewer sediments. Once these sediments erode, MPs may get released into water flow along with other pollutants. However, very little is known about the contribution of sewer sediments to the quality of CSOs regarding MP pollution. This study will investigate MP content in sewer sediments for the first time. The obtained results will be used to improve the assessment of CSO contribution into plastic pollution. These two objectives aim to increase the knowledge on MPs occurrence and fate in the sewer network – WWTP continuum and their pathways in urban waters before entering the environment.

4.2. Materials and methods

4.2.1. Study site

Paris City - a part of the Parisian megacity – is chosen for this case study. This catchment has a high population density (20,000 inhabitants per km^2) with intense commercial and service activities. Impervious surface constitutes about 70 % of the total area. The sewer systems serving Paris City is fully combined; in which, Seine River is the recipient of treated wastewater from WWTPs or CSOs in wet weather conditions. The Parisian sewer network has experienced the formation and accumulation of sewer deposits. Therefore, sand chambers, which behave as sediment traps, were installed to reduce adverse impacts of these deposits on hydraulic conveyance efficiency. About 100 sand chambers were located over the network (Rocher et al., 2004). Most of them have the same width as the sewer, but they are deeper. This induces abrupt decrease in the flowrate of wastewater when passing through, thereby allowing particles to detach from water phase and settle down. Sand chambers are regularly cleaned up as a part of the system maintenance. Extracted sediments are then sent to specialized centers for proper treatment.

Twelve sediment samples were collected from sand chambers inside the Parisian sewer network during regular cleaning-up activities between March and August 2021. Samples from primary sewer pipes were named P1, P2 and P3, with letters 'a' and 'b' indicating samples from the same sewer pipe. Samples from secondary sewer pipes were listed as S1 to S7. The map of sampling points is shown in Figure 4-1, and the exact addresses with the coordinates of each site are provided in Table A-5. After collection, samples were stored in glass jars at 5 °C until further analysis. Sediments in sand chambers showed similar characteristics to sewer deposits found along the network (Rocher et al., 2004).

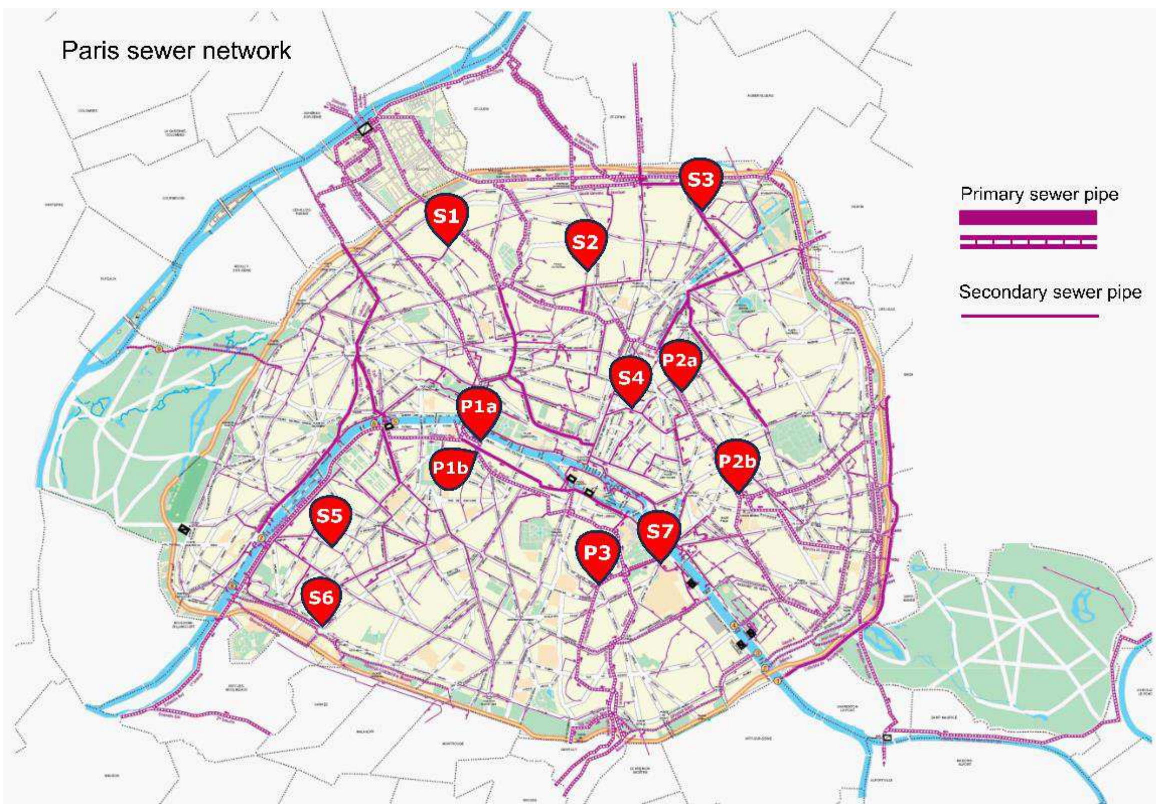

Figure 4-1: Sampling location of sewer sediments (modified from the map of the Parisian sewer network provided by Hotel de Ville Paris)

4.2.2. Sample preparation & analysis

Samples were first freeze-dried to remove water content and then sieved through 5 mm metallic mesh for removal of large items. A subsample of 0.5 g dw sediment underwent chemical oxidation with H_2O_2 30 % at 45 °C for 24 hours. Sample was then filtered through 10 µm metallic filters to remove degraded organic matter. Remaining material was subjected to density separation using sodium iodide solution (>1.6 g/cm^3) for 24 hours before going through another oxidation step with H_2O_2. After treatment, samples were deposited on Anodisc filters (Ø 25 mm; pore size of 0.2 µm) before being analyzed with the automated µ-FTIR imaging in transmission mode (Nicolet iN10 MX, Thermo Scientific, 25×25 µm pixel resolution). Data processing was carried out later with the siMPle software (version 1.1.β, developed by Aalborg University, Denmark and Alfred Wegener Institute, Germany) to identify the polymer composition of the particles. Besides, particulate organic carbon (POC) was measured in all samples.

4.3. Results and discussion

4.3.1. Microplastic content in sewer sediments

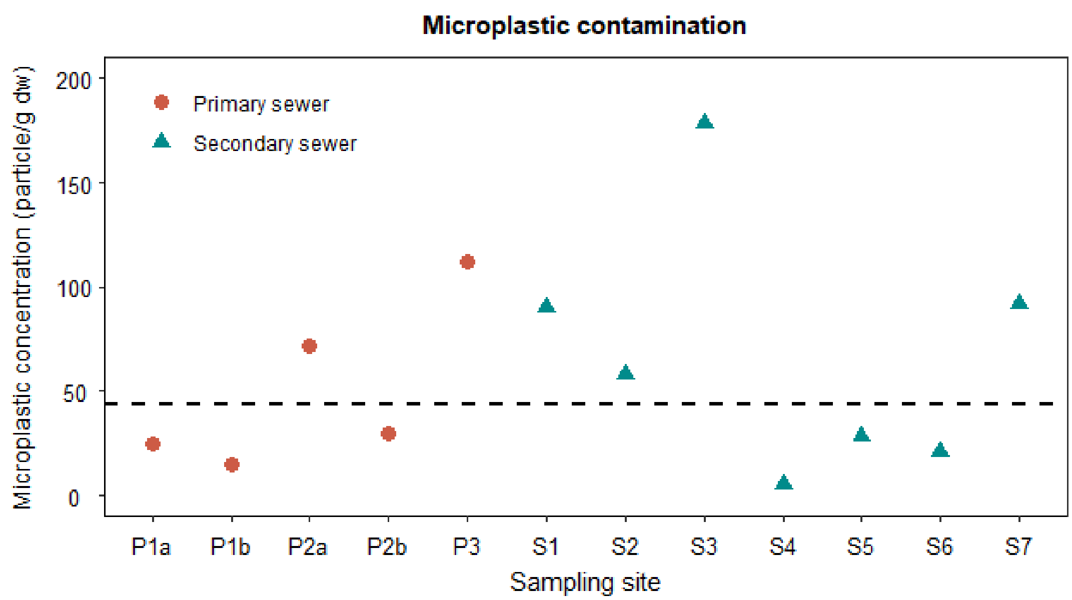

Figure 4-2: MP concentration detected in in-sewer deposits from primary sewer pipes (red point) and secondary sewer pipes (blue triangle) of the Parisian sewer network. The dashed line shows the median concentration

MPs were encountered in all samples analyzed in this study. The highest concentration up to 178×10^3 particle/kg dw was found in S3. It is 30 times higher than the lowest concentration detected in S4 with 5×10^3 particle/kg dw. The median concentration of all samples was 4.4×10^4 particle/kg dw (n=12). It can be observed that MP contamination level in sewer sediments varied among different sampling sites, regardless of their localization on primary or secondary pipes. The variability between samples from the same pipe (i.e., P1a-P1b and P2a-P2b in Figure 4-2) increased with the distance between sampling points. The characteristics of wastewater can be considered homogenous in a large-scale sewer system like Paris, thus, MP content in in-sewer deposits may mostly be related to the accumulation of these sediments inside the system. The accumulation depends generally on different factors such as water flowrate, sewer structure and cleaning frequency.

MP content found in sewer sediments and sewage sludge are in the same order of magnitude (Table 4-1). This indicates that a considerable amount of MPs in wastewater remains within the sewer network instead of reaching WWTPs. In other words, the findings in this study highlight a major stock of MPs in sewer sediments and the associated risk of downstream transfer during wet weather events due to the resuspension of these sediments.

Table 4-1: MP contamination in raw sludge at WWTPs in Europe. Values in the table are presented as min-max or average with standard deviations depending on the available data. The unit is particle per gram of dry-weight sample matrices.

Location	Sample type	Level of contamination (particle/g dw)	Reference
France	Sewer sediment	5 - 178	This study
UK	Raw sludge	107.5	Harley-Nyang et al. (2022)
Italy	Primary sludge	1.67	Pittura et al. (2020)
	Secondary sludge	5.3	
Spain	Raw sludge	133±59	Edo et al. (2019)

4.3.2. Polymer composition and size distribution

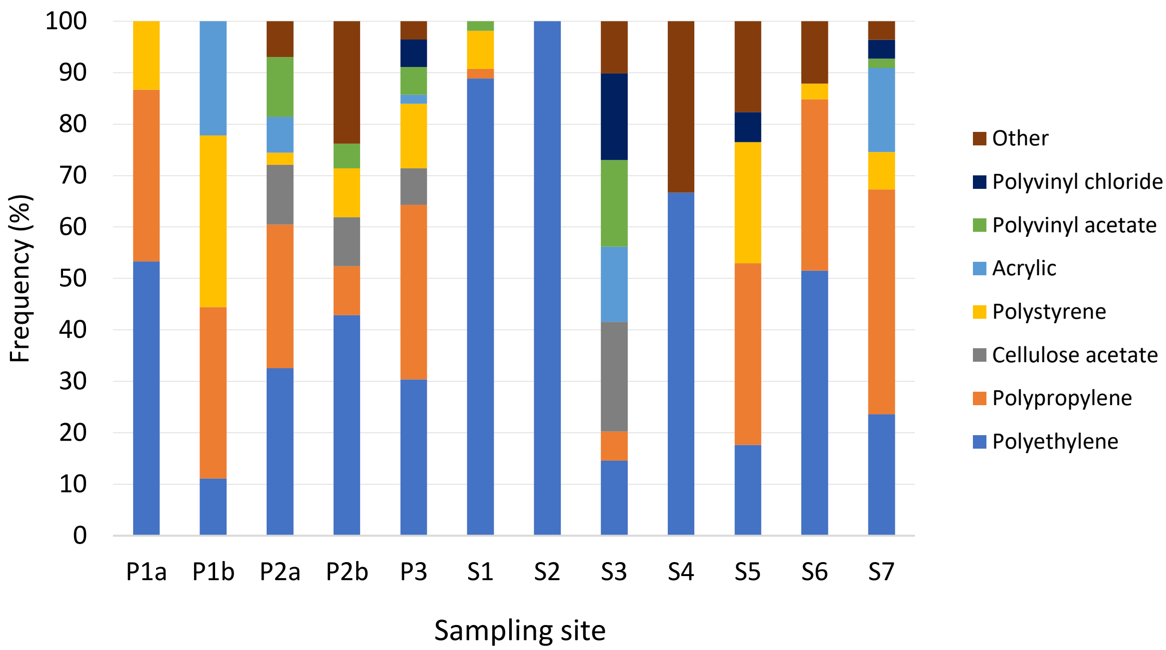

Figure 4-3: Polymer composition of MPs in sewer sediments. The frequency was calculated based on the number particle of each polymer type over the total number particles in each sample. 'Other' group includes ABS, Polyamide (PA), Polyesters (PEST), Polyurethane (PU) and Vinyl copolymer (VC)

Twelve different polymer types were found in sewer deposit samples. Polyethylene (PE) is the most common, present in all samples, ranging from 11 % in P1b to 100 % in S2. Polypropylene (PP) and polystyrene (PS) are also common, found in 10 and 9 out of total samples, respectively. Cellulose acetate (CA) was found abundant in S3 up to 21 %, whist being absent in most samples. The similar phenomenon was observed for Acrylic, Polyvinyl acetate (PVAC) and PVC. Other polymers including ABS, PA, PEST, Polyurethane (PU) and Vinyl copolymer (VC) were present with much less frequency, only accounting for 7 % of all detected plastic particles (n=424).

