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• Review Article

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Occurrence of microplastics in municipal sewage treatment plants: a review

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Municipal sewage treatment plants (STPs) are thought to be important point sources of microplastics in freshwater systems and many peer-reviewed articles have been published on this issue since mid-2010s. In this review, we summarize existing literature on the occurrence of microplastics in STPs and experimental methods used for isolation and identification of microplastics. The number concentrations of microplastics in STP influents were 15.1-640 L^{-1}, whereas those in the STP effluents were highly variable and ranged from not detectable to 65 L^{-1}. For most of cases, conventional STPs are removing microplastics very effectively. Fragments and fibers are dominant shapes of microplastics. Thermoplastics (polyethylene and polypropylene) and polyester are the predominant materials recovered. Although further research is needed, size distribution of microplastics in STPs is likely to follow a power law, implying that different studies using different size cutoffs may be compared after establishing a power law relationship.

Keywords: microplastic identification; sewage treatment plants (STPs); material type; shape; size distribution

INTRODUCTION

Heavy use of plastic products inevitably ends up with small-sized plastic particles in the environment. Plastic particles less than 5 mm in size are called “microplastics” [1]. Recent studies revealed that microplastics are accumulating in the oceans [2-6] as well as in the terrestrial environments [7-12]. Because identified level of microplastics in the environment is much lower than estimated from mismanaged flows of plastic products [5,13,14], the actual level of microplastics including unidentified is suspected to be much higher than observed [4,13]. The increasing level of microplastics in the environment as well as biota has drawn great attention from researchers and general public with increasing evidences of adverse effects of microplastics [15-18].

The origins of microplastics are suspected to be engineered small plastic particles in products such as microbeads in cosmetics and other consumer products or breakage of bigger plastics into smaller particles via various weathering processes [19,20]. Due to long degradation half-life of plastics (often estimated over 100 years [21,22]), microplastics, once formed, may travel long distance and spread over the world like persistent organic pollutants [23-26]. Among many potential sources of microplastics to the environment, sewage treatment plants (STPs) are regarded as important point sources to the freshwater environments and released microplastics may ultimately reach to the oceans via river flows [19,20,27-41]. Thus, it is crucial to evaluate the contribution of STPs as sources of microplastics to the natural waterways.

In order to estimate the level of microplastics entering into and leaving from STPs, it is required to have reliable and reproducible experimental methods to count microplastic particles in sewage influent and effluent. Many researchers tried to isolate and quantify microplastics from wastewater influent and/or effluent [27,29,31-33,35-38,42-48]. In those studies, occurrence of microplastics is usually expressed in the units of number of plastic particles per volume of water [31-33,35-38,43-48]. The number concentration of microplastics in the STP influents was between 15 and 640 particles L^{-1} [32,35,37,38,44,46] and that in the effluents was much lower, but varied over 4 orders of magnitude [31-33,35-38,43-48]. Many different meth-

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Table 1. Summary of studies in which microplastics were identified in STP influents and effluents.

