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# **Removal of microplastics and nanoplastics in water treatment processes: A systematic literature review**

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## **Abstract**

Understanding the dynamics of microplastics (MPs) and nanoplastics (NPs) in water treatment processes is crucial, particularly in the context of waterworks, as they are recognized as emerging contaminants in aquatic systems. This review explores into the complex interplay between the MPs and waterworks systems, which are crucial components in treating and delivering treated water to consumers. In this paper, recent research articles were conducted on 26 water treatment plants (WTPs) across 12 countries worldwide, along with an analysis of 103 studies encompassing 54 articles undertaken. This review focused on the characteristics and removal of MPs and NPs in water treatment processes compared to their abundance in the influent, effluent, and sludge. Subsequently, a quantitative analysis was conducted to compare the removal efficiency of MPs and NPs in various critical treatment technologies based on existing data. The paper extensively discussed the detailed mechanisms of critical treatment technologies, covering primary settling treatment with screening, coagulation-flocculation-sedimentation, filtration, advanced technology, biological treatment, disinfection, and water distribution. Currently, it appears that there are still gaps in the research on the removal of MPs and NPs in water treatment. This review suggests guidelines for future studies that should be undertaken to address these gaps.

**Keywords:** Micro- and nano-plastics, removal efficiency, Water treatment, Tap water, Water distribution system, Sludge management

## **1. Introduction**

Microplastics (MPs), which are fragment plastic particles with the size range of less than 5 mm, and their smaller likeness known as nano-plastics (NPs), usually referred to the particles with the size dimension of 1 to <1000 nm [1], have become an emerging pollutant in freshwater resources [2]. Due to their ubiquitous occurrence, environmental persistence, and potential

toxicity, they pose a risk to aquatic habitats, biodiversity, and human health [3, 4]. The increasing uses of plastics in daily life is contributing to the contamination of MPs in various environmental bodies including raw water sources, in which they can pollute drinking water during the production process, increasing a possibility of harmful toxic effects on humans [5, 6]. Since accessible clean drinking water is one of the Sustainable Development Goals of the United Nations [7], it is of utmost importance to prevent any associated risks to humans from the consumption of drinking water contaminated with MPs and NPs.

MPs and NPs can be orally intaked to human body via a huge variety of food and beverages [8-10]. It was found that ingestion of MPs through drinking water could be up to 4,000 MPs per person per year [6, 11]. After entering human body, MPs can be distributed through endocytosis in gastrointestinal tract and translocation in tissues and organs, of which the degree depends on the size of MPs [11]. High concentration of MPs can induce the toxic effects in digestive, respiratory, and nervous systems by up-regulating the gene expressions for inflammatory responses, morphological alteration, apoptosis-relating processes, and oxidative stress responses [6, 12]. According to previous research, human health risks from MPs in drinking water are linked to variation in shape and size and the presence of chemicals released from or absorbed onto MPs [13]. Even though the chemical toxicity arising from MPs has been thought to be low, the number of studies undertaken in this area are still limited due to that fact that they were mostly conducted in vitro and with the concentrations up to higher than the detected level in raw and potable water [14]. Therefore, to determine the risk levels related to human exposure to MPs through drinking water consumption, further studies focusing on both MPs detection and toxicity determination are required.

Previous MPs studies have focused on wastewater treatment systems as wastewater was considered the principal source of MPs contamination [15]. Evidence showed that the fragmentation and decomposition of MPs occurs during wastewater treatment processes [16], which would significantly increase the content of smaller-size MPs and NPs in treated waters. This fragmentation could be transported further to drinking water treatment processes. Even though attention of MPs contamination in drinking water systems has been increasing, only few studies have been conducted on MPs in water distribution system [17-19], and more research is required for the optimization of each water treatment process for MPs removal [20, 21]. Additionally, the fate of MPs during drinking water treatment processes is not yet fully understood partly due to a lack of standard methodology of sample handling, collection, identification, and classification of MPs and their adsorbed contaminants [22-24], leading to the limitation on MPs and NPs studies in drinking water treatment systems. At present, the conventional water treatment processes are not designed for microplastic removal, and the occurrence of MPs may affect the whole drinking water treatment process such as coagulation, filtration, and disinfection [21].

This review aims to provide a comprehensive and systematic literature review on the removal of MPs and NPs by different water treatment processes. The specific objectives of this review are: 1) to update the current knowledge on MPs and NPs contamination in water treatment systems up until the water distribution systems, and 2) to analyze the efficiencies of each water treatment process for removing MPs and NPs. By elucidating the intricate mechanisms and performance of water treatment systems for removing MPs and NPs, we seek to shed light on the gaps for further research on the development of water treatment systems for MPs and NPs removal in order to successfully address this contemporary environmental challenge and protect water quality.

## 2. Methods of Systematic Literature Review (SLR)

The systematic search of this study followed Preferred Reporting Item for Systematic Reviews and Meta-Analysis (PRISMA) guidelines. Firstly, a combination of search keywords “Water treatment + Microplastics” was applied to screen the related articles. Articles published in English language were acquired from 2019 until January 2023 via different search engines, resulting in 1,166, 721, 763, and 18,300 related articles acquired via Scopus, Web of Science, PubMed, and Google Scholar, respectively. Then, the inclusion and exclusion criteria (Table 1) were used to extract final publications. Consequently, 54 articles were extracted for this study, comprising 2 articles from 2019, 13 articles from 2020, 22 articles from 2021, 13 articles from 2022, and 4 articles from 2023, as illustrated in Fig. 1.

**Table 1** Inclusion and exclusion criteria

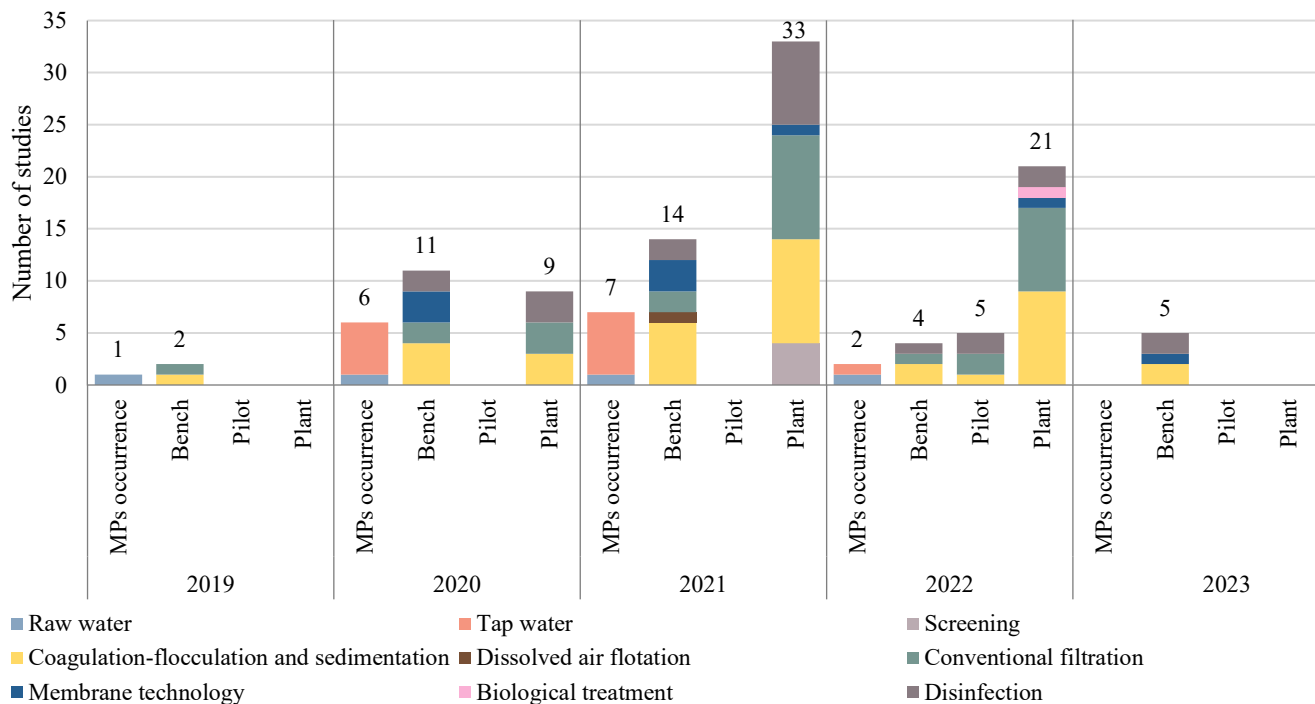
Inclusion	Exclusion
1. Keywords <ul style="list-style-type: none"><li>- Tap water</li><li>- Removal technology</li><li>- Treatment technology</li><li>- Environmental toxicity</li><li>- Nano-Plastic</li><li>- Fiber MPs</li><li>- Number concentration</li><li>- Sludge</li><li>- Water distribution</li></ul>	1. Keywords <ul style="list-style-type: none"><li>- Wastewater treatment</li><li>- Groundwater</li></ul> 2. Review paper3. Short communication4. Book chapter

## 3. Result

### 3.1. Global overview of the database

All the selected articles can be characterized into three characteristics (Table S1 – Table S3, supplementary data): the studies on MPs overall removal efficiency throughout the WTPs (14 articles, Table S1), the studies on MPs information relating to water treatment process in

experimental scale (27 articles, Table S2), and the studies on MPs occurrence in drinking water, tap water, and water supply systems (13 articles, Table S3).



**Fig. 1** Number of studies in various water treatment processes and MPs occurrence

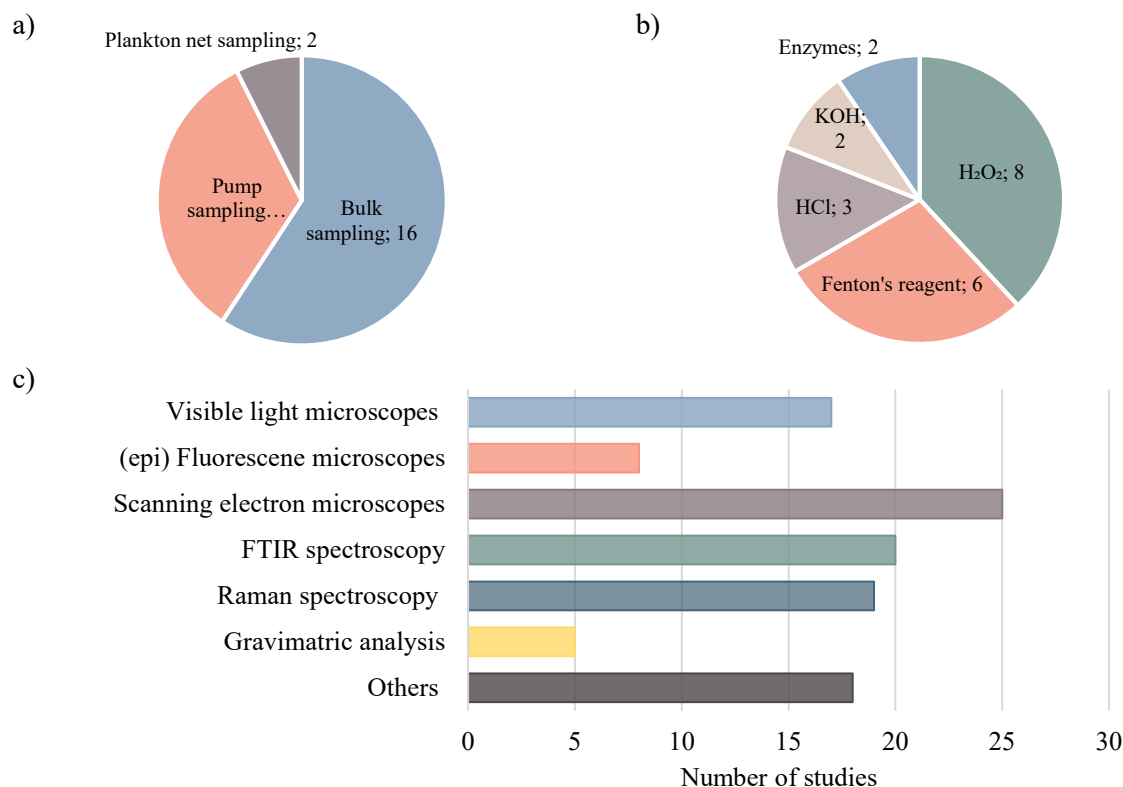
According to Fig. 1, 16 studies reported the occurrence of MPs, in which 25% focus on the presence of MPs in raw water, and 75% concentrate on treated water including drinking and tap water. Moreover, the highest number of studies related to MPs are conducted in water treatment (104 studies). This outcome indicates that the water treatment has been the center of interest for the studies of MPs impact and removal efficiency in recent years. The studies in WTPs were highly increased in 2021 whereas the pilot scale was initially conducted in 2022. The research in the field of MPs in (drinking) water is still in its developing stage compared with in wastewater treatment. Most of the recent studies focused on investigation of MPs removal efficiency of the existing water treatment plants followed by finding mechanisms and factors of the removal and discovering the occurrence of MPs in the water supply systems, including tap water and sludge.

In detail, MPs removal techniques in water treatment fall into two major categories: coagulation-flocculation-sedimentation (CFS) and filtration processes. Approximately 36% of the studies primarily focused on CFS whereas about 36% investigated filtration techniques, which can be further divided into 28% of conventional filtration and 8% of membrane technology. The others included screening, dissolved air flotation, biological treatment, and disinfection. Additionally, 4 studies investigated MPs occurrence in sludge.

### **3.2. Concentration and characteristics of MPs detected in drinking water supply system and tested in experimental scale**

MPs were detected along the drinking water supply system from raw water to tap water and sludge. From the review, bulk sampling is mostly used to collect the water samples (Fig. 2a), since it requires simple sampling tools like glass or stainless bottles. Depending on the origin of the sample, it is sometimes necessary to remove environmental matrix before conducting MPs analysis, especially organic matter, which interferes the signal of the analysis or lead to under- or overestimation of MPs [25, 26]. Thus far, hydroxide-based treatments are typically applied to remove the organic matter by using  $H_2O_2$  alone or in combination with  $Fe_2SO_4$  catalyst (Fenton's reagent) as shown in Fig. 2b. In order to detect and characterize MPs, microscopic and spectroscopic techniques are mainly utilized in the recent studies so far (Fig.2c). More details of the sampling methods, sample treatments, and MPs analysis techniques performed in the reviewed articles were provided in section S2.

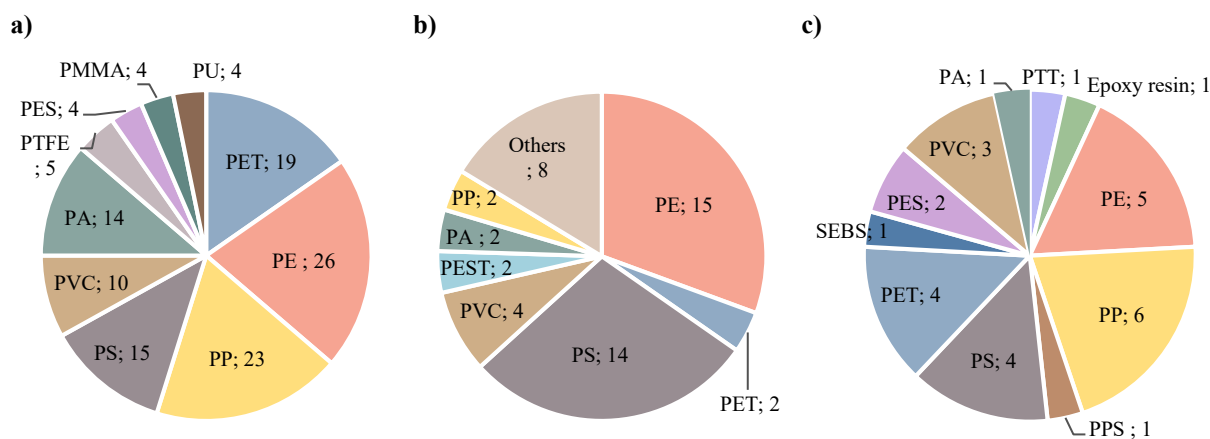




**Fig. 2** Microplastic investigation techniques applied in the reviewed articles: (a) sampling methods, (b) reagents for samples organic matter clean-up, and (c) microplastic analytical techniques enumerated by number of studies

The reviewed studies in WTPs reported a wide range of MPs concentrations found in influent from  $0.55 \pm 0.15$  to  $6614 \pm 1132$  particles/L (Table S1). Only one study reported mass concentration with the value of  $93.2 \mu\text{g}/\text{m}^3$  [27]. Based on our literature review, over 33 types of plastic were detected (Table S1 and Table S2). Fig. 3 a presented the most abundant types of MPs in 26 WTPs of 17 articles showing that polyethylene (PE), polypropylene (PP), polyethylene terephthalate (PET), polystyrene (PS), polyamide (PA), and polyvinyl chloride (PVC) were the most common plastics in the influent of the WTPs, respectively. These results are in accordance with the global and European plastics production in 2019 – 2023 provided by Plastics Europe [28]. In terms of the particle size, 1 – 5000  $\mu\text{m}$  of MPs were found in the influent (Fig. 4a), in which the size range of 50 – 224  $\mu\text{m}$  was commonly detected. With respect to the shape of the MPs (Fig. 5),

fibers are the highest abundance of MPs shape indicated in most of the reviewed articles (13 WTPs, 9 articles) followed by fragments (6 WTPs, 4 articles) as shown in Table S1 and Table S3. A reviewed study in surface water as source of drinking water production also reported similar results of the MPs characteristics [29].