Polymer composition differs from sample to sample (Figure 4-3). While some samples (e.g., P2a, P3, S3) contained 8 to 9 different polymers, S2 was composed of PE exclusively. Researches on sewage sludge also found PE and PP abundant in their samples, which is consistent with the results in this study (Vollertsen & Hansen 2017; X. Li et al. 2018; Liu et al. 2019; El Hayany et al. 2020). However, they also found a large amount of PA or nylon in sludge which might stem from laundry activity of textile products. PA was present in sewer sediments in this study, however, only in 4 out of 12 samples, accounting for less than 2 % of total detected particles. Apart from PP, PE and PA that are considered as low-density polymers, high-density types such as PEST, CA and PVC were detected in sewer sediments, which is different from polymer composition of sewage sludge. This can be attributed to lack of harmonization in methodology. For instance, some studies only perform chemical characterization for a small part of suspected particles due to limit of contemporary technological advance (Gies et al., 2018; X. Li et al., 2018). Some applied NaCl (1.2 g/cm^3) in density separation which did not allow to recover all polymer types (X. Li et al., 2018; Liu et al., 2019). Besides, the diversity of polymers found in this study suggests that different and complex mechanisms might contribute to the accumulation of MPs in sewer deposits rather than gravity sedimentation only. For example, the presence of organic particulate matter in wastewater might facilitate the development of biofilms on the surface of plastic debris, thereby altering the physicochemical properties of these particles (Kelly et al., 2021; Martínez-Campos et al., 2021; He et al., 2022). The exposure to different forces during conveyance with wastewater inside sewer systems can also influence the sinking behavior and subsequent transport modes of plastic particles (Aghilinasrollahabadi et al., 2021; Jiang et al., 2023).

The variability between different samples was high, both in terms of MP number concentration and polymer composition. Additionally, MP content showed no relationship with POC (R^2 <0.01, Figure A-6). This reflects the heterogeneity of sewer sediments as an environmental matrix. High variability was also frequently observed for different substances in urban water pollution studies, for example, polycyclic aromatic hydrocarbons and metals in stormwater and CSOs (Gasperi et al., 2011; Zgheib et al., 2011). For more accurate insights into MP occurrence and its potential emission during wet weather, future studies are recommended to have a larger sample size across the entire sewer network. This will help identify hotspots and temporal variations of MP pollution, thereby enabling the formulation of mitigation strategies.

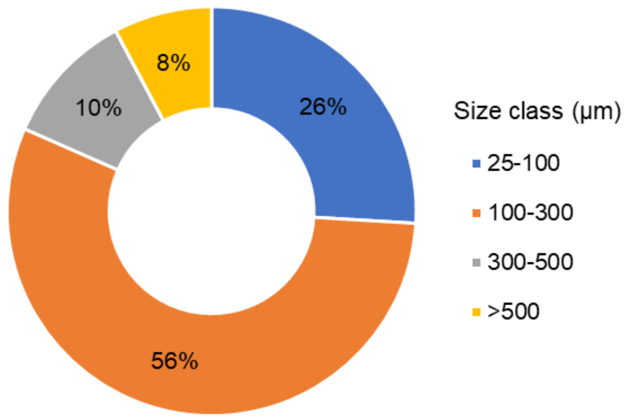

Figure 4-4: Size distribution of MPs in sewer sediments. A total of 424 MPs were detected in this study

Particle size distribution of MPs found in this study is illustrated in Figure 4-4. More than 20 % of total detected particles were smaller than 100 µm and most of them are smaller than 300 µm. These findings are in agreement with previous works studying MPs in sewage sludge (Liu et al., 2019; El Hayany et al., 2020). The smaller particles are, the more likely they attach on the surface of suspended solids or organic particulate matter in wastewater during transport and then accumulate in sewer sediments (Liu et al., 2019).

4.3.3. Estimated contribution of sewer sediments to microplastic contamination in CSOs

Previous studies have indicated the significant contribution of suspended particulate matter from in-sewer remobilization to high contamination levels in wet weather flows. Given the abundance of MPs in sewer sediments, water flow during these intensive occurrences is expected to exhibit high concentrations of MPs (Ashley et al., 2004; Chebbo et al., 1995; Gasperi et al., 2010). Equation 1 was applied to estimate the amount of MPs released from sewer sediments and discharged along with CSOs into surrounding water bodies. Kafi et al. (2008) reported that suspended solids in water samples collected at Clichy, one of two major CSO outfalls in Paris, ranged from 203 to 343 mg/L. While, it is estimated that 20-80 % of suspended solids in CSOs originate from sewer sediments (Gromaire et al., 2001; Passerat et al., 2011; Hannouche et al., 2014). The estimation showed that the resuspension of sewer sediments can result in a minimum concentration of 203 particle/m^3 up to 4.8×10^4 particle/m^3 in CSOs. Stormwater, a main source of pollutants in CSOs, also contained an equivalent amount of MPs, ranging from 3×10^3 to 12.9×10^4

particle/m^3 according to Treilles et al. (2021). These values are in the same range as MP level reported in effluents of WWTPs worldwide, from 250 to 22.5×10^4 particle/m^3 (Leslie et al., 2017; Ziajahromi et al., 2017; Gündoğdu et al., 2018; Gies et al., 2018; Akarsu et al., 2020). Thus, CSO discharges can act as a potential pathway transporting MPs from urban area into the environment besides treated wastewater.

$$[MP]_{CSO}^{SS} = [MP]_{SS} \times C_{CSO}^{SS} \times [Suspended\ solids]_{CSO} \text{ (Equation 1)}$$

In which,

$[MP]_{SS} - MP\ concentration\ in\ sewer\ sediments\ (particle/kg\ dw)$;

$C_{CSO}^{SS} - Fraction\ of\ sewer\ sediments\ in\ CSO's\ suspended\ solids$ (%);

$[Suspended\ solids]_{CSO} - Concentration\ of\ suspended\ solids\ in\ CSO\ (mg/L)$

$$Min\ of\ [MP]_{CSO}^{SS} = 5 \times 10^3 \times 20\ \% \times 203 \times 10^{-3} = 203\ (particle/m^3)$$

$$Max\ of\ [MP]_{CSO}^{SS} = 178 \times 10^3 \times 80\ \% \times 343 \times 10^{-3} = 4.8 \times 10^4\ (particle/m^3)$$

4.4. Conclusion

Part of MPs in urban wastewater entering sewer systems is not transported to treatment facilities, but temporarily stored in sewer sediments. The resuspension of these sediments might release trapped MPs into the water flow. During intensive rainfall events, particulate matter and associated pollutants originating from the erosion of sewer sediment can be carried by CSOs into surrounding water bodies (Verbanck, 1992; Ahyerre et al., 2000; Gromaire et al., 2001). MPs accumulated in in-sewer deposits can also be discharged along with CSOs into the recipients. Thus, this study highlights the necessity of monitoring MP content in sewer sediments to better understand the occurrence and fate of these plastic particles in the sewer network, as well as assess the role of CSOs as a pathway for releasing MPs from urban wastewater into the environment.

Chapter 5

Combined sewer overflows – a neglected pathway of urban-based microplastics to the environment: Case study of the Parisian sewer network

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Abstract

Polluted with microplastics (MPs) due to high density population, urban wastewater threatens ecological health of surrounding water bodies regarding plastic pollution. As untreated wastewater, combined sewer overflows (CSOs) are expected to transport significant number of MPs from urban areas to the environment during intense wet weather events. However, up to date, limited data have been published to confirm this. By analyzing CSO samples during several occurrences within Paris region, this study found out CSO discharges were highly polluted with MPs (25–500 µm), ranging from 67.7 to 391.5 particle/L. Based on event volume discharge, about 6×10^{8} - 8×10^{10} MP particles are released into the Seine River via each CSO event. At an annual scale, CSOs discharge a massive MP load equivalent to treated wastewater, despite much lower discharge flow. This study thereby emphasizes the need of implementing environmental management practices to reduce the impacts of CSOs to surrounding water bodies regarding plastic pollution.

Highlights

- High MP level up to 391.5 particle/L found in CSOs
- An overflow event can release about 6×10^{8} - 8×10^{10} MPs into surrounding water bodies

- Annual contribution of CSO discharges is equivalent to treated wastewater
- PE, PP and PEST are the most common polymers in CSOs
- Most of detected MPs are in the size range <300 µm

Keywords: wastewater, microplastics, combined sewer overflows, freshwater, runoff, sewer sediments

5.1. Introduction

Plastics play an indispensable role in different aspects of human daily life owing to its features. The pervasive use of plastics, especially single-use plastics, and poorly adapted solid waste management systems lead to the introduction of plastic items into the environment. Microplastics, plastic debris <5 mm, have emerged as a contaminant of concern due to their presence being reported in most environmental compartments. Riverine environments have been identified as a crucial recipient of land-based MPs and a potential transport way of these particles to seas and oceans (Horton et al., 2017a; Eo et al., 2019).

Untreated sewage from urban areas constitutes a potential and major source of MPs entering freshwater systems, especially riverine environments. Urban areas are typically equipped with wastewater management systems, which can be either a combined sewer - stormwater system, or a separate system designed to convey sewage and storm water independently. Combined wastewater management systems are commonly found in old town centers, where wastewater and storm water are transported through the same pipe to treatment facilities. During intensive events, the capacity of these systems might become insufficient for conveying water and managing the incoming inflow. As a result, excess water is discharged into receiving water bodies to prevent the overload of the system and to protect treatment facilities. Combined sewer overflow refers to untreated discharges into the environment due to the overload of the combined sewer system. While, literature has documented that high contamination levels in CSOs adversely impact the ecological health of receiving waters (Gasperi et al., 2011; Passerat et al., 2011; Phillips et al., 2012; Launay et al., 2016), research on the quality of CSOs concerning plastic pollution and its associated impacts is limited, with only a few studies on MPs (Dris, 2016; H. Chen et al., 2020; Sun et al., 2023).

MPs have been found abundant in wastewater, runoff and sewer sediments, which are known as sources of pollutants in CSOs. According to Talvitie et al. (2017b), MP concentrations of 390-900 particle/L in were detected in 24-hour composite samples taken from the influent of a WWTP in Finland. The observed variation in MP levels may correspond to the daily activities in households and commercial buildings. MPs have also been detected in industrial wastewater, which is occasionally conveyed through the same network as domestic wastewater; for instance, in discharges from textile wet-processing mills (Chan et al., 2021) or polymer processing plants (Bitter & Lackner, 2020). Besides, stormwater is found to be contaminated with MPs from various sources. During wet weather events, plastic particles can fall from the atmosphere and be carried along with runoff into sewer systems (Dris et al., 2016). The authors estimated that approximately 3.5×10^{10} - 7.6×10^{10} MPs, mainly fibers, fall from the atmosphere on the Paris agglomeration annually. Runoff water also carries particles that have settled on different impervious surfaces, including tire-wear particles, vehicle-derived debris, and fragments of road marking paints, into the sewer systems (Horton et al., 2017b; Treilles et al., 2021). Piñon-Colin et al. (2020), who investigated plastic pollution in stormwater runoff in Tijuana (Mexico), found several hundred MPs for different land uses. Meanwhile, Sun et al. (2023) documented high MP contamination up to over 5,000 particle/L in urban surface runoff from residential areas. In addition, high levels of MPs up to 178 particle/g dw were discovered in sewer sediments inside the Parisian combined sewer systems (Nguyen et al., submitted). The resuspension of these sediments during wet weather events is expected to release plastic particles into the water flow, leading to their discharge into the environment in the event of CSOs. Sun et al. (2023) also highlighted the potential contribution of in-sewer deposits to MP contamination in wet weather flows. Thus, CSOs are expected to be an important pathway for the transport of MPs from urban areas to surrounding freshwater environments.

