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# Occurrence, removal and potential threats associated with microplastics in drinking water sources

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### ABSTRACT

Microplastics (MPs) have gained increasing attention as an emerging contaminant in drinking water. However, there is no definitive conclusion on the deleterious effects of MPs on human health. Herein, we consider the potential threats of these anthropogenic particles that have been increasingly found in drinking water sources (DWSs) based on reviewing their occurrence and removal from a water treatment perspective. As revealed by 53 publications on MP presence in conventional DWSs, bulk sampling can better reflect the current knowledge on the pollution in DWSs; the median MP concentration in conventional water sources was  $2.2 \times 10^3$  items m<sup>-3</sup> with the size of particles identified usually >50 µm. Next, the removal efficiency of MPs across multiple barriers in drinking water treatment plants was also elaborated. Almost all MPs (>10 µm) were removed after coagulation, sedimentation and filtration processes. For smaller MPs (>1 µm), removal rates of >80 % were typically observed. Two size-dependent threats associated with MPs in DWSs were identified: 1) the increased probability of the accumulation of potential pathogenic bacteria and the transmission of antibiotic resistance genes where larger MPs in DWSs may serve as important carriers; 2) the release of nanoplastics and dissolved organic carbon from the photodegradation process of MPs. Additionally, MPs in alternative DWSs were given special attention due to their potential to accumulate MPs. The review provides new information for practitioners and scientists alike with respect to the potential threats posed by MPs in DWSs.

### 1. Introduction

Microplastics (MPs) have emerged as a contaminant present in many different water sources [1]. In general, plastic particles less than 5 mm are defined as MPs [2]. The smallest fraction of MPs defined as nanoplastics (NPs) are those plastics <100 nm or <1000 nm in size, with the lower size limit under scientific debate [3]. Human exposure to MPs is thought to primarily occur from inhalation and consumption in food and drinks [4]. MPs have been detected at low concentrations in some tap water samples [5–7]. As a result, humans can be exposed to MPs from drinking water through ingestion or accidental inhalation while showering. Humans are known to consume MPs as these particles have been detected in human stools [8]. Consequently, understanding the health impacts of human exposure to MPs in potable water has therefore gained considerable attention from the global media and general public [5].

There is some debate amongst scientists as to the importance of human exposure to MPs in drinking water. Some believe that MP exposure as an important issue due to their potential particle toxicity and the chemical hazards associated with substances found in the plastic polymers or adsorbed onto plastic surfaces [9-11]. Others argue that the expected low exposure concentrations in drinking water make the risks negligible [12], especially when compared to other contaminants present in water sciences (for example micro-organisms). A recently published report from WHO [13] concluded that there is no reliable evidence to suggest that human exposure to MPs from drinking water is of concern and routine monitoring on MPs is unnecessary. However, the report identified a number of areas that require future attention, including the requirement for more reliable data on human health risk assessments from MPs, and the need for more data on the occurrence and fate of MPs in drinking water systems. This should also be viewed in the overall context of the increased frequency of MP detection in the

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Nomenclature		NOM NPs	natural organic matter
			nanoplastics
Acronym	S	PA	polyamide
ARGs	antibiotic resistance genes	PAM	polyacrylamide
DAF	dissolved air flotation	PE	polyethylene
DBP	disinfection by-product	PES	polyester
DLVO	Derjaguin-Landau-Verwey-Overbeek	PP	polypropylene
DOC	dissolved organic carbon	PS	polystyrene
DR	direct re-use	PVC	poly(vinyl chloride)
DWSs	drinking water sources	RGFs	rapid gravity filters
DWTPs	drinking water treatment plants	RO	reverse osmosis
EPS	expanded polystyrene	SUVA	specific ultraviolet absorbance
FTIR	Fourier transform infrared spectroscopy	THM	trihalomethane
HANs	Haloacetonitriles	TOC	total organic carbon
HGT	horizontal gene transfer	UV	ultraviolet
IR	indirect water re-use	WWTPs	wastewater treatment plants
MPs	microplastics		

environment [14]. The total amount of plastic entering into the environment since the 1950s has been estimated to be 4,900 million metric tons as a result of significant increases in the mass manufacture of plastic and improper waste management [15,16]. MPs in water also are released from the degradation of larger plastic debris and thus have become progressively more prevalent in water sources over time [17]. As a result, an inevitable consequence is that MPs have increasingly been found in water sources used for drinking and in the treated water itself.

Nearly one hundred field studies have been published that consider the occurrence of MPs in water sources, typically sampling from a single location using one specific sampling method. This research has revealed, either directly or indirectly, that MP pollution is present in many water sources used for drinking. However, only limited attempts have been made to link MP presence in drinking water sources (DWSs) with the method of plastic detection and quantification [18-20]. As a result, there is still a significant gap in our understanding and awareness of MP pollution in DWSs. Therefore, a focused review on current MP environmental monitoring from a water treatment perspective is needed to provide more useful information on MP occurrence in DWSs. In addition, only once a comprehensive understanding of MP characteristics and prevalence is determined can we begin to effectively understand how drinking water treatment plants (DWTPs) are able to perform with respect to MP removal. DWTPs are designed to remove a whole range of particulate and dissolved material [21]. This includes particles that are of the same size as MPs and NPs [22]. However, there are unknowns associated with how plastic particles behave in natural water sources and how they might react on exposure to chemicals and shear stresses found in water treatment systems. From the available data, MP removal efficiencies across DWTPs can be estimated, providing useful information on the concentrations and sizes of MPs that are challenging to treat.

This review has therefore focused on MP occurrence, removal and threats in DWSs. We have critically assessed the approaches used to monitor MPs in freshwater monitoring to illustrate how current approaches often miss the MPs that are important from a water treatment perspective. Next, the removal efficiency of MPs across multiple barriers in drinking water treatment plants (DWTPs) was considered, alongside a discussion on how plastic particles behave compared to other particulate contaminants. On the basis of the above, two potential threats of MPs in DWSs were proposed and explored: 1) the potential size-dependent risks of MPs in water sources, and 2) emerging threats associated with MPs in alternative water sources (Fig. 1). The study aimed to draw to the attention of practitioners and research scientists alike the potential threats of increasing MPs in DWSs in the Anthropocene.

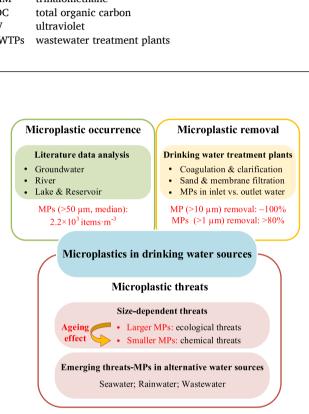


Fig. 1. The flowchart of the article to show the research approaches.