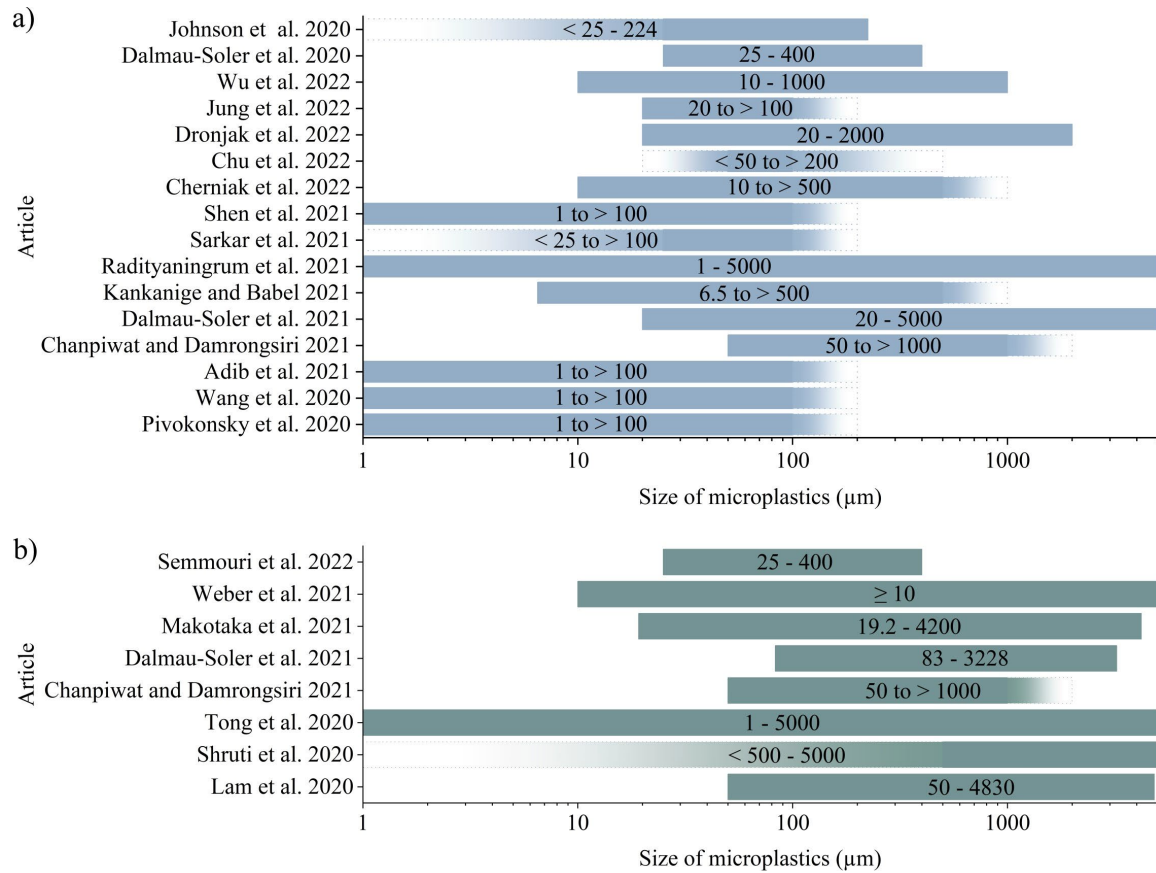


**Fig. 3** Number of studies showing types of MPs: (a) detected in water treatment plant, (b) tested in experimental scale, and (c) detected in tap water

In tap (drinking) water, MPs were detected up to 440 particles/L. The main types of plastic found in the tap water were PP, PE, PS, PET, and PVC (Fig. 3c), which are in general consistent with the MP types in the raw water. However, these plastics were also released from the water distribution system rather than from the remaining MPs in treated water in some studies (see section 3.5). Regarding the size, 1 – 5000  $\mu\text{m}$  particles were collected to investigate MPs occurrence in the tap water, and the MPs of 83 – 400  $\mu\text{m}$  were the most frequently detected (Fig. 4b). Same as in raw water, fragments, fibers, spheres, pellets, and films were observed in tap water (Fig. 5), in which fragments and fibers are the most common MPs shape (Table S3).

The colors of MPs are seldom employed to signify the potential origins of microplastic pollution, as colors are impermanent and susceptible to alteration through weathering and bleaching processes as well as discoloration during MPs sample preparation [1, 30, 31]. Based on

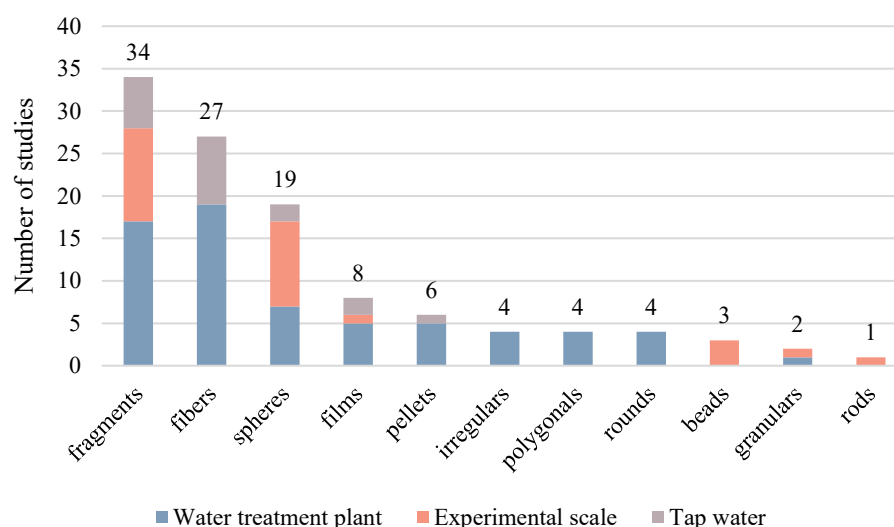
the present review, only 4 articles examined the colors of MPs in the water supply systems, which are transparent/white, black, blue, red, yellow, and green MPs. In raw water, black is the dominant color in a study [16], while another study found mostly transparent/white MPs [32]. Two other studies focused on tap water discovered that transparent [33] or black MPs [34] are in highest proportion followed by blue MPs as the second abundance in both articles.



**Fig. 4** Size ranges of MPs detected in (a) water treatment plant and (b) tap water

In an experimental scale, various MPs concentrations and characteristics were applied to study the removal efficiency or water treatment mechanisms (Table S2). The MPs concentrations were prepared mainly in mass concentration in range from 0.1 – 7,500 mg/L with the number concentration of  $10 - 2 \times 10^{11}$  particles/L tested in the studies. The main types of MPs conducted in the experimental scale were PE and PS (Fig. 3b). Similar to the concentrations, the sizes of tested

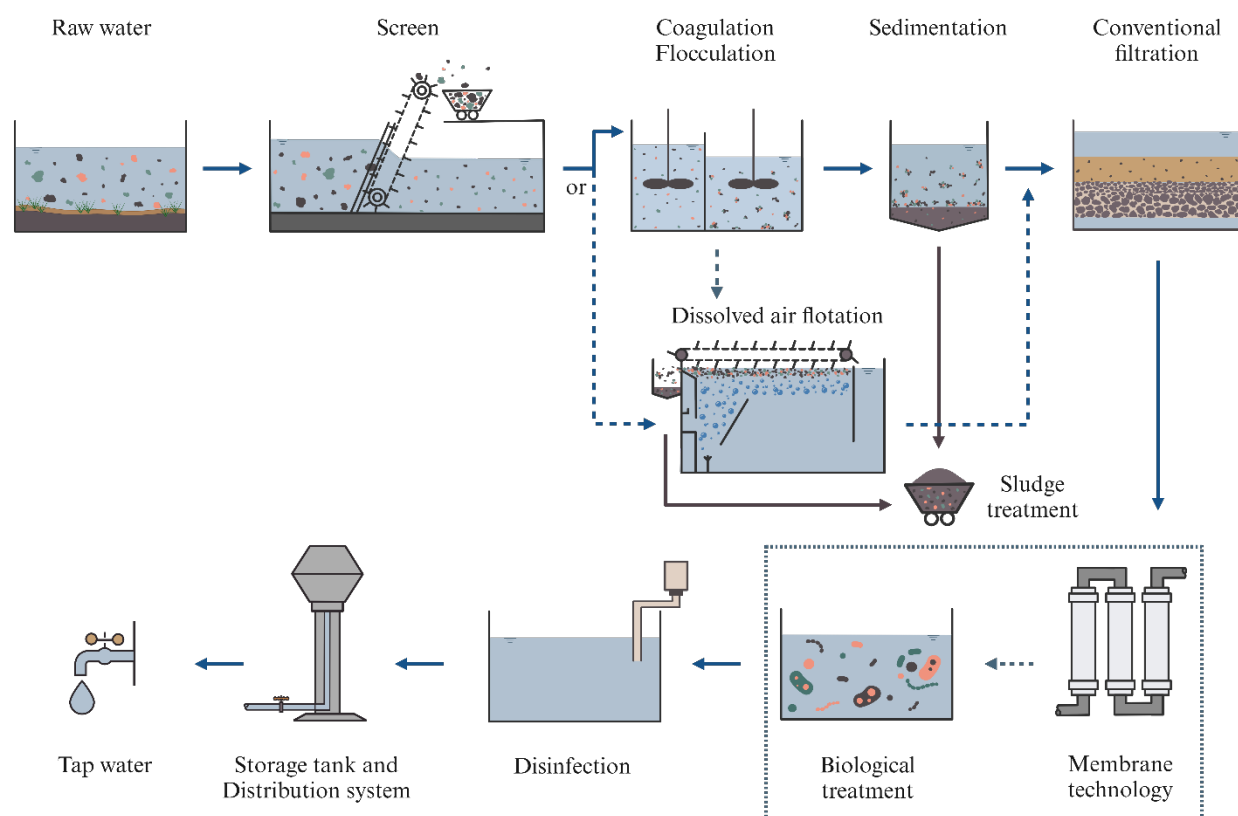
MPs were also varied from the threshold micro-scale (5,000  $\mu\text{m}$ ) down to nano-scale (0.3 nm). In terms of shape, fragments and spheres were commonly applied as illustrated in Fig. 5 although spheres were not the most typically found in raw water or water supply systems. With spherical plastic particles, the detection, observation, or result evaluation are less complicated to handle compared to fibers or fragments due to the certain dimension and smooth surface. Besides, this is also due to their commercial availability and particles standardization [35, 36].



**Fig. 5** Number of studies reporting shapes of MPs detected in water treatment plant, used in experimental scale, and detected in tap water

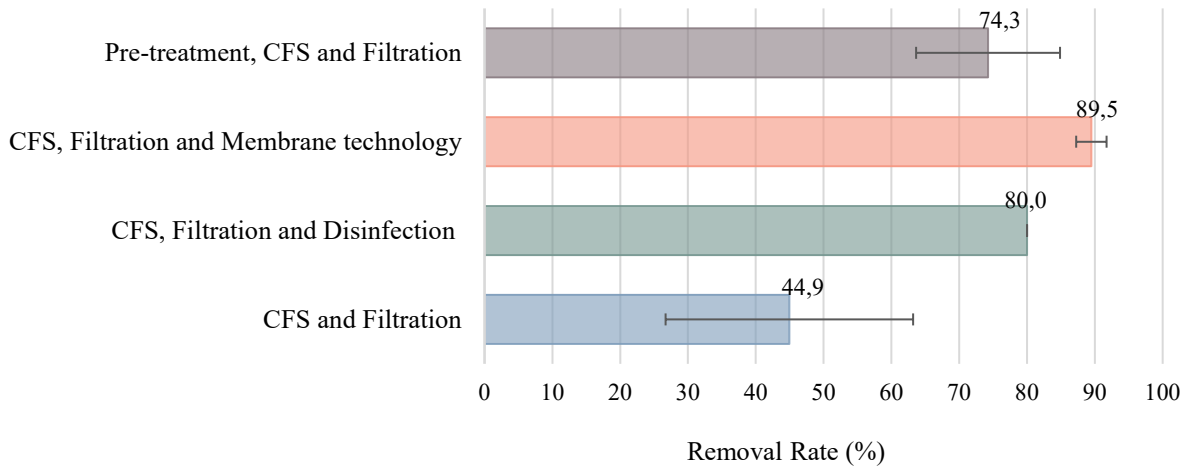
### 3.3. Water treatment system and microplastics removal efficiency

Water treatment processes play a critical role in removing MPs and providing clean and safe water for human uses. This section focusses on exploring the efficiency of MPs removal in water treatment systems as shown in Fig. 6, which illustrates various stages in the treatment, highlighting the critical points where the MPs removal efficiency is of particular interest.



**Fig. 6** Schematic diagram of water treatment processes applied in the reviewed studies: the solid line is the main processes, the dash line represents alternative process of CFS, and the optional processes for drinking/water treatment are enclosed in the dotted square.

Generally, a series of treatment steps including CFS combined with conventional filtration are designed to eliminate impurities from raw water. Other treatment units such as screening or disinfection are added according to specific requirements. Although the primary objective of these processes is to clarify water and make it suitable for various applications, it is noteworthy that they also exhibit a remarkable capability to remove MPs effectively as illustrated in Fig. 7, which presents the average MPs removal efficiency of water treatment plants.



**Fig. 7** MPs removal efficiency of water treatment plant. Pre-treatment includes screening, aeration, pre-sedimentation, pre-ozonation, screening, biological pretreatment, pre-disinfection. The native data can be found in Table S1.

In further subsections, the efficiency of MPs removal will be thoroughly discussed for individual and overall process levels, including bench-scale studies, pilot plants, and water treatment plants. The summarization and the evaluation of treatment steps will be presented including screening, coagulation-flocculation-sedimentation, dissolved air floatation filtration, sludge management, filtration, biological treatment, and disinfection.

### 3.3.1. Screening

Screening is an integral component of the preliminary water treatment plant to efficiently remove large solids and debris from the intake water and to protect downstream equipment. Although it is commonly used in water treatment plants, the studies on MPs removal by screening process seem to be limited thus far. Based on the literatures reviewed, screening was found to be able to remove MPs from water. Kankanige and Babel found that the removal performance of screening is high for MPs in the size of  $\geq 500 \mu\text{m}$  and  $300 - 500 \mu\text{m}$  while it does not effectively remove MPs smaller than  $300 \mu\text{m}$  ( $<10\%$  removal) [37]. To overcome this limitation, the authors

suggested that decreasing the fine-screen pore size could largely help minimizing the number of MPs entering the drinking water treatment plant (DWTP) during screening process.

### **3.3.2. Coagulation – Flocculation – Sedimentation (CFS)**

CFS process is common water treatment techniques used in water treatment systems to remove small particles [38]. During coagulation, chemicals are added to destabilize suspended particles. This is followed by flocculation, which is a gentle mixing process to promote the formation of aggregates of destabilizes particles called flocs. The latter then settle in the sedimentation step, facilitated by gravity force (a common technique) or specialized equipment.

Although CFS have a significant potential to remove dispersed particles, the emerging contamination of MPs in water systems poses as a new challenge for the CFS systems, accelerating research and the development of technologies to enhance particle removal performance and make them suitable for microplastic elimination. The following subsections summarize recent studies on MPs removal efficiency by CFS processes along with factors affecting their removal performance. Details of the studies in experimental scale and water treatment plant are provided in the supplementary data (Table S4 and Table S5, respectively). From the literature review, it could be noticed that the removal efficiency spans from 1 – 100% in experimental studies and from -356 – 100% in water treatment plants. In addition, details on the coagulation mechanisms related to MPs can be found in the supplementary section S3 (Coagulation mechanisms).

#### **3.3.2.1 Coagulants**

The main coagulant/flocculant commonly used nowadays are inorganics salts, which act through charging neutralization or sweep flocculation depending on various operational conditions [39], and synthetic organic polymers, used alongside inorganic salts, as providing in a comprehensive review of polymers for water purification applications [40]. Recent literatures have

also revealed new types of coagulants produced from natural materials [7, 41-43]. Depending on the type of coagulant used, either one or a combination of the different mechanisms promote destabilization and/or agglomeration of the MPs (section S3, supplementary data).

Since the use of organic as inorganic coagulants may lead to the release of metal ions or monomers [44, 45], new types of coagulants are of natural or environmental-friendly coagulants are actually developed and tested against MPs [7, 41-43]. These natural polymeric coagulants are mainly derived from starch, chitosan or cellulose/lignin. They have sometimes been tested on the removal of MPs in conjunction with metal salts. Peydayesh et al. [42] used lysozyme amyloid fibrils on the removal of PS MPs by increasing charge neutralization, and their removal rate is up to 97.9%. More recent research of Hu et al. [44] showed that a starch-based coagulant combined with polysilicic acid efficiently remove 0.1 – 5  $\mu\text{m}$  MPs with removal rates between 88 and 98% depending on their size. The efficiency of natural coagulants assisted with salts or polymers is due to the combination of charge neutralization and bridging effect. Although the application of bio-coagulants/bio-flocculants remains limited and scattered due to factors such as the complex extraction process, limited availability of raw materials, and variations in the characteristics of water to be treated, there is significant potential for their development as sustainable and eco-friendly technology for water treatment processes.

The dosage for coagulant varies a lot depending on the type of MPs, coagulant, and water (Table S4). It should also be mentioned that for some studies two coagulants are used simultaneously involving different coagulation mechanisms at the same time. In this case, even if the zeta potential of MPs is far from being null, an efficient removal can be achieved [44]. Tabatabaei et al. [46] found that the existence of polyacrylamide (PAM) affects the PE removal; under an appropriate and high concentration range, the MPs removal efficiency remains almost



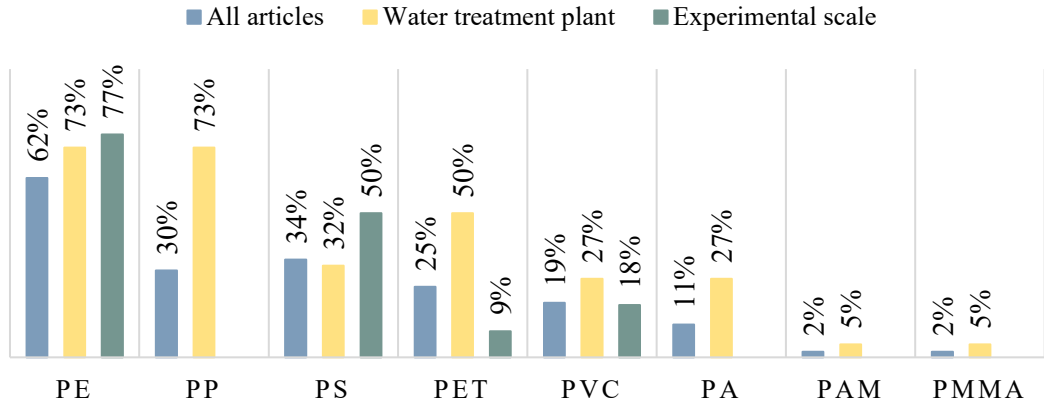
constant while it is decreased under lower concentrations. This uncovering is aligned with the observations by Ma et al. [47], which reported increased MPs removal rates of PE smaller than 0.5 mm when adding PAM. Thus, it is quite difficult to draw clear and straight conclusions about the coagulant dosage. Nevertheless, as underlined by Tang et al. [48] and also by Shahi et al. [49], if charge neutralization is not the main mechanism involved, generally increasing the coagulant dose allows an improvement of the removal efficiency of MPs [41].

### **3.3.2.2 Concentration and characteristics of MPs in CFS process**

According to the reviewed articles (Table S5 and Fig. S1), the CFS can remove MPs typically within the range of 25 – 60%, and its effectiveness does not appear to be particularly concentration dependent. However, there is still not enough studies investigating the removal efficiency of different concentrations of an identical microplastic in term of type, size, and shape. Therefore, the dependence of MPs removal efficiency on its concentration cannot be evaluated and draw any conclusion yet. Another point worth mentioning is that in the case of primary treatment, negative removal efficiencies arise. This is most likely due to the fact that coagulant dispersion operations are very often carried out at high shear rates (high velocity gradient) and this results in the breakage of larger MPs, especially the fibers making the number concentration increase. To our knowledge, no study has evaluated this impact. It would be interesting to know how many pieces of these microplastics are broken into and whether or not the smallest pieces generated remain detectable by the analytical techniques.