Literature have documented an increase in MP pollution levels in various receiving waters due to CSO discharges. For example, Forrest et al. (2022) reported a sevenfold increase in MP level in watercourses downstream of a combined sewage outfall during one storm event in Ottawa, Ontario, Canada, compared to ambient conditions. Similarly, Rowley et al. (2020) established a strong relationship between MP contamination in the water column of the Thames River and CSO discharges from a nearby wastewater pumping station. While these findings illustrate the contributing role of CSOs to MP contamination level in freshwater environments, they are insufficient to estimate the quantity of MPs discharged via this pathway into the environment. To achieve a more accurate quantitative assessment, direct analysis on CSO samples is required. To the best of our knowledge, only a few studies have followed this approach: Dris (2016) collected water samples from a CSO outfall in Paris during three separate events; H. Chen et al. (2020) and Sun et al. (2023) analyzed wet weather flow samples collected from pumping stations in Shanghai during overflow events. Consequently, there remains a knowledge gap regarding MP contamination in CSOs.

In addition to MPs, larger plastics items coined as ‘macroplastics’ also make their way into sewer systems from different sources. Sanitary products containing plastics like wet wipes, sanitary pads and tampons, which are often flushed down toilets, can be found at the screening step of WWTPs. Besides, stormwater also carries plastic litter and debris from streets and other open spaces into sewer systems during wet weather events. Plastic bags, packaging materials and cigarette butts are common items found in runoff. In case of CSO events, these macroplastics can be discharged into the environment through sewer networks. Gasperi et al. (2014) reported a substantial amount of macroplastics, reaching up to 5.1 % by weight, in debris-retention booms deployed in the Seine River. Similarly, large plastic items are found abundant on coarse screens at the pretreatment plant for wet weather flows in Paris (personal communication). While such infrastructure can efficiently remove macroplastics from CSO discharges when equipped, it might not be true MPs due to their small size. To date, no data are available to on the quantity of macroplastics in wastewater, nor on their quantity entering the environment via CSOs.

The sewerage system in the Paris megacity was initially constructed dating as early as 1370 and underwent main development during the 19th century. Expanding and upgrading over time to accommodate the growing population in the agglomeration, the system now encompasses more than 2,600km of pipelines, conveying over 300 million m^3 of rainwater and wastewater per year. With four reservoir tunnels and eight storage basins, the system is capable of storing up to 900,000 m^3 of wastewater before discharging it into the Seine River as CSOs. However, there are still approximately 20 to 40 CSO events occurring per year, causing about 21 million m^3 of untreated wastewater released into the river (Di Nunno et al., 2021). In this study, MP levels in CSOs discharged from the Parisian sewerage into the Seine River during wet weather events were quantitatively assessed. The results aimed to elucidate the contribution role of CSOs in transporting MPs from urban wastewater into surrounding water bodies compared to other point sources, mainly WWTP effluents.

5.2. Materials and methods

5.2.1. Study site

With a population exceeding 10 million inhabitants, Paris megacity is one of the megalopolises in the world. As a local watercourse running through Paris, the Seine River endures intense anthropogenic pressures stemming from agricultural, industrial, and urban activities within this megacity. The flow rate of the Seine in Paris is modest compared to other rivers in France, with an average value of 310 m^3/s (measured at Austerlitz bridge). During low-flow period in summer, the flow rate decreases to approximately 80-100 m^3/s. Downstream of Paris, the average WWTP effluent discharge flow is around 25 m^3/s, contributing to 25-31 % of the downstream Seine River flow. The river receives not only regular treated wastewater from WWTPs, but also CSOs during intensive wet weather events. There are two major outfalls in Paris, namely La Briche and Clichy, located downstream of the city. During CSO events, untreated sewage discharged from Clichy outfall to Seine River averaged 30 m^3/s (data for 2022) or reached up to 40 m^3/s during the event on August 7th, 2008 (Passerat et al., 2011), thereby strongly impacting the flow dynamics of the Seine River.

The Seine represents an inland water system profoundly affected by urban wastewaters generated within their catchment area. Therefore, Paris region with Seine River provide a good case study to investigate the transport pathway of MPs from urban areas into freshwater environment, especially in the context of CSO discharges.

5.2.2. Sample collection

Sampling was carried out at La Briche from April to October 2022, allowing 16 CSO samples collected during different heavy rain events, named chronologically Event 1 to Event 16. Seven samples were additionally taken at Clichy during event 1, 5, 6, 7, 10, 11 and 16 for comparison. Available data on the main characteristics of the sampling events and the global parameters of water samples are provided in Table A-6.

One composite sample was collected per rain event, combining several 100-mL aliquots taken within an interval of 5 minutes using an automatic sampler Hach™ AS950 or an interval of 15 minutes with Hach™ SD900. The composite sample is expected to be representative for the whole event.

5.2.3. Sample processing and sample analysis

About 1 L of collected samples were first filtered through a 500 µm metallic mesh for fractionation. The mesh containing the fraction larger than 500 µm was stored in a Petri dish, while a subsample of the smaller fraction was kept and concentrated on 10 µm metallic filter (Figure 5-1). These two fractions were then processed separately as follows.

5.2.3.1. Size fraction 500–5000 µm

The 500 µm stainless steel mesh was inspected under a stereomicroscope (Leica M125C, 8-100x magnification). Each potential plastic particle was photographed to document its shape and size with the help of Histolab software (version 11.5.1). These particles were then analyzed for composition identification using FTIR spectroscopy. As complex refraction may occur when particles with irregular shapes are irradiated under reflection mode, leading to the production of uninterpretable spectra (Harrison et al., 2012), transmission mode is generally preferred to analyze MP samples. However, particles larger than 500 µm are unsuitable for FTIR measurement in transmission mode because of their thickness. Therefore, ATR mode was employed. The measurement was performed on a Nicolet iS5 spectrometer coupled to an iD7 ATR–Diamond accessory. The wavenumber ranged from 4000 – 400 cm^{-1}, and 16 scans were carried out with a spectral resolution of 4 cm^{-1}. The data were then analyzed with the help of the software OMNIC (Thermo Fischer Scientific), in which the obtained spectra was compared to reference database with a score out of 100 returned as the goodness of match. A particle was considered to be successfully identified when the match was higher than 70.

5.2.3.2. Size fraction 25–500 µm

Material retaining on 10 µm filter underwent a sequencing enzymatic treatment with sodium dodecyl sulfate (SDS), protease and cellulase for MPs extraction. The protocol was adapted based on the study of Löder et al. (2017), aiming to gently remove organic matter in CSO matrix without harming plastic particles. In detailed, sample was first incubated with SDS at 45 °C for 24 hours to break down biological residues in sample matrix, thereby enhancing the efficiency of the following treatments with an increase in contact surface. Protease treatment was then applied to facilitate decomposition of proteins, in which the enzyme was mixed with TRIS buffer in a ratio of 1:1 (v/v) to achieve the optimum pH condition suggested by the manufacturer (i.e., pH 9.0). Next, cellulase treatment was performed targeting cellulose-based residues in sample, and acetate buffer was added to maintain pH at 4.5. Treated samples were then deposited on Anodisc filters (pore size 0.2 µm) and analyzed using an automated µ-FTIR imaging (Nicolet iN10 MX, Thermo Scientific). Due to the 25×25 µm pixel resolution of the detector used in this technology, particles down to 25 µm were analyzed. Data processing was carried out later with the siMPle software (version 1.1.β, Aalborg University, Denmark and Alfred Wegener Institute, Germany) to identify the polymer composition of the particles.

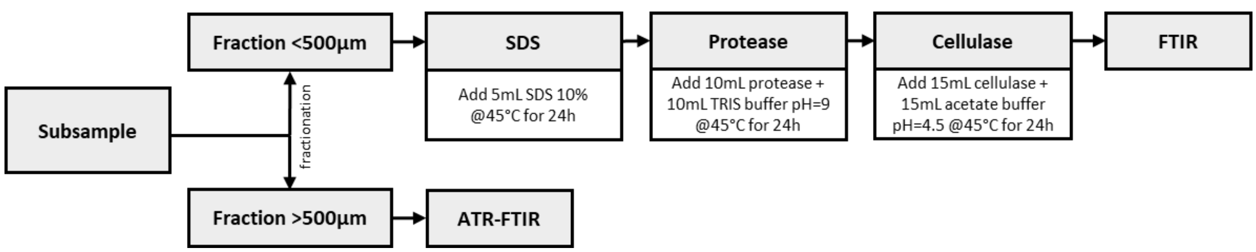


Figure 5-1: Pretreatment protocol for MPs extraction

5.2.4. Blank control

To minimize sample contamination during lab processing, cotton lab coats and nitrile gloves were worn. Samples were handled under a clean bench with laminar flow and covered with aluminum foils whenever possible. Glass apparatus were burned at 525 °C for 2 hours and metalware were rinsed thoroughly with ultrapure water before use. All solutions in direct contact with sample (i.e., enzymes and buffers) were filtered through GF/D (pore size 2.7 µm). In addition, two procedural blanks for the fraction <500 µm were carried out to monitor potential contamination. Only two PP particles were detected, showing negligible contamination.

5.3. Results and discussion

5.3.1. Microplastics >500 µm in CSOs

Analysis of size fraction >500 µm detected MPs in 11 out of 23 CSO samples. Only 14 particles were determined to have polymer composition among potential particles sorted out from 500 µm mesh. Number of particle ranged from 1 to 3 MPs per sample, corresponding to a concentration of 0.9-3.2 particle/L. Detected MPs>500 µm were transparent or in color such as green, red, black and yellow. The particle size ranged from 1200 to 4700 µm with fragments as the most common shape. The images of some MPs>500 µm are shown in Figure 5-2. Several fibers were also observed entangled on the mesh, making it infeasible to sort out individual fibers for chemical identification.

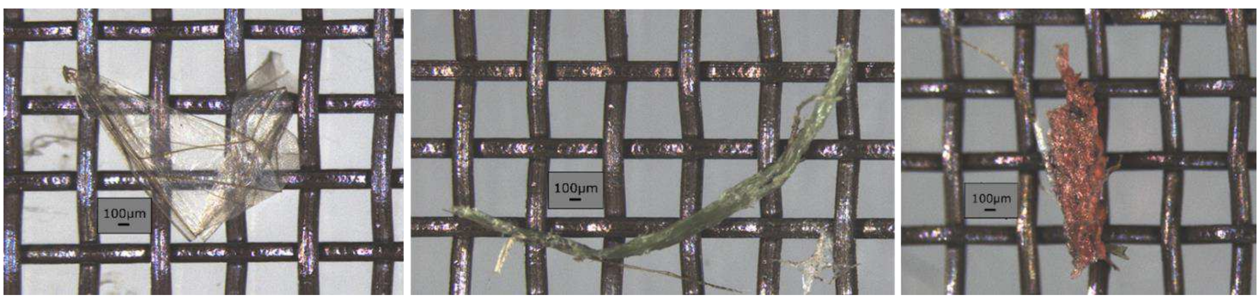
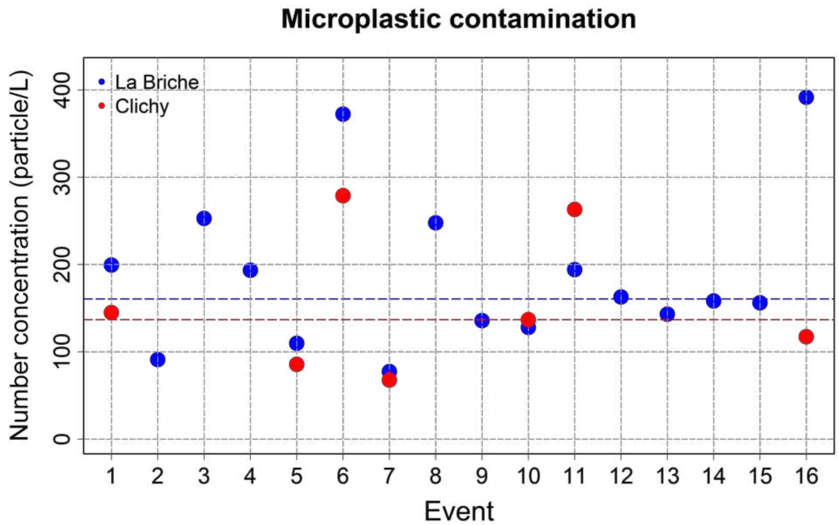


Figure 5-2: Exemplars of MPs>500 µm detected with ATR-FTIR. The black bar has a scale of 100 µm. From left to right: PA, PE and PP

5.3.2. Microplatics <500µm in CSOs

Most MPs were detected with µFTIR with a total of 1973 particles for all samples. Polymer composition of these particles was determined by comparing their spectrum to a reference library of different polymer types which is integrated in the siMPle software. The particle size was also estimated based on the resolution of the detector, with a pixel size of 25×25 µm. Since the amount of MPs <500 µm provides a sufficient sample size for data analysis, they were used for visualization and discussion of polymer composition and size distribution later on.

