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Municipal sewage sludge as a source of microplastics in the environment

Charles Rolsky[1,2], Varun Kelkar[1,3], Erin Driver[1,3] and Rolf U. Halden[1,3]

Abstract

Wastewater treatment plants are known to contribute to microplastic (MP) pollution in freshwater and terrestrial environments, but studies on MP abundance in sludge are scarce. This study aimed to (i) conduct a literature review to assess the number and extent of MPs in sludge worldwide, (ii) determine extraction and analytical techniques used to isolate and identify these materials, and (iii) assess the fate and transport of these materials in the environment as a result of sludge disposal and reuse. Research in this area has increased as 12 countries have now reported and quantified MPs in sludge. This study highlights the need to assess the temporal and spatial differences in MP pollution in sludge, this relationship to land-applied biosolids, and the risk to human and ecological health.

Addresses

[1] Biodesign Center for Environmental Health Engineering, The Biodesign Institute, Arizona State University, 1001 S. McAllister Avenue, Tempe, AZ, 85287-8101, USA

[2] School of Life Sciences, Arizona State University, 427 E Tyler Mall #320, Tempe, AZ, 85281, USA

[3] School of Sustainable Engineering and the Built Environment, Arizona State University, 660 S. College Avenue, Tempe, AZ, 85281, USA

Corresponding author: Halden, Rolf U. (Rolf.Halden@asu.edu)

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Microplastics, Sewage sludge, Plastic pollution, Biosolids, Wastewater treatment plant.

Introduction

Plastic pollution is a well-documented threat to ecosystems around the world, ever increasing as plastic production intensifies [1]. Microplastics (MPs), in particular, have emerged as a source of concern because of their small size (<5 mm) and interactions with dangerous contaminants [2]. These small plastics are now being discovered in sewage sludge around the world. Despite their high removal ratio in some cases (84–99%), differences in water treatment strategies among wastewater treatment plants (WWTPs) and characteristics of the MPs still allow for a sizable amount of small plastics to pass through the plants within solid materials or sludge [3]. Sludge once treated, termed biosolids, is sent to a landfill, incinerated or land applied, thus providing three pathways for MPs to enter the environment. Once in the environment, MPs present health threats to a variety of biota if ingested, sometimes negatively affecting factors such as species growth and reproduction [3]. Many countries use biosolids as an agricultural amendment and MPs have been found in soil which received biosolids from WWTPs, prompting more attention to the role of WWTPs as contributors to the release of environmental MPs as in some cases, MPs are able to leach from soils into the environment [4]. In addition to damage inflicted by MPs to the wastewater treatment process, such as inhibition of sludge hydrolysis and reduction of important microorganisms, their environmental implications present even more significant dangers [5,6]. Chemical and physical threats have been associated with MPs because of their hydrophobicity and chemical composition [7]. The degradation of MPs can trigger the release of both manufactured additives in plastics (e.g. phthalates) and adsorbed contaminants (e.g. persistent organic pollutants) which can concentrate on the high MP surface area, up to a million times stronger than levels within the surrounding environment [7,8]. If ingested, the distribution and toxicity of chemical contaminants may increase and concentrate up the food chain, threatening humans and animals similarly [8]. Environmental exposure can occur directly as primary MPs, where manufacturing creates these sizes for a particular use (e.g. microbeads for cosmetic purposes), or as secondary MPs that are the results of larger plastics fragmenting into the target size range over time [9].

Owing to these factors, research chronicling the presence of MPs in sludge has increased. New information has emerged aiming to better understand the existence of MPs in sludge, including morphological characteristics, their fate after water treatment, and suggested impact on the environment. These plastics have been shown to leach from landfills, linger within agricultural soils, and contribute to atmospheric pollution once incinerated, suggesting that the presence and fate of MPs in sludge must be further studied [4,10,11]. Thus, we aim to assess the literature related to MPs in sewage sludge globally, including reported numbers, extraction and analytical methods, and fate and transport in the environment. In addition, this review offers suggestions for future research to improve the study of sludge-borne MPs.