The reviewed studies primarily conducted MPs removal experiments using PE, followed by PS, PVC, and PET (Fig. 8). Further details are available in Table S4. This aligns with the types of MPs typically found in raw water and the CFS systems of WTPs, which are summarized in Table S5. The results indicate that PP, PE, PET, and PS are the most common types of MPs in these

systems; however, experimental study of PP microplastics removal in the water treatment system is still neglected compared to the other common MPs (Fig. 8).



**Fig. 8** Types of MPs used or detected in the CFS reviewed studies

Various size ranges and shapes of plastics were detected in the water treatment plant. These size ranges were used to analyze the relationship between size and its impact on removal efficiency, as illustrated in Fig. S2. Surprisingly, there was no substantial evidence to support the idea that the size of MPs significantly affects the removal rate. Most studies in CFS process showed the removal rate consistently exceeded 60% irrespective of the microplastics' size. Furthermore, the size of the MPs has been still considered in the studies analyzed: the largest MPs tested were 5,000  $\mu\text{m}$  [47], while the smallest size was 0.1  $\mu\text{m}$  [44]. It is important to note that evaluating the size of fragments can be challenging, and authors often do not provide comprehensive characteristics of the MPs used. However, some studies, like Lapointe et al. [50], offer more detailed specifications; for instance, they indicated that their polyester (PEST) fibers measured 14  $\mu\text{m}$  in width and 580  $\mu\text{m}$  in length, corresponding to an equivalent diameter of 90  $\mu\text{m}$ .

The shape of the particles is somewhat important since it can provide different specific surface areas for MPs, which can have impact on the interactions between MPs and other contaminants or flocculants such as adsorption bridging. However, more studies on this aspect are required since

most existing studies conducted experiments using commercially available standard plastic beads in sphere form [51-53] and milled fragment MPs [46, 54, 55], while only one study examined the removal performance of CFS in removing fiber MPs [56].

According to the literature review, pristine MPs were mostly used to study the conditions affecting MPs removal efficiency of CFS process [47, 51, 54, 55] while weathered MPs, which are real properties of MPs in natural environment, were used in only few studies [56, 57]. In environmental samples, weathering processes could lead to plastics becoming more porous and changing their surface chemistry. The study of Lapointe et al. [56] investigated the removal efficiency of pristine PE and weathered PE showing that 99% of weathered PE could be eliminated while 89% of pristine PE was removed (Table S4). This research indicated that coagulant efficiency increased when the plastic surface was weathered due to greater surface area of MPs and the presence of new anchoring points for coagulants on the PE material, such as  $-OH$ ,  $-COOH$ , and  $-C=C-$ , after being photo-oxidized by UV lamp resulting in the increased potential for surface interactions with coagulants and flocculants.

### **3.3.2.3 Water characteristics**

The removal of MPs from water by CFS is influenced not only by the characteristics of the particles themselves but also by the properties of the water in which they are suspended. The reviewed studies used a range of water characteristics in the experiments including ultra-pure with low ultraviolet absorbance (HPLC) grade water, demineralized (DI) water, tap water, and raw surface water. Some studies also adjusted certain parameters, such as turbidity, by adding natural clay like bentonite or kaolin. Additionally, humic acids (HA) was used to modify natural organic matter (NOM) in the water (see Table S4). The primary objectives of these studies were to determine the optimal conditions for eliminating MPs from the water as well as to identify which

parameters influence the removal efficiency; however, a few studies did not provide information about the water characteristics used in their experiments. The following summarizes the findings of the parameters playing a role in the removal efficiency of the reviewed studies.

In the aspect of evaluating the impact of turbidity on MPs removal by CFS, Lapointe et al. [56] conducted a study to explore the influence of coagulant type and concentration on both turbidity and the MPs removal. The results showed that at the optimal coagulant concentrations for turbidity treatment, the removal efficiency for PEST fiber reached an impressive 99%, while PE removal reached up to 82%. This trend was in consistent with the removal of turbidity. Additionally, Skaf et al. [58] delved into various parameters including zeta potential, pH, humic acid, turbidity induced by kaolin, and alum concentration. The results confirmed that effective conditions for removing turbidity are also effective for eliminating model microplastic spheres and microfibers.

The importance of pH in the MPs removal has been a prominent focus in many studies as it is clear that the initial pH has a significant impact on zeta potential, and consequently, it affects the mechanism of aggregation or the chemical form of the coagulant. However, it might not be possible to conclude what the suitable range for coagulation and flocculation is since some researchers propose the use of slightly acidic conditions [47, 57, 59], while others suggest basicity or neutrality [46, 52, 54] as optimum. The results showed that higher pH (from 7 – 9) leads to a greater size of created flocs, and consequently, more PE microplastics can be trapped into the flocs and settle due to the heavier agglomerated mass. Moreover, zeta potential analysis revealed that as pH increases, zeta potential decreases, indicating reduced particle stability at higher pH levels [54]. Additionally, Skaf et al. [58] found that kaolin exhibits greater stability in tap water compared to MPs; moreover, the MPs were less stable under acidic conditions. Interestingly, the alum dose has a similar impact on zeta potential as observed with kaolin and MPs. It is important to note that the

pH effect on MPs removal can be slightly different, which may vary depending on various factors such as the specific type of coagulant used and the characteristics of the MPs themselves. Therefore, the interplay between pH and MPs removal is a complex area that requires careful consideration in water treatment processes.

The presence of NOM can impact the removal of MPs during coagulation and flocculation processes. HA are frequently used to imitate NOM in water as they contain compounds commonly found in surface waters and have high molecular weight and colloidal properties. Their presence can influence the behavior of MPs during coagulation-flocculation which may either hinder or promote the aggregation and settling of MPs [47, 60]. Lee et al. [53] found that the presence of HA in the water enhanced the efficiency of PE removal by using ferrate coagulant, particularly at low concentrations. According to the mechanism of the study, the presence of HA primarily influenced charge neutralization, which played an important role in controlling particle destabilization. On the other hand, Baiwen et al. [47] conducted research on the efficiency of PE removal and explored various factors such as ionic strength, turbidity level, and HA concentration. Their findings indicated that these factors had minimal influence on PE removal because of PE's stable chemical properties.

### **3.3.3. Dissolved air flotation (DAF)**

DAF offers an effectively alternative solution for dealing with microplastic contamination, especially considering the unique challenge posed by the low density of these particles compared to water and other suspended solids. To enhance the DAF process, coagulants can be added to increase floc formation. Based on the literature review, Wang et. al [61] indicated that MPs larger than 15  $\mu\text{m}$  could be removed up to 100% while Posi-DAF is more effective to remove MPs especially the smaller ones (Table S6), since long molecular chain structure of the coagulants

(CTAB and PDADMAC) can increase the collision and adhesion probability of micro-bubble and MPs. However, this report is retrieved from one study; therefore, further intensive research is needed to obtain better understanding of MPs removal of DAF.

#### **3.3.4. Sludge management**

Water treatment plants generate water treatment sludge (WTS) as a by-product during the water clarification process. Sources of MPs in WTS are flocculant, e.g., polyamine, polyester, polyacrylamide [62], and MPs in raw water. Thus far, only few studies investigated MPs concentration in WTS.

Johnson et al. [63] reported that the values for MPs found in the waste sludge in Water Treatment Works in the UK were extremely variable, from below the limit of detection to 86,000 particles/g (Dry Weight, DW). PE, PET and PP are mainly found from the concentration up to 5,589 particles/g DW, 60,357 particles/g DW, and 19,783 particles/g DW, respectively. Siegel et al. [64] studied the identification and quantification of MPs in raw surface water and WTS. The total number of MPs was  $196 \pm 42$  particles/m<sup>3</sup> found in the sludge samples from Rostock DWTP in Germany. The detected MPs consisted of 12 different polymer types: PE, PP, PS, PET, PVC, polyoxymethylene, polymethyl methacrylate (PMMA), polysiloxane, acrylonitrile-butadiene-styrene (ABS), acrylic resin, epoxy resin and silicone rubber. PE dominated in all samples with an average proportion of 37.8%, followed by PP (31.0%) and PS (24.4%). MPs sizes investigated ranged from 50 – 500 µm, and the most abundance size fraction is 101 – 300 µm. Regarding MPs morphology being classified into irregular shaped fragments, spherules and fibers, fragments were the most abundant MP shape, accounting for 83.3%. Fibers constituted 16.7% and spherules did not occur. In addition, the number of MPs in WTS did not show any significant difference between surface raw water and sludge, which could be implied that most of MPs in water were trapped in

sludge during water treatment process. Kankanige et al. [37] found 0.3 – 1.2 particles/g of MPs in sludge from drying bed and filter press-mud of a conventional WTP in Thailand during dry and rainy seasons. MPs in the size range of 6.5 – 53  $\mu\text{m}$  were present in the highest concentration while  $\geq 500$   $\mu\text{m}$  particles were found the least in sediment. Dronjak et al. [65] examined the MPs concentration in dry sludge and water from sludge centrifugation unit. Water from the sludge centrifugation unit had a MPs concentration of 194 particles/L dominated by particles of sizes ranging from 2,000 – 1,000  $\mu\text{m}$ , in which fibers were more than 75% of MPs detected in water from the sludge centrifugation unit. In dry sludge, the MPs concentration was 35,680 particles/kg DW. The highest concentration of particles was between 500 and 100  $\mu\text{m}$ , followed by a fraction of 100 – 50  $\mu\text{m}$ . Similar to the water from sludge centrifugation, fibers were also the most abundant morphology in dry sludge; however, it can be observed that their concentration decreases from larger size ranges (2,000 – 1,000  $\mu\text{m}$ ) to the smallest (50 – 20  $\mu\text{m}$ ) while abundance of film and fragment increase significantly. High fiber content found in the sludge was correlated with high removal efficiency from the effluent [65].

Recently, the sludge management plan has been designed to overcome environmental impact based on harmfulness from coagulant compositions [66-68]. However, with more concern of MPs as an emerging pollutant that occurred in WTS, the way of sludge management has to be improved.

### **3.3.5. Filtration**

#### **3.3.5.1 Conventional filtration**

Following the sedimentation process, the filtration aims to eliminate any remaining fine particles with various filter media including sand, gravel, anthracite, and activated carbon. The efficiencies of MPs removal through conventional filtration at both experimental and full-scale water treatment were assessed and shown in Table S7 and Table S8, respectively. Experimental

studies often utilized filter columns with various media types and pore sizes to replicate filtration conditions and assess the efficiency across different filtration rates and MPs characteristics. Key factors influencing filtration efficiency include media type, pore size, filtration rate, filter bed depth, water quality (particle size and concentration), and operational conditions (pressure, backwash, media regeneration). Understanding and optimizing these parameters are essential for effective MPs removal and overall filtration performance.

Based on our comprehensive review, MPs particle size significantly affected the performance of filtration system regardless of plastic type. However, the results seem to be contradictory. Shao et al. [60] showed a microplastic removal rate of up to 100% in the sand filter process after CFS stages. According to Zhang et al. [51], the removal efficiency of MPs in filtration processes is significantly influenced by size of particles with a non-proportional relationship. This study revealed the lowest removal efficiency (approximately 97%) for MPs in the size range of 10 – 20  $\mu\text{m}$ . Cherniak et. al. [69] noted limitations in the removal efficiency by conventional treatment for microfibers <45  $\mu\text{m}$ . Nevertheless, filtration is highly effective for removing NPs and MPs, especially those larger than 100  $\mu\text{m}$ , aligning with their limited presence in treated water. However, particles in the size range of 1  $\mu\text{m}$  to 10 – 20  $\mu\text{m}$  were still detected. The particle removal involves mechanisms like straining, interception, sedimentation, diffusion, and attachment/detachment, with the overall rate shaped by factors such as particle sizes, bed porosity, and flow rates. Similarly, Na et. al. [57] demonstrated complete removal of 45 and 90  $\mu\text{m}$  MPs, but for 10 – 20  $\mu\text{m}$  particles, the removal rate was only around 15%. The transport of MPs in porous media was influenced by their size. The two-site model revealed that attachment and detachment, as well as straining, predominantly determined MPs (PS) retention in the sand column. These mechanisms depend on the collective attributes of the collector and particle, with the attachment influenced by single-



collector collisions and particle size variation, while straining operates based on the explicit ratio between particle and collector diameters.

Furthermore, Pulido-Reyes et. al. [70] explored the behavior of NPs during sand and activated carbon (AC) filtration. Their study emphasized that slow sand filtration exhibited the highest NPs removal efficiency due to biofilms formed in both rapid and slow sand filtration that can effectively retain NPs. NPs retention in the filter column ranged from 22% – 60% depending on flow conditions, with higher flow rates decreasing the retention. This decrease was attributed to shorter NPs residence times and increased drag forces. Anticipated improvements in NPs removal in slow sand filters involve increased filter depth. Surface characteristics of filter media, influenced by biofilm formation, also impact NPs retention by modifying the filtration medium's surface properties, suggesting potential enhancements in NPs removal. In the same way, Arenas et. al. [71] demonstrated that NPs removal through sand and granular activated carbon (GAC) filtration achieved an overall efficiency of 88.1% even without coagulation. The effectiveness of sand filtration was further enhanced by coagulant addition, increasing efficiency from 54.3% – 99.2%. This enhancement suggests substantial removal of PS nanoplastics through sand filtration. Coagulant addition likely promoted the formation of larger NPs aggregates, improving their retention in the filter media by reducing repulsive forces. GAC filtration surpassed sand filtration in NPs removal due to its porous structure and strong adsorption capacity. Despite electrostatic repulsion,  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  ions facilitated NPs adsorption onto GAC surfaces and pores, introducing new adsorption sites. However, granular filtration may not entirely eliminate all nanoparticles. Further research is needed to detect naturally-occurring NPs using advanced analytical techniques. Concerning the choice of media, no distinct advantage was apparent between GAC and anthracite under identical conditions.

Translating MPs removal research from controlled laboratory setting into practical applications involves evaluating the role of various factors on the MPs removal efficiency including media characteristics, filtration unit configuration, and MPs' properties, as presented in Table S8. Unfortunately, predicting filtration efficiency proves challenging due to fluctuating effectiveness. Wang et al. [72] studied at the Yangtze River Delta's major drinking water treatment plant and found that sand filtration has an insignificant role in MPs removal. The conventional treatment process achieved a removal efficiency of 58.9% – 70.5%, with the complete removal of MPs larger than 10  $\mu\text{m}$ . Coagulation, sedimentation, and GAC filtration were the dominant removal mechanisms, influenced by shapes and compositions of MPs. GAC filtration also effectively removed MPs in ozonation effluent. Integrating ozonation and GAC filtration enhanced MPs removal by 17.2 – 22.2%, primarily targeting those sized 1 – 5  $\mu\text{m}$ .

Contrary to some studies, recent research emphasizes the crucial role of filtration processes, including sand, silica, anthracite, GAC filtration, or multistage media filtration, in MPs removal. Dalmau-Soler et. al. [73], Kankanige and Babel [37], and Dronjak et. al. [65] highlighted that sand filtration as pivotal, achieving up to 99% elimination in the initial treatment stages of coagulation, sedimentation, and sand filtration. Strategies to enhance smaller-sized MPs retention include reducing the lower sand layer's pore size and increasing filter medium thickness. Pivokonský et. al. [74] noted that clay-based and GAC filtration material commercially available as Filtralite significantly contributed to the MPs removal, accounting for approximately 50% of the additional efficiency compared to CFS. Deep-bed filtration, whether using Filtralite post-sedimentation or GAC later, significantly aids microplastics removal. This aligns with the findings of Wang et al. [72] and Jung et al. [75], resulting in a slight shift in particle size distribution. This shift may be

attributed to the fragmentation of microplastics caused by friction with the filter media, as suggested by Radityaningrum et al. [76].

Turning to the single sand filtration, Wu et. al. [77] found that the removal efficiency for fibrous microplastics (100  $\mu\text{m}$  – 5 mm) was 71.7%, lower than the efficiency for granular microplastics in the same size range (87.8%). Notably, PE granules were significantly severely broken during sand filtration, consistent with findings by Na et al. [57]. The decrease in microplastic size led to denser hydroxyl groups on their surfaces, forming stronger hydrogen bonds with silica grains. Consequently, smaller MPs adhered more strongly to the silica bed, making them challenging to clean during backwash procedures [16, 78]. The fragmentation of MPs poses a potential hazard to sand filtration effectiveness, requiring further simulation work to assess concealed risks.

MPs leakage from the filtration unit could be attributed to turbulence in water flow, displacing particles trapped in the filter media [76]. Further MPs removal can be achieved by membrane technology such as ultrafiltration and reverse osmosis demonstrated higher removal rates compared to the upgraded conventional treatment.

### **3.3.5.2 Membrane technology**

Macromolecules, colloids, and suspended particles can all be concentrated, purified, or separated from solution using the pressure-driven separation technique known as microfiltration (MF) with pores ranged 0.1 – 1.0  $\mu\text{m}$  with applied pressures range of 0.1 – 2 bar. According to Table S7, MPs removal efficiency of MF membranes in water treatment were almost complete. However, the decline in removal efficiency observed during the membrane filtration process was attributed to both membrane cleaning and aging. This led to the rupture of the filter membrane, subsequently causing an increase in the number of MPs/NPs [79]. As shown in the previous

sections that conventional DWTPs are not very effective at removing small-sized MPs (average size of 100  $\mu\text{m}$ ), this portion of small MPs can be further removed by using MF membrane. Even though the pore size of MF membranes was smaller than that of MPs, it was revealed in the study by Pizzichetti et al. [80] that MPs were still present in the permeate of the MF membrane after filtration.

Due to its low energy consumption, high separation efficiency and small plant size, ultrafiltration (UF) is one of solutions for water treatment since it enables the attainment of high-quality drinking water in a good economic consideration. Using asymmetric UF membranes with pores between 1 – 100 nm, it is a low-pressure (1 – 10 bar) method that may reject macromolecules and particles, e.g., proteins, fatty acids, germs, protozoa, viruses, and suspended solids. In water treatment plants, UF can replace the processes of CFS treatment and/or sand filtration and chlorination.

Nowadays, one of the primary water treatment technologies in the advanced water treatment plants in water is the UF process incorporated with the coagulation stage. These technologies, however, were not originally designed to remove MPs as the process is typically employed for removing ions such as NOM. NOM constitutes a complex matrix of organic substances with diverse chemical characteristics, chemical composition, and molecular weight, thus the article indeed presented the removal of NOM [53]. Thus far, few studies have examined the removal of MPs for the manufacture of drinking water using the coagulation and UF processes. Ma et al. [47] studied on the PE removal behavior in UF and coagulation processes using Fe-based as a coagulant. PE is the most prevalent plastic contaminant found in water, and because of its density (0.92 – 0.97  $\text{g}/\text{cm}^3$ ), which is nearly water density, it is challenging to remove MPs using water treatment techniques. Enfrin et al. [81] reported that MPs are hydrophobic, as a result, MPs and

hydrophobic or hydrophilic surfaces of membranes interact in differently. Since surface charges of membrane are varied, then the repulsion is caused by the opposite charge of the membrane surface, resulting in effectiveness of removal [60, 82, 83]. Furthermore, Xiong et. al. [84] found that MPs with biofilm covering on the surface can accumulate on the membrane surface, causing the biofouling layer to become more hydrophobic with a dense and compact surface due to the accumulation of extracellular polymeric substances.

