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## Abundance of Microplastics in Wastewater Treatment Sludge

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## Abstract

Wastewater treatment plants (WWTPs) have been regarded as feasible interceptors of microplastics, which have now contaminated all spheres of the environment. The microplastics removed from WWTPs are frequently trapped in the sludge produced. This article aims to examine the abundance of microplastics in the sludge of WWTPs in different regions by reviewing the relevant scholarly papers published between 2016 to 2022. The papers were retrieved from three databases, namely Web of Science, Scopus, and ScienceDirect. WWTPs receive influents containing microplastics whose abundance differs significantly in relation to the population density, urbanization level, and land use of the areas served. Depending on the water treatment strategies and the efficiencies of the WWTPs, a large proportion of the microplastics in the influents is trapped in the sludge, giving a common sludge-to-influent abundance ratio of 0.02 to 0.77, though a ratio of 3.4 has been reported for recycled activated sludge. Sludge treatment affects the final abundance of microplastics therein. However, a relationship between the abundance of microplastics in influents and that in sludge cannot be established. The reutilization of sludge as fertilizer introduces microplastics back into the environment and significantly compromises the potential of WWTPs as interceptors and sinks of microplastics. This review calls for strategies and technologies to reduce, if not remove microplastics in the sludge of WWTPs. It suggests that future research could take this path to optimize the role of WWTPs as sinks of microplastics.

Keywords: Influents; Microplastics; Population; Sinks; Sludge; Urbanization.

## **1. Introduction**

Microplastics have now been found in every compartment of the environment [1, 2]. Figure 1 shows that plastic waste and primary microplastics are released in all regions of the world, leading to their ubiquity in the environment (Figure 1). Water treatment plants have been considered a feasible way of removing microplastics from the environment [3]. Water treatment plants can be broadly categorized into two types, namely wastewater treatment plants (WWTPs) which treat industrial, agricultural, or domestic wastewater, as well as drinking water treatment plants (DWTPs) which produce water of drinking quality from surface water, groundwater, or seawater [4, 5]. WWTPs are further categorized into industrial WWTPs treating industrial waste streams to an acceptable quality for final discharge and sewage treatment plants (SWPs) treating domestic or municipal wastewater generated, for instance, from residential, commercial, and administrative premises [6]. In some instances, SWPs might receive pretreated industrial waste streams contaminated by fertilizers, pesticides, crop residues, and animal excreta, which are high in chemical and biological oxygen demands [7]. Their designs could be similar to industrial WWTPs or purpose-built to lower operational costs and cater for certain types of waste streams [8].

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Figure 1. Amounts of plastic waste and primary microplastics generated worldwide [9]

In most cases, WWTPs receive influents with significantly higher pollutant loading than DWTPs, and the microplastics levels in the influents of industrial WWTPs are frequently higher [10, 11]. Since the abundance of microplastics is somehow correlated to human activities and WWTPs are built to treat wastewater generated by various human activities, the microplastics in the wastewater bound for WWTPs are expected to be more concentrated than the natural waterbodies feeding the DWTPs [12, 13]. Typically, WWTPs comprise primary treatment, secondary treatment, and optional tertiary treatment [7]. A pre-treatment unit, consisting of course screens and grit chambers, could come before primary treatment to remove large objects such as stones and plant residues from the influent [7]. The influent cleared of large debris and floatables then undergoes primary treatment involving gravity separation of solids in primary sedimentation tanks, with or without skimmers to strip off floating films and fine solids [7]. The primary effluent is subsequently subjected to secondary treatment where soluble and suspended organic matters are removed via biological processes involving the degradation of the organic matters by microorganisms to simpler compounds such as carbon dioxide and water [6]. Biological treatment can be achieved through trickling filters, activated sludge processes, oxidation ponds, and rotating biological contactors. A trickling filter is fundamentally a stone-filled bed over which wastewater is sprayed. Organic matters in the wastewater are taken up and degraded by the microorganisms gathering on the stones, resulting in the reduction of biochemical oxygen demand. After passing through a trickling filter, the wastewater is channeled to a secondary clarifier to remove the microorganisms [6].

Activated sludge system employs an aeration tank instead of a filter bed where compressed air is diffused from the bottom of the tank and rises to the surface, providing oxygen and the mixing required for biodegradation of dissolved organics by activated sludge therein [4]. The mixture then flows to a secondary clarifier for separation of activated sludge, part of which is recycled back to the aeration tank [4]. Oxidation pond, on the other hand, retains wastewater in shallow ponds. The growth of microorganisms, algae and the presence of sunlight facilitate the breakdown of organic matters in the wastewater, leaving sludge deposits in the ponds [4]. Rotating biological contactor is another alternative of secondary treatment involving the use of a horizontal shaft attached with large plastics where bacteria grow. The bacterial films metabolize the organics upon contacting the primary effluent [8]. Tertiary treatment is optional to produce cleaner final effluent and the technologies used range from granular media filters, membrane filtration, nitrification-denitrification, ozonation, chlorination-dechlorination and ultraviolet irradiation, depending on the purposes of treatment [8].