### 2. Literature data analysis based on published studies: the occurrence of MPs in drinking water sources

Surface water and groundwater are the most accessible and widely used DWSs globally. MPs have been found in freshwater sources across the world, including in Africa, Europe, Asia, and America [18]. In this review, data of MPs in freshwater from studies up until June 2020 was collected from the available literature. Only studies that considered these freshwaters as DWSs were considered. The literature before August 2018 was taken from a review carried out by Koelmans et al. [23]. The final list (Table S1) contained 55 records (1 groundwater, 19 lakes and reservoirs, and 35 rivers) from 53 studies, adding more than 64 % new studies since that review. The polymer type, shape, abundance and size distribution of MPs were documented in each record. In addition, information on the sampling and analysis methods were gathered. The records used for statistical analysis are listed in Table 1, which included 1439 data points from 42 studies in total. Where available the raw data was used from the research paper. If this was not available, MP prevalence was interpolated from figures and other available data in the paper.

### Table 1

### The references used for the statistical analysis.

Counting unit	Category	References	The number of references	The frequency of data (items)
	Groundwater bulk sampling	[24]	1	16
	Lake & Reservoir bulk sampling	[25-30]	6	218
items per m <sup>3</sup>	River bulk sampling	[28,31-44]	15	340
	Lake & Reservoir net sampling	[25,45-48]	5	43
	River net sampling	[31,32, 48–56]	11	591
items per m <sup>2</sup>	Lake & Reservoir net sampling	[26,49, 57–63]	9	172
	River net sampling	[64,65]	2	59

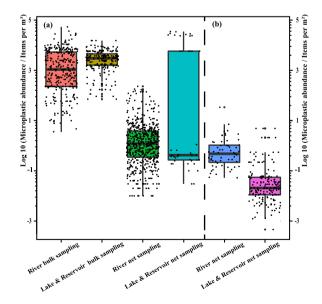
The separation of qualitative and quantitative detection of MPs is common in sampling because of the difficulty in identification of MPs. Nearly 20 % of studies in this review directly identified MPs with the naked eye or under microscope, with some of these verifying putative MP particles based on their response to hot needles (Table S1). Both of the above are known to have a high degree of error associated with them, even for skilful operators [66]. Over 80 % of the research studies identified the MP polymer type, mainly through Fourier Transform Infrared Spectroscopy (FTIR), or Raman spectrometry (Table S1). Polypropylene (PP) and polyethylene (PE) were the most frequently detected polymer types in the majority of studies (Table S1). These polymers are widely used as packaging material, are mass-produced, and are used for products that have short life cycles [15], causing their dominance in the environment. Other common types of MPs were detected in several locations. For example: poly(ethylene terephthalate) (PET) in the Hudson river [67], Min River [35] and Jinze Reservoir [45]; polystyrene (PS) in the Lower Rhine [68] and Wuliangsuhai Lake [36]: expanded polystyrene (EPS) in the Feilaixia Reservoir [46]; poly(vinyl chloride) (PVC) in western Lake Superior [62]; polyamide (PA) in the Pearl River [37]; and cellophane in Taihu Lake [26].

Almost all studies recorded the shape of MPs by visual sorting with a stereomicroscope or dissection microscope (Table S1). MPs were categorised as fibres, fragments, films, and pellets. However, many different terms have been used to describe the shapes of the MPs in these studies (Table S1). For example, pellets have been described as beads, spheres, spherules, and granules. Some researchers regarded these as being the same shape but others made finer distinctions between them. The plastic shape and colour (documented by some studies) can provide information on the sources and residence time of MPs in different environments [28]. For example, MPs may be defined as being from primary or secondary sources. Primary MPs are those materials that have been manufactured specifically to be within this small size range. For example, plastic microbeads used to exfoliate or scrub in cleansers, cosmetics, and toothpaste can be released from wastewater to aquatic environments [69]. Secondary MPs are formed by the ageing and fragmentation of larger plastic particles under the influence of external forces after they enter the environment. Fragments are often regarded as being representative of secondary MPs. Fibres were the most abundant shapes found in the Ottawa River [70], Pearl River [38], Yangtze River Basin [30], Jinze Reservoir [45], Great Lake tributaries [54], and Orange-Vaal River [71], and accounted for over 70 % of MPs in these locations. Some studies focused on only a single shape [67,72] or polymer type [68]. Additionally, shapes may have possible associations with specific polymer types; for example, a high correlation between fibres and PET has been observed [62]. There was no significant difference observed between the most common types and shapes of polymer in DWSs compared to those seen in other aquatic environments [23, 73]. This reflects the high production and persistence in the

environment of several kinds of plastics such as PP, PE and PET.

The abundance of MPs in DWSs is summarised according to source type, sampling method and counting unit. Each study used distinct methods for sampling, pre-treatment, and recording, highlighting the lack of standard methods for the environmental monitoring of MPs. Net and bulk sampling were the two main sampling strategies deployed, each having different units of measurement. In net sampling, the concentration of MPs was reported as either the number of particles per water volume or area sampled. Manta trawling and plankton nets were widely used in net sampling. A manta trawl is typically applied in marine environments and requires a large sampling area. Plankton nets are more practically applied in smaller lakes and rivers and typically have smaller mesh sizes. In contrast, bulk sampling refers to the extraction of water samples directly from the water body into a container or vessel, and is considered only in volumetric terms. Most studies reduce the volume of the bulk sample by filtering the water sample *in-situ* instead of transporting it to a laboratory. The volume-reduction methods [74] make it possible to collect greater volumes of water. For example, volumes of up to 1,500 L are reported in the literature [43]. Net sampling was used in 60 % (32/53) of the studies, while bulk sampling was used for 55 % (29/53) – note that some studies used both net and bulk water sampling methods. This analysis has also shown that there are differences in the representation methods used for quantifying MPs in rivers and in lakes & reservoirs with net sampling. For example, the abundance of MPs is more frequently reported 'per unit volume' for rivers where net sampling was used, whereas most data were reported using 'per water surface area' for lakes & reservoirs (Table 1). The difficulties in comparing different sampling types and methods of reporting data will continue in the future when conducting meta-analyses and comparing studies, particularly given the lack of standard sampling methods and reporting units.

MP concentrations spanned five or six orders of magnitude  $(10^{-4} \text{ to } 10^1 \text{ items m}^2 \text{ or } 10^{-2} \text{ to } 10^4 \text{ items m}^3)$  across all individual samples, mainly from Asia, Europe, and North America (Fig. 2). There was only one case for groundwater with a limited sample volume (2 L) was



**Fig. 2.** Box and whisker plot showing the abundance and range of MPs in groundwater, lake & reservoir water, and river environments determined through either bulk or net sampling (Table S1). The units are expressed on a logarithmic scale for items per  $m^3$  (left axis, a) and items per  $m^2$  (right axis, b). The values for some bulk samples were scaled up (generally items per litre), while most values were scaled down for surface area-based samples (generally items per km<sup>2</sup>). Note that the sum of cases (n = 49) exceeds the total number of studies (n = 42), because several studies used different sampling methods or water types.