Reverse osmosis (RO) is applied in municipal and industrial water treatment systems to purify water by removing salts, pollutants, heavy metals, and other impurities using nonporous or nanofiltration membranes (pore size  $>2$  nm). It is operated by passing a concentrated water solution under high pressure (10 – 100 bar) through a semipermeable membrane, leaving all the other components essentially in a highly concentrated water solution. Recently, there are very few studies focusing on the efficiency of MPs removal from drinking water treatment via RO technology [85]. The presence of MPs in surface waters has potential to form fouling, which is one of the most important factors and a significant obstacle to membrane performance of RO technology, leading to the flux reduction and increase its pressure. In order to keep the flux rates constant and reduce the membrane fouling, a pretreatment stage is necessary that can enhance the membrane life and reduce the membrane cleaning in the process (CIP). The chemicals and reagents including coagulants, antiscalants, oxidizing agents, and disinfectants are a typical part of conventional pretreatment of RO procedure. Other methods including cleaning and surface modification could be employed to reduce fouling and maintain the membrane performance during operation. Nowadays, the desalination process uses RO pretreatment along with a UF pretreatment to provide consistent performance in terms of water quality and flux. Ashfaq et al. [86] revealed a rising trend in the use of mixed RO-UF systems for desalination on an industrial scale. Concerning

the presence of MPs as a potential fouling, pre-treatment of the influent either by ordinary coagulation-sedimentation or membrane filtration is strongly required.

### **3.3.6. Biological treatment**

Biological water treatment is driven by two major processes: biodegradation by active microorganisms and biosorption onto biomass. Biodegradation of MPs by microorganisms is effective in laboratory scale, subjective to the type of MPs, and is still ongoing in research [87, 88]. Data showed that the biological contact oxidation tank as a pretreatment unit of DWTP could not remove 5  $\mu\text{m}$  – 5 mm granular MPs; on the other hand, the concentration was increased from 3444.7 particles/L in raw water to 7789.1 particles/L in the effluent of the biological treatment [77]. For this reason, biofiltration that relies on filtration and biosorption should be considered a more effective system for MPs removal than the conventional biological treatment. Normally, GAC filtration exhibited certain efficiencies in MPs removal [72]. After operation, GAC filtration will be transformed to biological activated carbon (BAC) filtration due to biofilm formation, which increased the effectiveness of pollutant removal including MPs [89]. Wu et al. [77] reported that the BAC filtration was able to significantly remove more than 60% of the granular MPs  $\leq 20 \mu\text{m}$ . However, the effluent of BAC filtration showed a slight increase in the number of fibrous MPs. This could be due to the fragmentation of MPs (abrasion and collision) resulting in the breakdown of MPs and generating more numbers in water [16]. Since there is the intrinsic force of adhesion between microorganisms and physicochemical surface of MPs [90], so biofilm of microorganisms can occur on these small pieces of MPs, contributing to more biosorption by filtration process. Zhang et al. [51] observed the removal of MPs and NPs ranging between 180 nm to 125  $\mu\text{m}$  by granular filtration combined with CFS for drinking water treatment. The results showed that biofilm could rapidly form on MPs, and this biofilm formation on MPs significantly increased the

efficiency of CFS from < 2.0% to 16.5% in MPs removal. Since biological treatment alone might not be effective enough for MPs removal in water treatment, the combination with other physicochemical processes should be aimed for future applications.

### **3.3.7. Disinfection**

In drinking water treatment, a disinfection process must be performed to ensure the absence of pathogenic agents in the drinking water. Generally, chlorination, ozonation and ultraviolet irradiation are the most frequently employed disinfecting techniques. Disinfection is also applied as a pre-oxidation treatment to facilitate the following water treatment processes. The reviewed articles indicated that the pre-treatment by disinfection removed no notable microplastic abundance [75, 78]. Several conventional pre-oxidants (potassium permanganate, sodium hypochlorite, and ozone) used in pre-oxidization changed the hydrophobicity, specific surface area, and functional groups of PE surface exposing to water phase. However, these changes had only minor effect on the removal of the PE microplastics by subsequent CFS and filtration process [79]. In contrast, using the pre-ozonation process before biological pretreatment can generate carbonyl groups on the plastic surface, enhancing the microbial mineralization of MPs [77], which may lead to further biodegradation of the MPs in WTPs.

Based on the articles reviewed in the present study, ozonation is the most typical technique used for disinfection (Table S1 and Table S2). In terms of MPs elimination, ozonation did not significantly improve MPs removal efficiency [69, 74]. Most studies found an increased number of MPs in the effluent of the ozonation tank, especially small MPs in size of 1 – 20  $\mu\text{m}$  [57, 77]. The reason for the higher number of MPs is possibly because MPs were first weakened by oxidation, and then fragmented due to external forces like the shearing force of the water flow and aeration, which led to the generation of smaller MPs. The fragmentation and oxidation could be

accelerated by the presence of aged MPs, which were long term degraded in natural water before entering into WTPs [91]. The MPs breakdown was proved to be dependent on ozone exposing time and dosage. An increase in ozone dosage and the reaction time enhanced the degree of oxidation [92]. Another study observed that ozonation did not result in fragmentation or agglomeration of the NPs [70]; however, the surface charge of the NPs decreased with increasing ozone doses. The authors concluded that this alteration facilitated the transport of NPs in the porous media. To improve the MPs removal efficiency, the use of ozonation combined with GAC filtration revealed an increase in elimination of small MPs in size range of 1 – 5  $\mu\text{m}$ , 10 – 50  $\mu\text{m}$ , and fiber up to 22% [72, 74]. Since the subsequent GAC filtration could effectively remove MPs in the effluent of ozonation, further studies on the removal mechanism and operating parameters adjustment of the GAC filtration (e.g., GAC size, filtration rate, and backwashing cycle) should be conducted in the future.

By treating water with chlorination, reduction of MPs abundance has been reported up to 28% [37] and up to 71% [76]. Chlorination can also cause the alteration of physical characteristics and structure of MPs by performing with high dose and contact time of chlorine, but at higher level than the typical applied ones [76]. For the disinfection by UV-based oxidation, a study compared the degradation of MPs by UV photolysis with and without the addition of  $\text{H}_2\text{O}_2$  into the treating water (UV and UV/ $\text{H}_2\text{O}_2$  treatment), the results presented that UV/ $\text{H}_2\text{O}_2$  treatment promoted the MPs fragmentation, MPs alterations in physical properties (i.e., size and morphology), and chemical leaching more significantly than only UV treatment. Both UV and UV/ $\text{H}_2\text{O}_2$  treatment caused the remarkable increase of the smaller MPs size ranges and the total particles number concentration [57]. Thus far, there is still a lack of knowledge of MPs removal and mechanism in disinfection processes.



### 3.4. Water distribution system

Water distribution systems consist of interconnected pipes, pumps, and valves aimed at transporting treated water to consumers. The primary objectives are maintaining water quality standards, preventing leakage, minimizing water loss, and safeguarding water quality. Emphasis is placed on addressing pipe corrosion issues to avoid discoloration and microbial contamination in drinking water, ensuring it remains clean and safe for consumption [17]. Nowadays, it was discovered that water distribution systems can release MPs through a variety of mechanisms, including pipe corrosion and abrasion of fittings [17, 37, 73]. After purification, the drinking water is stored in tanks or directly fed into the distribution system and transported to the consumers. Once released, the MPs can directly contaminate the tap water and can be a potential risk to human health. Most possible sources of MPs in water distribution system are the component materials of pipe and fitting. Generally, pipes in water distribution systems in DWTPs or households are frequently made of PE, PVC, PP, or cast iron, with PA (nylon) fittings widely used to connect pipes in plumbing system [18, 27]. Besides, iron storage tanks and some cast iron pipe have an inner layer of epoxy resin to avoid corrosion [79]. Even though plastic is a material that is known for its durability and resistance, it is susceptible to abrasion. Pipe materials, along with water flow rate, temperature, and pH, significantly play a role in the growth and formation of biofilms in the pipe [93]. These pipewall biofilms provide a source of microbes to form on the surface of MPs and NPs in drinking water, and the microbial communities of the MPs-biofilms can change over time from the hydraulic conditions in the pipe, which might be harmful to humans if they are opportunistic pathogens [94, 95].

The following studies provided evidence indicating that MPs are being released from water distribution systems. Dalmau-Soler J. et al. [73] studied the existence of MPs in drinking water

along the water distribution supply network in Barcelona, Spain. The study found that MPs in the size range of 83 – 3,228  $\mu\text{m}$  were present in both fibers (58%) and fragments (42%) shapes, which is consistent with the results observed in the outlet of the DWTP. This study showed that PE has not been detected in any samples analyzed, which could be indicated that there is no contribution of the PE pipes even if PE mostly constitutes the pipe material of each sector along the network. This result is contrasted with the results observed in their previous study. The authors indicated that the age of pipelines may be a contributing factor to the presence of MPs, with pipes at the end of their operational lifetime being the potential source of MPs, while pipelines in good condition may not exhibit these behaviors. Conversely, the assessment recently performed by Kristien et al. [19] on a Swedish drinking water distribution system concluded that the age of pipes had no significant impact on MPs load. Additionally, we found in the study of Dalmau-Soler J. et al. [96] that there appears to be no clear relationship between the concentration of detected MPs and the linear distance from the DWTP, which supports their previous hypothesis. The contamination and removability of  $\geq 6.5$   $\mu\text{m}$ -sized MPs in conventional water treatment in Thailand have been examined by Kankanige and Bable [37], who found that the accessories used in the treatment units, such as distribution pipes and fittings made of PE, PP, PA, and PVC, can be the causes of additional MPs contamination due to abrasion. In the study of Chu et al. [79], the MPs in the water distribution system were examined by sampling the water from pipe scales from three sections of the burst metal pipes (approximately 20 years in service) of two water distribution systems (pipes 1 and 2 were obtained from the same water distribution system). The occurrence of MPs ranged from 569.99 – 751.73 particles/kg in the pipe scale samples. For the MPs characteristics, the authors found that most of detected MPs were  $<100$   $\mu\text{m}$  in size, and 70.91% of the MPs in pipe 3 were  $<50$   $\mu\text{m}$  in size. Overall, fragments were the most abundant in the samples, accounting for

38.61 – 64.55% of the MPs in the pipe scale samples followed by MPs in spherical shape ranging from 13.76% – 45.13%. The most common colors of the MPs were brown, black, and transparent. Considering the type of the polymers, PVC (42.86%) was mainly found followed by PE, PP, and nylon. Furthermore, there is a possibility of MPs contamination in water distribution system from atmosphere. Cherniak et al. [69] studied the performance and impacts of ozone on MPs removal in the full water treatment plant in Ontario, Canada. The results found that the number of MPs particles increased during clearwell, indicating deposition of airborne contaminants occurred in the open-air vented clearwell for particles <300 µm and especially for 10 – 125 µm particles. In contrast, in the distribution system using PVC pipe 1 km away from the plant, the number of particles reduced due to particles settling or adsorption onto biofilm in the service pipe. A study provides direct evidence that pipe scales can adsorb a large amount of MPs, indicating that adsorption can influence their occurrence and distribution in water supply systems [79]. However, the MPs in pipe scales may be released with changes in the hydraulic conditions, increasing the risk to contaminate MPs to tap water.

Apart from MPs occurrence and release in water distribution system, preliminary migration tests of microplastics from materials used in the DWTP performed by Dalmau-Soler J. et al. [73] indicated that some old and worn elements from the DWTP could be a potential source of MPs. The study found that PE water supply pipe released several PE fibers and fragments in the migration experiments, but PE was not detected in the final treated water. Due to this uncertainty, the authors strongly suggested to conduct successive migration experiments to elucidate whether materials used in treatment system can act as a potential source of microplastics in waterworks including water distribution system.

In conclusion, this review highlights the significant role of water distribution systems in the contamination of drinking water with MPs, which poses a risk to human health. While evidence suggests the occurrence and release of the MPs in water distribution systems, the extent of their contribution to contamination remains unclear. Thus, further in-depth research is still necessary to fully understand the impact of water distribution systems on MPs contamination and to develop effective strategies for mitigating this issue.

### 3.5. Occurrence microplastics in tap water

The presence of MPs in tap water has been established by scientific research in various locations. However, there remains a dearth of information regarding the extent of microplastic pollution in tap water globally. This is largely due to significant variations in reported MPs concentrations as well as differences in sampling methods, sample handling, and sample analysis, which have made it challenging to directly compare findings across studies. To address this knowledge gap, several studies have investigated the levels of MPs in tap water in different regions. In this section, the overview of these studies that have been conducted and summarize their findings. Furthermore, to facilitate comparisons between studies the abundance of MPs detected in tap water based on information obtained from the reviewed literature were summarized in Table 2.

**Table 2** Occurrence of MPs in tap water

Sample location	MPs concentration (particles/L)			Reference
	Min	Max	Average	
China	0	1247	440	[97]
Changsha, China	254	446	343.5	[98]
Tianjin, China	NA	NA	13.23	[79]
Chiba, Japan	1.9	225	39	[99]
Tokyo, Japan	4	81	29.75	
Kanagawa, Japan	10	14	12	
Yamagata, Japan	2	11	7	

California and Nevada, U.S.	11	80	46	
Helsinki, Finland	25	61	47	
Paris, France	46	131	97	
Munich, Germany	22	79	53	
Germany	5	74.5	17.28	[100]
Barcelona, Spain	0.02	0.13	0.04	[96]
Essen, Belgium	0	0	0	[101]
Kluizen, Belgium	0	0	0	
Egenhove, Belgium	0	0	0	
Oelegem, Belgium	0	0.01	0.01	
Puttebos, Belgium	0	0	0	
Torreele, Belgium	0.0	0.06	0.05	
Blankaert, Belgium	0	0.05	0.05	
De Gavars, Belgium	0	0	0	

In a study of Shen et al. [98], MPs in tap water were examined in April, May, June, and July 2019 with the detected MPs concentrations of  $267 \pm 11$ ,  $321 \pm 11$ ,  $381 \pm 18$ , and  $405 \pm 30$  particles/L, respectively. This indicated that the MPs abundance in tap water was not significantly different from the treated water. Considering the type of MPs in the tap water, the content of PE, PP, PET, and PA was not significantly change compared with the treated water, while the abundance of PVC obviously increased. In this study, the water pipes in households are made of PVC materials, which may be a plausible explanation for the increase of PVC in tap water and showing that drinking water supply chains might contribute to PVC contamination in tap water. This finding is similar to the study by Tong et al. [97] that indicated a higher concentration of MPs in tap water compared with that of the outlet from water treatment plant in this study. In contrast, Chu et al. [79] found lower MPs concentration in tap water (13.23 particles/L) than that in the effluent (95.63 particles/L). This decrease in MPs abundance could be possibly due to the influence of adsorption in the drinking water distribution system.

PVC pipe as a potential source of MPs in tap water was also studied in the research of Mukotaka et al. [99] due to a significant number of PVC microplastics detected in tap water. A

total of 42 tap water samples collected from 12 cities and states from 5 developed countries demonstrated that the concentrations of the MPs at each site ranged from 1.9 to 225 particles/L with the size range of 19.2  $\mu\text{m}$  to 4.2 mm. Fragments were the predominant shape in these tap water samples while fibers and spheres were also observed. For the polymer class, 29 types of MPs were detected, in which PS is the most abundant followed by Styrene ethylene butylene styrene (SEBS), PP, PES, PE, and other plastic types such as PVC, respectively. However, the contamination source of the first three major plastic classes were not clearly indicated. The authors deduced that the tap water samples may be contaminated with MPs in the water distribution system by deriving them from the air or water pipes.

Aside from PVC pipe, sealing materials like PTFE and silicone were concerned as the source of MPs in tap water. The study of Dalmau-Soler et al. [96] investigated 21 tap water samples along a large distribution water supply network. MPs were detected in 38% of samples with a mean concentration of 0.01 particles/L and the polymers identified were PP, polysulfone (PES), and PA, which are related with the polymer type detected in the outlet of DWTP from their previous study [73]. Moreover, polytetrafluoroethylene (PTFE) particles were also detected in 29% of the samples ranging from 0.02 to 0.08 particles/L, even though PTFE is not used in the pipelines of the drinking water supply network, and it was not detected at the outlet of the DWTP. The authors pointed out that this PTFE contamination is related to the tape used in the junctions and plugs within the maintenance works in the sampling cabinets to avoid water leaking. However, PTFE can act as a hypothetical source of MPs, since the tape is widely used to prevent leakage from screwed connections. Apart from PTFE, sampling cabinets released silicone, which was found in two samples at concentrations of 0.13 and 0.07 particles/L. This material was used in these two cabinets to strengthen junctions and to avoid water leaks and clearly was leached to water. The results from

734 this research showed that there is no contribution of PE water distribution pipes as PE was not  
735 detected in these tap water samples.

736 Release of MPs from the water distribution system to tap water was not also noticed in study  
737 of Semmouri et al. [101], who studied MPs in drinking water focusing on 1 – 5,000 µm from  
738 different sources, including groundwater, surface water, and treated sewage water in Flanders,  
739 Belgium. They found that the average concentration of MPs in DWTP and tap water samples was  
740 5.59 and 3.76 particles/L, respectively, which do not differ significantly from each other.  
741 Additionally, no significant differences were observed in MPs concentrations among the samples  
742 from different water sources. PP and PET were the most common polymer in the samples.  
743 Furthermore, Weber et al. [100] indicated that the treatment in DWTPs and the transport in a  
744 regional and municipal supply network do not lead to an entry of microplastics  $\geq 10$  µm in the  
745 samples. The authors investigated MPs  $\geq 10$  µm in the drinking water of a German city. The  
746 samples were randomly collected from three house connections, one transfer station, and five  
747 consumption taps in an educational institution, an apartment, a single-family house, a residential  
748 building, and a commercial enterprise. From this study, the determined number of MPs of the  
749 house connections, the transfer station, and four out of five consumption tap samples was not  
750 significantly above the average blank value. Besides, no MPs were detected in the samples from  
751 the consumption tap of the educational institution.

752 Based on the present literature review, most of MPs can be removed by existing water  
753 treatment plants prior to being distributed. However, there are some MPs residues in tap water that  
754 still have been detected without clear major source of MPs contamination after the water was  
755 treated from WTPs.

#### 4. Conclusions, future perspectives, and suggestions

This article highlights the importance of standardizing experimental protocols and analytical methods for extracting, separating, and characterizing MPs in water treatment. It emphasizes the susceptibility of MPs, especially fibers, to breakage during treatment, potentially leading to higher concentrations at the outlet. Consequently, understanding fragmentation mechanisms is also essential, and further research on weathered plastics is needed to reflect actual conditions. Moreover, expressing MPs concentration in consistent units, considering their varied sizes and shapes, is crucial for effective comparison and analysis.