Microplastics are progressively removed through the water treatment processes, with primary and secondary treatments capable of removing a combined 78.1 to 99.9% of microplastics depending on the technologies used [10]. However, a microplastics removal of 99.9% is rarely achieved with only primary and secondary treatments alone, unless tertiary treatment is also incorporated [14]. Most of the microplastics are removed through sedimentation processes in the primary and secondary treatments which generate the raw and activated sludge respectively [15]. As only a fraction of the activated sludge is recycled, most of the sludge is bound for disposal, thus reintroducing the

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microplastics back to the environment [16]. Prior to disposal, the sludge is treated to reduce the weight and volume, hence the transportation and disposal costs. Sludge treatment typically consists of thickening, dewatering and digestion either aerobically, anaerobically or by composting [17]. Treated sludge called biosolids are disposed to landfill, incinerated or reused as fertilizer [18]. Their disposal and reuse reintroduce microplastics back to the environment and counteract the feasibility of WWTPs as interceptors of microplastics [19].

Current studies on the prevalence of microplastics in sludge are regional. For instance, Mahon et al. examined the microplastics in the sludge samples of WWTPs in Ireland treated with different methods [17]. Microplastics in the sludge samples from WWTPs in China have also been profiled [20, 21]. Zhang et al. quantified two types of microplastics in the sludge of sewage treatment plants in the United States [22]. Corradini et al. revealed a correlation between the frequency of sludge application on agricultural soils and the microplastics levels of the soil samples [23]. This was echoed by van den Berg et al. who found the microplastics loads of agricultural soils in Spain with sludge added were significantly higher than those without sludge added [24]. Multiple studies have pointed to sludge as a source of microplastics in soils and there are studies examining the contents of microplastics in regional sludge samples. However, there is a lack of review of the microplastics in sludge samples in different regions. This review aims to compare the abundance of microplastics in the sludge samples of WWTPs in various regions of the world and the sludge-to-influent ratios of microplastics abundance of the WWTPs.

## 2. Method

This review retrieves articles from three main journal databases namely Web of Science, Scopus and ScienceDirect related to the prevalence and abundance of microplastics in the sludge of WWTPs [19, 25, 26]. Keywords such as prevalence and abundance, coupled with microplastics and sludge were entered into the search engines of the respective databases. For an article to be included in the review, it must be published between year 2016 and 2022 and it must contain data related to the abundance of microplastics in sludge samples taken from WWTPs. As this review focuses on WWTPs, articles on microplastics in the sludge of DWTPs were excluded. The articles included should be peer-reviewed and written in English (Figure 2). Information related to the concentrations of microplastics, as well as the daily abundance of microplastics was not available, it was calculated from the concentrations of microplastics of the WWTPs were used in the calculation where water treatment rates during the study periods were not provided. The ratios of microplastics in sludge to those in influents were calculated to provide an indication of the amounts in microplastics trapped in sludge in comparison to those received during wastewater treatment.



Figure 2. Flowchart of the article selection process for review

## **3. Results and Discussions**

Table 1 and Figure 3 show highly variable average microplastics concentrations in the influents of the WWTPs in different countries ranging from 0.28 number of microplastics per liter of influent (n/L) in China to 910 n/L in the Netherlands. The significant variability of microplastics concentrations was also observed among WWTPs influents in

the same countries, e.g., 68 n/L versus 910 n/L in the Netherlands, and 57.68 n/L versus 686.7 n/L in Finland. The levels of microplastics in the influents may be affected by a number of factors, particularly population density and degree of urbanization of the areas serviced by the WWTPs [27]. Areas of high population density and urbanization tend to produce waste streams containing more microplastics [12]. Zhang et al. also revealed that tertiary industry is a major contributor of microplastics in the influents of WWTPs. Tertiary industry, otherwise known as the services sector, comprises hotels, financial institutions, education centers as well as food and beverages sector [28]. Zhang et al. reported a potential correlation between tourist activities and microplastics levels in waste streams where the presence of more tourists increased the activities of the tertiary industry, resulting in more microplastics generated by the industry [28].