Location	Information of STP		Sampling method			Analytical Method			Number concentration[a]		Ref.
	Daily process capacity (×10^3 m^3 d^{-1})	Population Equivalent (×1,000)	Sampling equipment	Sample volume (L) Influent	Sample volume (L) Effluent	Pretreatment	Analytical equipment	Size cut-off (mm)	Influent (MP L^{-1})	Effluent (MP L^{-1})	
Scotland	261	650	Steel buckets (10 L) Steel sieve (65 μm)	30	50	filter (11 μm)	dissection microscope & FT-IR	11	15.7 ± 5.23	0.25 ± 0.04	[38]
Sweden		14	Plankton net (mesh size; 300 μm) Suction pump	2	1,000		stereo microscope & FT-IR	300	15.1 ± 1.54	0.0082 ± 0.0017	[44]
USA			Steel sieve (400, 180, 45 μm)		189,000-232,000		microscope & FT-IR	45		0	[27]
Germany	0.52-35	7-210	Moble pumping device (10 μm stainless steel filter)		390-1,000	EM[b] WPO[c]	micro FT-IR	10		0-0.05[d] 0.01-9[e]	[43]
Netherland			Glass jar	2	2	filter (0.7 μm)	light microscopy & FT-IR	0.7	73 ± 13	65 ± 67	[32]
USA	189	680	Steel sieve (5, 1, 0.355, 0.125 mm)			WPO	dissection microscope & micro FT-IR	125			[42]
Finland		310-800	Pump-filter device (200, 100, 20 μm)	0.3	30-285		stereo microscope	20	610	13.5 ± 2.9	[35]
Finland	270	800	Pump-filter device (300, 100, 20 μm)	0.1[e] 0.1[f]	1,000[e] 10.5-13.5f		stereo microscope & FT-IR	20	568[f] ± 165 640[g] ± 255	2.5[f] ± 1.5 0.6[g] ± 0.2	[37]
Australia	13-308	67-1,227	Stainless steel sieve (500, 190, 100, 25 μm)		3-200	WPO	dissection microscope & FT-IR	25		0.21-1.5	[31]
USA	2.3-382	3.5-56,000	Tyler sieves (355, 125 μm)		4,847	WPO	dissection microscope	125		0.05 ± 0.024	[33]
USA	2.3-310		Stainless steel sieve (355, 125 μm)			WPO	dissection microscope	125		0.022-0.13	[45]
Finland	10		Stainless steel bucket sieve (0.25, 5 mm)	4-30		WPO	optical microscope & FT-IR	250	57.6 ± 32.8	1 ± 1.1	[46]
France	240		Automatic sampler			filter (1.6 μm)	stereomicroscope	100	293	35	[47]
Finland	14-88		Pump-filter device (300, 100, 20 μm)				stereomicroscope & FT-IR	20		0.04-1.2	[36]
Canada			ISCO peristaltic pump 100 μm nylon mesh		100	WPO	stereomicroscope	100		0.07	[48]

[a]mean value ± standard deviation; [b]enzymatic maceration; [c]wet peroxide oxidation; [d]MP size > 500 mm; [e]MP size < 500 mm; [f]grab sampling; [g]24-h composite sampling.

ods for sampling, isolation and identification of plastic particles from wastewater samples were tried [27,31-33,35-38,42-48]. Thus, it is unclear that the differences in number concentration of microplastics in wastewater is due to the difference in the level of plastic contamination or due to the difference in sampling and analytical methods used.

In this mini-review, we summarize existing peer-reviewed articles on microplastics in STPs. Because a few reviews and reports have been published in a broader context [19-20,39-41], we narrowed the scope to microplastics in STP influents and effluents. The reported variations in the number concentrations, types, and size distribution of microplastics in STP influents and effluents are compared with experimental methods used for isolation and identification of microplastics. Percent removal of microplastics are also assessed based on reported data. Finally, we propose future research needs on the refined assessment of the microplastics in STPs.

METHODS FOR ISOLATION AND IDENTIFICATION

Table 1 summarizes recent peer-reviewed publications in which microplastics were identified in STP influents and/or effluents since mid-2010s [27,31-33,35-38,42-49]. As shown, researchers have used different methods of isolating, recovering, purifying, identifying, and counting methods.

Glass bottles or steel buckets were used for sampling STP influents that contain high concentration of microplastics and sampling volumes were from 0.1 to 30 L [32,35,37,38,44,46]. Larger volumes of STP effluents were required to isolate microplastics, ranging from 2 to 232,000 L [27,31-33,35,37,38,43,44,48]. Various sampling devices were used, including simple steel buckets [38,46], glass jars [32], commercial metal sieves [27,31,33,38,42,45], plankton nets [44], or custom-made pump-filter systems [35-37,43,44,47,48]. Pore size of filtering devices also highly varied. The smallest pore size was 0.7 μm [32] and the largest size cutoff was 300 μm [44]. Different size cutoffs inevitably lead to great variations in identified number concentration of microplastics.

In order to remove organic matters other than synthetic polymers, wet peroxide oxidation (WPO) method was predominantly used [31,33,42,43,45,46,48]. Reaction temperature and time varied depending on the concentration of organic matter. For some effluents, microplastics were isolated by simple filtration without any chemical treatment such as WPO [33,45,47,48]. Staining microplastics using fluorescent dyes such as Nile red are suggested for better detection of smaller microplastics [50-52].