assessed (Panno et al., 2019); thus, MP abundance in groundwater was excluded from the following analysis. Independent-sample t-tests were carried out with SPSS software (version 24, SPSS Inc.) to determine whether different continents and water types sampled by the same method had significantly different MPs concentrations. The data showed that Asian rivers, lakes and reservoirs had more MP pollution per unit volume than in sources from Europe and North America (p < 0.05). The median MP concentrations for the Asian water sources were at least an order of magnitude higher than those seen for other parts of the world (Table 2). Additionally, the number of MPs in North American surface waters was significantly lower than those seen in DWSs in Europe and Asia when considered by MPs per area unit (p < 0.05, Table 2). As can be seen from Fig. 2, the abundance of MPs in lakes and reservoirs  $(4.8 \times 10^{-1})$  $^{1}/8.4 \times 10^{-1}$  items m<sup>-2</sup>, median/interquartile range, both here and below) was significantly lower than that seen in rivers ( $2.1 \times 10^{-2}/4.4 \times$ 10<sup>-2</sup> items m<sup>-2</sup>) when net sampling has been used and data was reported by area unit (p = 0.03). However, two Asian lakes and reservoirs were detected very high level of MPs ( $5.9 \times 10^3$ – $3.6 \times 10^4$  items m<sup>-3</sup>) by net sampling [45,47], which significantly increased the MP concentrations for these sources (Fig. 2). In contrast, there was no significant difference in the MP concentration from bulk sampling for rivers and lakes & reservoirs (p = 0.99). While the median MP values where higher in lakes and reservoirs  $(3.0 \times 10^3/3.0 \times 10^3 \text{ items m}^{-3})$  compared to rivers using bulk sampling  $(1.1 \times 10^3/5.2 \times 10^3 \text{ items m}^{-3})$ , there was a much broader range of MP concentrations observed in river sources, with the highest values also being observed in these sources (Fig. 2). There was therefore no firm conclusion as to whether one source water type can be considered more problematic than another. The results infer that river water contains more pollution of larger MPs. In the case of lakes and reservoirs, accumulation of pollution may occur due to the lower mobility of flows out of these systems. As a result, MPs may persist and degrade into smaller particle sizes in these water bodies.

Typically, a recommended drinking water intake depth of 1.5–2.0 m below the surface and 0.5-1.0 m above the bottom of the waterbody instead of surface layer is used. While net sampling usually allows larger sampling volumes, only the water in the surface layer can be sampled. In contrast, bulk water sampling usually uses lower sampling volumes but allows the position in the water column from where the sample is taken to be selected. According to the analysis above, the result of bulk sampling is more representative of the MPs in DWSs than net sampling. The median abundance of MPs in net sampling (1.2 items  $m^{-3}$ ) was 3-4 orders of magnitude lower than bulk sampling  $(2.2 \times 10^3 \text{ items m}^{-3})$ when data was reported by volume (Fig. 2). One explanation for this gap is the difference in the initial interception size employed in the two sampling methods. There was a significant relationship (p < 0.05) between the MP abundance and a decrease in the pore size of the filter or mesh size of net (Fig. 3a). The meshes in net sampling ranged in size from 80 to 335 µm (Fig. 3). Mesh sizes over 300 µm were more common, used in 64 % (18/28) of studies. The remaining 36 % of studies used meshes that were in the range of 75–153  $\mu$ m. For bulk sampling, the largest filter or sieve size was 100 µm apart from one 300-µm filter, with most studies using filters in the 0.45-50 µm range. It should be noted that 50 µm is the minimum size that can be easily identified by eye or picked for further identification [32,43]. However, most studies using

Table 2

The abundance of MPs in	drinking water	sources in diffe	rent continents.
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Abundance /Median	Bulk sampling	Net sampling	
(interquartile range)	Items m <sup>-3</sup>	Items $m^{-3}$	Items $m^{-2}$
Asia	$2.6 imes10^3$ (4.9 $ imes10^3$ )	2.5 (7.9)	$3.7 imes 10^{-1}$ (9.2 $ imes 10^{-1}$ )
Europe	$1.4 imes10^2$ (4.0 $ imes10^2$ )	$3.2  imes 10^{-1}$ (2.1)	$6.1 imes 10^{-2}$ (2.2 $ imes 10^{-1}$ )
North American	$2.7\times10^2$ (5.1 $\times10^2$ )	1.0 (2.7)	$1.4\times10^{-2}$ (1.9 $\times$ $10^{-2}$ )

net sampling failed to quantify MPs in this range; this is an important omission since particles in this size range may account for a large proportion of the total by frequency. When selecting a mesh size or a pore-diameters, there is a trade-off between the sampling volume and the minimum size of particle that can be captured, since the higher fluxes necessary to sample large volumes can only be achieved with larger mesh size. Therefore, filtering surface water using sieves with decreasing mesh size [31,44] are recommended. On the other hand, sampling locations and the different sampling volumes used for the two sampling methods may also influence the observed MP abundance. As a result, the current view is that the bulk sampling provides a better description of MPs > 50  $\mu$ m in DWSs.

Additionally, the measurement of MP abundance depends largely on the detection limit. In bottled water, almost 80 % of all MP particles were smaller than 20  $\mu$ m when the detection limit was 5  $\mu$ m [75], while over 80 % of the detected MPs were between 1–5  $\mu$ m when the detection limit was reduced to 1  $\mu$ m [76]. The latter study even detected 2649–6292 MPs per litre in mineral water contained in plastic or glass bottles [76]. Thus, the abundance of the total MPs in DWSs is very likely to be underestimated due to the non-detection of smaller-sized MPs. In the future, more information on smaller-sized MP pollution in DWSs is required to guide the assessment of exposure levels, treatability and to aid in the development of standards for the protection of water sources.

### 3. The removal of MPs in drinking water treatment plants

### 3.1. Coagulation and clarification processes

Coagulants aim to remove suspended particles and colloidal substances from raw water through charge neutralization, adsorption, and enmeshment (sweep flocculation). Aluminium (Al) and iron (Fe) salts are the most commonly used coagulants. There are limited studies on the removal of MPs by coagulation, however Ma et al. [77,78] investigated the removal efficiency of large PE particles using Al and Fe coagulants in jar tests. The size of the plastic particles was of a diameter <5 mm, with the smallest size range classified as <0.5 mm. The density range was between 0.92–0.97 g cm<sup>-3</sup>. The results revealed that Al-based salts performed better than Fe-based salts with increasing dose. For example, the results for the smallest particle class (<0.5 mm), showed that the PE removal efficiency remained stable at approximately 13 % after the dose of FeCl<sub>3</sub>·6H<sub>2</sub>O was increased to concentrations of 1 mM and above. In the case of AlCl<sub>3</sub>·6H<sub>2</sub>O, the removal of the small PE MPs reached 25 % at doses of 1 mM and increased to 40 % using 15 mM of AlCl<sub>3</sub>·6H<sub>2</sub>O (equivalent to 405 mg  $L^{-1}$  Al) [78]. Large MPs (>0.5 mm) were less well removed, with removal of <10 % for both Fe and Al coagulants. When the coagulants were applied at doses typical of those used in DWTPs, they both performed in a similar way. For example, the removal of PE <0.5 mm was 8.24  $\pm$  1.22 % in the presence of 0.5 mM FeCl<sub>3</sub>·6H<sub>2</sub>O and  $8.28 \pm 1.06$  % with 0.5 mM AlCl<sub>3</sub>·6H<sub>2</sub>O at pH = 7, respectively [78].