_	Average microplastics concentration in influent (n/L)	Average microplastics concentration in sludge (n/kg)	Average microplastics in influent (n/day)	Average microplastics in sludge (n/day)	Ratio of microplastics in sludge to influent	Wastewater treatment rate (L/day)	Country	Ref.
	1.00	1,000	1,510,000,000	1,090,000,000	0.72	1,510,000,000 (capacity)	United States	[15]
	587.00	Not specified	79,832,000,000	Not specified	Not available	136,000,000 (capacity)	United States	[29]
	879.00	Not specified	19,953,300,000	Not specified	Not available	22,700,000 (capacity)	United States	[29]
	585.00	Not specified	8,190,000,000	Not specified	Not available	14,000,000 (capacity)	United States	[29]
	686.70	186,700	193,649,400,000	151,000,000,000	0.77	270,000,000	Finland	[30]
	68.00	660	Not specified	Not specified	Not available	Not specified	Netherlands	[31]
	910.00	510	72,800,000	Not specified	Not available	80,000	Netherlands	[31]
	31.10	14,900	14,040,000,000	3,506,849,315	0.25	493,000,000	Canada	[32]
	57.68	170,965	620,982,880	460,000,000	0.74	10,766,000	Finland	[33]
	29.85	14,895	605,726,028	298,849,315	0.49	35,000,000 (capacity)	South Korea	[34]
	16.45	9,655	1,309,397,260	585,315,069	0.44	110,000,000 (capacity)	South Korea	[34]
	13.87	13,200	1,496,575,340	732,630,138	0.49	130,000,000 (capacity)	South Korea	[34]
	79.90	404,300	1,598,000,000	Not specified	Not available	20,000,000	China	[35]
	0.28	2.30	33,600,000	1,940,000	0.06	120,000,000	China	[35]
	2.50	113,000	1,000,000,000	3,400,000,000	3.4	400,000,000	Italy	[36]
	11.80	7.91	566,400,000	12,165,580	0.02	48,000,000	Australia	[27]
	92.00	56,300	11,960,000,000	5,067,000,000	0.42	130,000,000	Australia	[37]
	98.00	51,200	6,370,000,000	3,655,680,000	0.57	65,000,000	Australia	[37]
	55.00	48,500	8,250,000,000	4,850,000,000	0.59	150,000,000	Australia	[37]
	16.00	2,920	4,800,000,000	315,000,000	0.07	300,000,000	China	[38]
	171.00	133,000	4,788,000,000	2,191,780,820	0.46	28,000,000	Spain	[39]
	126.00	36,300	75,600,000,000	21,205,479,500	0.28	600,000,000 (capacity)	China	[40]
	8.72 (shared	6 908	1.264.400.000	Not specified	Not available	100,000,000	China	[28]
	influent)	0,200	1,201,100,000	The specifica		45,000,000	China	[28]
	1.70	2,190	170,000,000	Not specified	Not available	100,000,000	China	[28]
	0.70	234.7	28,000,000	Not specified	Not available	40,000,000	China	[28]
	533.00	1,401,000	165,784,320,000	267,983,280,000	1.6	311,040,000	Sweden	[41]
	77.00	26,300	9,240,000,000	Not specified	Not available	120,000,000 (capacity)	Thailand	[42]
	72.60	32,000	29,000,000,000	3,222,000,000	0.1	400,000,000	Turkey	[43]
	Not specified	10,900	Not specified	Not specified	Not available	2,500,000	Mauritius	
	Not specified	2,575	Not specified	Not specified	Not available	38,000,000	Mauritius	

Note: In some articles, the wastewater treatment capacities of the WWTPs are provided instead of the wastewater rates during the sampling periods. Therefore, the word 'capacity' is stated in bracket to differentiate the wastewater treatment capacities from the water treatment rates.



Figure 3. Abundance of microplastics in the influents of WWTPs by country

The concentrations of microplastics in the sludge of WWTPs also varied considerably from as high as 1,401,000 number of microplastics per kg of dry sludge (1,401,000 n/kg) in Sweden to as low as 2.3 n/kg in China (Table 1 and Figure 4). The highest sludge microplastics concentration recorded in China was 404,300 n/kg (Figure 4) which shows a significant contrast to the lowest sludge microplastics concentration recorded there (2.3 n/kg), indicating high variability even for WWTPs in the same country. The variability is likely attributed to the wastewater and sludge treatment processes employed, their efficiency and partly due to the microplastics concentrations in the influents. The conventional water treatment process of a WWTP in Italy consisting of pre-, primary, secondary and tertiary treatments yielded dry sludge with 113,000 n/kg microplastics [36]. However, a WWTP employing primary and secondary treatments in the United States produced sludge which contained only 1,000 n/kg microplastics with most of the microplastics in the influent removed during the primary skimming of floating solids [15]. To further illustrate the differences in the abundance of microplastics in sludge, two WWTPs adopting similar treatment processes, namely one consisting of primary clarifier, A<sup>2</sup>O and secondary clarifier in China [35] and another consisting of primary settling tank, A<sup>2</sup>O and secondary settling tank in Korea [34], produced sludge with microplastics abundance of 404,300 n/kg and 14,895 n/kg respectively. In some instances, primary settlement alone yielded sludge with higher microplastics abundance than that from both primary and secondary settlements [32, 34]. Besides, the doses and types of flocculants used during primary and secondary settling can potentially affect the number of microplastics removed from wastewater, hence their abundance in the sludge formed [44]. Therefore, it is challenging to single out a major factor causing the widely differing levels of microplastics in sludge. Nonetheless, each kg of sludge generally contains more microplastics than each L of wastewater [14].