Choosing an appropriate sampling volume is very crucial to obtain reliable number concentration of microplastics especially for analyzing influent samples in which concentration often exceeds 100 particles L^{-1} since identifying plastic particles under infrared spectroscopy is time-consuming. Performing preliminary tests would be helpful to decide an appropriate sampling volume for a given size cutoff. WPO is frequently used for isolating plastic particles from organic-rich water samples. Although it was proven to be reliable [42], this also requires long digestion time and needs further refinement.

OCCURRENCE OF MICROPLASTICS IN STP INFLUENTS AND EFFLUENTS

As summarized in Table 1, occurrence of microplastics in STP influents and effluents was expressed on the basis of the number concentration. Further details such as treatment

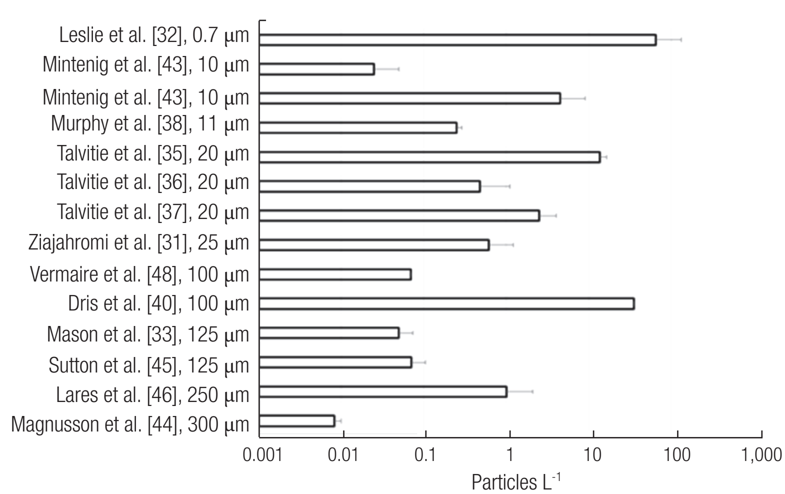

Figure 1. Relationship between the number concentration of microplastics in STP effluents and size cutoff. Mean values from literature are shown with error bars representing standard deviations.

methods of the investigated STPs, year of sampling campaign, sources of the influent are in the web-only Supplementary data file. Although it is difficult to directly compare literature values due to different size cutoffs, the reported number concentrations were not much different among STP influents [32,35,37,38,44,46], ranging between 15.1 and 640 L^{-1}. It is not surprising that the greatest value was obtained when the influent was filtered through a 20 μm filter [37]. Unlike relative invariance among different STP influents, the reported number concentration in STP effluents varied from not detectable to 65 L^{-1}. There is a general trend that the reported median number concentration increases with decreasing size cutoff (Figure 1).

The removal efficiency of STPs could be estimated when the number concentrations in both the influent and the effluent were reported although they are not based on the conservation of mass. It is not surprising that conventional STPs are very efficient for removing microplastics. The calculated removal efficiency was 98.3-99.9% [35,37,38,44,46] except for one study in the Netherlands [32], supporting that microplastics are easily removed during the conventional sewage treatment. Because microplastics are thought to be removed by settling to sewage sludge, recovering microplastics from STP sludge would be important to complete the mass balances of microplastics in STPs. This will help us understand the fate of microplastics entering STPs.

MATERIAL TYPES AND SHAPES OF MICROPLASTICS

The term "plastics" in the polymer industry often refers one of five forms of synthetic polymers, namely fibers, elastomers, plastics, adhesives, and coatings [53]. Plastics are further divided into thermoplastics and thermosets depending on the ease of reprocessing after molten plastics solidify into a shape [54]. However, the term "microplastics" is often used to include all types of anthropogenic polymers [1,55].