5.3.2.1. Concentration level



Site	Min - Max	Average	Median
La Briche (n=16)	77.3 - 391.5	188.3	160.5
Clichy (n=7)	67.7 - 278.8	156.3	136.8

Figure 5-3: Concentration of MPs (25-500 µm) in CSOs (dashed line: median value; unit: particle/L)

Figure 5-3 indicates the abundance of small-sized MPs in all CSO samples with a mean concentration of 178.6 particle/L and a median concentration of 156.2 particle/L for all events regardless of sampling site. The highest MP concentration was recorded at La Briche for the event on October 17th, 2022 with 391.5 particle/L. It is nearly 6 times higher than the lowest value documented at Clichy on June 30th, 2022 with 67.7 particle/L. The slight difference in MP levels in CSOs between two outfalls can be attributed to various factors, including their distinct catchment areas, the proportion of wastewater and runoff in the water flow, the antecedent dry periods before the events, and the strategies implemented in the systems' operation, which are determined by SIAAP in this case.

Dris et al. (2016) reported 35 – 3100 particle/L of discharge for three events at the same outfall (i.e., La Briche). While sample collection and sample processing were similar in both studies, methods used for particle detection and characterization were different. This inhibits the comparison of results between two studies. In addition, the discrepancies in obtained data may result from the differences in the specific characteristics of each studied event (e.g., rain intensity, precedent dry days, etc.). Dris et al. (2016) assumed a long period of heavy rainfall before sampling was attributed to a decrease in the amount of MPs detected.

MP level in this study was found to be higher compared to the findings in Treilles et al. (2021), which focused on rainwater overflows from a separate sewage system serving a suburban area in the Paris region. The concentrations reported in Treilles et al. ranged from 3 to 129 particle/L (min-max) with a median of 29 particle/L. This highlights the significant contribution of wastewater to plastic pollution in sewage overflows stemming from combined systems. However, this study employed a lower limit detection of 25 µm, compared to 80 µm in Treilles et al. (2021), allowing the detection of smaller-sized particles. Using a similar methodology in sample collection and identification technology, a study in Shanghai (China) documented an abundance of MPs up to 8,500 particle/L in CSOs (H. Chen et al., 2020), which is nearly two orders of magnitude higher than the findings of the present study. The combined sewer system in Shanghai city not only receives daily a large amount of domestic wastewater from over 24 million habitants, but also industrial wastewater, which is not the case of Paris region. In addition, the difference in consumption habit, products available in household environment and waste management between two studied sites may lead to different MP levels found in CSO discharges. However, it should be noted that the variance in the methodology used for sample processing in the two studies limits the comparability of the reported results.

5.3.2.2. Polymer composition

A total of 14 different polymer types were detected in CSO samples. PE and PP were the most common, accounted for 45 % and 38 % of total particles, respectively. PEST and PS were found with much lower frequency. Some other polymers were also detected, including PA, ABS, acrylic, alkyd, CA, pan_acrylic fiber, PU, PVAC, PVC and VC. These polymers are combined in 'other' group, constituting 6 % of total particles.

The diversity in sample composition ranges from 4 polymers at Event 8 to 12 polymers at Event 15, both at La Briche. PE and PP were present in all samples, accounting for 64 % (Event 10 at Clichy) up to 94 % (Event 4 at La Briche). There are similarities in composition of samples from two sampling sites, except PEST were found more frequently in Clichy than La Briche. It can be attributed to the difference in the fraction of domestic wastewater in CSOs between two sampling sites.

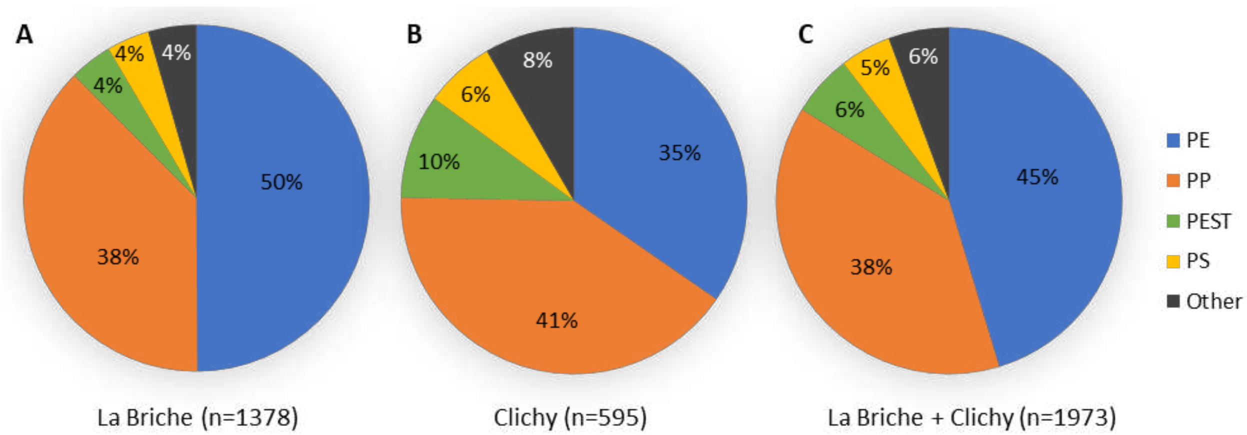


Figure 5-4: Polymer composition in CSOs for the events at La Briche (A), Clichy (B) and for all events (C)

PE, PP and PEST were dominant in present study, which is consistent with the findings of previous studies on CSOs (Sun et al., 2023); raw wastewater (Lares et al., 2018; Simon et al., 2018); and runoff water (Piñon-Colin et al., 2020). However, much smaller amount of PEST was found compared to Sun et al. (2023). This can be attributed to the difference in methodology used between two studies.

5.3.2.3. Size distribution

Particles smaller than 300 µm accounted for the largest proportion in CSO samples. In which, most of these particles are in a range of 25-100 µm (up to 59 % on average). There are similarities in size distribution of particles found from both studied sites. Despite the difference in methodology, the abundance of small-sized MPs under 500 µm up to 50 % was also reported in Sun et al. (2023).

The findings in this study showed that MPs in CSOs which came from different sectors of the Parisian sewer system were similar in number concentration, polymer composition and size distribution. This demonstrates the homogeneity of water flow in the system regarding plastic pollution level. It could be advantageous in the process of developing a model to predict the emission flux of MPs from the entire system into the receiving water.

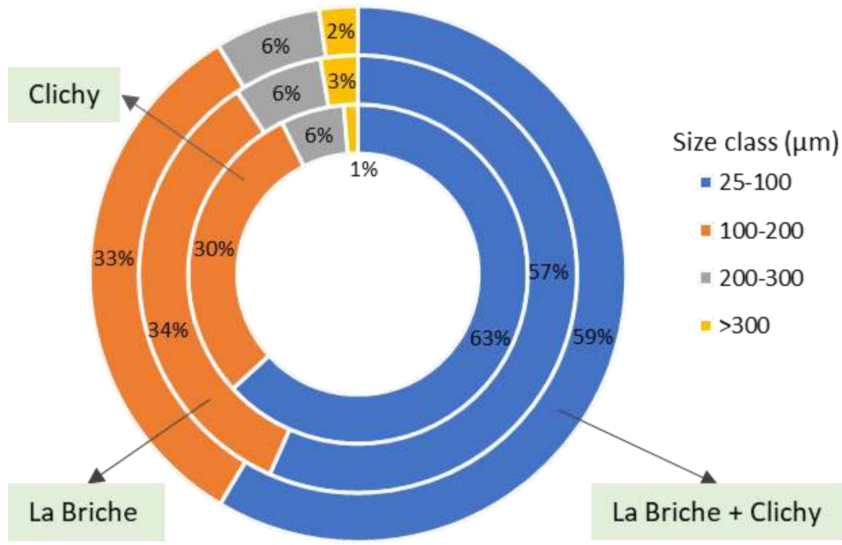


Figure 5-5: Size distribution of MPs (25-500 µm) in CSOs

In addition, a negligible amount of MPs >500 µm was documented in CSO samples compare to the smaller fraction. This suggests that a larger volume of water samples need to be inspected in future studies to gain more insights on the occurrence of MPs >500 µm.

5.3.3. Relation between microplastic levels and wastewater – stormwater proportion in CSOs

The proportion of surface runoff and wastewater in CSO discharges were estimated for each event based on the electrical conductivity (Table A-7). The estimation was carried out based on the hypothesis that the conductivity of wastewater ranges from 1050 to 1170 µS/cm, while that of stormwater varies from 80 to 150 µS/cm. This approach was applied in several studies on (micro)pollutants in CSOs such as Passerat et al. (2011), Gasperi et al. (2012) and Launay et al. (2016). Electrical conductivity appeared as an appropriate parameter for estimating the mixture ratio of wastewater and runoff thanks to its conservative nature (Gasperi et al., 2012; Launay et al., 2016). Estimated fraction of wastewater in CSO samples in this study ranged from 15-78 % for hypothesis 1 and 23-90 % for hypothesis 2, showing the variation of runoff/wastewater in CSOs from different rain events. Besides, the correlation between MP content in CSOs and different parameters (e.g., total of discharge volume, max discharge flow rate, COD, etc.) were established to identify the impacts of rain event on plastic pollution level in CSOs (Figure A-7).

The results did not reveal a clear pattern between MP content and proportion of wastewater in CSOs, nor with other parameters. This suggests the intricate interplay of various factors on MP contamination levels in CSOs, making it complicated to establish a direct correlation between rain event and MP levels. Indeed, the quality of CSOs is influenced by a combination of pollutant sources, including wastewater, runoff and resuspension of sewer sediments. While wastewater reflects the characteristics of the sewer catchment (e.g., population, land-use, economical status, etc.), the other sources are dependent on weather conditions, the extent of impervious areas within the catchment, the design of sewer systems and the global wastewater management practices carried out by operators. In particular, the resuspension of sewer sediments during wet weather events is expected to contribute a substantial part of MPs in CSOs according to the findings in Nguyen et al. (submitted), which reported high accumulation of MPs in the in-sewer deposits inside the Parisian sewer system.

5.3.4. Microplastic fluxes discharged via CSOs at the scale of Parisian megacity

Given an approximate volume of 21 million m^3 of CSOs discharged from the Parisian sewer system into the Seine River per year during wet weather events, an annual MP flux discharged via CSOs was estimated based on the concentration values (min - max) found in this study, ranging from 1.4×10^{12} to 8.2×10^{12} particle/year.

Treilles et al. (2022), which reported an increasing MP flux from upstream to downstream of the Seine River up to 6.2×10^{14} particle/year, highlighted the significant contribution of WWTP effluents and untreated stormwater as sources of MPs entering the river. However, the findings in this study indicate the significant role of CSOs as another input of plastic pollution in the Seine.

Extrapolations from the literature, based on per capita MP load, allowed the estimation of the annual MP flux discharged via WWTP effluents into the Seine River within the Paris megacity. These estimates ranged from approximately 2.4×10^{11} to 2.3×10^{13} particle/year (Table A-8), providing insights into the equivalent contribution of CSOs to plastic pollution levels in the receiving water, along with WWTP effluents. While discharges from treatment facilities are regular with relatively low concentrations, CSOs are more intermittent, short-lived and emit higher concentrations, which may induce acute impacts on the receiving water bodies. Previous studies also emphasize the significant role of CSOs into MP levels in the recipient compared to WWTP effluents. For example, H. Chen et al. (2020) reported the total annual MP released via CSOs was significantly higher than that discharged from WWTP effluents in Shanghai (China), with figures of 8.5×10^{14} particle/year compared to 1.43×10^{14} particle/year. In another study in Nanning (China), the highest peak of MP abundance in CSOs was found to be 10 times higher than that in treated wastewater from WWTPs (Zhou et al., 2023).

In addition, data obtained in Stratmann et al. (in preparation) enabled the estimation of an annual suspended MP flux in the downstream of the Seine River downstream at approximately 1.1×10^{13} particle/year. Since minimum impacts from overflows were recorded on sampling days, this estimated value can be considered as the background level. Thus, with an annual MP flux up to 8.2×10^{12} particle/year, CSO discharges can contribute up to 70 % of the MP flux level in the Seine River, assuming no sedimentation occurs.

5.4. Conclusion

This study found that CSOs, untreated discharges from the Parisian combined sewer systems, were highly contaminated with MPs. During intense wet weather events, a substantial quantity of MPs was emitted along with CSOs into the Seine River. At an annual scale, the contribution of CSOs to MP pollution in the river is estimated to be equivalent to that of WWTP effluents. Together, they serve as major sources of MPs entering the freshwater environment. Therefore, the maintenance and upgrading of sewer systems are required to prevent CSO events and their impacts, especially on the level of plastic pollution in receiving waters. In addition to MPs, the inclusion of data on macroplastics is essential for a comprehensive quantitative assessment of plastic waste released into the environment. These large items make a substantial contribution to the overall mass flux and play an important role in the generation of smaller plastic debris later on.