Another important finding from this article is the crucial need to develop standard water treatment techniques for MPs and NPs removal. Our review revealed that a range of treatment technologies, spanning from conventional to advanced methods, might be unable to entirely eradicate these MPs in the ultimate effluents. However, among the conventional methods, water clarification processes that encompasses CFS have shown a great potential for future development. Even though the removal efficiency remains highly variable and difficult to predict depending on operating conditions, these processes are able to remove a significant portion of MPs both in bench-scale experiments and in water treatment plants. Different mechanisms are simultaneously involved in the removal of MPs via coagulation-flocculation. The relative contribution of each of these mechanisms depends on various factors, including the characteristics of the coagulant/flocculant, the MPs themselves (concentration, size, form, zeta potential), and water characteristics (turbidity, pH, NOM). At the present time, there is an increasing number of studies concerning the natural coagulant. Their environmentally friendly, safe, biodegradable, and sustainable features make them a real and credible alternative to traditional coagulants. In addition, optimizing the retention time in sedimentation can significantly enhance MPs removal. Therefore,



future studies should investigate appropriate retention times for various MPs and treatment conditions, assess reality applicability, and explore innovative sedimentation designs.

Apart from the conventional processes, advanced treatments such as ultrafiltration and reverse osmosis demonstrate effectiveness in the removal of MPs. The integration of these separation technologies with other approaches has the potential to achieve efficient separation of MPs and seems to be higher when coupled with biological treatments. Therefore, applications of these advanced techniques for water treatment should also be considered in the decisions regarding selection of effective MPs removal processes.

Finally, while treating water in treatment plants seems feasible, there remain two challenges that research needs to tackle: the management of sludge and the optimization of water distribution systems. The type of MPs found in tap water certainly come from pipe and tap materials also including fitting and sealing; however, the source identification of MPs occurred in tap water need further comprehensive study of materials used for the whole water supply system along with the environment surrounding the system. Future research inquiries must also include understanding the transportation of MPs in sludges, particularly the disposal of sludge laden with MPs to agricultural fields and its potential impact on soil and groundwater contamination. Therefore, the fate of MPs during the drinking water supply chain should be valued and given attentions, and standards of the content of microplastics in drinking water should be formulated.

## 5. Abbreviations

Abbreviations	Definitions
ABS	Acrylonitrile butadiene styrene
AC	Activated carbon
AR	Alkyd resin
CA	Cellulose Acetate
CTAB	Cetyltrimethyl ammonium bromide

DWTP	Drinking water treatment plant
EDS	Energy - dispersive X-ray spectroscopy
FITR	Fourier transform infrared spectrometer
GAC	Granular activated carbon
LDPE	Low density polyethylene
MPs	Microplastics
NPs	Nanoplastics
NOM	Natural organic matter
PA	Polyamide
PA12	Polylauro lactam
PAC	Poly Aluminum Chloride
PAM	Polyacrylamide
PE	Polyethylene
PES	Polysulfone
PEST	Polyester
PET	Polyethylene terephthalate
PMMA	Polymethyl methacrylate
PolyDADMAC, PDADMAC	Polydiallyldimethylammonium chloride
PP	Polypropylene
PPS	Polyphenylene sulfite
PRISMA	Preferred Reporting Item for Systematic Reviews and Meta-Analysis
PS	Polystyrene
PTFE	Polytetrafluoroethylene
PTT	Polytrimethylene terephthalate
PU or PUR	Polyurethane
PVC	Polyvinyl Chloride
PVDF	Polyvinylidene fluoride
SEBS	Styrene ethylene butylene styrene
SEM	Scanning electron microscope
SLR	Systematic Literature Review
VINYL	Vinyl chloride-vinyl acetate copolymer
WTP	Water treatment plant
XPS	X-ray photoelectron spectrometer

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1106 **Supplementary data for Removal of microplastics and nanoplastic in water treatment**  
1107 **processes: A systematic literature review**

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1122 **This supplementary data contains the following sections, tables and figures:**

1123 **S1** Summary of studies carried out to date on MPs and NPs in drinking water treatment

1124 **S2** MPs sampling, treatment, and analysis techniques

1125 **S3** Coagulation mechanisms

1126 **S4** Relationship between MPs' characteristics and the removal efficiency of the CFS process

1127 **S5** The MPs removal efficiency of coagulation-flocculation-sedimentation process

1128 **S6** The MPs removal efficiency of filtration process

1129 **S1 Summary of studies carried out to date on MPs and NPs in drinking water treatment**

1130 **Table S1** Microplastics' overall removal efficiency including analytical techniques and their characteristics throughout water treatment plant

Countries	Name of DWTPs	Systems	MPs Analytical techniques	MPs type	MPs size (μm)	MPs shape	Inlet MPs conc. (particles/L)	Total removal efficiency (%) <sup>a</sup>	Reference
Yangtze River Delta, China	ADWTP	Coagulation/flocculation, Sedimentation, Sand filtration, and ADV treatment units: Ozonation combined with GAC filtration	μ- Raman, SEM	PET, PE, PP, PA, and others	1 to >100	fibers, fragments, sphere	6614 ± 1132	88.6	[72]
Czech Republic	Milence	Coagulation/flocculation, Sand filtration, Disinfection	μ- Raman, SEM	CA, PET, PVC, PE, PP, EVA, PBA, PTT, and PTFE	1 to >100	fragments, fibers	23 ± 2	80	[74]
	Plzeň	Coagulation/flocculation, Sedimentation, Sand filtration, Ozonation + GAC filtration, UV absorption, Disinfection	μ- Raman, SEM	CA, PET, PVC, PE, PP, EVA, PS, PA6, PEO + PEG, and others	1 to >100	fragments, fibers	1296 ± 35	92	
Catalonia, Spain	Sant Joan Despi	Coagulation/flocculation, Sedimentation, Sand filtration, Ozonation and GAC filtration, Ultrafiltration and Reverse Osmosis	stereo microscope, μ-FTIR	PE, ABS, PVA, ER, PTFE, PA, AR, PAN, PS, PES, and PP	20 – 5000	fibers, fragments	0.96 ± 0.46	93.96	[73]
China	Changsha	Aeration, Coagulation/sedimentation, Sand filtration and GAC filtration	stereo microscope, SEM, μ-FTIR, μ-Raman	PA, PVC, and PS	1 to >100	fragments, fibers, spheres	2173 ± 112 to 3998 ± 246	87.22	[98]
Surabaya, Indonesia	DWTPs I	Aeration, Pre-sedimentation, Coagulation, Flocculation-Sedimentation, Filtration, Disinfection	dissecting microscope, FTIR	cellophane, PE, PP, LDPE, polytrimoliticamide, PA 6, PA 6.6, and PES	1 – 5000	fibers, fragments, films, and pellets	26.8	40.58	[76]
	DWTPs II	Aeration, Pre-sedimentation, Coagulation, Flocculation-Sedimentation, Filtration, Disinfection	dissecting microscope, FTIR	cellophane, PE, PP, LDPE, polytrimoliticamide, PA 6, PA 6.6, and PES	1 – 5000	fibers, fragments, films, and pellets	35	75.71	

1132 **Table S1** Microplastics' overall removal efficiency including analytical techniques and their characteristics throughout water treatment plant  
1133 (cont.)

Countries	Name of DWTPs	Systems	MPs Analytical techniques	MPs type	MPs size (μm)	MPs shape	Inlet MPs conc. (particles/L)	Total removal efficiency (%) <sup>a</sup>	Reference
South Korea	DWTPs (a)	Pre-ozonation, Coagulation/flocculation, Sedimentation, and Sand Filtration	μ- FTIR, SEM	PE, PP, PET, PMMA, PA, PS	20 to >100	irregulars, polygons, rounds	2.37	99.70	[75]
	DWTPs (b)	Pre-ozonation, Coagulation/flocculation, Sedimentation, and Sand filtration	μ- FTIR, SEM	PE, PP, PET, PMMA, PA, PS	20 to >100	irregulars, polygons, rounds	2.13	99.58	
	DWTPs (c)	Pre-ozonation, Coagulation/flocculation, Sedimentation, and Sand filtration	μ- FTIR, SEM	PE, PP, PET, PMMA, PS, SBR	20 to >100	irregulars, polygons, rounds	2.28	99.16	
	DWTPs (d)	Coagulation/flocculation, Sedimentation, and Sand filtration	μ- FTIR, SEM	PE, PP, PET, PMMA, PS, SBR	20 to >100	irregulars, polygons, rounds	*	*	
Bangkok, Thailand	WTP (Dry season)	Screen, Clarification (Coagulation, flocculation, Sedimentation), Dual Media Filtration (rapid sand/ anthracite coal), Chlorination	fluorescence microscope, optical microscopy, ATR-FTIR, confocal Raman	PE, PP, PET, PA, PVC, and others	6.5 to >500	fibers, fragments, films, and pellets	1385	67.6	[37]
	WTP (Rainy season)	Screen, Clarification (Coagulation, flocculation, Sedimentation), Dual Media Filtration (rapid sand/ anthracite coal), Chlorination	fluorescence microscope, optical microscopy, ATR-FTIR, confocal Raman	PE, PP, PET, PA, PVC, and others	6.5 to >500	fibers, fragments, films, and pellets	1796	57.2	
Jiaxing, China	DWPT	Pre-ozonation, Biological pretreatment, Coagulation and Sedimentation, Ozonation, Biological Activated Carbon (BAC) filtration, Micro-flocculation, Sand filtration	Metalloscope, μ-Raman	PP, PE, PVC, PA, VINYL, VINYON	10 – 1000	granules, fibers	3738.36 ± 461.29	81.18	[77]

1135 **Table S1** Microplastics' overall removal efficiency including analytical techniques and their characteristics throughout water treatment plant  
1136 (cont.)

Countries	Name of DWTPs	Systems	MPs Analytical techniques	MPs type	MPs size (μm)	MPs shape	Inlet MPs conc. (particles/L)	Total removal efficiency (%) <sup>a</sup>	Reference
Tianjin, China	DWTP	Coagulation/sedimentation, and Membrane filtration	stereomicroscope, μ-FTIR, SEM	Nylon, PEST	< 50, 50 – 100, > 200	fragment, fiber	134.79	49.69	[79]
Kolkata, India	DWPT	Pre-disinfection, Coagulation-flocculation, Pulse clarification, Sand filtration and Post-disinfection	optical microscope, ATR FT-IR	PET, PE, PP, PS, others	<25 to >100	fibers and films/fragments	17.88	84.6	[78]
Ontario, Canada	DWPT	Coagulation, Flocculation, Anthracite-sand filtration, and Chlorination	μ-Raman, stereo microscope	PET-PEST, PP, PU, PE, PBT, PAN, Acrylic, Silicone	10 to >500	fibers, fragments, films	42 ± 18	52	[69]
Tehran, Iran	DWTP 1	Screening, Coagulation and Flocculation, Sand filtration and Disinfection	SEM, μ-Raman	PP, PET, PE, PS, PTFE, PU, others	1 to >100	fibers, fragments, sphere	2808±80	50.1	[102]
	DWTP 2	Screening, Coagulation and Flocculation, Sand filtration and Disinfection	SEM, μ-Raman	PP, PET, PE, PS, PTFE, PU, others	1 to >100	fibers, fragments, spheres	1996±268	48.4	
	DWTP 3	Coagulation and Flocculation, Sand filtration and Disinfection	SEM, μ-Raman	PP, PET, PE, PS, PTFE, PU, others	1 to >100	fibers, fragments, spheres	2172±119	55.2	
Bangkok, Thailand	DWTP (Western)	Coagulation, Flocculation, Sedimentation, Sand filtration, and Chlorination	μ-Raman	PP, PE, PS, PVC, PET	50 to >1000	fragments, fibers	0.55 ± 0.15	-12.7	[32]
	DWTP (Eastern)	Coagulation and Flocculation, Sedimentation, Sand filtration, and Chlorination	μ-Raman	PP, PE, PS, PVC, PET	50 to >1000	fragments, fibers	0.94 ± 0.49	27.7	
Catalonia, Spain	DWPT	Clarifier, Sand filter, Carbon filter	stereoscopic microscope, μFTIR, ATR - FTIR	PES, PP, PA, PE, PET, PS, others	20 – 2000	fibers, fragments, films	4.230 ± 1.263	98.3	[65]

1137 <sup>a</sup>Maximum efficiency, \* No information provided

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1139 **Table S2** Information of microplastics' studies relating to water treatment process in experimental scale

Experimental Scale	Systems	MPs analytical techniques	MPs type	MPs size (μm)	MPs shape	tested MPs concentration (particles/L)	Removal efficiency (%) <sup>a</sup>	Reference
Bench	Coagulation, Filtration	optical microscope, camera, weighing	PE	<500 – 5000	fragment	$1.0 \times 10^{-4}$ g (0.1 mg/L)	40	[47]
Bench	Coagulation	light microscope	PE, rayon, PES	1– 5, 100, 900 – 1300, 500 – 1000, 3175	sphere, and fiber	5 and 25 mg/L	99	[58]
Bench	Coagulation/flocculation, Sedimentation (CFS), Ozonation, Filtration, and Chlorination	Epi-fluorescence microscope	PE, PS	0.18, 1.2, 10 – 20, 45 – 53, and 106 – 125	beads	1800, 2000, 9500	< 0.1 - 99.9	[51]
Bench	Membrane	FTIR-ATR, Raman, SEM, Dynamic Light Scattering, Nanoparticle tracking analysis (NTA)	PE	13 - 690	fragment	$2 \times 10^{11}$	*	[81]
Bench	Membrane	SEM	PVC	< 5	*	10	~ 100	[103]
Bench	Coagulation/flocculation, Settling	SEM, ATR- FTIR	PE, PS, PEST	10 – 20, 125 – 150, 130 – 150, 105 – 1325	sphere, and fiber	500	99	[56]
Bench	Pre-oxidization and following by Coagulation, Sedimentation, and Filtration	SEM, Raman	PE	250 – 300	fragment	*	*	[60]
Bench	Coagulation/Sedimentation, Sand filtration, and UV-based oxidation	Raman, weighing	PS	10, 20, 45, 90	bead	54 μg/mL (54 mg/L)	100	[57]
Bench	Coagulation/flocculation, Sedimentation	epi-fluorescence microscope	PS	3, 6, 25, 35, 90	sphere	223.4 ± 22.7, 215.1 ± 13.4, 239.2 ± 20.1, 88.8 ± 13.2, 60.6 ± 25.4	99	[104]
Bench	Continuous-flow ultrafiltration	SEM-EDS	PE	40 – 48	rod	4.21 g/L (4210 mg/L)	N/A	[84]
Bench	Ultrafiltration	fluorescence microscope, SEM	PS	0.1, 1, 10, 18	sphere	0.1 – 1 mg/L	100	[83]
Bench	UV-disinfection, Chlorine disinfection	μ- FTIR	PE, PA	10, 50	granular, and fiber	0 – 100 mg/L	*	[105]

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1142 **Table S2** Information of microplastics' studies relating to water treatment process in experimental scale (cont.)

Experimental Scale	Systems	MPs analytical techniques	MPs type	MPs size (μm)	MPs shape	tested MPs concentration (particles/L)	Removal efficiency (%) <sup>a</sup>	Reference
Bench	Membrane	SEM, optical microscope, fluorescence spectrophotometer, fluorescence microscope	PS	0.08 – 1	sphere	*	99.3	[106]
Pilot	GAC pilot filter	μ-Raman, stereo microscope	PET-PEST, PP, PU, PE, PBT, PAN, Acrylic, Silicone	10 to >500	Film, Fragment, Fiber	12.72 ± 0.39	-102.12 – 17.53	[69]
Bench	Dissolved air flotation	SEM, high speed camera	PE, PET, PA	2 to >25	fragment	0.1 g/L (100 mg/L)	100	[61]
Bench	Coagulation	Weighing, SEM, FITR, particle size analyzer	PS, PE	<500 – 5000	fragment	500 mg/L	98.5	[54]
Bench	Coagulation-Flocculation	Light microscope, Cryo-SEM	carboxylated PS	0.5	sphere	100	97.9	[42]
Bench	Coagulation/flocculation, and Sedimentation	optical microscope	PS	100	bead	10 mg/L	98.9	[59]
Bench	Coagulation	SEM, TOC analyzer	PVC	> 50	sphere	18 × 10 <sup>6</sup> (10 mg/L)	84.8	[52]
Bench and pilot	Ozonation, Rapid sand filter, Activated carbon filter, Slow sand filter	ICP- MS	a core/shell structure with a polyacrylonitrile core labeled with palladium (Pd) and a polystyrene shell	163.5 nm ± 0.3 nm to 214.7 nm ± 1.4 nm	sphere	1.7 and 141.6 mg/L	99.97	[70]
Bench	Coagulation-flocculation, and Sedimentation	Weighing	PE	150 – 200	fragment	500 mg/L	80.0	[55]
Pilot	Coagulation-Sand filtration, Ozonation, GAC filtration	SEM, nanoparticles tracking analysis	PS nanoplastic particles with amidine functional groups (PS-NH-NH <sub>2</sub> <sup>+</sup> )	124 ± 38 nm and 110 ± 25 nm	sphere	10 mg/L	99.4	[71]

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1145 **Table S2** Information of microplastics' studies relating to water treatment process in experimental scale (cont.)