The relatively poor removal of MPs by coagulation was likely explained by the large size of the plastic particles and their high buoyancy, factors that would prevent MPs from being captured in flocs. Pristine MPs are typically electronegative in near-neutral pH environments [77] and the charge will change depending on the character of the water matrix. The mechanism of removal of MPs during coagulation will be the same as for any particle through charge neutralization and adsorption when coagulants are added into water and hydrolyse into electropositive hydroxyl complexes. Increasing coagulant concentrations enabled further sweep flocculation to occur, improving entrapment of MP. The average size of the flocs formed by Fe and Al were 258.6  $\pm$  20.8  $\mu m$  and 474.8  $\pm$  25.6  $\mu m$  respectively when the dose was 0.5 mM and the pH was 7.0. Consequently, while the larger MPs were in the mm size range, the formed flocs were only several hundred  $\Box m$  in diameter. Such dimensional differences between the MPs and the floc is a critical feature that influences the efficacy of coagulation, as it would be very hard to capture MP particles of this size into flocs. The Al flocs were

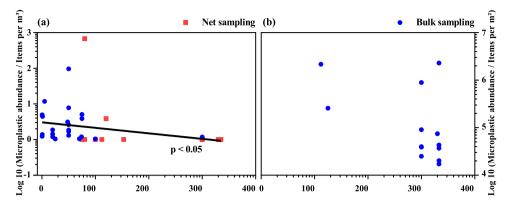


Fig. 3. Relationship between the mesh size of nets in net sampling or pore size of filters in bulk sampling and average abundance of microplastics in drinking water sources as (a) items per m<sup>3</sup> and (b) items per m<sup>2</sup>.

smaller and had a larger specific surface area than the Fe flocs at 286.8  $\pm$  11.2 m<sup>2</sup> g<sup>-1</sup> and 254.1  $\pm$  16.3 m<sup>2</sup> g<sup>-1</sup>, respectively. As a result, the larger specific surface area may have provided more opportunities for capture of the smaller MPs (<0.5 mm), which may have explained why Al coagulants removed more of the MPs [78]. Nevertheless, the removal of 40 % of the MPs was achieved using very high coagulant doses, far higher than those used in real DWTPs. In the case of both coagulants, such high concentrations would not be feasible due to the quantities of sludge produced and the health concerns associated with high residual metal in the treated water [22].

The removal of smaller MPs by coagulation has also been studied using coagulation jar tests and much higher removal was observed [79]. Their results showed that removal of MP spheres (1-5 µm diameter, density 1.3 g cm<sup>-3</sup>) and PE fibres (5 µm diameter, 0.1 mm length, density 0.96 g cm<sup>-3</sup>) were similar to kaolin when aluminium sulphate was used as a coagulant. The turbidity of a 5 mg  $L^{-1}$  MP solution was reduced by 99 % reduction and to less than 1.0 NTU at coagulant doses of between 5–10 mg L<sup>-1</sup> Al. Similar removal was seen for PE fibres [79]. The coagulation performance was not significantly improved by changing the zeta potential of the MPs using a surfactant. This indicated that enmeshment of MPs during sweep flocculation was the dominant mechanism by which these small MPs were removed. In this case, as the coagulant dose was increased above 10 mg L<sup>-1</sup>, there was higher residual turbidity [79]. The increased volume of aluminium hydroxide at high alum doses decreased the average number of microspheres in a cluster, and resulted in colloidal protection at these higher doses [79]. Although much higher removal was seen here, the residual turbidity could still equate to a high number of MPs in treated water (estimated at  $6 \times 10^5$ items m<sup>-1</sup>). However, this needs to be considered with caution due to the very high load of MPs spiked into the water, much higher than would be typically seen in DWS.

Additional understanding from these jar test studies was the role that polymers could play in improving MP removal when dosed with conventional coagulants, particularly for the larger particle sizes. In many DWTPs, polymer is added to aid particle removal in flocculation processes [80]. For the removal of MPs, studies have shown that anionic polyacrylamide (PAM) performed better than cationic PAM for the additional removal of mm-size MPs when compared to dosing of Fe and Al coagulants alone [77,78]. For example, the removal of PE (2–5 mm) with 5 mM AlCl<sub>3</sub>·6H<sub>2</sub>O at pH = 7 was 4.27 % (no PAM), 5.83 % (15 mg L<sup>-1</sup> cationic PAM), and 18.34 % (15 mg L<sup>-1</sup> anionic PAM) [78]. The same phenomenon has been observed for the removal of clay impurities in DWTPs [81]. Other factors, such as pH, ionic strength, the concentration of NOM and turbidity had a little or no effect on MP coagulation performance [77–79], but this conclusion needs further verification from more complex matrices than those used in the jar test trials.

MPs are a set of bio-refractory solids with different shapes, sizes, and chemical composition, similar to many other small particles removed by coagulation processes. However, like many other types of particulate, they will not be effectively entrapped in a floc if they are too large or have a significant density difference to that of water [79,82]. Small MPs in the low micron range appear to be effectively removed by coagulation processes and, while larger MPs were poorly removed, downstream processes (such as clarification or filtration) are expected to be highly effective barriers for their removal.

After coagulation and flocculation, particles are typically removed in clarification treatment processes, based on either sedimentation or flotation. To date there has been limited published research on MP removal in clarification processes, but evidence can be obtained with data from real DWTPs and WWTPs. Data from a Chinese DWTP identified that 40.5–54.5 % removal efficiency of MPs (>1 µm) was obtained across coagulation and a sedimentation clarification process [83]. The highest removal efficiency was observed for the removal of MPs that were defined as being  $>10 \,\mu m$  [83], particles closer in size to floccules. In addition, it was observed that agglomeration of MPs into flocs was more likely for fibres when compared with spheres and fragments of plastics, with around 50.7-60.6 % removal efficiency in coagulation/sedimentation process compared to less than 40 % for spheres and fragments, respectively [83]. PET accounted for 59.1-68.7 % of the MPs removed by coagulation/sedimentation, a reflection of their high prevalence in raw water and because many of the fibres were present as this polymer type [83]. Sludge blanket clarifiers may offer enhanced opportunities for removal of MPs because removal can be increased by contact flocculation and filtration through sludge blankets. Flotation is another effective process method available to remove matter of small particle size and low density. For example, algae and organic matter can be effectively removed by flotation processes [84,85]. There is limited data available for MP removal using flotation processes implemented at DWTPs. However, Talvitie et al. [86] investigated a dissolved air flotation (DAF) process as an advanced final-stage treatment technology at Paroinen WWTP, Southern Finland. The DAF process removed 95 % of MPs (>20  $\mu$ m) from an influent containing a concentration of 2.0 to 0.1 M P items  $L^{-1}$  [86]. Given that many MPs have a low density, flotation should be considered as an effective method for their removal. The use of microbubbles [87] or nanobubbles [88] for MP removal should also be explored in the future to determine whether these more novel flotation systems offer enhanced removal of plastic materials.