Figure 4. Abundance of microplastics in the sludge of WWTPs by country

While the abundance of microplastics in influent may be one of the factors contributing to higher abundance of microplastics in the sludge of a WWTP, Figure 5 reveals a very weak relationship between the abundance of microplastics in influents and that in sludge ( $R^2 = 0.1549$ ). This means that higher concentrations of microplastics do not always lead to higher removal of microplastics and their entrapment in sludge. In addition, the microplastics removal efficiency of WWTPs may partly account for the amount of microplastics trapped in sludge, and removal efficiency may vary considerably between WWTPs. Calculation of the ratios of microplastics abundance of sludge to that of influents showed a wide range of 0.02 to 3.4 (Table 1). A value higher than 1 indicates that sludge contains more microplastics than influents on a daily basis and this seems to imply that it is possible for more microplastics to be trapped than channeled into WWTPs. In fact, activated sludge had been recycled in a WWTP with a ratio > 1, resulting in repeated exposure to different batches of influent [36, 41]. The estimated activated sludge generated daily was 30 tons (dry weight) but with recycling, the activated sludge might have been used over an extended period. In other WWTPs reviewed, the ratios were mostly < 1, indicating that the daily abundance of microplastics in sludge was less than the daily abundance of microplastics in the influents. Some sludge trapped more microplastics than the others, thus, aligning with the observations that the abundance of microplastics in the sludge of WWTPs varies considerably and the microplastics removal efficiency of the WWTPs also varies. However, it is also possible that sludge treatment may lead to degradation of microplastics in the dry sludge. Mahon et al. reported potential decline in the abundance of microplastics in anaerobically digested sludge while lime stabilization sheared microplastic particles into smaller sizes, thus increasing the abundance of small-sized microplastics in sludge [17].



Figure 5. Relationship between the abundance of microplastics in influents and that in sludge

The use of treated WWTPs sludge or biosolids as agricultural additives reintroduces the microplastics trapped in the biosolids back into the environment. Norway and Ireland are known to utilize 82% and 63% of the biosolids generated from WWTPs, whereas the Netherlands and Korea incinerated 99% and 55% of the biosolids, respectively [18]. The contamination of agricultural soil with microplastics leads to the contamination of crops and agricultural products, and the entry of microplastics into food chains [2, 16]. It was estimated that sludge reutilization had resulted in the entry of 3,500 microplastic particles per kg of agricultural soil [23]. Therefore, removal of microplastics from sludge dictates the future research in reutilization of sludge from WWTPs.

## 4. Conclusion

WWTPs act both as interceptors and sources of microplastics because they remove microplastics from waste streams through water treatment processes, particularly settling and sedimentation. It has been reported that primary sedimentation alone could remove most of the microplastics. The microplastics removed from sedimentation are trapped in sludge and are returned to the environment when the sludge is used as fertilizer. The abundance of microplastics in the influents of WWTPs varies considerably and is influenced by factors such as the extent of urbanization, population density, and land use of the areas served by the WWTPs. As WWTPs employ different

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wastewater treatment strategies, they tend to have different efficiencies in removing microplastics from the waste streams. Distinct treatment strategies and facilities, as well as microplastics removal efficiencies, result in a highly variable abundance of microplastics captured by the sludge formed. Recycling of activated sludge for repeated secondary wastewater treatment could contribute to elevated levels of microplastics in the sludge. No particular relationship has been observed between the abundance of microplastics in the influents of WWTPs and that in the sludge generated from WWTPs needs to be properly treated to reduce the microplastics content as much as possible if it is intended for reutilization. It brings attention to the current sludge treatment of WWTPs, which lacks the efficiency to significantly reduce or eliminate the microplastics content. As a large proportion of the microplastics in waste streams is retained in sludge, and sludge has been applied to agricultural soil as fertilizer in some countries, it calls for more studies into the removal of microplastics from sludge effectively to reduce soil pollution with microplastics.

## **5. Declarations**

## 5.1. Data Availability Statement

No new data were created or analyzed in this study. Data sharing is not applicable to this article.

#### 5.2. Funding

The author received no financial support for the research, authorship, and/or publication of this article.

#### 5.3. Institutional Review Board Statement

Not applicable.

### 5.4. Informed Consent Statement

Not applicable.

## 5.5. Declaration of Competing Interest

The author declares that there is no conflict of interests regarding the publication of this manuscript. In addition, the ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/or falsification, double publication and/or submission, and redundancies have been completely observed by the author.

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