Quantification of MPs in sludge

Fourteen articles collected and quantified MPs in biosolids, which include data from 12 different countries (Table 1). Typically, MPs are reported within sludge by particle number per unit mass and reported particle numbers vary considerably between locations. For instance, the Netherlands had the lowest particle counts at 0.45 ± 0.2 MP g^{-1}, whereas Italy reported the highest at 113 ± 57 MP g^{-1}. Countries that have been surveyed for MPs in sludge cover a wide range of populations, which likely contribute to the variability seen between samples.

Particles were characterized by particle type using 16 different classifications of MP morphological descriptions, the most common label being fiber (100%), followed by fragment (71%), and sphere (35%) (Figure 1a). A fraction of the shapes reported had rather ambiguous names, which were up for interpretation, such as ‘line’ or ‘shaft’. These nonstandardized shape delineations could hinder future comparisons.

In some cases, MPs in sludge were organized by size, that is, >500 or <500 μm [12] or average count, but total dimensional data were not reported. All published articles report MP particle counts, which do not provide information on total weight of the MP particles within that particular environment. As there can be a 20 times difference between the smallest and largest ‘MPs’, reporting only particle number does not allow for meaningful direct comparisons between locations from different studies. With varying abundances, the mass of MP (μg/g of biosolids) should be considered as a reporting unit, rather than particle number [13]. Without being able to directly compare, researchers may be losing the ability to assess broad-scale occurrences related to MPs, which may provide information pertinent to ecosystem health.

Extraction and identification methods of MPs

Extraction protocols for MPs in sludge are varied and often use a combination of methods [14–18], including the mixing of sludge with high-density solvent, such as sodium chloride or zinc chloride [10,19] for buoyancy separation, followed by capture via sieves or vacuum filtration [19]. The particular laboratory practices used for extraction and quantification were a function of research goals. For instance, Carr et al. [14] chose a variety of sieve sizes from 400 to 200 μm to isolate a range of possible MP sizes, whereas Zubris and Richards [20], who were specifically seeking out plastic fibers, used a much smaller sieve (0.45 μm) when vacuum filtering their supernatant. Elutriation columns were also used to separate MPs from more dense materials [10]. Nonstandardization of extraction steps may translate into variable MP recovery between methods, which can contribute to differences in MP loading numbers for identical sites. For example, in regards to density separation, each solvent has a different density, which could alter the fraction of MPs reaching buoyancy. Aiming to add solvent enough to reach an optimal density to catch the most common polymers could mean missing out on other plastics whose density is higher than the optimal number. Contamination has been reported in virtually every study chronicling MPs in sludge. Thus, steps must be taken to evaluate this incidental occurrence. For example, researchers have found success integrating blank experiments to assess any airborne microfiber contamination [21].