### 3.2. Sand filtration and membrane filtration processes

Rapid gravity filters (RGFs) can intercept suspended and colloidal particles to improve the safety and hygiene of drinking water [89]. Particles can be strained by the void spaces in the filter when the particle to media diameter ratio is greater than 0.15; for example, an effective media diameter of 0.5 mm will strain particles with a diameter of 75  $\mu$ m on the filter surface [90]. MPs larger than this equivalent size should

therefore be strained from the water using sand media grain sizes typical of those used in DWTPs, including those that have not been removed in coagulation and clarification. However, the aim of RGFs is to remove particles throughout the depth of the bed, through their attachment onto the surface of filter grains. As a result, filtration can remove particles much smaller than the gaps between filter grains. For example, Cryptosporidium oocysts that are 4-6 µm in size can be reduced by 2-log through their interaction with filter grains [91]. The attachment mechanisms that exist between particles and media are fundamentally associated with particle charge interactions. As such. Derjaguin-Landau-Verwey-Overbeek (DLVO) theory, which considers van der Waals and electrostatic double layer interactions (extended DLVO theory also includes Lewis acid-base interaction), can be used to assess filtration performance [92]. Particles can be removed when attraction is high enough to overcome the repulsive forces. A series of sand column experiments were carried out to investigate the transportation and retention of MPs in a filter using media with a diameter of  $0.45 \pm 0.03$  mm [93]. The results showed that when PS microspheres with diameters of 0.1-2.0 µm were in deionised water, there was very limited removal of the plastic particles. When the ionic strength was increased by placing the MPs in brine water, near complete removal was observed. In this case, the increased ionic strength reduced the double layer thickness, allowing repulsive forces between particles to be reduced. A similar effect is observed through the addition of coagulant. As a result, water is typically coagulated prior to filtration with or without a clarification stage.

Data from operational filtration systems is limited. Studies from wastewater filtration sites have reported high levels of removal of MPs. For example, an RGF was found to have 97.1 % removal efficiency of MPs (> 20  $\mu$ m) from secondary effluent in a tertiary WWTP, where MPs were detected using a stereomicroscope and FTIR [86]. In the case of DWTPs, lower removal has been observed for smaller MPs. For example, the removal efficiency of MPs (> 1  $\mu$ m) by sand filtration was between 29.0–44.4 % for water treated by coagulation/sedimentation in a DWTPs [83]. In this case, all MPs >10  $\mu$ m were almost completely removed by the sand filter.

Membrane processes, including porous membranes and diffusional membranes, remove particles in different ways compared to RGF process despite their similar objective [90]. Porous membranes retain particles that are larger than the pore size of the membrane by a straining mechanism [90]. These membrane technologies, especially ultrafiltration (membrane pore size:  $\sim 20$  nm [94]), are often retrofitted to upgrade DWTPs to facilitate enhanced removal of contaminants [95]. Ma et al. [78] reported no breakthrough of MP particles during ultrafiltration of water treated by coagulation. A looser cake layer on the membrane surfaces was observed during the experiments when the influent contained larger MPs in the range of 0.5–5 mm, which in turn would depress the fouling of the membranes [78].

In the case of diffusional membranes with smaller pore sizes, applications tend to be focused on desalination. Over two-thirds of the installed desalination capacity worldwide is reverse osmosis (RO) and the low end of the nanofiltration [96]. Desalination by these membranes can produce almost pure water, removing molecules much smaller than MPs, thus there should be no concerns about MPs in treated water using this technology. However, in the case of RO, treated water may contain very low mineral content [97], water that is not recommended for human consumption. In these cases, RO permeate water may be blended with conventionally treated in order to remineralize the water [95]. There is therefore potential for MPs to be added back into the water. Overall, however, while empirical data is lacking for MP removal by membranes, these processes are expected to be highly effective.

### 3.3. The comparison of MPs in inlet and outlet water of DWTPs

Much less research has been carried out determining the fate of MPs in DWTPs than have been carried out for WWTPs and in natural water

bodies. As far as the authors are aware, only three peer-reviewed research papers have investigated the presence of MPs in the inlet and outlet water of DWTPs using surface water and groundwater as sources. The different detection limits used in these three articles contributed to the varying MP abundance reported. In one assessment of a groundwater used as a drinking water source, the concentration of MPs (>20 µm), ranged from 0 to 7 items  $m^{-3}$  in groundwater wells and tap water [98]. Such low levels of MPs were insufficient to enable calculation of a removal efficiency. The MP particles that were identified had a size distribution between 50 and 150 µm and were comprised of PE, PA, PES, PVC, and epoxy resin [98]. The authors supposed that the MP particles might have derived from the abrasion of plastic materials in the water treatment system [98]. Similar speculations were made from the results obtained from surveys of surface water DWTPs [83,99]. Some water treatment chemicals may also appear as MPs. For example, PAM used as a coagulant aid has been observed to increase the MP count in treated water [83,98].

In an assessment of four DWTPs that had different water sources. consistent removal of MPs of between 81-86 % was observed in three DWTPs (#2-4) that had extensive treatment stages, even though varying concentrations of MPs were observed in their influent and effluent (Fig. 4). The lowest removal was observed across DWTP #1 (70 %), which was a simpler flowsheet without sedimentation/flotation and granular activated carbon filtration (Fig. 4). In all of these cases, MPs were quantified down to particle sizes of 1 µm. In the DWTPs supplied with surface water, MPs with diameters of  $>1 \,\mu m$  were most abundant in the source water. In addition, Pivokonsky et al. [99] identified that PET particles were the most abundant polymer type, consisting of between 60–68 % of the particles quantified in the source water. PET plastic also dominated in the raw and treated water from a DWTP in China [83]. The PP and PE plastics only made up a maximum of 30 % of the plastic particles found in the source and treated water for all four DWTPs [83, 99]. However, a wider variety of polymer types were observed in the river water compared to the reservoirs [99]. Both of these studies suggested that MPs greater than 10 µm were either completely or nearly entirely removed from the drinking water sources.

Much greater removal was observed in an assessment of eight DWTPs in the UK, treating a range of water sources (river, groundwater and reservoir sources) [100]. Although an array of different treatment technologies was deployed, removal across all DWTPs was very high, with removals typically around 99.9 %. In this analysis, MP particles greater than 25  $\mu$ m were assessed. Much lower concentrations of MPs in the influent to the DWTPs were detected, with an average of 4.9 item-s·L<sup>-1</sup>. Levels in the treated water were around an average of 0.00011 items·L<sup>-1</sup>. These lower concentrations are likely to be a reflection of the focus on larger MPs in this assessment. This is a point highlighted by the abundance of MP particles that were <10  $\mu$ m in the source and treated water in the studies of Wang et al. [83] and Pivokonsky et al. [99]. In these studies, the small MPs <10  $\mu$ m comprised more than 80 % of the particles characterised.

From the small number of studies carried out, it was evident that MP removal efficiency was highly dependent on the size range of the MPs assessed in the investigation. The larger MPs (>10  $\mu$ m) can be removed completely during treatment, while there was a great uncertainty on the removal of smaller MPs, particularly those particles that were <1  $\mu$ m. More focus should be placed on understanding and controlling the fate of different sized particles through our drinking water systems as well as continued efforts to harmonise robust MP sampling to ensure that results can be more comparable.