Chapter 6: Limitations, conclusions and perspectives

This work aimed to enhance the knowledge on the occurrence and fate of MPs in the Parisian wastewater management system. Various compartments of the system were therefore investigated, namely the sewer network and WWTPs with a focus on the sludge-line treatment. MP analyses were carried out for various environmental matrices, from sediment, sludge to wastewater. The same identification methodology was applied, enabling a deep comparison of the obtained data. However, methodological limitations were encountered throughout the study, from sampling, pretreatment to analysis.

6.1. Limitations of the methodology

6.1.1. Sample collection

Grab sampling was the method employed to collect sewer sediments and sewer sludge in this study. Sediments samples, extracted from twelve different locations within the Paris City's sewer network, were assumed to be representative of the in-sewer deposits throughout the entire system. Similarly, sludge samples were presumed to reflect the general characteristics of the matrix under normal operating conditions of the studied plants. It is important to note that WWTP is a complex system with multiple treatment units operating in parallel at each stage. Thus, the representativeness of the obtained samples for the overall studied treatment step was assumed.

Regarding CSOs, composite water samples were collected using autosamplers, capturing the quality of the discharge throughout the entire studied events. However, sample volumes for MP analysis were limited due to the need of sample fractions for multiple control measurements.

The majority of samples were collected by technicians at the study site, following an agreement with the author regarding both the selection of sampling points and the sampling methods. This arrangement was necessary due to technical constraints and restricted access to sampling sites. Even though it was the best solution, these constraints limited the study design.

6.1.2. Sample processing

This study encompassed various sample matrices, including sediments, sludge and wastewater. Each matrix possessed specific characteristics, but they generally contained high levels of organic matter. Therefore, various protocols were applied for sample pretreatment, aiming to reduce the organic and inorganic content in the sample matrix. Given the absence of a standard protocol for MP analysis in environmental samples, this study adapted existing protocols from previous studies and incorporated recommendations from the literature to achieve reasonable sample purification without causing harm to plastic particles. For instance, a temperature below 50°C was maintained during sample processing, and the use of strong base/acid was avoided. However, the recovery rate of these protocols was not addressed; only blanks were carried out to monitor potential contamination during lab processing.

For sewage sludge, a single protocol was applied for different sludge types to ensure data comparability. However, the protocol appeared to be less efficient at reducing impurities in the sample matrix of the final treated sludge compared to other sludge types. This resulted in a smaller amount of successfully analyzed treated sludge, indicating the need for modifications in the extraction protocol for treated sludge in future studies.

6.1.3. Sample analysis

The detection of plastic fibers was limited with the methodology applied in this study. While the analysis of the fraction <500 µm using µ-FTIR and siMPle software did not allow to distinguish between fragments and fibers, identifying particles in the larger fraction >500 µm, especially fibers, was also challenging. Because of their thin and long shape, fibers were difficult to handle for ATR measurements. The obtained signal was weak due to the small contact surface between the fiber and the crystal. Only a few fibers were successfully identified. Different analysis methods used for two size fractions 25-500 µm and 500-5000 µm inhibited the combination of two obtained datasets. Visual sorting and analysis of particles one by one particle with ATR are a manual work which is highly dependent on the operator, potentially introducing human bias. In contrast, the application of µ-FTIR imaging reduces human bias through automatic analysis, however, this method is time-consuming. Because of this, only a small subsample could be analyzed, especially for complex matrices such as sewage sludge. This causes uncertainties in extrapolating the obtained data due to questions regarding the representativeness of the subsamples.

In addition, technical errors occurred occasionally during the analysis using µ-FTIR and siMPle software. One of the observed issues was the false assignment of PE particles, as illustrated in Figure 6-1. Samples experiencing this issue were considered unsuccessful, consequently, they were discarded. Reprocessing these samples was limited due to time constraints, leading to a reduction in the sample size in this study. The problem with false PE signals was predominantly observed in sludge samples, which can be attributed to the inefficiency of the sample processing step in reducing impurities in the sample matrix.

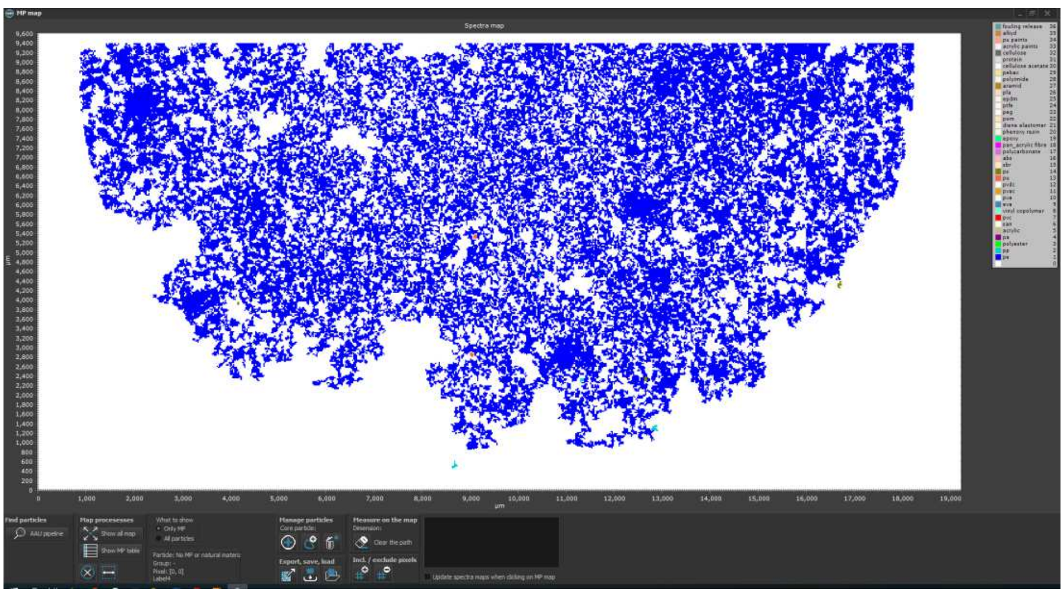


Figure 6-1: PE false signals observed from the analysis of a dewatered sludge sample (siMPle interface)

6.2. Conclusions

This section summarizes the main findings of this PhD study (Table 6-1) with the aim of reducing knowledge gaps on MP occurrence and fate within the wastewater management system at the Paris megacity scale. It therefore provides insights into the contribution of urban wastewater to plastic pollution in the surrounding environments. The three primary objectives of this study were addressed, and their findings were encapsulated in this thesis as follows.

Objective 1

Investigate the occurrence of MPs in sludge-line treatment at WWTPs in the Greater Paris area, from raw sludge to final treated sludge, using various treatment technologies

Assess the impacts of these treatment technologies on MPs

Evaluate the potential of treated sludge as a pathway for MPs entering the surrounding terrestrial environment

MP occurrence throughout the sludge-line treatment at three different Parisian WWTPs were investigated. At all the studied plants, which employ various treatment technologies for high treatment capacity, MP concentration levels in sewage sludge changed insignificantly after passing through these treatment steps. However, impacts of sludge treatment on plastic particles were observed. During thickening and dewatering processes, a portion of sludge-based MPs was released from the sludge back into the water phase, which was then returned to the system along with reject water. Anaerobic digestion showed insignificant impacts on MP particles, both in terms of particle number and particle size. In contrast, high temperatures during thermal treatment appeared to promote the fragmentation of plastic particles, leading to a significant reduction in size, from 120.1 µm in raw sludge to 80.9 µm in the final treated sludge. Sludge treatment may also affect the properties of plastic particles, which was not only observed through a reduction in polymer diversity, especially in the final treated sludge, but also through changes in the spectra of the measured particles, particularly for PE as reported in Chapter 3.

The remaining of MPs in the final treated sludge in this study, following all treatment steps, indicated the transfer of these particles into the environment via sludge disposal. For example, MPs can be released into the terrestrial environment via soil application in agriculture, with an estimated 3.2×10^{13} particles emitted via this pathway within the Paris region yearly. In summary, it can be concluded that current sludge management systems are inefficient at completely removing MPs from sludge, and sludge disposal acts as an important source of MPs entering the environment.

Objective 2

Understand transfer of MPs to wastewater treatment plants by monitoring their accumulation in sewer sediments inside the Parisian sewer network

Assess indirectly the contribution of in-sewer process to the quality of CSOs concerning plastic pollution

The presence of MPs in sewer deposits within the Parisian sewer system was detected, with high contamination levels up to 178×10^3 particle/kg dw. This study confirmed the separation of MPs from the water phase when travelling through the sewer network, which led to the settling down of MPs with other particulate matter in wastewater and their temporary accumulation within sewer deposits.

The increase in pollution levels in water flow, resulting from the resuspension of in-sewer sediments during wet weather events, has been documented in the literature. Since in-sewer sediments appear as a significant stock of MPs within the sewer system with the findings of this study, the release of MPs with other pollutants during wet weather events is expected. This may not only increase the influent load arriving at WWTPs, but also cause downstream emissions in case of the untreated wastewater's discharge during intensive events. An attempt to estimate the contribution of in-sewer processes to the quality of CSOs concerning plastic pollution, based on suspended solids, was carried out. However, the precision of this estimation is limited due to the large variability of suspended solids which are shared by CSOs and in-sewer sediments.

Objective 3

Investigate the emission of MPs along with CSOs into the Seine River during intensive wet weather events

Evaluate the contribution role of CSOs to the level of plastic pollution in receiving water bodies compared to other point sources, mainly WWTP effluents, in the scale of Greater Paris area

MP pollution levels in CSO discharges from the Parisian sewer systems into the Seine River during 16 intensive rain events were monitored. The findings showed high concentrations ranging from 67.7×10^3 to 391.5×10^3 particle/m^3 of discharge in CSOs. It is estimated that up to 8×10^{10} particles could be released into the river during one event. An annual flux is estimated to reach 1.4×10^{12} – 8.2×10^{12} particle/year. The obtained results underscored the contribution role of CSOs to plastic contamination level in the river, along with WWTP effluents, despite their much lower volume of discharge. In summary, the study confirmed that CSOs are a major input of MPs from urban areas into the receiving environments.

The finding of the study does not allow to conclude the relation between MP levels in CSOs and MPs release from the resuspension of sewer sediments. Although resuspension of in-sewer deposits is expected to contribute significantly to pollutants including MPs in CSOs, solid concentration of MPs in sewer sediments were found to be much lower than that in CSOs. However, this comparison is limited due to the difference in particle size distribution in two sample matrices. The highest frequency of MPs>300 µm was observed in sewer sediments, showing gravity sedimentation plays a role in the accumulation of MPs inside the sewer network. This is consistent with the abundance of high-density polymers detected in sewer sediments, such as PVC and CA. In contrast, much higher frequency of MPs<100 µm was found in CSOs. This indicates the small-sized particles are favored to be swept away with water flow. It can also be hypothesized that there are changes in particle size of MPs during their stay inside the sewer network under various influencing factors: water flow, humidity and microorganism growth. However, the mechanism of sedimentation – resuspension of in-sewer particulate matter is much more complex while the similarity in behavior of MPs and other particles in wastewater is still unclear.

Table 6-1: Summary of the findings on MP occurrence and fate in this thesis

Sector	Sewer network		WWTP – Sludge-line treatment				
Sample matrix	Sewer sediments	Combined sewer overflow	Raw sludge	Digested sludge	Dewatered sludge	Final treated sludge	Reject water
Concentration level (min-max)	5.0 – 178.0 (particle/g)	67.7 – 391.5 (particle/L)	170.9 – 488.7 (particle/g)	239.1 – 360.6 (particle/g)	231.8 – 493.3 (particle/g)	86.5 – 450.2 (particle/g)	52 – 1120 (particle/L)
Converted value	–	130.8 – 1506.7 (particle/g)	–	–	–	–	18.7 – 402.3 (particle/g)
Size distribution 25-100 µm 100-200µm 200-300µm >300µm	 25.9% 37.5% 18.2% 18.4%	 58.6% 32.7% 6.4% 2.3%	 37.8% 33.9% 16.8% 11.5%	 39.5% 35.8% 12.8% 11.9%	 43.4% 35.6% 10.2% 10.8%	 67.7% 21.5% 6.5% 4.3%	 36.6% 37.8% 12.9% 12.7%
Dominant polymers	PE, PP, PS, acrylic, CA, PVC	PE, PP, PEST, PS	PE, PP, PEST, PS, PVC	PE, PP, PEST, PS	PE, PP, PEST, PS	PE, PP, PS, PVAC, PU	PE, PP
Polymer diversity (types)	12	15	12	13	13	7	11

Based on the results obtained in this study, the annual MP fluxes entering the environment via different pathways from the Paris megacity were estimated, as illustrated in Figure 6-2. The estimation for the MP flux via sewer sediment management were derived for the scale of Paris City only, ranging from 6.0×10^{9} to 4.7×10^{11} particle/year.