Figure 2 describes the relative abundance in percent of material-types of microplastics in STP influents and effluents. Representative thermoplastics (polyethylene (PE), polypropylene (PP), and polystyrene (PS)) and polyester are major materials. The relative abundance agrees with the reported production volumes [56]. PE is the most largely produced plastic material in the world and it has density lower than water [56]. The higher abundance of polyester is characteristic in STPs and is different from the material types of microplastics identified in the oceans and beaches, mainly PE, PP, and PS [55]. Because polyester is used as synthetic fibers in garments, sewage water may contain large amount of micro-sized polyester fibers from laundry effluents [28,57-59]. For example, more than 6 million microfibers may be released from a typical 5 kg polyester fabrics during domestic washing conditions [59].

Figure 3 describes morphology of microplastics in STPs. Fibers forms, mainly from synthetic fibers for fabrics, are the most dominant followed by flakes/fragments. This suggests that microplastics entering STPs are mainly those used as synthetic fibers and fragmented secondary microplastics. Less than 10% are films, pellets, and foams. Because STPs in the United States, Europe, and Australia were studied, further investigation in other regions would provide different abundance patterns owing to different culture and life-styles.

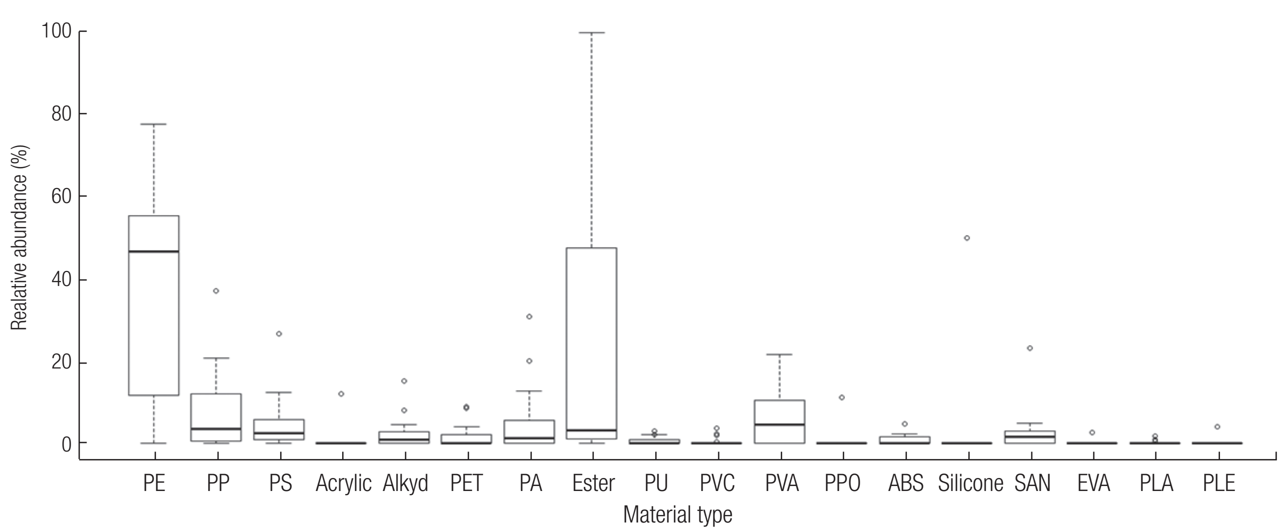

Figure 2. Relative abundance in percent of material-types of microplastics identified in STP influents and effluents. Minimum, 25 percentile, median, 75 percentile, and maximum values from ref 27, 32, 34, 36, and 39 are presented in the box plot. (Abbreviations: PE=polyethylene, PP=polypropylene, PS=polystyrene, PET=polyethylene terephthalate, PA=polyacrylate, PU=polyurethane, PVC=polyvinyl chloride, PVA=polyvinyl alcohol, PPO=polyphenylene oxide, ABS=acrylonitrile butadiene styrene, SAN=styrene acrylonitrile, EVA=ethylene-vinyl acetate, PLA=polylactic acid, PLE=polyaryl ether).