## 4. The potential size-dependent threats of MPs in drinking water sources

### 4.1. The potential ecological threats of larger MPs

The current environmental monitoring has shown the ubiquity of

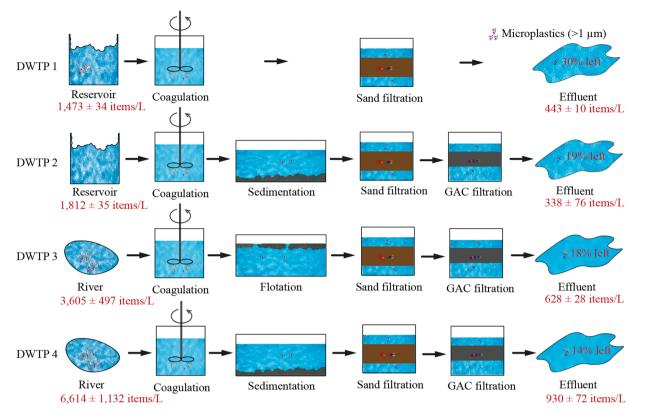
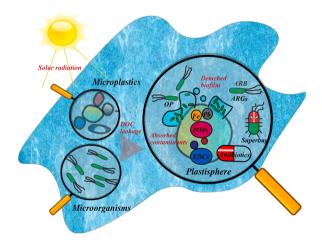


Fig. 4. Microplastics in the influent and effluent of four drinking water treatment plants. Data source: Pivokonsky et al., 2018 (DWTP 1, 2 and 3), Wang et al., 2020 (DWTP 4).

larger MPs (>50 µm) in DWSs. These larger MPs serve as a possible substrate on which biofilms may grow in aquatic environments [101]. Their hydrophobicity and high surface area to volume ratio makes them favourable for the attached growth of microorganisms. MP particles are often assumed to be bio-refractory, but many hydrocarbon-degrading microorganisms are found on MPs [102,103]. An increasing number of microbes that are capable of degrading MPs have been discovered, including fungi (e.g. Zalerion maritimum [104]) and bacteria (e.g. Enterobacter asburiae YT1 and Bacillus sp. YP1 [105]). This shows that plastic can provide energy for biofilms to grow on. Additionally, as an ecological niche [106], MPs have a longer life and greater mobility than many natural substrates. Heavy metals and organic pollutants may also accumulate in biofilms on the surface of MPs. For example, a positive relationship between the accumulation of heavy metals on MPs and the amount of biofilm attached to MPs has been observed [107]. This evidence shows that the biofilms that grow on MPs are different from those growing on other natural substrates. For example, in marine and freshwater environments, microbial communities attached to MPs have been found to be significantly different from those in the surrounding water and sediments [108]. Researchers have therefore proposed that the sum of all living things on MPs and their surroundings should be named the "Plastisphere", as a new ecosystem due to its uniqueness and omnipotence [103]. The opportunity for plastispheres increases as more MPs enters DWSs (Fig. 5). In turn this might exert a subtle influence on the microbial composition in plastic impacted which may ultimately influence organisms in drinking water, particularly in circumstances where water is consumed untreated or partially treated.

Opportunistic pathogens have been identified on MPs [109]. This includes organisms such as *Vibrio* spp. [110], which can then be spread further into the environment with the dispersion of MPs in DWSs. Some other potentially harmful microorganisms may also be selected for when biofilms develop on MPs. For example, Oberbeckmann et al. [111] identified that *Sphingolomonas* and *Enterobacter sphingolomonas* were the



**Fig. 5.** Potential ecological risks of microplastics in drinking water sources. DOC: dissolved organic carbon; OP: opportunistic pathogen; ARB: antibiotic resistant bacteria; ARGs: antibiotics resistance genes; POPs: persistent organic pollutants; EDCs: endocrine-disrupting chemicals.

dominant bacteria in the MP biofilm. These species are known as being resistant to many disinfecting and oxidative chemicals used in drinking water systems [112]. Microorganisms in MP biofilms also display some unique properties at the genetic level. This is demonstrated by gene exchanges that are more likely to occur between different species of microorganisms through horizontal gene transfer (HGT) [113]. Lagana et al. [114] found that the frequency of plasmid pKJK5 transfer in bacteria on MP surfaces was 1000 times that of suspended bacteria, suggesting that MP biofilms greatly increased the frequency of HGT. Integrons are gene capture systems that can carry recombinant gene boxes into transposons or plasmids, and therefore play an important role in HGT [115]. Eckert et al. [116] have shown that the content of class I integrons on the surface of MPs is significantly higher than that in the surrounding surface water. HGT increases the risk of the inter-population spread of detrimental genes such as antibiotic resistance genes (ARGs) and metal resistant genes, and the risk may be heightened in the plastisphere. Bacteria enriched on the surface of polystyrene sheets (macroplastics) deposited off the Antarctic coast were found to form biofilms and be resistant to multiple antibiotic drugs [114]. Yang et al. [117] found that antibiotic and metal resistant genes were significantly more abundant in MP-associated microbiota than in seawater-associated microbiota, and noted that MP biofilms in the ocean were a repository for antibiotic resistance genes (ARGs). MPs selectively enrich microbial communities in the environment, and even long-term impacts on the carbon or nitrogen cycle [118,119] could also be realised by increasing plastispheres.

The presence of MPs in DWSs have complex impacts on aquatic microbial communities. Moreover, bacteria can be detached from biofilms through abrasion, grazing, erosion and sloughing [121], and therefore may result in a planktonic state in the water column. Abrasion occurs when particles in the water collide with the biofilm, causing bacteria on the surface to enter the water. Grazing refers to the predation of microorganisms in the biofilm by protozoa in the water, which causes the bacteria to fall off. MP ingestion by zooplankton has been widely recorded in marine environments [122]. Most researchers have focused on the toxic eff ;ects of MPs on zooplankton (for example, studies showing how MPs aff ;ect their development and reproduction) [122], and the related biofilm detachment has been ignored. Erosion of biofilms occurs by hydraulic action, which causes a small amount of material in the biofilm to continuously enter the water. Sloughing is similar to erosion, but refers to a part, or all, of the biofilm entering the water. The detached component retains some of the structure of the biofilm, including the presence of extracellular polymeric substances. Additionally, microorganisms may enter the water at any time by desorption and dispersion at the stage of biofilm formation and development [123]. The release of bacteria from MP biofilms may allow opportunities for pathogenic organisms to enter the water, and thus bring public health and ecological risks to DWSs. Similar health concerns have been focused on drinking water distribution systems for a long time, which supply much larger surface than that offered by the MP particles. However, further understanding of the magnitude of this risk is still required considering that more complex microbial composition exists in DWSs than that in the chlorinated water environment.

### 4.2. The potential chemical threats of smaller MPs

MPs <10  $\mu$ m have been typically less well studied in the investigations of DWSs, usually due to analytical limitations. However, as has been identified, these are the MPs that have the lowest removal potential across DWTPs and are therefore more likely to be found in final treated drinking water. It is also important to consider that further degradation of MPs has the potential to change the composition, character and treatability of a water body given that plastics typically have a carbon content >90 % [124].