Among the various sludge disposal options available in the Paris megacity, land application and composting are expected to transfer MPs from WWTPs into terrestrial ecosystems. Based on the available data on sludge management in SIAAP's WWTPs, approximately 61,900 tons of treated sludge from Seine Aval directly spread in agricultural fields each year. Given the MP levels found in this study, the estimated MP flux ranges from 5.4×10^{12} to 2.8×10^{13} particle/year. In addition, about 32,000 tons of produced sludge are composted before being used for soil amendment, leading to an estimated MP flux of 2.8×10^{12} to 1.5×10^{13} particle/year into the environment. The annual MP flux via CSOs is estimated to range from 1.4×10^{12} to 8.2×10^{12} particle/year.

To provide an overview of other MP fluxes within Paris megacity, which were not investigated in this study, estimates were carried out based on data from the literature. In detail, the annual MP flux in the Seine River was estimated to range from 4.1×10^{11} to 2.0×10^{13} particle/year, as reported by Stratmann et al. (in preparation). MP levels in the river water were monitored at three different locations in Paris megacity during three sampling campaigns conducted from November 2021 to July 2022. These campaigns took into account the impacts of Paris City and treated wastewater from WWTPs, with little to no impact from CSO discharges documented. In addition, the annual MP flux via WWTP effluents of the Paris megacity were estimated based on MP levels reported in WWTP effluents worldwide, taking into account the treatment capacity and population equivalent of each studied plant. The estimated MP flux ranges from 2.4×10^{11} to 2.3×10^{13} particle/year.

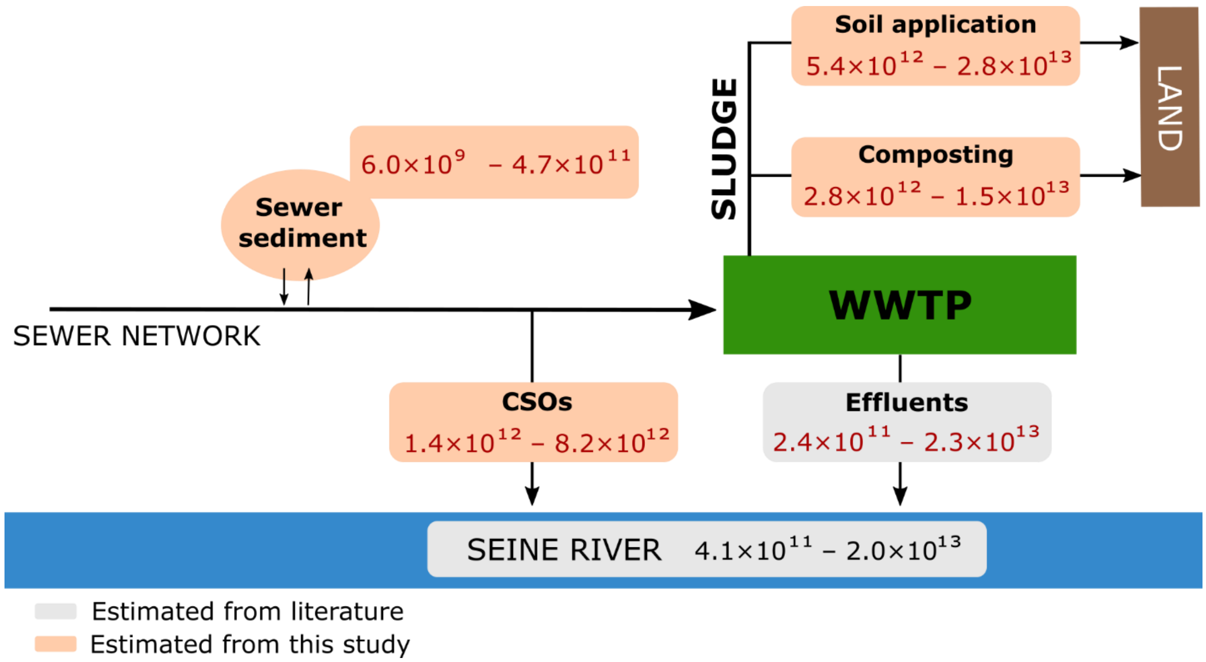


Figure 6-2: The estimated annual MP fluxes emitted from Paris megacity (unit: particle/year)

6.3. Research outlooks and perspectives for future work

The retention of MPs in sewer sediments during transport in the combined sewer system was reported for the first time. This finding implies the complex transport of MPs within sewer systems before reaching WWTPs, which has not been well investigated in the literature. Sewer sediments serve as a stock of MPs within the sewer network, with nearly 3,000 tons of these materials removed during regular maintenance activities per year. However, the efficiency of treatment for these wastes in terms of MPs remains largely unknown, raising concerns about potential emissions into the environment if not handled properly. Since estimation of the total amount of MPs accumulated within the system is limited due to a small sample size and the large variation of MPs level observed in studied samples, future work with a larger sample size is recommended to obtain a precise assessment of the MP budget inside sewer system. In addition, it is necessary to investigate the contributions of different inputs to MP levels within in-sewer deposits, including wastewater and runoff, while taking into account various levels of the sewer network, from municipal collection in secondary sewers to conveyance in primary sewers before reaching WWTPs. This data will be valuable in understanding the distribution of MP accumulation within the system and facilitate appropriate clean-up activities and mitigation measures for reducing MP emissions into the environment.

The contribution of in-sewer deposits' resuspension to CSO quality compared to other inputs like domestic wastewater and runoff water has not been addressed in this study. However, it is important to investigate the downstream transfer risk, particularly via wet weather flows when they are discharged without proper treatment. In other words, further study is required to elucidate the occurrence and fate of MPs inside the sewer network.

The study has confirmed the significant contribution of CSOs to the MP levels in the receiving waters during wet weather events. Therefore, reducing the frequency and the volume of the CSO events are vital to diminish the amount of MPs release into the environment via this pathway. Maintenance activities are essential to reduce operational problems, such as leaking pipes, power outages at pumping stations, and to prevent blockage problems due to the accumulation of sewer sediments.

Other runoff source control management approaches, such as nature-based solutions, infiltration, and storage, can also be promising in significantly reducing the volume of wet weather flows, thereby preventing CSOs and the emission of associated pollutants, including MPs. For example, in case of Paris region, 955,000 m^3 of storage tanks and reservoir tunnels were built (Tabuchi et al., 2020), and a basin with a capacity of 70,000 m^3 is under construction at Clichy site to increase the volume of excess water stored in wet weather conditions. Additionally, WWTPs are operated in a wet-mode, for instance, with application of ballasted flocculation processes at the Seine Aval WWTP, allowing larger volume of water flow treated. Moreover, the operation of a real-time control system (MAGES - Modèle d'aide à la gestion des effluents du SIAAP) provides better control of water pollution caused by CSOs via managing temporary storage systems and optimizing treatment capacity of WWTPs (Tabuchi et al., 2020).

CSO discharges, together with WWTP effluents, represent major inputs of MPs from urban areas into the surrounding freshwater environment. In particular, due to the intensive discharge during short-time periods, CSOs may have acute impacts on the ecological health of the recipient regarding plastic pollution. Future work is recommended to elucidate this hypothesis. Apart from CSOs, other urban discharges, which were not included in this study, should also be investigated regarding MP transfer into the environment; for instance, industrial wastewater, for which data is often lacking.

Although a part of MPs in wastewater remains inside the sewer network, a substantial amount of them reaches WWTPs. This is demonstrated by the high concentration of MPs found in raw sludge, which is separated from the water phase during various water treatment stages. This result is consistent with previous findings (Edo et al., 2019; Alavian Petroody et al., 2021; Harley-Nyang et al., 2022), thereby highlighting the important role of sludge-line treatment in dealing with MPs arriving at WWTPs. The current sludge treatment appears to concentrate MPs rather than removing them. Indeed, while water and biomass in the sludge are reduced through different sludge treatment technologies, MPs particles in this matrix appear to remain abundant, with insignificant changes in their concentrations. However, a reduction in particle size was observed. This may result in particle falling below the size detection limit, leading to the false implication of MP degradation or removal in WWTPs. This finding also highlights the impacts of treatment technologies that increase the number of small-sized particles released into the environment, even on the nano-scale.

Due to the lack of data on MP levels in WWTP effluents in Greater Paris area, the contribution of the two main discharges from WWTPs cannot be directly compared. However, based on the estimated values from the literature, it is likely that sludge disposal via soil application is responsible for the higher annual MP flux emitted into the environment (Figure 6-2). Plastics tend to persist over extended periods in the environment due to their resistant nature. In addition, the transfer of MPs from agricultural soils to other environmental compartments is limited, as indicated by Schell et al. (2022). The authors found that the infiltration capacity of MPs into deeper soil layers or underground water was low, and surface runoff had a negligible influence on transferring these particles into surrounding watercourses. In this context, rapid accumulation of MPs in terrestrial ecosystems, leading to soil contamination, is expected. Therefore, considering other options for managing final treated sludge is required to reduce the number of

MPs released into the terrestrial environment. Incineration may be the best option to completely remove MPs. According to Geyer et al. (2017), incineration can ultimately convert polymers into carbon dioxide and other mineral elements at high temperatures up to 800 °C, thereby eliminating completely plastics. However, it should be noted that incineration is a high-cost option for sludge disposal, which averaged around 75 EUR/ton in wet weight and 290 EUR/ton in dry mass according to A. Long et al. (2021).

Besides sewage sludge, waste from screening and grit-grease removal steps are also byproducts of water treatment, which contain MPs and may act as other inputs of MPs into the environment. Since not many studies have paid attention to these wastes, future work is recommended to investigate this topic. The data will be valuable in calculating the MP budget within WWTPs, thereby identifying significant point sources from WWTPs.

Overall, the removal of MPs from urban wastewater appears to be a complex and inefficient process. Upgrading current management systems demands significant investments in terms of time, effort and resources. The most efficient and effective approach to address the issue is source control, which entails reducing and ultimately eliminating the entry of plastic waste into wastewater systems.

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Annex

Annex I – Supplementary materials of Chapter 3

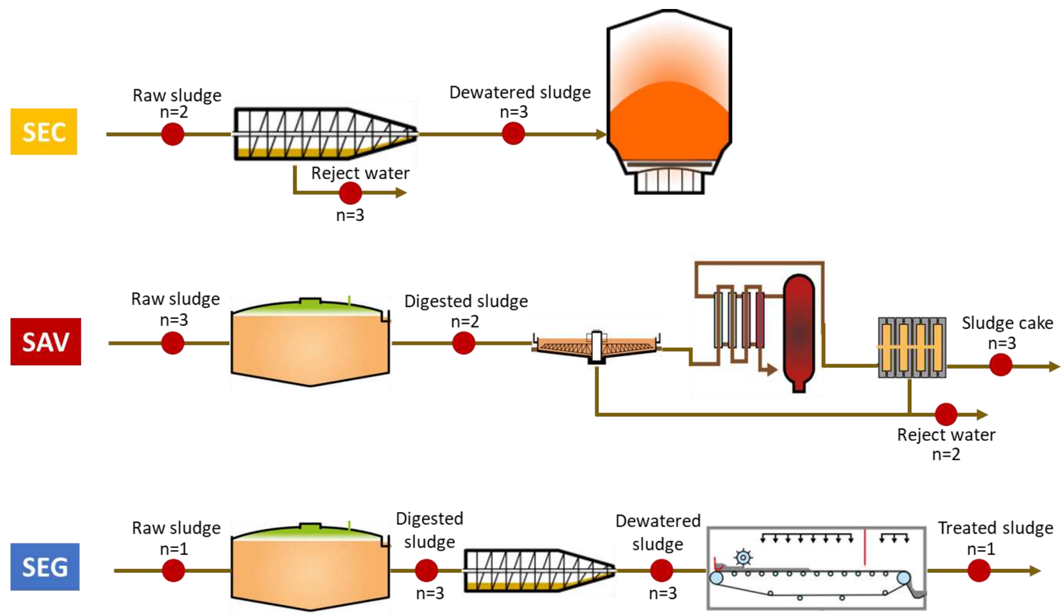

Figure A-1: Scheme of sludge-line treatment of the three studied WWTPs with sampling points

Table A-1: Data on characteristics different sludge types from the three studied WWTPs (mean ± standard deviation values modified from Mailler et al., 2017)