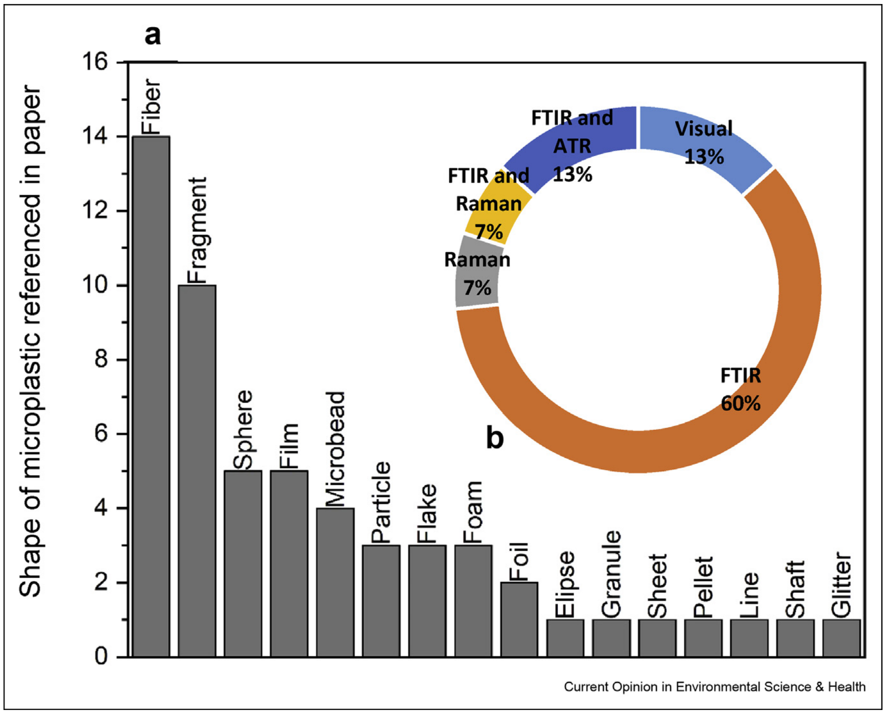
Table 1

Counts of microplastics reported per gram of sludge (dry weight) and associated WWTP data.

Country	Population (million)	Sludge produced (MMT/year)	Average MPs (#/g)	Number of WWTPs sampled
Italy [38]	60	1	113	1
Germany [31]	80	2	40.1 ± 24	6
Finland [22]	5	0.1	27.3	1
Sweden [15]	10	0.2	17	1
Canada [24]	37	0.7	9.65 ± 5.2	2
Ireland [10]	4	0.004	8.5 ± 1.6	8
China [28]	1400	35	8.03 ± 8	29
US [14,20]	332	6	2.5 ± 1.5	2
Korea [27]	77	4	2.2 ± 0.3	3
Scotland [16]	5	0.1	1	1
Norway [46]	5	0.1	0.8 ± 0.4	10
Netherlands [19]	17	0.6	0.45 ± 0.2	3

WWTPs, wastewater treatment plants; MPs, microplastics; MMT, million metric tons.

Figure 1



Reported shapes and identification techniques of MPs in sludge. (a) The number of articles reporting certain classifications of MPs and (b) types of analytical tools used to identify the MPs. *Note FTIR = fourier-transform infrared spectroscopy; ATR = aattenuated total reflection; MPs, microplastics.

After MP extraction, polymers were then searched visually, using a microscopic source and often times, distilled water was added to help break up the organic material and avoid static electricity upon MP extraction [23]. Suspected plastic particles were confirmed most often via Fourier-transform infrared spectrometry (FTIR) (60%), followed by FTIR combined with attenuated total reflectance (ATR) (13%), then by visual identification (13%) (Figure 1b). Raman analysis was also used as a standalone technique (7%) and in conjunction with FTIR (7%). Both FTIR and Raman spectroscopy remain the most popular methods of MP identification [23]. It is often difficult to confirm a particle of interest to be plastic using a microscope alone. Gies et al. [24] found that of all particle of interests initially isolated and extracted using light microscopy, only 32.4% were confirmed to be plastic polymers via FTIR. There are tradeoffs to these analytical methods. With higher numbers of MP particles, it becomes more feasible to analyze a subset of particles allowing for an underestimation of reported numbers. In addition, identification techniques such as ATR–FTIR have reported issues identifying fibers due to the inability to differentiate plastic fibers from natural materials [23]. The attachment of organic materials, the presence of additives, or the use of oil may result in an incomplete match to an FTIR or Raman database, making the identity of the plastic material more difficult to pinpoint [22,24].

Most MPs entering a conventional WWTP are sequestered in sludge. Mahon et al. [10] demonstrated that approximately 99% of MPs can persist in sludge, even after several treatment stages, such as lime stabilization or anaerobic digestion, aimed at degrading organic matter within a conventional WWTP. Another research study found that larger MPs are sequestered in sludge at in higher numbers than smaller particles, while other studies have demonstrated that smaller MPs have an increased chance of remaining in sludge because their size allows them to traverse the treatment processes [10,25]. A range of MP removal efficiencies within WWTPs exist within the literature but these numbers were contingent upon several factors including temporal variations and/or disparities between WWTP practices. Studies have shown removal techniques, such as membrane bioreactor treating and rapid sand filters, have the greatest impact on MP removal, but not all WWTPs use these removal technologies [26].