Experimental Scale	Systems	MPs analytical techniques	MPs type	MPs size (µm)	MPs shape	tested MPs concentration (particles/L)	Removal efficiency (%) <sup>a</sup>	Reference
Bench	Coagulation	Turbidity meter, SEM	PS PMMA PVC PE	0.1, 0.5, 1, 5 0.5 0.5 2 – 10	sphere	8 mg/L	98.95	[44]
Bench	Ultraviolet (UV), chlorine and UV/chlorine disinfections	SEM, µ - FTIR, XPS	PP, PS, PVC	6.5	fragment	*	*	[107]
Bench	Coagulation-membrane filtration process	µ - FTIR, ATR – FTIR, laser diffraction	PE, PET	40 – 48, 300	fragment	10 mg/L	98.4	[53]
Bench	Coagulation	SEM, EDS, FTIR, Raman spectroscopy	PS	120 – 1000	fragment	7500 mg /L	98.0	[108]
Bench	Coagulation and Disinfection	FTIR, light scattering (SLS) particle size analyzer, SEM - EDX	PE	40 – 70 µm, 70 – 100 µm, 10 – 45 µm	Fragment, fragment, sphere	0.100 g/L	58.19	[46]

1146 <sup>a</sup>Maximum efficiency, \* No information provided

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1155 **Table S3** Reported MPs occurrence in drinking water, tap water, and water supply systems including treatment, distribution, and sludge

Countries	Focus	Analytical techniques	MPs type	MPs size	MPs shape	Concentration (particles/L)	Reference
Mexico	Metro station free drinking water fountain	Epi-fluorescence microscope, SEM-EDS, $\mu$ -Raman	PTT, epoxy resin	<500 – 5000	fibers, fragments	$18 \pm 7$ ( $5 \pm 2 - 91 \pm 14$ ) <sup>c</sup>	[33]
China	Investigated the presence of microplastics in tap water	fluorescence microscope, $\mu$ -Raman	PE, PP, PPS, PS, PET, others	1 – 5000	fragments, fibers, and spheres	440 (0 – 1247) <sup>c</sup>	[97]
China	Occurrence in surface water	dissecting microscope, $\mu$ -Raman, SEM	PP, PS, PE	48 – 5000	fibers, fragments, pellets, and styrofoams	$2594 \pm 3875$ particles/ m <sup>3</sup> (467 - 15,017) <sup>a</sup>	[29]
England and Wales	Report MPs that contaminate in water treatment for each process	FTIR	PE, PET, PP	<25 – 224	N/A	0 – 113 <sup>a</sup>	[63]
Southern Brazil	Reported MPs occurrence in raw and drinking water	fluorescence microscope	*	*	fibers, pellets, films	$330.2$ (0 – 940) <sup>a</sup> $105.8$ (2 – 459) <sup>d</sup>	[109]
Hong Kong	Microplastics in 110 surface water-sourced tap water samples	Stereomicroscope assisted by Rose Bengal Staining $\mu$ - Raman	*	50 – 4830	fibers, and films	$2.181 \pm 0.165$ (0.000 - 8.605)	[34]
Germany	Focusing on sludge after pre-ozonation, coagulation/flocculation with sedimentation, treatment		PE, PP, PS, PET, PVC, polyoxymethylene, PMMA, polysiloxane, ABS, acrylic resin, epoxy resin and silicone rubber	50 – 5000	fragments, fibers, and spherules	$196 \pm 42$ /m <sup>3</sup>	[64]

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1158 **Table S3** Reported MPs occurrence in drinking water, tap water, and water supply systems including treatment, distribution, and sludge (cont.)

Countries	Focus	Analytical techniques	MPs type	MPs size	MPs shape	Concentration (particles/L)	Reference
Japan	Analyzing 42 tap water samples collected from 5 countries	FTIR	PS, SEBS, PP, PES, PE, PVC, and others	19.2 – 4200	fragments, fibers, and spheres	39 ± 44 (6.7 ± 4.3 to 97 ± 45)	[99]
Norway	MPs in water supply system	pyrolysis-GCMS	PE, PA, PET, PP, PS, PVC	*	*	93.2 µg/m <sup>3</sup> <sup>a</sup>	[27]
Germany	MPs in drinking water	Raman	PE, PET, PP, PS	≥ 10	fragments, fibers	5 – 74.5 (17.28) <sup>b</sup>	[100]
Thailand	Abundance and characteristics of microplastics in freshwater and treated tap water	µ - Raman	PE, PET, PP, PS, PVC	50 – 999 <sup>a</sup> 50 – > 1000 <sup>c</sup>	fragments, fibers	0.43 ± 0.03 to 1.52 ± 0.53 <sup>a</sup>  0.68 ± 0.12 to 0.62 ± 0.38 <sup>c</sup>	[32]
Spain	MPs throughout a tap water supply	Stereomicroscope, µ - FTIR	PP, PES, PA	83 – 3228	fibers, fragments	0.02 – 0.05	[96]
Flanders, Belgium	Investigated the concentration of MPs in drinking water, samples were collected from both drinking water treatment plants (DWTP) and tap water (TW)	FTIR	PP <sup>d</sup> , PE <sup>d</sup> PET <sup>c</sup> , PP <sup>c</sup> , PVC <sup>c</sup> , PE <sup>c</sup>	25 – 550 <sup>d</sup> 25 – 400 <sup>c</sup>	* *	0.02 ± 0.03 <sup>d</sup> , 0.01 ± 0.02 <sup>c</sup>	[101]

1159 <sup>a</sup> MPs in raw water, <sup>b</sup> the concentrations are lower than in blanks, <sup>c</sup> MPs tap water, <sup>d</sup> MPs in drinking water, \* No information provided

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## **S2 MPs sampling, treatment, and analysis techniques**

The assessment of MPs levels in water treatment systems typically relies on water sampling. Studies showed two main sampling approaches: bulk sampling or volume-reduced sampling (Fig. 2a). Bulk samples refer to samples where the whole volume of the sample is taken without reducing it during the sampling process: water collection in a container (a glass bottle, a HDPE bottle, or a stainless-steel bottle) is mostly used as a sampling procedure (16 studies), in which 1 L of water was typically collected [33, 34, 72, 74, 77, 79, 97, 109]. In contrast, volume-reduced samples come from higher volumes ( $20^{[29]}$  –  $1,300^{[100]}$ L) of collected water filtered with a pump (9 studies, [27, 29, 63, 73, 77, 96, 100, 101]). Only a small number of studies used a plankton net to collect particles from water of intake points [75] and from inlet and each treatment stage [78]. Only a few studies determined the presence of MPs in sludge. For sludge samples, only grab sampling with glass Kilner jars [63], stainless steel container [37], glass bottle [65], and metal scoop [64] were used.

Depending on the origin of the sample, it is sometimes necessary to remove organic matter before conducting MPs analyzes (Fig. 2b). To that end, oxidizing agents were mainly utilized namely  $H_2O_2$  [27, 29, 72, 75, 77-79, 99] and Fenton reagents ( $Fe_2SO_4$  and  $H_2O_2$ ) [32, 65, 76, 84, 98, 102]. Several studies also applied acid digestion by HCl [47, 60, 100] or alkaline digestion using KOH [101, 109]. A few studies applied an enzymatic digestion, e.g., cellulase [63], and protease [27]. Apart from organic matrix, some minerals aggregates could be found naturally or formed by coagulation-flocculation processes of water treatment. In order to disaggregate them, samples were acidified using diluted  $H_2SO_4$  [74], HCl [97, 98]. Furthermore, several studies performed density separation to remove samples impurities mainly minerals particles either as a single step cleaning, or as a subsequent step after organic matter removal. For this, salt solutions like NaCl [32, 109] and  $ZnCl_2$  [65, 73, 75, 78, 98, 102] were used.

Prior to MPs examination, samples were commonly filtered to collect overlaid particles. This filtration leads to cut-off size of analyzed MPs. In the reviewed studies, the filter with a pore size of  $0.2\ \mu m$  was mostly used. To examine MPs in water treatment systems, distinct kinds of devices have been used depending on the goals of investigation. The analytical methods dedicated to MPs are summarized in Fig. 2c (full list in Table S1 – Table S3). The analysis techniques stated as ‘Others’ in Fig. 2c included camera, metalloscopy, Dynamic Light Scattering, Nanoparticle tracking analysis, particle size analyzer, TOC analyzer, ICP-MS, Turbidity meter, XPS, laser diffraction, and fluorescence spectrophotometer.

Basically, MPs have been detected and reported in color, shape, size, and number concentration by using visual observation method (17 studies, visible light microscope class in Fig. 2c) namely compound microscope (e.g. [42, 55, 58]) and stereo microscope (e.g. [34, 65, 73, 110]) or more advance technique like

fluorescence microscope (5 studies, e.g. [109] and [83]) and epifluorescence microscope (3 studies, [33, 51, 104]), which are more precise due to only plastics could be visibly detected facilitated by fluorescent staining. These traditional techniques have been widely used since they are useful pre-screening tools for identifying and enumerating MPs particles and are a comparatively quick and low-cost method; however, these devices are not able to identify the type of MPs. To achieve the chemical characterization objective, Raman spectroscopy (e.g. [37, 72, 77]) and FTIR spectroscopy (e.g. ATR-FTIR [78, 81], and ( $\mu$ -)FTIR [73, 79, 107]) have been used in 19 and 20 studies, respectively, to confirm microplastic particle counts by providing chemical confirmation of material type.

Aside from the microscopes coupled or not with spectroscopic techniques, detection of microplastics have been analyzed by camera [47, 61], and nano-ranged particles were investigated by TOC analyzer [52], Turbidity meter [44], fluorescence spectrophotometer [106], and ICP-MS [70] (for metal-doped nanoplastics). In addition, the particles size distribution has been determined by Dynamic Light Scattering (DLS) [46, 81], particle size analyzer [54], laser diffraction [46, 53], and Nanoparticle Tracking Analysis (NTA) [71, 81].

Considering the analytical techniques, scanning electron microscope (SEM) is the most widely used device (25 studies) to determine size, shape, and surface morphology of MPs, making possible the enumeration of MPs that can be distinguished from surrounded particles or membrane surface [102]. Moreover, SEM imaging can enhance better understanding of MPs removal mechanism, i.e., formation of floc with MPs in CFS processes [54, 71, 77], and formation of MPs fouling on membrane surface [81, 103, 109]. To observe changes of MPs elemental composition between treatment processes such as coagulation [108], disinfection [46], and ultrafiltration [84], SEM coupled with energy-dispersive X-ray spectroscopy (SEM-EDS or SEM-EDX) were used. The compositional data obtained via EDS was also used to distinguish plastics (carbon based) from other non-organic particles [33]. Apart from SEM-EDS, the element composition on the MPs surface has been analyzed by X-ray photoelectron spectrometer (XPS) [107], and metalloscope [77].

While MPs detection is mainly examined by number concentration, some studies reported the MPs abundance in mass concentration by gravimetric analysis [47, 54, 55, 57] or together with chemical characterization by using pyrolysis-GC/MS [27]. From a regulatory perspective, mass concentration serves as a crucial parameter for assessing the presence of MPs. It is appropriate for the routine and recurrent assessment of MPs as part of monitoring efforts and evaluating the effectiveness of measures taken to combat plastic pollution. Furthermore, it has to be taken into account that a few large particles are more significant in terms of mass than many small particles.

### **S3 Coagulation mechanisms**

The MPs removal can be explained by the classical coagulation mechanisms such as charge neutralization, adsorption bridging, and sweep flocculation [47, 111]. A straightforward scheme of those mechanisms available in Figure 2 of the work of Tang et al. (2022) [48] and can be concisely summarized as follow.

#### **Charge neutralization**

The majority of the MPs carry a negative surface charge, the electrostatic repulsion gives the system a relative dynamic stability. By adding cations, the ionic strength of the suspension is increased and according to the DLVO theory the electric double layer becomes denser/compressed leading to a decrease of the zeta potential. Particles can then come more easily into contact to each other and gather to form aggregates. This mechanism is well documented [47, 111] and is known as the charge neutralization. On a practical point of view, the addition of coagulant such as metal salts that hydrolyze to cations in water leads to a neutralization of the surface charge of the MPs, reducing the repulsion between them. For example, Skaf et al. (2020) [58] observed that the zeta potential of polymer microspheres comes close to zero by the addition of alum and the removal efficiency reaches then a maximum. As for the other types of natural colloidal particles, if the coagulant dose is too excessive, a re-stabilization of the MPs occurs as mentioned by Yang et al. (2016) [112].

#### **Adsorption bridging**

Bridging generally occurs when using high molecular weight (generally greater than  $10^6$ ) polymer adsorb simultaneously on several particles. This bridging effect is all the more intense as the molecular weight is high. The bridges formed among the MPs and also other contaminants allow their connection and agglomeration into large flocs. However, it must be kept in mind that when the dosage of the polymer is too high, a kind of protective effect arises [48] preventing the agglomeration of MPs. Indeed, when the surface covering is elevated, the mutual repulsion of charged polymers can limit the physical proximity of MPs, hindering their aggregation.

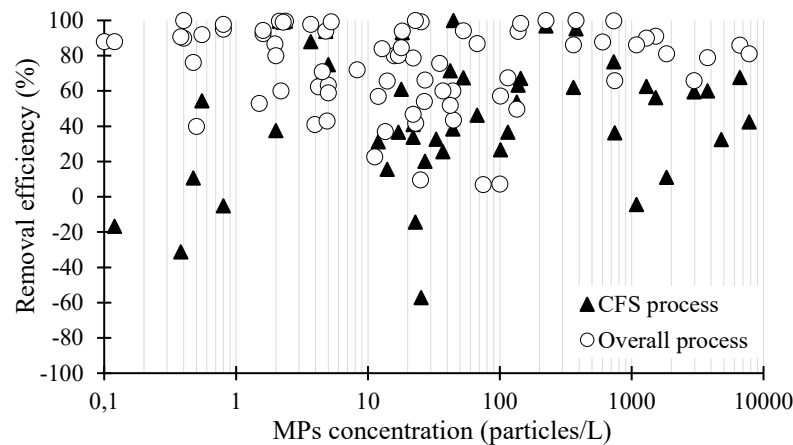
#### **Sweep flocculation**

Under suitable pH and concentration conditions, the addition of multivalent metal salts such as aluminum or ferric chloride leads to the cationic hydrolysis and/or to the precipitation of coagulant hydroxide. The later have a rather high mesh surface structure capable to incorporate MPs (and particles, bacteria or others...) by an enmeshment mechanism also called sweep flocculation.

**S4 Relationship between MPs' characteristics and the removal efficiency of the CFS process**

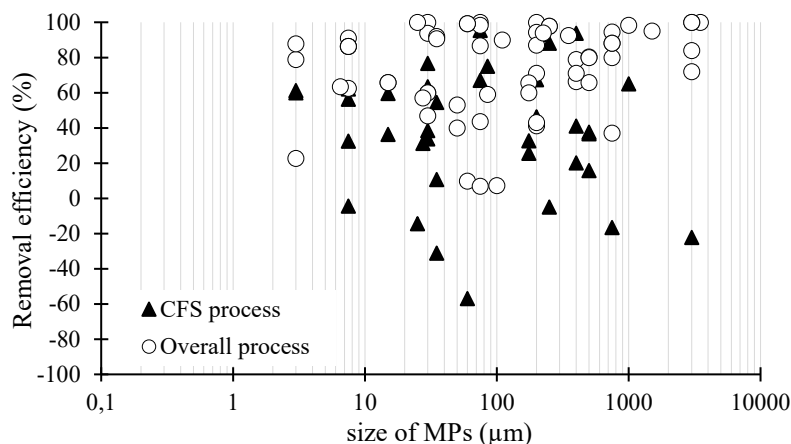
The relationship between the characteristics of MPs and the removal efficiency of the CFS process is a critical aspect of microplastics research. This relationship helps us understand how the properties of microplastics, such as their size, shape, and composition, affect their removal during the coagulation, flocculation, and sedimentation processes in water treatment. By examining this relationship, researchers can develop more effective strategies for removing microplastics from water sources.

To begin, the relationship between the concentration of MPs and the removal efficiency of the CFS process, as well as the overall water treatment plant process, can be illustrated in Fig. S1. The dots correspond to the different studies of Table S5, when a concentration range is mentioned, the average concentration is used in the figure. Also, when a range of removal efficiency is mentioned in the table, the average value is plotted.



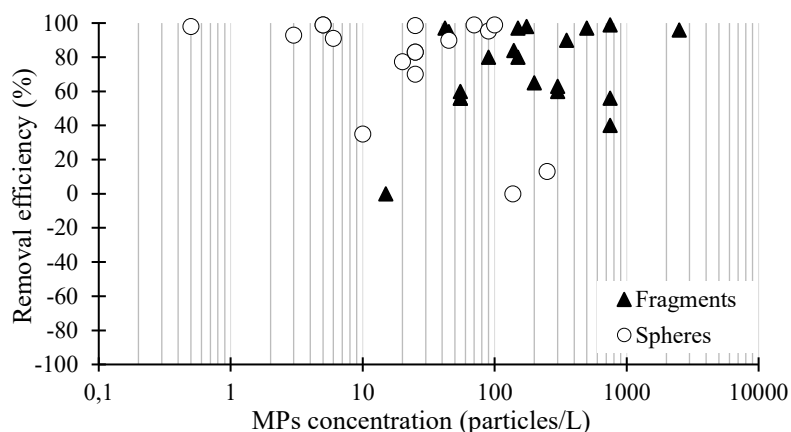
**Fig. S1** Relationship between MPs concentration and removal efficiency in water treatment plant

Fig. S2 presents data from Table S5 as well to demonstrate how the size of microplastics affects removal efficiency. In the case of treatment by CFS, it is evident, as shown in the concentration data (Fig. S1), that the removal rate can sometimes be negative. Importantly, it indicates that regardless of the size of the microplastics, the removal rate generally does not exceed 80%.



**Fig. S2** Relationship between size of MPs and removal efficiency in water treatment plant

The results of Table S4 regarding the relationship between microplastic size, morphology and removal rate are presented in the Fig. S3 for the CFS experiments. Before analyzing this graph, it is necessary to recall that the size of a fragment is often difficult to evaluate and that the authors do not generally mention all the characteristics of the microplastic used, as did Lapointe et al. (2020) [50], who indicate that their PEST fibers measure 14  $\mu\text{m}$  wide, 580  $\mu\text{m}$  long corresponding to an equivalent diameter of 90  $\mu\text{m}$ .