	Raw sludge	Digested sludge	Dewatered sludge	Sludge cake	Treated sludge	Reject water
DM (%)	4.7 ± 0.4	2.3 ± 0.1	25.9 ± 1.6	52.1 ± 2.9	89.9 ± 5.8	-
VM (% DM)	75.6 ± 3.0	59.0 ± 1.2	77.7 ± 3.2	38.5 ± 0.9	63.6 ± 10.3	-
TSS (mg/L)	-	-	-	-	-	1220 ± 345

```
        Kruskal-Wallis rank sum test

data:  list(n_size_SEC_RS, n_size_SEC_DeS, n_size_SEC_RW)
Kruskal-Wallis chi-squared = 12.706, df = 2, p-value = 0.001741

> dunn.test(list(n_size_SEC_RS,n_size_SEC_DeS, n_size_SEC_RW))
  Kruskal-Wallis rank sum test

data: x and group
Kruskal-Wallis chi-squared = 12.706, df = 2, p-value = 0


                           Comparison of x by group
                                (No adjustment)
Col Mean-|
Row Mean |          1          2
---------+----------------------
       2 |   2.487921
         |    0.0064*
         |
       3 |   0.195678  -3.420512
         |     0.4224    0.0003*

alpha = 0.05
Reject Ho if p <= alpha/2
```

Figure A-2: For Seine Centre WWTP, compared groups are the dataset on particle size distribution of MPs in raw sludge (1), dewatered sludge (2) and reject water (3)

```
        Kruskal-Wallis rank sum test

data:  list(n_size_SAV_RS, n_size_SAV_DiS, n_size_SAV_TS, n_size_SAV_RW)
Kruskal-Wallis chi-squared = 37.172, df = 3, p-value = 4.231e-08

> dunn.test(list(n_size_SAV_RS,n_size_SAV_DiS,n_size_SAV_TS,n_size_SAV_RW))
  Kruskal-Wallis rank sum test

data: x and group
Kruskal-Wallis chi-squared = 37.1721, df = 3, p-value = 0


                           Comparison of x by group
                                (No adjustment)
Col Mean-|
Row Mean |          1          2          3
---------+---------------------------------
       2 |   0.764359
         |     0.2223
         |
       3 |   5.908235   5.528653
         |    0.0000*    0.0000*
         |
       4 |   0.617234   0.263282  -3.161623
         |     0.2685     0.3962    0.0008*

alpha = 0.05
Reject Ho if p <= alpha/2
```

Figure A-3: For Seine Aval WWTP, compared groups are the dataset on particle size distribution of MPs in raw sludge (1), digested sludge (2), sludge cake (3) and reject water (4)

```
        Kruskal-Wallis rank sum test

data:  list(n_size_SEG_RS, n_size_SEG_DiS, n_size_SEG_DeS, n_size_SEG_TS)
Kruskal-Wallis chi-squared = 12.042, df = 3, p-value = 0.007242

> dunn.test(list(n_size_SEG_RS,n_size_SEG_DiS,n_size_SEG_DeS,n_size_SEG_TS))
  Kruskal-Wallis rank sum test

data: x and group
Kruskal-Wallis chi-squared = 12.0416, df = 3, p-value = 0.01


                           Comparison of x by group
                                (No adjustment)
Col Mean-|
Row Mean |          1          2          3
---------+---------------------------------
       2 |   1.717337
         |     0.0430
         |
       3 |   2.233296   0.333839
         |    0.0128*     0.3693
         |
       4 |   3.193744   2.451596   2.382485
         |    0.0007*    0.0071*    0.0086*

alpha = 0.05
Reject Ho if p <= alpha/2
```

Figure A-4: For Seine Grésillons WWTP, compared groups are the dataset on particle size distribution of MPs in raw sludge (1), digested sludge (2), dewatered sludge (3) and treated sludge (4)

Table A-2: Calculations for the estimated annual MP budget at different sludge treatment processes

Seine Centre				
Centrifugation		IN raw sludge	OUT dewatered sludge	OUT reject water
Microplastic number concentration		$[MP]_{raw\ sludge}$ particle/g	$[MP]_{dewatered\ sludge}$ particle/g	$[MP]_{reject\ water}$ particle/L
Measured data at WWTP	Flow	$Q_{raw\ sludge}$ m³/year		$Q_{reject\ water}$ m³/year
	Dry matter	$DM_{raw\ sludge}$ % or g/L		
	Mass in dry weight		$m_{dewatered\ sludge}$ ton/year	
Calculated data	Total particle number	$m_{raw\ sludge} = Q_{raw\ sludge} \times DM_{raw\ sludge}$ $\sum MP_{raw\ sludge} = m_{raw\ sludge} \times [MP]_{raw\ sludge}$ $\sum MP_{dewatered\ sludge} = m_{dewatered\ sludge} \times [MP]_{dewatered\ sludge}$ $\sum MP_{reject\ water} = Q_{reject\ water} \times [MP]_{reject\ water}$		
Budget balance equation		$\sum MP_{\ raw\ sludge} = \sum MP_{\ dewatered\ sludge} + \sum MP_{\ reject\ water}$		

Seine Aval				
Anaerobic digestion		IN Raw sludge	OUT Digested sludge	
Microplastic number concentration		$[MP]_{raw\ sludge}$ particle/g	$[MP]_{digested\ sludge}$ particle/g	
Data at WWTP	Mass in dry weight	$m_{raw\ sludge}$ ton/year	$m_{dewatered\ sludge}$ ton/year	
Calculated data	Total particle number	$\sum MP_{raw\ sludge} = m_{raw\ sludge} \times [MP]_{raw\ sludge}$ $\sum MP_{digested\ sludge} = m_{digested\ sludge} \times [MP]_{digested\ sludge}$		
Budget balance equation		$\sum MP_{\ raw\ sludge} = \sum MP_{\ digested\ sludge}$		
Thermal drying		IN Digested sludge	OUT Sludge cake	OUT Reject water
Microplastic number		$[MP]_{digested\ sludge}$	$[MP]_{\ sludge\ cake}$	$[MP]_{reject\ water}$

concentration		particle/g	particle/g	particle/L
Data at WWTP	Mass in dry weight	$m_{digested\ sludge}$ ton/year	$m_{sludge\ cake}$ ton/year	NA
Calculated data	Total particle number	$\sum MP_{digested\ sludge} = m_{digested\ sludge} \times [MP]_{digested\ sludge}$ $\sum MP_{sludge\ cake} = m_{sludge\ cake} \times [MP]_{sludge\ cake}$		
Budget balance equation		$\sum MP_{\ raw\ sludge} = \sum MP_{\ dewatered\ sludge} + \sum MP_{\ reject\ water}$		

Seine Grésillons			
Anaerobic digestion		IN Raw sludge	OUT Digested sludge
Microplastic number concentration		$[MP]_{raw\ sludge}$ particle/g	$[MP]_{digested\ sludge}$ particle/g
Data at WWTP	Mass in dry weight	$m_{raw\ sludge}$ ton/year	$m_{digested\ sludge}$ ton/year
Calculated data	Total particle number	$\sum MP_{raw\ sludge} = m_{raw\ sludge} \times [MP]_{raw\ sludge}$ $\sum MP_{dewatered\ sludge} = m_{dewatered\ sludge} \times [MP]_{dewatered\ sludge}$	
Budget balance equation		$\sum MP_{\ raw\ sludge} = \sum MP_{\ dewatered\ sludge}$	

Table A-3: Polymer composition of MPs in three studied WWTPs

Seine Centre	Raw sludge	Dewatered sludge	Reject water
PE	30.8	40.4	25.4
PEST	7.4	8.0	3.2
PP	46.0	25.9	63.3
PS	6.3	11.7	3.5
PU	2.9	2.9	0.6
PVAC	3.4	2.0	1.2
PVC	0.5	2.7	1.2
ABS	0.0	0.7	0.6
Acrylic	0.5	1.3	0.3

Alkyd	0.5	2.8	0.3
Cellulose acetate	1.1	0.0	0.0
PA	0.5	1.4	0.6
Polymer diversity	11	11	11

Seine Aval	Raw sludge	Digested sludge	Sludge cake	Reject water
PE	46.3	32.4	26.6	6.7
PEST	7.0	6.7	2.5	0.0
PP	22.0	34.6	37.3	93.3
PS	10.8	13.3	18.3	0.0
PU	0.6	3.1	0.0	0.0
PVAC	2.0	2.6	9.7	0.0
PVC	7.5	1.1	4.1	0.0
Vinyl copolymer	0.4	0.0	0.0	0.0
ABS	0.2	0.3	0.0	0.0
Acrylic	1.0	3.0	1.4	0.0
Alkyd	1.9	0.7	0.0	0.0
Cellulose acetate	0.0	1.3	0.0	0.0
PA	0.0	0.4	0.0	0.0
Pan_acrylic fibre	0.2	0.4	0.0	0.0
Polymer diversity	12	13	7	2

Seine Grésillons	Raw sludge	Digested sludge	Dewatered sludge	Treated sludge
PE	42.7	40.4	38.7	16.0
PEST	2.8	6.6	11.7	4.0
PP	34.8	38.1	25.2	16.0
PS	7.3	9.0	11.7	56.0
PU	1.1	1.5	3.2	8.0
PVAC	1.7	0.0	1.6	0.0
PVC	0.6	0.3	1.6	0.0

Vinyl copolymer	0.0	0.0	0.0	0.0
ABS	0.6	0.3	0.2	0.0
Acrylic	5.6	2.8	1.8	0.0
Alkyd	0.0	0.5	1.8	0.0
Cellulose acetate	0.6	0.0	0.4	0.0
PA	2.2	0.5	1.2	0.0
Pan_acrylic fibre	0.0	0.0	0.8	0.0
Polymer diversity	11	10	13	5

Table A-4: Microplastic contamination in reject water from different mechanical dewatering processes

Reference	Findings	Size limit
Alvim et al. (2022)	450 ± 212 MPs/L (a portion of 98 ± 2 % fibers and 2 ± 2 % fragments)	>150 µm
Nakao et al. (2021)	650 MPs/L centrifugation effluent 600 MPs/L thickened effluent (gravity thickening method)	>20 µm
Salmi et al. (2021)	gravimetric thickening was on average 475 MPs/L 10,400 MPs/L centrifuge reject	>20 µm
Talvitie et al. (2017b)	12,866.7 MPs/L reject water from dewatering	>20 µm

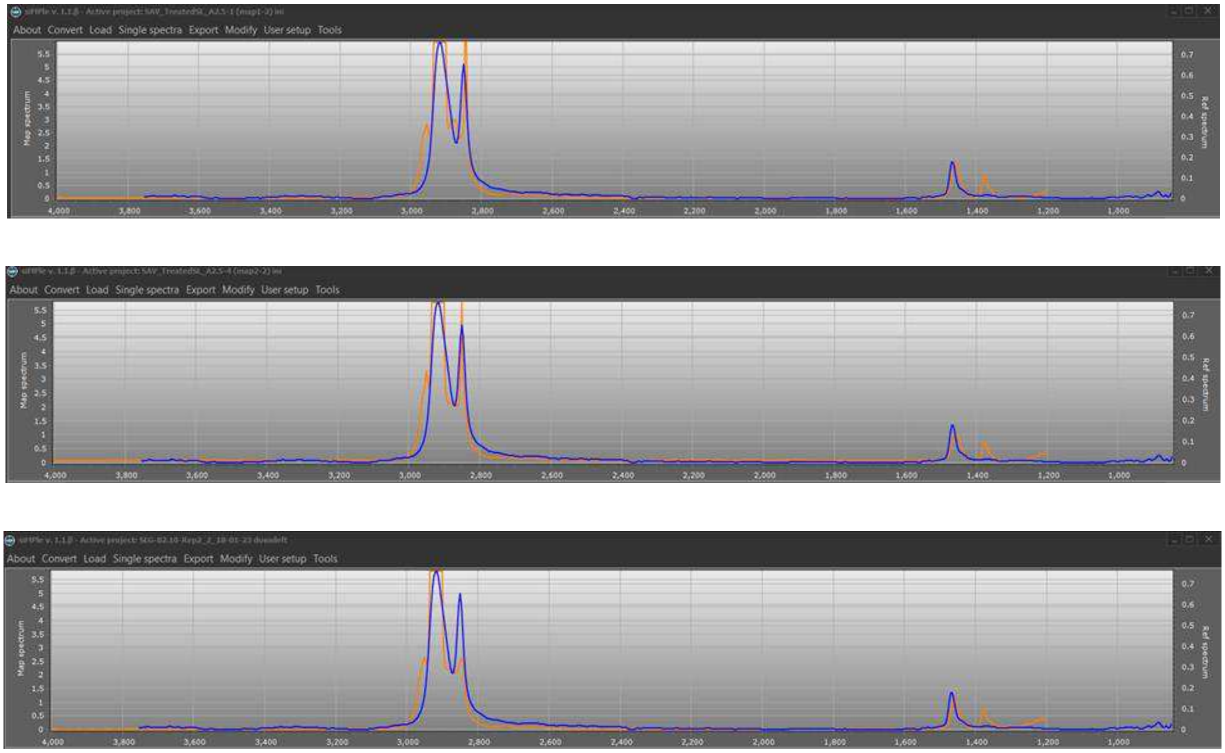

Figure A-5: Spectra of PE found in sludge cake/treated sludge after thermal treatment (in orange) compared to the reference spectra (in blue)