Seasonality can also play a role in MP variability within sludge. Lee and Kim [27] found that during a three-month period of high precipitation, the amount of MPs in sludge increased. Sociality is also considered to have an effect on sludge MP concentration. In China, for example, an increase in infrastructure and industrial activities were positively correlated with higher concentrations of MPs found in sludge [28]. Li et al. [28] found MP concentration in sludge to be also positively correlated with more infrastructure and increased industrial activity, as well as smaller areas of afforested land. There are variations in the amounts of MPs in sludge, some suggest differences in seasonality, urbanization, and treatment processes play a role, but better geographical data coverage are needed to better understand how these and likely other processes contribute to the accumulation of MPs in sludge.

A variety of analytical methods are used when seeking to isolate, extract, and identify MPs in sludge. Density separation is very commonly used to isolate MPs, and FTIR remains the most common method of identification. The presence of MPs in sludge is not surprising as they have been shown to survive multiple removal stages and degradative mechanisms therein. The amount of MPs discharged from WWTPs in sludge can be influenced by several factors, including seasonality and urbanization.

Pathways and mechanisms of exposure

Treated sewage sludge or biosolids have a range of endpoints including, but not limited to, beneficial reuse as agricultural amendments and soil composting, as well as disposal mechanisms, including landfilling and incineration (Table 2). All three of these disposal paths present opportunities for sludge-borne MPs to penetrate the environment. Specific MPs sent for disposal via incineration are destroyed; however, harmful contaminants such as dioxins and polychlorinated biphenyls can be emitted during their destruction [29]. It is thought that MP disposal by landfilling should sequester this material; however, MPs have been found in landfill leachate, with the ability to migrate into groundwater and disrupt freshwater ecosystems [10,30]. Biosolids, when land applied, increase soil fertility, create more favorable soil properties, and contribute to the ability of the soil to recycle nutrients, owing to the addition of nutrients such as sulfur, magnesium, and sodium, present in the material [31,32]. MPs have been found within soil that was the recipient of sludge application and were also shown to undermine the positive aspects of biosolids by negatively affecting the water holding capacity, microbial activity, and the bulk density of soils [33]. Owing to their ability to survive microbial assimilation, MPs delivered via biosolids can spend years accumulating on land in high numbers, between 125 and 850 tons MP/million inhabitants are added annually to European agricultural soils alone [35]. Atmospheric circulation is thought to aid in the remobilization of MPs away from fields, with shapes such as fibers, which have a lower removal efficiency than other MPs, penetrating porous soils more easily, suggesting a mechanism of environmental release postland application [6,20,57]. Some studies have suggested it is unlikely that MPs in soil will undergo relevant disintegrating or degradation but much is still unknown regarding the movement or weathering of MPs within agricultural soils [33].

Chemicals linked to the presence and degradation of MPs have also been shown to pose a serious threat. For example, plasticizers, which are emollient additives to the plastics, have been linked to endocrine disruption in several animal species [34]. Many studies have demonstrated the dangerous interaction between MPs and surrounding contaminants. Toxic chemicals such as polychlorinated biphenyls have been shown to attach to the surface of MPs because of their mutual hydrophobicity [35]. Thus, WWTPs present an opportunity for this interaction to intensify because of the presence of contaminants such as heavy metals or persistent organic pollutants [36]. Studies have even shown MPs to act as reservoirs for antibiotic resistant genes, which may have

Table 2

Reported percent fraction of biosolids usage type per country. The remaining biosolids use described as ‘other’. Total land application includes both agriculture and soil/compost.