**Fig. S3** Relationship between shape of MPs and removal efficiency in CFS experiments

1281 **S5 The MPs removal efficiency of coagulation-flocculation-sedimentation process**

1282 **Table S4** The MPs removal efficiency of coagulation-flocculation-sedimentation process (Experimental scale)

Study Scales	Microplastic Conditions						Coagulant type	Water Conditions (selected appropriate)			MPs removal efficiency (%)	Reference
	MPs' characteristic	MPs type	Shape	MPs size (µm)	MP concentration (particles/L)	Zp (mV)		Coagulant dose (mg/L)	pH	Alkalinity		
Jar Test + Deionized water	Pristine	PE particles issuing from mechanical disintegration followed by sieving		2000 – 5000 1000 – 2000 500 – 1000 <500	0.1 g/L		AlCl <sub>3</sub> or FeCl <sub>3</sub>	130 mgAl/L (calculated from the data given in the article Al : coagulation performance compared to FeCl <sub>3</sub>	[6;8]		Max 40% <sup>(a)</sup> for the smallest PE particles (<500 µm)	[47]
Jar Test Tap Water + Kaolin + MPs	Pristine	Polymer PE Rayon Polyester	Microspheres	Microspheres : 1-5 µm diameter Microfibers : PE : 5µm diameter - 100µm long and 15 µm diameter – 900 or 1300 long Rayon : 8.7 or 20.6 µm diameter, cut to 0.5 of 1.0 mm lengths Polyester : 17.5 to 50.6 µm, cut to 3.175 mm lengths	Microspheres : 90,000	- 42.0 ± 4.2 (!microsphere alone : from -14mV at pH=5 to -35mV at pH=10	Al <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub>	0-10	5.0 - 10.0		Up to 99%	[58]
Jar test Surface water + MPs	Pristine	PS	Sphere	0.18 1.2	9,500 9,500	*	Al <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub>	20	7.5 ± 0.1	*	N/A	[51]
	Pristine	PE	Sphere	10 – 20 45 – 53 106 – 125	7,000 2,000 1,800	*	Al <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub>	20	7.5 ± 0.1	*	1.0 ± 0.2 13.6 ± 6.8 2.1 ± 0.6	

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Study Scales	Microplastic Conditions						Coagulant type	Water Conditions (selected appropriate)			MPs removal efficiency (%)	Reference
	MPs' characteristic	MPs type	Shape	MPs size (μm)	MP concentration (particles/L)	Zp (mV)		Coagulant dose (mg/L)	pH	Alkalinity		
Jer Test Surface Water + MPs	Pristine	Polyethylene (PE)	Sphere	10 – 20 125 – 150	500	*	Alum, Aluminum chlorohydrate (ACH)  Flocculent (cationic polyacrylamide: PAM)	0.45 – 3.64 mg Al/L	7.0 ± 0.1	*	89	[56]
								0.05 – 0.30 mg/L			82	
		Polystyrene (PS)	Fiber	130 - 150		*				*	84	
		Polyester (PEST)		width: 12-16 μm and length: 105 - 1325 μm		*				*	97	
	Weathered (UV)	Polyethylene (PE)	Sphere	35 – 102  MPs degradation and fragmentation during weathered process		*		0.45 – 2.73 mg Al/L	7.0	*	99	
Beaker DI + Humic Acid + kaolin	Pristine (pre-oxidation)	Polyethylene (PE)	Sphere	250 – 300	5 g/L	*	Polyaluminum choride (PACl)	3 mg	7.0 ± 0.2	*	12.5 – 18.2 <sup>(a)</sup>	[60]
Jar Test + Deionized water or raw water	Weathered (UV)	PS Microbeads Amindine PS microbeads	Beads	10 ; 20 ; 45 ; 90 μm	10 <sup>4</sup> 5,800 1,000 120		AlCl <sub>3</sub> or FeCl <sub>3</sub>	10 – 50	[4.3 ; 8.5]		77.4-95.3% for 20, 45 and 90 μm 33-41% for the 10 μm	[57]
Jar Test Surface Water + MPs	Pristine	Carboxylated PS	Sphere	1 μm 3, 6, 25 (microscopy)	223.4 ± 22.7, 215.1 ± 13.4, 239.2 ± 20.1	-10, -25, -35	Al <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub>	30 – 50 mg Alum/L	8.0	*	92-95, 88-93, 98-99	[104]
				45, 90	88.8 ± 13.2, 60.6 ± 25.4	-42, -63	Alum	50	8.0		90, 80	
Jar test	Pristine	PS, PE	Fragment	< 500 500 – 1000 1000 – 5000	500 mg/L	-15.77, -14.55	PAC	60, 90	7.0	*	76, 50 98.5, 6 96.5, 2.5	[54]
	Pristine	PS, PE	Fragment	< 500 500 – 1000 1000 – 5000	500 mg/L	-15.77, -14.55	FeCl <sub>3</sub>	60, 90	7.0	*	63, 16 98.5, 7 96, 5.5	
Jar test	Pristine	PS	Beads	0.5 μm		*	lysozyme amyloid fibril	0 – 40 mg/L	[2;12]		Up to 97.9%	[42]

1287 **Table S4** The MPs removal efficiency of coagulation-flocculation-sedimentation process (Experimental scale) (cont.)

Study Scales	Microplastic Conditions						Coagulant type	Water Conditions (selected appropriate)			MPs removal efficiency (%)	Reference
	MPs' characteristic	MPs type	Shape	MPs size (µm)	MP concentration (particles/L)	Zp (mV)		Coagulant dose (mg/L)	pH	Alkalinity		
Jar Test with tap water	Pristine	PS dyed with red acrylic paint	Beads	100	100 <sup>(b)</sup>	*	Alum	3.4 mg Al/L	[1;13]	NTU=0.8	Up to 98.9%	[59]
Jar Test HPLC grade water	Pristine	PVC	Sphere	<50	18x10 <sup>6</sup> (10 mg/L)	*	Without	-	5 – 8	1 mmol/L of NaHCO <sub>3</sub>	68.5 – 70.5	[52]
	Pristine	PVC	Sphere	<50	18x10 <sup>6</sup> (10 mg/L)	*	Fe <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub> .9H <sub>2</sub> O	20	7	1 mmol/L of NaHCO <sub>3</sub>	82.5 – 83.5	
	Pristine	PVC	Sphere	<50	18x10 <sup>6</sup> (10 mg/L)	*	Alum	40	7	1 mmol/L of NaHCO <sub>3</sub>	84.5 – 84.8	
Jar test DI mixed with Kaolin	Pristine	PE	Fragment	150 – 200	500 mg/L	-55.4	PAC mixed with APAM	25 mg/L + 4 mg/L	7.5 ± 0.1	*	80.0	[55]
Jar test	Pristine	PS, PMMa, PVC, PE	Mainly beads	0,1 – 10µm	8 mg/L	-30	Etherified starch based coagulant assisted by polysilicic acid (PAC)	Starch : [0.25 – 2.5 mg/L] - optimal dose : 1mg/L PAC : [0-15 mg/L] - optimal dose : 6mg/L	[5;9]		Up to 98.95%	[44]
Jar Test DI water	Pristine	PE	Irregularly polygonal	40 – 48	10 mg/L	-3.36 ± 0.85	Ferrate (FeCl <sub>3</sub> +NaOCl)	1.8 – 3.6	6.8 – 7.2	*	92.3 – 98.4	[53]
		PET	Irregularly polygonal	300	10 mg/L	-0.84± 1.83	Ferrate (FeCl <sub>3</sub> +NaOCl)	1.8 – 3.6	6.8 – 7.2	*	51.2 – 75.0	
Jar Test DI + HA	Pristine	PE	Irregularly polygonal	40 – 48	10 mg/L	-35.8 – -37.2 ± 0.68	Ferrate (FeCl <sub>3</sub> +NaOCl)	2.7 – 3.6	6.8 – 7.2	*	96.7 – 98.2	
		PET	Irregularly polygonal	300	10 mg/L	-35.8 – -37.2 ± 0.68	Ferrate (FeCl <sub>3</sub> +NaOCl)	1.8 – 2.7	6.8 – 7.2	*	51.2 – 74.2	
Jar test 18.25 MΩOcm	Pristine	PS	Fragment	120 – 150 150 – 250 250 – 425 425 – 500 500 – 1000	0.25 - 5 g/L	*	Poly aluminium sulfate (PAS)	0 – 25 g/L	7.0 ± 0.2	*	97.0 98.0 90.0 70 56	[108]
Jar test DI water + kaolin + 1 mg/L HA	Pristine	PE	Fragment	10 – 100	0.100 g/L	11.2 – 4.6 ± 1.3	PAC with anionic PAM	50 – 350 mg/L	5 – 9	*	Up to 58.19%	[46]
	Pristine	PE	Fragment	10 – 100	0.100 g/L	11.2 – 4.6 ± 1.3	Ferric chloride with anionic PAM	50 – 350 mg/L	5 – 9	*	Up to 56.37%	

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1291 **Table S5** The MPs removal efficiency of coagulation-flocculation-sedimentation process (Water treatment plant)

Countries	Name of DWTPs	System	Coagulant type	Microplastic conditions of influent				MPs removal efficiency (%)		Reference
				MPs type	Shape	MPs size (µm)	MPs concentration (particles/L)	Coagulation	The overall	
								Flocculation Sedimentation	process	
Yangtze River Delta, China	ADWTP	*	High Al-base salt	PET, PE, PP, PAM	Fiber, Sphere, Fragment	1 – 5	3760 ± 726	44.9 – 75	78.91 <sup>(b)</sup>	[72]
						5 – 10	1520 ± 258	56.25 <sup>(b)</sup>	91.05 <sup>(b)</sup>	
						10 – 50	731 ± 216	76.56 <sup>(b)</sup>	99.86 <sup>(b)</sup>	
						50 – 100	379 ± 117	95.31 <sup>(b)</sup>	100.00 <sup>(b)</sup>	
						>100	224 ± 126	96.88 <sup>(b)</sup>	100.00 <sup>(b)</sup>	
						Total	6614 ± 1132	67.64 <sup>(b)</sup>	82.1 - 88.6	
Czech Republic	Milence	*	*	Cellulose Acetate (CA), PET, PVC, PE, PP	Fiber, Sphere, Fragment	0.2 – 1	N/A	*	N/A	[74]
						1 – 5	11.23 <sup>(b)</sup>	*	22.66 <sup>(b)</sup>	
						5 – 10	4.21 <sup>(b)</sup>	*	62.50 <sup>(b)</sup>	
						10 – 50	2.19 <sup>(b)</sup>	*	60.00 <sup>(b)</sup>	
						50 – 100	1.97 <sup>(b)</sup>	*	86.67 <sup>(b)</sup>	
						>100	3.95 <sup>(b)</sup>	*	41.11 <sup>(b)</sup>	
	Plzeň	*	*	Cellulose Acetate (CA), PET, PVC, PE, PP	Fiber, Sphere, Fragment	Total	23 ± 2	*	41.71 <sup>(b)</sup>	
						0.2 – 1	N/A	N/A	N/A	
						1 – 5	601.75 <sup>(b)</sup>	61.22 <sup>(b)</sup>	87.76 <sup>(b)</sup>	
						5 – 10	362.28 <sup>(b)</sup>	62.03 <sup>(b)</sup>	86.27 <sup>(b)</sup>	
						10 – 50	137.54 <sup>(b)</sup>	63.39 <sup>(b)</sup>	93.75 <sup>(b)</sup>	
						50 – 100	143.68 <sup>(b)</sup>	67.09 <sup>(b)</sup>	98.29 <sup>(b)</sup>	
Catalonia, Spain	Sant Joan Despí	*	Aliminium salt	PES, PP, Ps ABS, PA	Fiber, Fragment	>100	52.81 <sup>(b)</sup>	67.44 <sup>(b)</sup>	94.19 <sup>(b)</sup>	[73]
						Total	1296 ± 35	62.58 <sup>(b)</sup>	87 – 92	
						20 – 200	0.4	*	90.00 <sup>(b)</sup>	
						200 – 500	1.6	*	92.50 <sup>(b)</sup>	
						500 – 1000	1.6	*	94.38 <sup>(b)</sup>	
						1000 – 2000	0.8	*	95.00 <sup>(b)</sup>	
China	Changsha	*	*	PE, PP, PS, PET	Fiber, Sphere, Fragment	2000 – 5000	0.4	*	100.00 <sup>(b)</sup>	[98]
						Total	4.8	*	93.96	
						1 – 10	*	*	63.37	
						10 – 50	*	*	9.73	
						50 – 100	*	*	6.85	
						>100	*	*	7.27	
						Total	*	*	87.22	

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1294 **Table S5** The MPs removal efficiency of coagulation-flocculation-sedimentation process (Water treatment plant) (cont.)

Countries	Name of DWTPs	System	Coagulant type	Microplastic conditions of influent				MPs removal efficiency (%)		Reference
				MPs type	Shape	MPs size (µm)	MPs concentration (particles/L)	Coagulation	The overall	
								Flocculation Sedimentation	process	
Surabaya, Indonesia	DWTPs I	Hydraulic mixing	Alum	PE, PP, LDPE	Fiber, Fragment, Film, Pellet	1 – 100	0.5	N/A	40	[76]
						101 – 350	4.5	N/A	71	
						351 – 1000	13.5	N/A	37	
						1001 – 5000	8.3	N/A	72	
						Total	26.8	N/A	54.10	
	DWTPs II	Hydraulic mixing	Alum	PE, PP, LDPE	Fiber, Fragment, Film, Pellet	1 – 100	1.5	N/A	53	
						101 – 350	4.9	N/A	43	
						351 – 1000	15.8	N/A	80	
						1001 – 5000	12.8	N/A	84	
						Total	35	N/A	75.71	
South Korea	DWTPs (a)	*	*	PE, PP, PS, PET, PMMA	Irregular, Polygonal, Round	10 – 20	*	*	*	[75]
						20 – 50	0.55	54.45 <sup>(b)</sup>	92.00 <sup>(b)</sup>	
						50 – 100	*	*	*	
						> 100	*	*	*	
						Total	2.37	99.70	99.70	
	DWTPs (b)	*	*	*	*	10 – 20	*	*	*	
						20 – 50	0.3819	-31.21 <sup>(b)</sup>	90.65 <sup>(b)</sup>	
						50 – 100	*	*	*	
						> 100	*	*	*	
						Total	2.13	99.58	99.58	
	DWTPs (c)	*	*	*	*	10 – 20	*	*	*	
						20 – 50	0.4750	10.69 <sup>(b)</sup>	76.21 <sup>(b)</sup>	
						50 – 100	*	*	*	
						> 100	*	*	*	
						Total	2.28	99.16	99.16	
	DWTPs (d)	*	*	*	*	*	*	*	*	

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1299 **Table S5** The MPs removal efficiency of coagulation-flocculation-sedimentation process (Water treatment plant) (cont.)

Countries	Name of DWTPs	System	Coagulant type	Microplastic conditions of influent				MPs removal efficiency (%)		Reference
				MPs type	Shape	MPs size (µm)	MPs concentration (particles/L)	Coagulation	The overall	
								Flocculation Sedimentation	process	
Bangkok, Thailand	WTP (Dry season)	*	*	PE, PP, PVC, PET, PS	Fiber, Fragment, Film, Pellet	6.5 – 53	44 ± 5	38.5 <sup>(b)</sup>	60.0	[37]
						53 – 300	33 ± 4	32.7 <sup>(b)</sup>	65.6	
						300 – 500	22 ± 4	41.1 <sup>(b)</sup>	78.8	
						>500	17 ± 2	36.6 <sup>(b)</sup>	80.2	
						Total	115 ± 11	36.6 <sup>(b)</sup>	67.6	
	WTP (Rainy season)	*	*	PE, PP, PVC, PET, PS	Fiber, Fragment, Film, Pellet	6.5 – 53	22 ± 3	33.8 <sup>(b)</sup>	46.9	
						53 – 300	37 ± 3	25.6 <sup>(b)</sup>	60.0	
						300 – 500	27 ± 3	20.2 <sup>(b)</sup>	66.2	
						>500	14 ± 2	15.7 <sup>(b)</sup>	65.7	
						Total	101 ± 12	26.7 <sup>(b)</sup>	57.2	
Jiaxing, China	DWTP – Coagulation and sedimentation	*	PAC, PAM (coagulant aid)	PP, PVC, PE, PA	Granular, Fiber	5 – 10	4788.18	32.46 <sup>(b)</sup>	86.24	[77]
						10 – 20	2972.34	59.47 <sup>(b)</sup>	65.82	
						20 – 100	25.31	-57.05 <sup>(b)</sup>	99.21	
						100 – 500	3.68	88.04 <sup>(b)</sup>	97.74	
						500 – 1000	0.10	-160.00 <sup>(b)</sup>	88.10	
						1000 – 5000	0.09	-22.22 <sup>(b)</sup>	100.00	
						Total	7789.69	42.50 <sup>(b)</sup>	81.18	
	DWTP – Micro-flocculation	*	PAC	PP, PVC, PE, PA	Granular, Fiber	5 – 10	1087.48	-4.37 <sup>(b)</sup>	86.24	
						10 – 20	743.98	36.30 <sup>(b)</sup>	65.82	
						20 – 100	5.27	-356.17 <sup>(b)</sup>	99.21	
						100 – 500	0.80	-5.00 <sup>(b)</sup>	97.74	
						500 – 1000	0.12	-16.67 <sup>(b)</sup>	88.10	
						1000 – 5000	0.07	-157.14 <sup>(b)</sup>	100.00	
						Total	1837.72	11.08 <sup>(b)</sup>	81.18	
						<50	22.91 <sup>(b)</sup>	-14.37 <sup>(b)</sup>	100.00 <sup>(b)</sup>	
Tianjin, China	DWTP	*	*	Nylon, PEST, PVDF, PET, PS, PP	Fragment, Sphere, fiber, others	50 – 100	44.48 <sup>(b)</sup>	100.00 <sup>(b)</sup>	43.59 <sup>(b)</sup>	[79]
						100 – 200	N/A <sup>(b)</sup>	N/A <sup>(b)</sup>	N/A <sup>(b)</sup>	
						>200	67.40 <sup>(b)</sup>	46.30 <sup>(b)</sup>	86.92 <sup>(b)</sup>	
						Total	134.79	53.71	49.69	

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1303 **Table S5** The MPs removal efficiency of coagulation-flocculation-sedimentation process (Water treatment plant) (cont.)

Countries	Name of DWTPs	System	Coagulant type	Microplastic conditions of influent				MPs removal efficiency (%)		Reference
				MPs type	Shape	MPs size (µm)	MPs concentration (particles/L)	Coagulation	The overall	
								Flocculation Sedimentation	process	
Kolkata, India	DWTP	Flocculation tank and Pulsator Clarifier	Al <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub>	PE, PP, PET, PS	Fibers, Films/fragments	<25	*	*	*	[78]
						25 – 50	*	*	*	
						50 – 100	*	*	*	
						>100	*	*	*	
						Total	17.88	60.90	84.6	
Ontario, Canada	DWTP	*	Alum	PET-PEST, PP, PU, PE, PBT, PAN, Acrylic, Silicone	Film, Fragment, Fiber	10 – 45	12 <sup>(b)</sup>	31.25 <sup>(b)</sup>	57	[69]
						45 – 125	5 <sup>(b)</sup>	75.00 <sup>(b)</sup>	59	
						125 – 300	18.2 <sup>(b)</sup>	93.13 <sup>(b)</sup>	94	
						300 – 500	4.80	93.75 <sup>(b)</sup>	71 <sup>(b)</sup>	
						> 500	2 <sup>(b)</sup>	37.50 <sup>(b)</sup>	80	
						Total	42	71.43 <sup>(b)</sup>	52	
Catalonia, Spain	DWTP	Pulsators	PACl (25 mg/L) and PolyDADMAC (0.8 mg/L)	PET, Synthetic cellulose, PP, PE, PA	Fiber, Fragment, Film	20 – 2000	4.230 ± 1.263	62.7 – 67.2	98.3	[65]

1304 Remarks: \* The result was not reported.

1305 <sup>(a)</sup> The number was reported by mass concentration.

1306 <sup>(b)</sup> Calculated from the data given in the article

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1313 **Table S6** MPs removal efficiency of DAF process

Study Scales	Microplastic Conditions						Chemical type	Water Conditions (selected appropriate)				MPs removal efficiency (%)	Reference
	MPs' characteristic	MPs type	Shape	MPs size (μm)	MP concentration (particles/L)	Zp (mV)		Surfactant dose (mg/L)	pH	Alkalinity	Pressure (MPa)		
Conventional DAF jar test	Pristine	PE, PET, PA	*	2 – 5 5 – 10 10 – 15 15 – 20 20 – 25 >25 Total	0.1 g/L	*	-	-	*	*	0.4 – 0.5	21 – 46.5 32.75 – 49.5 70.5 – 80 77 – 100 85.5 – 100 100 32.7 – 48.7	[61]
DI													
Positive DAF jar test	Pristine	PE, PET, PA	*	2 – 5 5 – 10 10 – 15 15 – 20 20 – 25 >25 Total	0.1 g/L	- 8	CTAB (coagulant)	0.8 – 1.2	*	*	0.4 – 0.5	29 – 71.5 41.5 – 63 77.5 – 93 84.5 – 100 98 – 100 97 – 100 43.8 – 68.9	[61]
DI													
	Pristine	PE, PET, PA	*	2 – 5 5 – 10 10 – 15 15 – 20 20 – 25 >25 Total	0.1 g/L		PDADMAC (coagulant)	0.6 – 1.0	*	*	0.4 – 0.5	32 – 85.5 46 – 78.5 81.5 – 100 89 – 100 94 – 100 100 46.3 – 80.7	[61]

1314 Remarks: \* The result was not reported.

1315 <sup>(a)</sup> The number was reported by mass concentration.