Firstly, a high concentration of MPs in DWSs can increase the total organic carbon (TOC) concentration. Hu et al. [124] have demonstrated that the TOC of river water increases significantly when NP plastics were added to the water (PS, 50–100 nm, 2.5 % w/v), rising from 9.78 to 11.43 mg L<sup>-1</sup>. Many NPs are small enough to pass through membrane filters typically used to determine dissolved organic carbon (DOC), which is technically defined as the fraction of TOC that can pass through 0.45  $\mu$ m filters [125]. As such, NPs may be measured as part of the DOC. Aside from NPs discharged into water bodies directly, and from fragmentation of larger parent particles, DOC can also be leached directly from the plastic into DWSs. Due to their low density, MPs usually float on the water surface and are therefore exposed to mechanical weathering from wave energy and are exposed to solar radiation. These abiotic

factors, particularly the ultraviolet (UV) wavelengths present in sunlight, reduce polymer molecular weight through chain scission, and/or crosslinking reactions, or oxidative processes [126]. As a result, NPs derived from polymer fragmentation [127] or polymers and chemical additives in the plastics [10,128] can be released from MPs. Romera-Castillo et al. [129] found that DOC was released from MPs (including PE and PP) in the range of 0.26–8.92  $\mu$ g DOC cm<sup>-2</sup> with or without artificial solar radiation during a 30-day exposure experiment in artificial seawater. From these results, the authors extrapolated that up to 10  $\pm$  0.3 % (23,600 t) of the DOC in the top 40  $\mu m$  surface microlayer of the sea originates from marine plastics [129]. In their experiments, there was no difference in the total DOC leached during tests carried out in the dark and under sunlight [129]. This was because  $60 \pm 26$  % of the DOC was released when MPs first contacted with water. However, after this instantaneous DOC release, MPs under radiation leached significantly more DOC than dark controls. Similarly, Zhu et al. [130] attributed the main cause of DOC leaching from MPs to photodegradation over a longer reaction time. In these experiments, the leached DOC from photochemical breakdown of MPs (including PE, PP, EPS, and environmental MPs collected from surface seawater) reached between 1.1–68.2 mg $\cdot$ (g-C)<sup>-1</sup> over 54 days of irradiation. No leaching was observed when the experiment was carried out in the dark for most of the MPs investigated. The exception was for environmental MPs, which had already been exposed to natural weathering and mechanical forces [130]. In these experiments, the MPs were cleaned prior to the experiments using H<sub>2</sub>O<sub>2</sub> and ultrasound treatment which removed the DOC that would have been released on immediate contact with the water [130]. The degradation through photo-degradation equated to the mass loss of EPS, PP, PE, and environmental MPs by  $6.8 \pm 0.1$  %,  $3.91 \pm 0.03$ %,  $1.1 \pm 0.2$  %,  $1.62 \pm 0.02$  % respectively [130]. Such high percentages suggested that the DOC released under continuous light exposure could only come from the breakdown of plastics. Leaching of DOC from a range of plastic polymer types and materials, ranging from shopping bags, nylon rope and plastic drinking vessels into freshwater reached up to 3350 mg kg of MP [131]. In this research, polyamide plastics released the highest amount of DOC, with most other materials being between 30–1000 mg kg of MP. Furthermore, the assimilable component of the DOC generated by MPs will have an impact on microbes in water environments. Romera-Castillo et al. [129] have reported that approximately 60 % of DOC released from MPs is available for microbes in less than 5 days. A wide scale survey on polymer materials and their subsequent degradation identified that 22 %-76 % of the DOC generated from the polymers were utilized by marine bacteria following a 92-day incubation period [130]. Microbial re-growth promoted by DOC of MP origin may therefore influence the water quality in drinking water systems.

In addition to consideration of DOC, UV absorption has been another useful surrogate measure of organic constituents in water quality monitoring of DWSs. The UV absorption of a substance depends mainly on the chromophores within the molecular structures. For example, lignin, tannin, humic substances, and various aromatic compounds in freshwater strongly absorb UV radiation due to the presence of unsaturated bonds [132]. The absorbance of UV can also therefore be important for plastic polymers, both with respect to their identification and in their potential for degradation. For example, polymers with unsaturated bonds (e.g. PS, PET) readily absorb UV light and undergo photo-oxidation reactions. However, the most common MPs (e.g. PE, PP) are examples of polyolefin materials, which only have C-C and CH- chains and therefore do not absorb UV light in their completely pure state. In most cases, though, plastics are not pure materials. The ability of the plastic material can be affected by the impurities that are present in the manufactured products, including the residual catalyst, additives, metal ions, carbonyl compounds and dye chemicals. As a result, different MP pollution levels in DWSs may interfere with UV absorption in raw water. MPs will therefore affect the calculation of secondary indicators of water quality in DWSs such as the specific

ultraviolet absorbance (SUVA), the ratio of UV absorption to the DOC concentration. This indicator is used to inform on the treatability of the water and the propensity for disinfection by-product (DBP) formation [133]. Our understanding of how leached DOC and the UV absorption from plastics interferes with such indicators is an area of microplastic research that needs further investigation.

With respect to DBPs, the DOC leached from plastics has been assessed with respect to its formation potential. In these tests, the reactivity of the DOC from plastics was similar to that seen for naturally derived organic matter (NOM). The yield of trihalomethane (THM) DBPs from DOC leached by plastic was between 11.3 and 158.9  $\mu$ g. (mg DOC)<sup>-1</sup>. This was a range similar to that seen by an assessment of DBP formation potential from a range of raw and treated water natural water sources in Scotland [134]. Haloacetonitriles (HANs) were also identified in concentration ranges between 2–9  $\mu$ g·L<sup>-1</sup>. Bromide was also released from some of the plastic materials, which favoured the formation of brominated THMs and HANs. These results therefore show that leached DOC from plastic may be an important precursor for DBPs in some heavily impacted water sources.

In summary, this analysis shows that MPs are another type of chemical organic matter that has an obvious difference in origin from that of NOM. However, there appear to be some similar risks associated with these two types of DOC with respect to their ubiquity and influence on water quality parameters, including DBP formation.

### 5. The potential threats of MPs in alternative water sources

Seawater, rainwater, and wastewater are alternative sources of drinking water that are being used to tackle the challenges of water shortages in an urbanising and water stressed world [135]. A higher abundance of MPs is typically seen in these water sources than in conventional DWSs. Seawater is a major source of drinking water in the Middle East and North Africa regions. For example, 50 % of drinking water originates from seawater in Israel, with some regional areas of the country reaching 80 % [136]. Seawater desalination is emerging in other countries, such as the U.S., China, and Australia where it may be blended with conventionally treated drinking water [137]. There are three types of water intake that are used in desalination that could influence the distribution of MPs in the source. These are surface/open seawater intakes, deep-sea water intakes, and under-the-seabed water intakes [138]. In seawater, plastic polymers tend to be more enriched in surface water layers compared to deeper waters due to the low density of MPs [138]. For example, the average abundance of MPs  $>20 \mu m$  in the surface (0–0.2 m), middle (3–27 m) and bottom layers of water (5–58 m) of South Korean coastal waters were 1,736, 423 and 394 items m<sup>-3</sup>, respectively [139]. However, MPs have still been detected in deep seawater, extracted from depths of 1,100-5,000 m [140]. In the Arctic ocean, deep seawater contained MPs at a concentration of between 0–104 items m<sup>-3</sup>, which was only slightly less than MPs found in the surface water layer (0-375 items m<sup>-3</sup>) [141]. Seawater taken from beach wells avoids the impingement and entrainment of marine ecology as a result of filtration of this water through the seabed [142]. It would therefore be expected that MPs would also be filtered out during this extraction. Although seawater has been identified as a water source that contains higher loads of MP pollution, the membrane treatment processes typically used to treat this water type have the capability of removing all of the MPs from the water. However, the influence of MPs on fouling and operation of membranes requires additional investigation for these DWSs [143].