Country	Total land application	Agriculture	Incineration	Landfill	Soil/compost
Finland [47]	94	5	0	3	89
Norway [48]	82	82	0	0	0
Scotland [16]	64	24	35	1	40
Ireland [49]	63	63	0	35	0
Sweden [50]	63	36	2	22	27
Korea [39]	0	0	55	0	0
US [51]	55	55	15	28	0
China [52]	45	45	4	35	0
Canada [53]	43	43	47	4	0
Italy [54]	27	1	6	17	26
Germany [55]	48	38	18	34	10
Netherlands [42]	0	0	99	0	0

dangerous ecological implications after release [56]. While in the treatment plant, MPs encounter physical and chemical degradative processes which can contribute to the adsorption of dangerous contaminants. For example, MPs have been found to exhibit a brittle surface, after treatment, along with an abrasive and 'hackly' surface, confirmed via scanning electron microscopy (SEM) [28]. These weathered MPs often have a negative charge, thus, have been shown to preferentially sequester heavy metals. Little is known about how degradative changes in the structure of MPs affects the efficiency of these materials in transporting chemical and microbiological contaminants. Kelkar et al. [37] found that during chlorination in the WWTP, the plastics' chemical structure can change, thereby increasing its toxicity.

Land-applied biosolids are an important use for many countries, which is a function of the regulations or laws in that given location (Table 2). Every country with reported MPs in sludge uses land application or landfilling of biosolids. Korea and Finland rely heavily upon composting, whereas Canada, China, and the US use approximately half of their biosolids for agricultural purposes [38–40]. Finland also reported one of the highest concentrations of MPs in sludge. Combining this information with their total use of land-applied biosolids presents a dangerous opportunity for large amounts of MPs to enter the environment and accumulate up the food chain [35]. Netherlands is an anomaly as around 99% of their biosolids are incinerated because of concerns over the presence of heavy metals but their reported number of MPs in sludge were the lowest of all countries surveyed [41,42]. China improperly disposes 80% of their total sludge, effectively increasing the total amount of 'land applied' biosolids and MPs therein [43]. The relationship between MP sequestration in sludge and the subsequent application of biosolids for agricultural purpose is very important to understanding the loading of these polymers in different environments and the resulting ecological effects of these practices. The fate of the MPs, once land applied, is not well understood. Studies have shown that plastic particles were identifiable in the soil column over 15 years after the initial application and it has also been suggested that they can last up to 100 years because of reduced light and oxygen, conditions which in higher amounts are normally associated with the degradation of MPs [20,29,44].

Future research directions

There are multiple knowledge gaps and areas of nonconsensus that need to be addressed so that the magnitude of plastic pollution stemming from MPs can be determined. Temporal and spatial trends in MPs must be studied to get a more comprehensive idea of annual MP deposits into sludge and biosolids. An estimate of annual variation of MPs in wastewater and the subsequent ability for WWTPs to adequately handle such flows has yet to be studied but is crucial to better understanding worldwide trends of MPs in sludge or biosolids [23]. Understanding the loading and transport of land-spread biosolids, MPs will shed further light upon the transfer of terrestrial MPs to freshwater ecosystems [10]. In addition, little is known about the ability of MPs to sequester and transport chemical and microbiological pollutants (including pathogens) across the landscape. Treatment plants contain a variety of harmful contaminants and pathogens, but the ability of MPs to adsorb them throughout all stages of the treatment process and thereafter are not well understood. Finally, there must be a standardization of reporting units for MP concentration. The particle size range of MPs varies from 100 nm to 5 mm, thus units of mass are the most accurate representation of MP contamination within a given sample, which would in turn allow for more efficient comparisons between sampling locations [45]. In addition, consensus on MP nomenclature would also help to identify shape, which will aid in elucidating fate and transport mechanisms.

Conclusion

Sewage sludge from around the world has been demonstrated to contain MPs. As the use of plastics continues to grow worldwide, MPs will only continue to be a problem to human and ecosystem health. Several analytical tools are used to confirm the identity of sludge-borne MPs, and a range of morphological classifications have been used to report them. Standardizing the research methodologies and reporting units could make comparisons between different studies more efficient. With this understanding, we can begin to crucially assess new technologies related to wastewater or biosolid treatment and subsequent biosolid application.

Conflict of interest statement

Nothing declared.

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