1316 <sup>(b)</sup> calculated from the data given in the articles

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1326 **S6 The MPs removal efficiency of filtration process**

1327 **Table S7** The MPs removal efficiency of filtration process (Experimental scale)

Study Scale	Microplastic conditions at inlet					Operation conditions				MPs removal efficiency (%)	Reference
	MPs' characteristic	MPs type	Shape	MPs size (μm)	MPs concentration (particles/L)	Setup/ Membrane unit cell	Pressure	Media/Material type	Filtration rate (m/hr.)		
Conventional filtration											
Bench	Pristine	PS	Sphere	0.18 1.2	9500 9500	Filter column	Gravity force	Filter media (648 um pore size) and anthracite	*	98.6 94.7	[51]
		PE	Sphere	10 – 20 45 – 53 106 – 125	6300 1700 1400					86.6 98.6 99.8	
Bench	Pristine (pre-oxidation)	Polyethylene (PE)	Fragment	250 – 300	4.21 g/L <sup>(b)</sup>	Sand filter column	*	Sand	9	100 <sup>(a)</sup>	[60]
Bench	Weathered (UV)	PS	Beads	10 20 45 90	*	Sand column (Cylindrical acrylic)	*	Quartz sand (range of grain size is 0.3 – 0.45 mm)	0.3	1.2 16.6 100 100	[57]
Pilot	Aged	PET-PEST, PP, PU, PE, PBT, PAN, Acrylic, Silicone	Film, Fragment, Fiber	Total	12.72 ± 0.39	GAC pilot filter (Calgon Carbon Filtrasorb® 300)	*	Anthracite, Sand, and GAC	*	-102.12 – 17.53	[69]
Bench	Pristine	a polyacrylonitrile core labeled with palladium (Pd) and a polystyrene shell	Sphere	214.7 nm ± 1.4 nm	1.7 mg/L	5 cm. inner diameter and height adjustable end-pieces (Diba industries inc, USA)	*	Pristine and aged sand (d <sub>50</sub> = 450 um) and AC (d <sub>50</sub> = 1050and 950 um)	0.28 or 1.02	22 – 60	[70]

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1330 **Table S7** The MPs removal efficiency of filtration process (Experimental scale) (cont.)

Study Scale	Microplastic conditions at inlet					Operation conditions				MPs removal efficiency (%)	Reference
	MPs' characteristic	MPs type	Shape	MPs size (µm)	MPs concentration (particles/L)	Setup/ Membrane unit cell	Pressure	Media/Material type	Filtration rate (m/hr.)		
Pilot	Pristine	a polyacrylonitrile core labeled with palladium (Pd) and a polystyrene shell	Sphere	163.5 nm ± 0.3 nm	141.6 mg/L	1.1 m diameter and total height of 2.65 m. (1.85 m height of the filter bed)	*	Sand and AC	1.05	Up to 99.5	
Pilot	Pristine	Surfactant-free polystyrene latex nanoplastics	Sphere	124 ± 38 nm and 110 ± 25 nm (NPs)	4.6 g/L	Dual media constituted of pumice (h = 50 cm.), quartz sand (h = 80 cm.) and support layer of gravel (h = 45 cm.) with a downward filtration.	*	Dual media consist of pumice and quartz sand, GAC filter consists of a mix of 75% bituminous coal and 25% coconut shell-based activated carbon	5.5 m/h	99.4	[71]

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1335 **Table S7** The MPs removal efficiency of filtration process (Experimental scale) (cont.)

Study Scale	Microplastic conditions at inlet					Operation conditions				MPs removal efficiency (%)	Reference
	MPs' characteristic	MPs type	Shape	MPs size (µm)	MPs concentration (particles/L)	Setup/ Membrane unit cell	Pressure	Media/Material type	Filtration rate (m/hr.)		
Membrane											
Bench	Pristine	PE	Sphere	2000-5000 1000-2000 500-1000 <500	*	UF stirred cell (Millipore 8400, USA)	maintained at 0.1 MPa with N <sub>2</sub> .	Flat sheet polyvinylidene fluoride (PVDF) Membrane	*	PE particles were completely rejected by the UF membranes and a cake layer was the main fouling mechanism.	[47]
Bench	Micro-nano plastic obtained from Facial scrub	PE	Fragment	13 – 690 nm	2 × 10 <sup>11</sup>	Cross-flow filtration (Sterlitech CF042AC)	2.25 Bar	Commerical PSF UF membrane			[81]
Bench	Pristine	PVC	*	< 5 µm	10	0.1 µm membrane pore size with 0.1 m² surface area (a Chiness membrane company)		Membrane module	2 L/hr	The membrane bioreactor system rejected almost all the microplas tics.	[103]

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1339 **Table S7** The MPs removal efficiency of filtration process (Experimental scale) (cont.)

Study Scale	Microplastic conditions at inlet					Operation conditions				MPs removal efficiency (%)	Reference
	MPs' characteristic	MPs type	Shape	MPs size (µm)	MPs concentration (particles/L)	Setup/ Membrane unit cell	Pressure	Media/Material type	Filtration rate (m/hr.)		
Bench	Pristine (pre-oxidation)	PE	Fragment	250 – 300	4.21 g/L <sup>(b)</sup>	*	*	0.45 µm filter membrane		The MPs were completely removed by the sand filtration before entering the filter membrane.	[60]
Bench	Pristine	PE	Rod	40 – 48 µm	18,000 particles/L or 1 mg/L	Two parallel of dead-end filtration cell of continuous-flow ultrafiltration units	0.2 kPa	A Flat sheet PVDF membrane (Ande, China) with molecular weight cutoff of 100 kDa, and effective area of 28.3 cm <sup>2</sup>	Membrane flux = 10 L/m <sup>2</sup> h	*** result is reported as membrane flux and fouling ***	[84]
Bench (Tap water)	Pristine	PS	Sphere	0.1, 1, 10, 18	0.1-1 mg/L	PVDF hollow fiber membrane module consisting of 22 membrane tubes	Max. 13.5 kPa	PVDF hollow fiber membrane (mean pore size of 0.03 µm)	1 L/h (20 L m <sup>-2</sup> · h <sup>-1</sup> )	All PS particles were rejected during the filtration.	[83]

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1343 **Table S7** The MPs removal efficiency of filtration process (Experimental scale) (cont.)

Study Scale	Microplastic conditions at inlet					Operation conditions				MPs removal efficiency (%)	Reference
	MPs' characteristic	MPs type	Shape	MPs size (µm)	MPs concentration (particles/L)	Setup/ Membrane unit cell	Pressure	Media/Material type	Filtration rate (m/hr.)		
Bench (Tap water + NOM)	Pristine	PS	Sphere	0.1, 1, 10, 18	0.1-1 mg/L	PVDF hollow fiber membrane module consisting of 22 membrane tubes	Max.35.5 kPa	PVDF hollow fiber membrane (mean pore size of 0.03 µm)	1 L/h (20 L m <sup>-2</sup> · h <sup>-1</sup> )	All PS particles were rejected during the filtration.	[106]
Bench (Raw water)	Pristine	PS	Sphere	0.1, 1, 10, 18	0.1-1 mg/L	PVDF hollow fiber membrane module consisting of 22 membrane tubes	Max. 74.0 kPa	PVDF hollow fiber membrane (mean pore size of 0.03 µm)	1 L/h (20 L m <sup>-2</sup> · h <sup>-1</sup> )	All PS particles were rejected during the filtration.	
Bench	Pristine	PS (fluorescent)	Sphere	0.08 - 1µm	*	Ti <sub>3</sub> C <sub>2</sub> Tx membrane followed by simple vacuum filtration using polymeric membranes as support matrix	1 k Pa	Holey Ti <sub>3</sub> C <sub>2</sub> Tx nanosheets (an average hole-size of 25 nm) with concentration of h-Ti <sub>3</sub> C <sub>2</sub> Tx 0.4 mg/ml (M1) to 1.2 mg/ml (M5)	196.7 L/ m <sup>2</sup> h to 68.9 L/ m <sup>2</sup> h	up to 99.3% under the present experimental conditions	
Jar Test DI water	Pristine	PE	Irregularly polygonal	40 – 48	10 mg/L	Millipore Amicon stirred cell (UFSC05001 5122, 50 mL for 44.5 mm filters)	0.5 bar	Polyethylene ether sulfone (PES) (pore size 0.22 µm), MF membrane	*	100	[53]
		PET	Irregularly polygonal	300	10 mg/L					100	
Jar Test DI + HA	Pristine	PE	Sphere	40 – 48	10 mg/L				*	100	
		PET	Irregularly polygonal	300	10 mg/L				*	100	

1344 Remarks: \* The result was not reported.

1345 <sup>(a)</sup> The number was reported by mass concentration.

1346 <sup>(b)</sup> Calculated from the data given in the article

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1349 **Table S8** The MPs removal efficiency of filtration process (Water treatment plant)

Countries	Name of DWTPs	System	Media type/ Material	Microplastic conditions at inlet				MPs removal efficiency (%)		Reference
				MPs type	Shape	MPs size (µm)	MPs concentration (particles/L)	Filtration	The overall process.	
Conventional filtration										
Yangtze River Delta, China	ADWTP	Sand filtration	Sand	PET, PE, PP, PAM	Fiber, Sphere, Fragment	1 – 5	*	26.56 <sup>(b)</sup>	78.91 <sup>(b)</sup>	[72]
						5 – 10	*	48.44 <sup>(b)</sup>	91.05 <sup>(b)</sup>	
						10 – 50	*	78.13 <sup>(b)</sup>	99.86 <sup>(b)</sup>	
						50 – 100	*	100.00 <sup>(b)</sup>	100.00 <sup>(b)</sup>	
						> 100	*	100.00 <sup>(b)</sup>	100.00 <sup>(b)</sup>	
						Total	*	29.0 - 44.4	82.1 - 88.6	
Czech Republic	Milence	Sand filtration	Sand (1-1.6 mm)	Cellulose Acetate (CA), PET, PVC, PE, PP	Fiber, Sphere, Fragment	0.2 – 1	*	*	N/A	[74]
						1 – 5	*	*	22.66 <sup>(b)</sup>	
						5 – 10	*	*	62.50 <sup>(b)</sup>	
						10 – 50	*	*	60.00 <sup>(b)</sup>	
						50 – 100	*	*	86.67 <sup>(b)</sup>	
						>100	*	*	41.11 <sup>(b)</sup>	
						Total	*	*	41.71 <sup>(b)</sup>	
	Plzeň	Deep-bed filtration	Filtralite HC 0.8–1.6 mm and Filtralite NC 1.5–2.5 mm (1:1)	Cellulose Acetate (CA), PET, PVC, PE, PP	Fiber, Sphere, Fragment	0.2 – 1	N/A	N/A	N/A	
						1 – 5	233.33 <sup>(b)</sup>	47.37 <sup>(b)</sup>	87.76 <sup>(b)</sup>	
						5 – 10	137.54 <sup>(b)</sup>	40.63 <sup>(b)</sup>	86.27 <sup>(b)</sup>	
						10 – 50	50.35 <sup>(b)</sup>	40.24 <sup>(b)</sup>	93.75 <sup>(b)</sup>	
						50 – 100	47.28 <sup>(b)</sup>	85.71 <sup>(b)</sup>	98.29 <sup>(b)</sup>	
						>100	17.19 <sup>(b)</sup>	71.43 <sup>(b)</sup>	94.19 <sup>(b)</sup>	
						Total	485.70 <sup>(b)</sup>	49.30 <sup>(b)</sup>	87 – 92	
Catalonia, Spain	Sant Joan Despí	Sand filtration	Sand	PES, PP, Ps ABS, PA	Fiber, Fragment	20 – 200	*	*	90.00 <sup>(b)</sup>	[73]
						200 – 500	*	*	92.50 <sup>(b)</sup>	
						500 – 1000	*	*	94.38 <sup>(b)</sup>	
						1000 – 2000	*	*	95.00 <sup>(b)</sup>	
						2000 – 5000	*	*	100.00 <sup>(b)</sup>	
						Total	*	*	93.96	
China	Changsha	Sand filtration	Sand	PE, PP, PS, PET	Fiber, Sphere, Fragment	1 – 10	*	*	63.37	[98]
						10 – 50	*	*	9.73	
						50 – 100	*	*	6.85	
						>100	*	*	7.27	
						Total	*	*	87.22	

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Countries	Name of DWTPs	System	Media type/ Material	Microplastic conditions at inlet				MPs removal efficiency (%)		Reference
				MPs type	Shape	MPs size (µm)	MPs concentration (particles/L)	Filtration	The overall process.	
Surabaya, Indonesia	SDWTPs I	Dual media rapid sand filtration	Silica, anthracite sand media	PE, PP, LDPE	Fiber, Fragment, Film, Pellet	1 – 100	1.0	N/A	88	[76]
						101 – 350	5.0	30	64	
						351 – 1000	8.5	33	N/A	
						1001 – 5000	4.0	31	18	
						Total	18.5	29.7	11	
	SDWTPs II	Dual media rapid sand filtration	Silica, anthracite sand media	PE, PP, LDPE	Fiber, Fragment, Film, Pellet	1 – 100	1.1	N/A	77	
						101 – 350	4.7	N/A	54	
						351 – 1000	8.9	N/A	73	
						1001 – 5000	9.6	5	78	
						Total	24.3	N/A	71	
South Korea	DWTPs (a)	Sand filtration	*	PE, PP, PS, PET, PMMA	Irregular, Polygonal, Round	10 – 20	*	*	*	[75]
						20 – 50	0.2505	82.44	*	
						50 – 100	*	*	*	
						> 100	*	*	*	
						Total	0.75	*	99.70	
	DWTPs (b)	Sand filtration	*	*	*	10 – 20	*	*	*	
						20 – 50	0.5011	92.88	*	
						50 – 100	*	*	*	
						> 100	*	*	*	
						Total	0.91	*	99.58	
	DWTPs (c)	Sand filtration	*	*	*	10 – 20	*	*	*	
						20 – 50	0.4242	73.36	*	
						50 – 100	*	*	*	
						> 100	*	*	*	
						Total	1.03	*	99.16	
	DWTPs (d)	Sand filtration	*	*	*	*	*	N/A	N/A	
Bangkok, Thailand	WTP (Dry season)	Dual media filter	*	PE, PP, PVC, PET, PS,	Fiber, Fragment, Film, Pellet	6.5 – 53	25 ± 2	26.8	60.0	
						53 – 300	22 ± 4	28.5	65.6	
						300 – 500	19 ± 2	52.8	78.8	
						>500	19 ± 2	60.7	80.2	
						Total	85 ± 8	34.8	67.6	
	WTP (Rainy season)	Dual media filter	*	PE, PP, PVC, PET, PS,	Fiber, Fragment, Film, Pellet	6.5 – 53	23 ± 2	12.7	46.9	
						53 – 300	23 ± 1	32.5	60.0	
						300 – 500	25 ± 2	47.9	66.2	
						>500	26 ± 2	52.1	65.7	
						Total	97 ± 9	30.9	57.2	

1355 **Table S8** The MPs removal efficiency of filtration process (Water treatment plant)

Countries	Name of DWTPs	System	Media type/ Material	Microplastic conditions at inlet				MPs removal efficiency (%)		Reference
				MPs type	Shape	MPs size (μm)	MPs concentration (particles/L)	Filtration	The overall process.	
Jiaxing, China	DWTP	Sand filtration	*	PP, PVC, PE, PA	Granular, Fiber	5 – 10 10 – 20 20 – 100 100 – 500 500 – 1000 1000 – 5000 Total	1135.04 473.88 24.04 0.84 0.14 0.18 1634.12	-9.20 <sup>(b)</sup> 93.09 <sup>(b)</sup> 90.48 <sup>(b)</sup> 64.29 <sup>(b)</sup> 66.67 <sup>(b)</sup> 16.68 <sup>(b)</sup> 16.68 <sup>(b)</sup>	86.24 65.82 99.21 97.74 88.10 100.00 81.18	[77]
Kolkata, India	DWTP	Sand filtration	Sand bed	PE, PP, PET, PS	Fibers, Films/fragments	<25 25 – 50 50 – 100 >100 Total	* * * * 6.99	* * * * -59.80	* * * * 84.6	[78]
Catalonia, Spain	DWPT	Sand filter	Sand (0.8-1.6 and 1.4-2.8 mm grain size)	PET, Synthetic cellulose, PP, PE, PA	Fiber, Fragment, Film	20 – 2000	1.388 - 1.577 ± 0.696	99.1	98.3	[65]
		Carbon filter	Granular activated carbon	PET, Synthetic cellulose, PP, PE, PA	Fiber, Fragment, Film	20 – 2000	0.038 ± 0.025	99.2	98.3	
Membrane										
Tianjin, China	DWTP	Membrane filtration	PVDF membrane	Nylon, PEST, PVDF, PET, PS, PP	Fragment, Sphere, fiber, others	<50 50 – 100 100 – 200 >200 Total	26.21 <sup>(b)</sup> 0.00 <sup>(b)</sup> 0.00 <sup>(b)</sup> 36.19 <sup>(b)</sup> 62.40 <sup>(b)</sup>	100 <sup>(b)</sup> N/A <sup>(b)</sup> N/A <sup>(b)</sup> 75.64 <sup>(b)</sup> -8.67	100.00 <sup>(b)</sup> 43.59 <sup>(b)</sup> N/A <sup>(b)</sup> 86.92 <sup>(b)</sup> 49.69	[79]
Catalonia, Spain	Sant Joan Despí	Ultra-filtration followed by Reverse osmosis	PVDF membrane (ultrafiltration), PP cartridge (reverse osmosis)	PA, AR, PAN, PS, PES, PP	Fragment, fiber	20 – 200 200 – 500 500 – 1000 1000 – 2000 2000 – 5000 Total	* * * * * *	* * * * * 54	90.00 <sup>(b)</sup> 92.50 <sup>(b)</sup> 94.38 <sup>(b)</sup> 95.00 <sup>(b)</sup> 100.00 <sup>(b)</sup> 93.96	[73]

1356 Remarks: \* The result was not reported.

1357 <sup>(a)</sup> The number was reported by mass concentration.

1358 <sup>(b)</sup> Calculated from the data given in the article