Rainwater is an important resource for drinking water in many countries and island communities [144,145]. For example, household rainwater tanks supply drinking water to up to 23 % of the population outside of major cities in Australia [146]. MPs have been found in rainwater and other precipitation, even in remote areas. More than 90 % of rainwater samples collected from the Colorado Front Range have been shown to contain MPs [147]. Snow from the Alps to the Arctic has been

shown to contain MPs at concentrations from between 0 to  $14.4 \times 10^3$  items·L<sup>-1</sup> [148]. The MPs in rain is thought to arise through interaction between water and plastic particles deposited in the air [149]. Typically, fibres are identified as the dominant MP type found in the air [150,151], an observation consistent with that seen in precipitation [147,148].

Rainfall has an impact on MPs in all types of water source. The number of MPs in receiving water has been shown to increase by more than 14 times from 539,189 to 7,699,716 items  $\text{km}^{-2}$  following heavy rainfall [152]. This is explained by the transfer of terrestrial MPs into water bodies by run-off, and from direct atmospheric wet deposition. Roof-harvested rainwater can be directly used as a DWS at household/community levels [153], while rainwater from urban stormwater runoff may influence DWSs through a more circuitous route. In this case, run-off water is typically collected into retention ponds or stored in storm water tanks and is then discharged into rivers or groundwater basins (with or without treatment) through artificial recharge [154]. Plastics on the road are washed into drainage systems, alongside other pollutants [152,155]. The median MP concentration found in Mexican stormwater runoff was 66–191 items L<sup>-1</sup> [156]. Levels of MPs in Danish receiving ponds have been reported to be between 490 to 22,894 items m<sup>-3</sup> and research has identified that PVC, PS, PP, PE, and polyester (PES) were the most common polymer types [157]. Run-off from industrial and commercial areas has been shown to contain even higher concentrations of MPs than that from highway and residential areas, 5,249-22, 894 versus 490-1,409 items L<sup>-1</sup> [157]. Pinon-Colin et al. [156] also recorded the highest abundance of MPs (median value: 191 items L<sup>-1</sup>) in run-off from an industrial land use site. These studies indicate that the concentration of MPs in run-off is closely related to human production activities.

The influence of wastewater discharges on DWSs has been extensively researched. This is becoming an ever more important area to consider as many DWTPs abstract water downstream from discharges of wastewater treatment plants (WWTPs) through indirect water re-use (IR) [158]. In addition, the direct re-use (DR) of wastewater for drinking water is being considered more widely. Currently, DR is only practised in a small number of locations, such as Windhoek, Namibia; Cloudcroft, New Mexico; and El Paso, Texas [159], but future trends are likely to see this increase significantly in line with growing water resource challenges. Treated wastewater is also used for groundwater recharge that is then subsequently used for drinking water production [160]. The fate of MPs in wastewater, from influent to effluent, has been studied more extensively than for drinking water and has been identified as an important source of environmental plastic pollution [161]. The median value of MPs in the effluent of wastewater treatment plants (WWTP) ( $\sim$ 100–1000 items m<sup>-3</sup>) [23] was observed to be similar to the range seen in lakes and rivers in the analysis here. However, the MP concentration data for wastewater spanned nearly eight orders of magnitude, a much broader range than that seen for DWS [23]. Fibres were identified as the most common shape of MPs in final WWTP effluent, and the most commonly identified polymers were PES (~28 %-89 %), PE (~4%–51 %), PET (~4%–35 %) and PA (~3%–30 %) [162]. In addition, as most MPs are hydrophobic, adsorption of organic compounds and heavy metals from the wastewater onto the plastic surface may occur [163,164]. Hence, a better comprehension of how contaminants adsorb onto the surface of MPs is also required. This may be more important in these alternative water sources, particularly those associated with wastewater where pollutants may be more prevalent.

A large proportion of seawater is used for industrial or agricultural applications, while rainwater and treated wastewater are usually used for potable water indirectly or for non-potable applications and for irrigation. However, as water resources are being placed under evermore pressure because of population growth and climate change, the use of alternative DWS for potable water has seen a significant increase [135,165]. As such, MPs in these DWSs should not be overlooked. In general, these DWSs have greater potential to be sources of MPs than surface water and groundwater. On the other hand, however, more

complex and advanced treatment processes are usually applied to treat these types of water source. For example, seawater typically goes through reverse osmosis or multi-stage flash distillation in order to obtain highly purified water [96]. Similarly, direct wastewater reuse schemes use advanced and multiple stages of treatment [159]. As a result, opportunities for high levels of MP removal are present. Overall, however, this analysis shows that the MP presence in DWSs is strongly dependent on the source water characteristics, particularly in the case of indirect reuse.

### 6. Conclusions and future directions

The potential threats posed by the presence of MPs in drinking water are of increasing concern in the Anthropocene. The understanding on MP occurrence, removal and transformation in DWSs and DWTPs is of great importance to understand their potential threats. We analysed 53 studies that were directly related to MPs in water sources, and found that bulk sampling can better describe the current MP presence in DWSs. The statistics showed the median MP abundance in conventional water sources was  $2.2 \times 10^3$  items m<sup>-3</sup> with the size usually larger than 50  $\mu$ m. These larger MPs can provide sufficient surface area for the growth of biofilm. Although data is limited, there are potential ecological threats presented by these plastispheres in DWSs, including the transmission of pathogenic organisms and antibiotic resistance. Additionally, components of MP associated biofilms may detach from MPs and become freeliving bacteria in the water, which may raise the risk of the spread of waterborne diseases in drinking water, particularly in circumstances where water is consumed untreated or partially treated. In contrast, the understanding with respect to smaller-sized MP exposure and the removal of MPs from DWSs remains limited. This is particularly the case for MPs that are smaller than 10  $\mu$ m. These particles are more likely to pass through treatment technology and therefore result in tap water and pose potential risks to the human body. Organic carbon leached from ageing MPs and NPs in DWSs may bring about cumulative effects on the ecology and carbon cycling in DWSs. Further understanding on the change of chemical water quality parameters, DBP formation and the influence of ageing MPs on microorganisms are required in future research. Moreover, the importance of alternative DWSs is growing worldwide as climate change and increasing population growth places more pressure on our water supplies. MP threats in alternative DWSs should also be considered with respect to water sources and treatment processes in the future.

#### **Declaration of Competing Interest**

The authors report no declarations of interest.

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### Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.jece.2020.104527.

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