STP AS SOURCES OF MICROPLASTCS TO FRESHWATER SYSTEMS

As summarized in Table 1, majority of studies on the occurrence of microplastics was from the United States, Europe and Australia. Because plastics are also massively used in the other geographic regions, it is expected that microplastics are widespread in other nations as well. If the volumetric flow rate and the population size are counted, it is possible to estimate the microplastics load per capita to the freshwater systems [27,33,37,38]. Murphy et al. estimated daily discharge of 6.5×10^7 microplastic particles per day from a secondary STP treating 2.6×10^5 m^3 d^{-1} and the population equivalent to 650,000 [38]. Large amount of microplastics daily discharge from STPs in spite of high removal efficiency, which denotes the requirement of further investigation on the contribution of STPs as point sources of microplastics.

Although no peer-reviewed publications were found yet for Asian countries, a few technical reports were accessible. In Korea, 0-2.2 particles m^{-3} were detected in river water [60], which were similar to the level in the United Kingdom and Austria [61]. In one Korean STP, the number concentrations in the influent and the effluent were 1.3-4.6×103 L^{-1} and 0.007-0.022 L^{-1}, respectively, indicating more than 99.99% removal [60]. In Japan, microplastic fibers were detected in STP influent and primary sludge by a research group in National Institute of Environmental Studies [62]. However, no number concentration was reported in this study [62].

STPs are regarded as one of the most important sources of microplastics in public waterways and a few studies quantitatively estimated the microplastic load via STPs [27,35,38]. However, it is still not clear whether STPs contribute predominantly compared to other routes of entrance (e.g., direct input to rivers and lakes, stormwater runoff, dry and wet deposition from air, etc.). Contributions by different routes of entrance warrant further investigations. In addition, published results about microplastics in STPs are mainly from developed countries where most of sewer is treated though STPs. However, percentage of sewage treatment is much lower in developing countries [63]. Thus, microplastic input to freshwater systems might be greater in developing countries because STPs are found to be able to remove microplastics very efficiently [35,37,38,44,46,60]. Another aspect to be considered is that the annual plastic consumption in developing countries is lower than developed countries. Further studies on the occurrence of microplastics in various geographic regions, especially in developing nations, would provide better estimates of global microplastic load to freshwater environment.

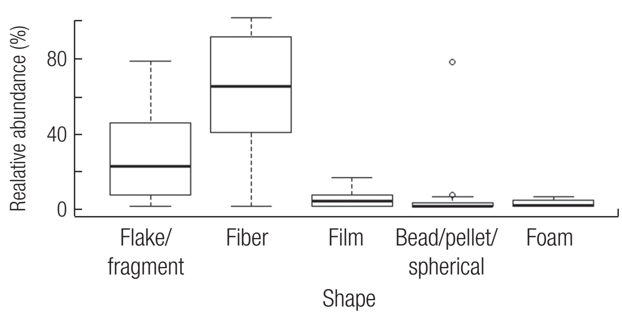

Figure 3. Relative abundance in percent of shapes of microplastics identified in STP influents and effluents. Minimum, 25 percentile, median, 75 percentile, and maximum values from ref 28, 29, 31, 33, 34 are presented in the box plot.