Annex II – Supplementary materials of Chapter 4

Table A-5: Information on the collection site of sewer sediment samples

Sample ID	Sampling date	Location	Latitude	Longtitude
S1	29/03/2021	The corner of Rue Cordinet and Rue Truffaut 75017	48.889071	2.316190
S2	22/06/2021	10, Rue Affre 75018	48.885700	2.355915
S3	16/08/2021	234, Rue de Crimee 75019	48.894557	2.373119
S4	16/03/2021	44, Rue de Turbigo 75003	48.865107	2.354860
S5	25/03/2021	79, Rue des Entrepreneurs 750015	48.844426	2.292023
S6	15/03/2021	5, Boulevard Lefebvre 75015	48.832748	2.289059
S7	07/04/2021	Interior courtyard of Austerlitz train station 75013	48.842614	2.363928
P1a	01/06/2021	5, Rue de Solferino 75005	48.860108	2.323630
P1b	13/04/2021	The corner of Rue de l'Universite and Rue de Solferino	48.859663	2.322772
P2a	19/05/2021	22, Bouvelard Jules Ferry)	48.868451	2.367996

P2b	11/06/2021	24, Rue Saint Bernard 75011	48.851902	2.381891
P3	15/03/2021	51, Rue Censier 75005	48.839362	2.350496

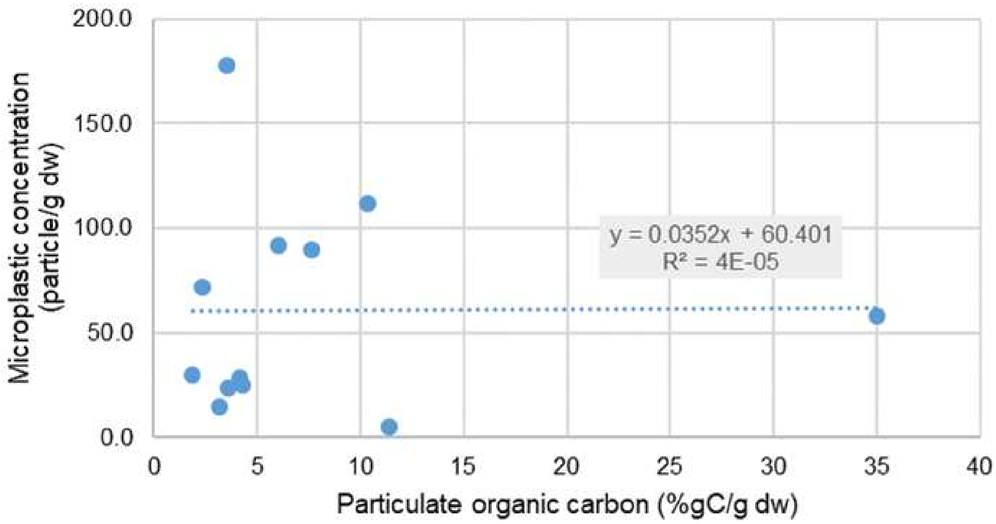

Figure A-6: Relationship between microplastic concentration and particulate organic carbon in sewer sediments

Annex III – Supplementary materials of Chapter 5

Table A-6: Characteristics of sampling events and several global parameters measured for water samples

Event	Date	Site	Discharge volume (m^3)	Duration (min)	Max flowrate (m^3/s)	Conductivity (mS/m)	Suspended solids (mg/L)	COD (mgO_2/L)
1	13/04/2022	La Briche	25483	285	3.35	52.9	328	513
		Clichy	35587	180	14.30	54.8	250	461
2	23/04/2022	La Briche	13933	160	3.35	64.1	696	1047
3	15/05/2022	La Briche	7913	340	5.17	49.3	676	811
4	20/05/2022	La Briche	63932	240	9.31	60.6	712	642
5	08/06/2022	La Briche	156782	560	9.52	32.7	255	368
		Clichy	463672	525	32.10	36.8	233	603
6	20/06/2022	La Briche	86754	305	10.23	35.5	247	656
		Clichy	105705	190	23.05	30.3	251	564
7	30/06/2022	La Briche	80786	340	7.72	43.1	292	372
		Clichy	155713	305	23.50	41.2	277	445
8	20/07/2022	La Briche	6060	160	7.72	61	610	807
9	14/08/2022	La Briche	24714	145	5.05	94.3	274	613
10	16/08/2022	La Briche	159448	305	14.94	35.4	270	328
		Clichy	612298	325	67.20	<30	241	258
11	17/08/2022	La Briche	8129	145	14.94	35.4	269	345
		Clichy	10955	60	67.20	<30	209	348
12	05/09/2022	La Briche	79510	270	9.18	94.6	467	636
13	07/09/2022	La Briche	4291	50	2.31	70.2	293	424
14	26/09/2022	La Briche	76332	295	8.46	38.4	475	589

15	02/10/2022	La Briche	27145	210	5.07	64.4	319	558
16	17/10/2022	La Briche	35664	405	5.07	42.8	272	431
		Clichy	77510	165	12.10	45.3	218	345

Table A-7: Estimated proportion of surface runoff and wastewater in CSO discharges

Event	Date	Site	Hypothesis 1		Hypothesis 2	
			% wastewater	% runoff	% wastewater	% runoff
1	13/04/2022	La Briche	0.37	0.63	0.46	0.54
		Clichy	0.39	0.61	0.48	0.52
2	23/04/2022	La Briche	0.48	0.52	0.58	0.42
3	15/05/2022	La Briche	0.34	0.66	0.43	0.57
4	20/05/2022	La Briche	0.45	0.55	0.54	0.46
5	08/06/2022	La Briche	0.17	0.83	0.25	0.75
		Clichy	0.21	0.79	0.30	0.70
6	20/06/2022	La Briche	0.20	0.80	0.28	0.72
		Clichy	< 0,15	> 0,85	< 0,23	> 0,77
7	30/06/2022	La Briche	0.28	0.72	0.36	0.64
		Clichy	< 0,15	> 0,85	< 0,23	> 0,77
8	20/07/2022	La Briche	0.45	0.55	0.55	0.45
9	14/08/2022	La Briche	0.78	0.22	0.89	0.11
10	16/08/2022	La Briche	0.20	0.80	0.28	0.72
		Clichy	< 0,15	> 0,85	< 0,23	> 0,77
11	17/08/2022	La Briche	0.20	0.80	0.28	0.72
		Clichy	< 0,15	> 0,85	< 0,23	> 0,77

12	05/09/2022	La Briche	0.78	0.22	0.89	0.11
13	07/09/2022	La Briche	0.54	0.46	0.64	0.36
14	26/09/2022	La Briche	0.23	0.77	0.31	0.69
15	02/10/2022	La Briche	0.48	0.52	0.58	0.42
16	17/10/2022	La Briche	0.27	0.73	0.36	0.64
		Clichy	0.30	0.70	0.38	0.62

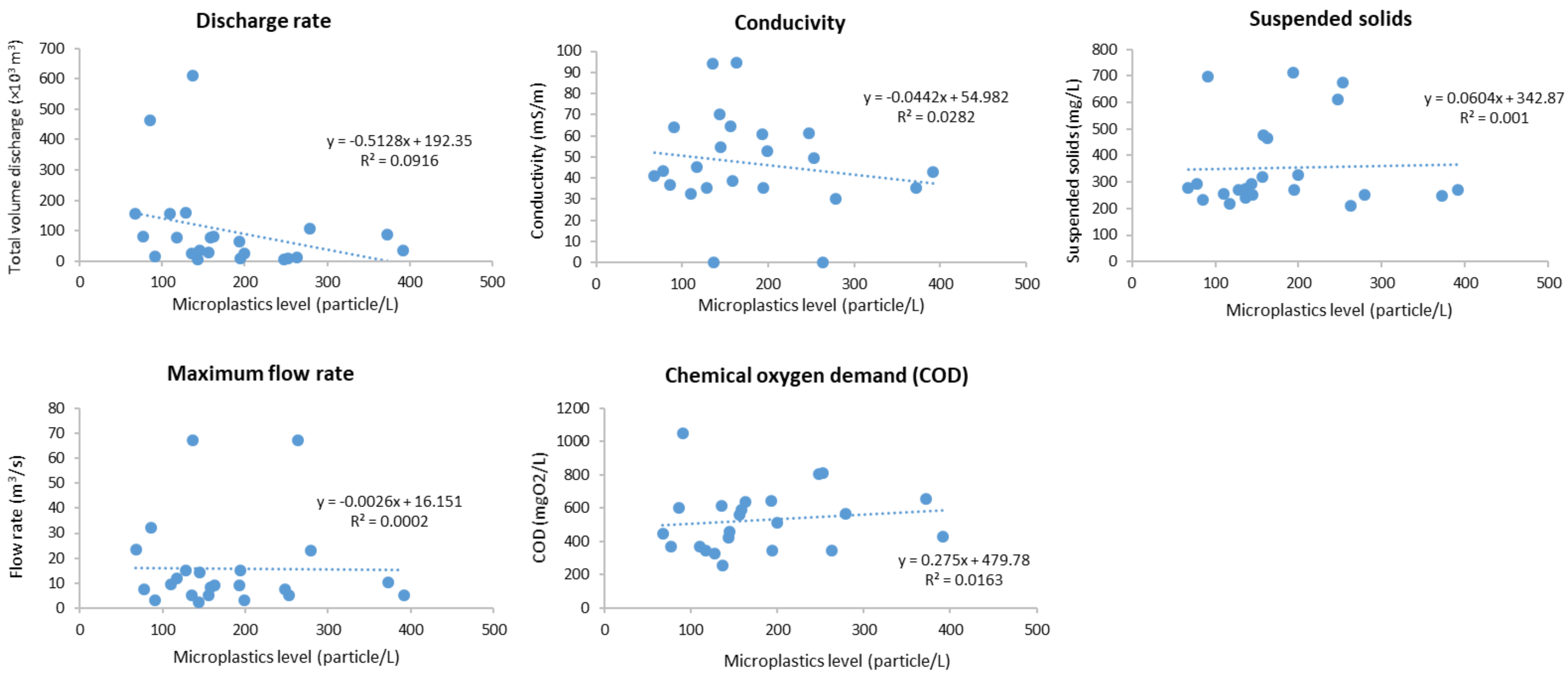

Figure A-7: Relation between microplastic levels in CSOs and different parameters related to CSO events

Table A-8: Per capita MP load discharged via WWTP effluents worldwide

Location	MP concentration (particle/L)	Population ($\times 10^3$ inhabitants)	Volume capacity ($\times 10^6$ L/day)	MP load (particle/per/day)	Size range (µm)	Reference
Australia						
Plant A	1.5	1,220	308	378.7	25-5000	Ziajahromi et al. (2017)
Plant B	0.48	67	17	121.8		
Plant C	0.21	151	48	66.8		
Canada	0.5	1,300	493	189.6	1-5000	Gies et al. (2018)
Italy	0.4	1,200	400	133.3	10-5000	Magni et al. (2019)
China	0.59	2,400	1000	245.8	50-5000	Yang et al. (2019)
USA						
Plum Island	3.7	180	83.3	1712.3	43-5000	Conley et al. (2019)
Rifle Range Road	17.6	53	18.9	6276.2		
Center Street	17.2	32	11.4	6127.5		
Spain	1.23	210	35	205.0	0.45-5000	Bayo et al. (2020)
Spain	10.7	300	45	1605.0	25-5000	Edo et al. (2020)
Australia						
Plant A	0.18	NA	130	NA	25-5000	Ziajahromi et al. (2021)
Plant B	0.96	234	65	266.7		
Plant C	0.91	700	150	195.0		

NA: information not available

According to the literature, the per capita MP load in the worldwide WWTP effluents ranged from 66.8 to 6276.2 particle/person/day. Thus, considering a population of 10 million inhabitants in Paris megacity, the annual MP flux discharged along with WWTP effluents into the Seine River within this region can be estimated as follows, approximately ranging from 2.4×10^{11} to 2.3×10^{13} particle/year.

$$Annual\ MP\ flux_{WWTP\ effluent}\ [particle/year] = MP\ load\ [particle/person/day] \times Population\ [persons] \times 365\ [day/year]$$