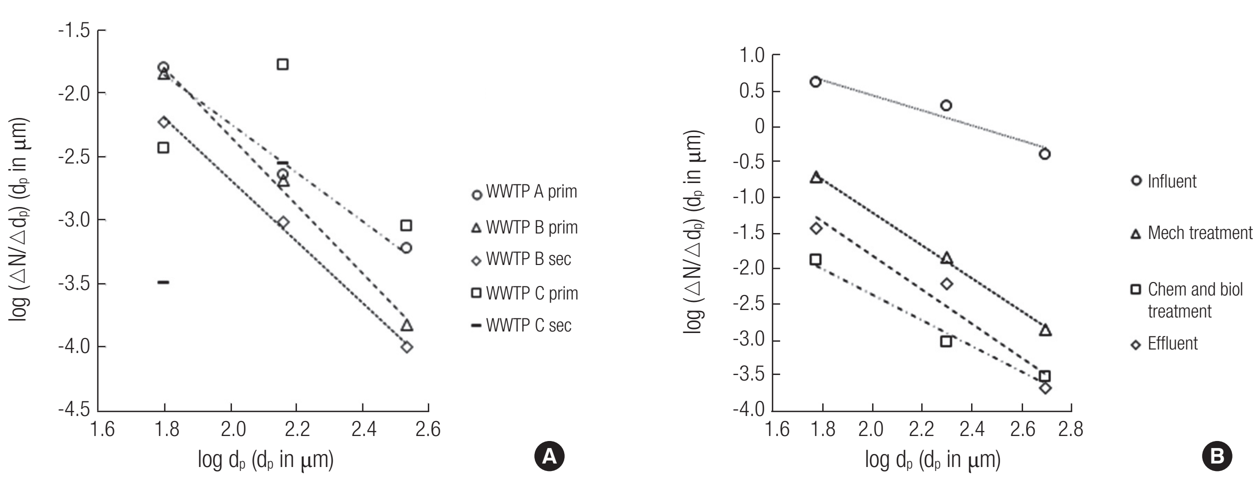

Figure 4. Microplastic particle size distribution in (A) primary and secondary effluents from three wastewater treatment plants (WWTP) by Ziajahromi et al. [31] and in (B) influent and effluents after mechanical treatment, chemical and biological treatment, and final effluent by Talvitie et al. [37]. Dashed lines are best-fit using linear regression.

SIZE DISTRIBUTION OF MICROPLASTICS

As shown in Table 1, researchers have used different size cutoffs for isolation of microplastic particles. Except for primary microplastics that are engineered to small sizes, microplastics are thought to be formed via various weathering processes from bigger plastic products. Although our understanding of these fragmentation processes is very limited, it may be assumed that the fragmentation of plastic particles to smaller ones is a scale-independent process within the size range of microplastics predominantly found (20 μm - 5 mm). Thus, the size distribution of microplastics should follow the power law [64]. Laboratory fragmentation study by Song et al. [65] provides a good support for this hypothesis. However, the size distribution of microplastics isolated from environmental samples did not always satisfy the power law relationship [4,45,66,67]. In a recent study using a novel Nile red staining method, more small-sized microplastics were found and the size distribution followed the power law [52].

Only a few studies reported size distribution of microplastics in STPs using size fractionation [31,37]. Figure 4 describes particle size distribution, log (△N/△dp) versus dp, where N denotes number concentration (particles L^{-1}) and dp is the median size in μm for the number concentration of microplastics reported by Ziajahromi et al. [31] (A) and Talvitie et al. [37] (B). The median values of reported dp were used to draw the size distribution. As shown, the particle size distribution in general follows the power law although the slopes were obtained from only 3 points. Interestingly, the slopes in Figure 4 did not vary much (-2.68 ~ -1.92 in Ziajajromi et al. [31] and -2.39 ~ -1.08 in Talvitie et al. [37]) among different STPs in two studies. It is also noteworthy that the slopes of the size distribution curve are smaller than those obtained in batch tests by Song et al. (-4.57 ~ -2.74) [65]. This might be because of the enhanced aggregation and removal of smaller plastic particles in the real environment that do not occur in a batch test.

The slope in the size-distribution of microplastics is important for the practical purpose of comparing experimental data using different size cutoffs. It is allowed to compare reported concentration data with different size cutoffs, if the size-distribution of microplastics in STPs or in other environmental media follows the power law with a specific exponent. It is also important to know the size distribution over which the power law is applicable. Studies on the size distribution of microplastics in environmental media warrants further investigation to estimate the current number and mass of microplastics in the environment.

CONCLUDING REMARKS AND FUTURE RESEARCH NEEDS

Recent investigations on microplastics in STPs show that microplastics are ubiquitously found all over the world. Although the occurrence of microplastics was investigated only in limited regions, secondary microplastics and synthetic fibers originated from garments are major source of microplastics in STPs [28,46,49]. Conventional STPs under typical operation conditions are found to be removing microplastics from their influents. However, the occurrence of microplastics in STPs in other geographic regions are needed to study for a better global estimation of microplastics load to freshwater systems. Further studies on the size distribution of microplastics in STPs are also needed to understand the fate of microplastics in STPs and to compare results from different studies using different size cutoffs.

CONFLICT OF INTEREST

The authors have no conflicts of interest associated with the material presented in this paper.

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