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Review

Microplastics in landfill leachate: Sources, detection, occurrence, and removal

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ABSTRACT

Due to the accumulation of an enormous amount of plastic waste from municipal and industrial sources in landfills, landfill leachate is becoming a significant reservoir of microplastics. The release of microplastics from landfill leachate into the environment can have undesirable effects on humans and biota. This study provides the state of the science regarding the source, detection, occurrence, and remediation of microplastics in landfill leachate based on a comprehensive review of the scientific literature, mostly in the recent decade. Solid waste and wastewater treatment residue are the primary sources of microplastics in landfill leachate. Microplastic concentration in raw and treated landfill leachate varied between 0–382 and 0–2.7 items L^{-1}. Microplastics in raw landfill leachate are largely attributable to local plastic waste production and solid waste management practices. Polyethylene, polystyrene, and polypropylene are the most prevalent microplastic polymers in landfill leachate. Even though the colors of microplastics are primarily determined by their parent plastic waste, the predominance of light-colored microplastics in landfill leachate indicates long-term degradation. The identified morphologies of microplastics in leachate from all published sources contain fiber and fragments the most. Depending on the treatment method, leachate treatment processes can achieve microplastic removal rates between 3% and 100%. The review also provides unique perspectives on microplastics in landfill leachate in terms of remediation, final disposal, fate and transport among engineering systems, and source reduction, etc. The landfill–wastewater treatment plant loop and bioreactor landfills present unique difficulties and opportunities for managing microplastics induced by landfill leachate.

1. Introduction

Over 335 million tons of plastic are produced annually globally, making it an indispensable component of modern life [1]. Plastics will be widely used despite initiatives to reduce their usage due to their portability, durability, and low cost. COVID-19 has boosted the usage of plastic in personal protective equipment (PPE), such as facemasks and gloves [2–4]. Plastic garbage enters aquatic and terrestrial areas, threatening ecosystems and possibly endangering biota and humans [5,6]. In the environment, plastics degrade with time through physical, chemical, and biological processes (i.e., hydrolysis, photodegradation, thermal oxidation, mechanical abrasion, and biodegradation) [7]. As plastics deteriorate, their size reduces, and microplastics (MPs) and nanoplastics (NPs) are generated, making the problem less evident but more harmful.

Plastics <5 mm are defined as microplastics, and nanoplastics are plastics with particle sizes ranging in sub-micron scale. Nevertheless, the precise scientific definition of NPs is still under discussion, with the size being described as either 100 or 1000 nm in one dimension. The first detection of MPs was made in oceans [8]. An estimated 5 trillion MP particles are weighing 243,000 tons floating in the water due to MP degradation and deposition, posing

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a lethal threat to marine life [9]. Several investigations have also identified NPs in the marine environment [10]. In addition to the ocean, the abundance of MPs has been identified in every other environmental media, including wastewater, drinking water, surface water, and landfill [1,9,11,12].

The toxic effects of MPs are well documented in the environment [6,13,14]. Due to the increasing concerns about the global impacts of MPs, many studies are going on, and documents are available on the occurrence and treatment of MPs in different environmental systems. Landfills receive a large number of MPs from several primary and secondary sources. Many other pollutants are also present in landfills that can be adsorbed by the MPs. MPs from landfill might infiltrate leachate via rainwater. If not properly engineered, MPs may percolate to groundwater or other terrestrial locations. Landfills have also been identified as a source of MPs input to the marine environment [15]. However, landfill as a contamination source and hotspot of MPs is less explored than other aquatic environments, as current research mostly focused on the fields of water (ocean, surface, and groundwater) and wastewater [16–21]. This study critically reviewed some available scientific literature on MPs in landfill leachate and provides state-of-the-science regarding the source, occurrence, and remediation of MPs in landfill leachate.

2. Source of microplastics in landfill leachate

The two principal sources of MPs in landfill leachate are solid waste and wastewater treatment plant residual, e.g., sludge and fat, oil, and grease (FOG), etc. [22]. Plastic waste dumped in landfills can go through several abrasive activities and produce secondary microplastics. MPs can also enter into landfill from numerous primary sources. Wastewater is also a significant collection point for MPs [20]. MPs can get entrapped in FOG and solid sludge during the treatment process. Upon disposal in landfills, sludge and FOG from sewage can augment the abundance of MPs in landfill leachate [23]. Fig. 1 demonstrates the sources of microplastics in the landfill leachate microplastics from solid waste.

At the end-of-life of plastic products, the best possible and most desired approach is recycling. However, only 15–20% of all plastic waste can be effectively recycled using conventional technologies worldwide, and approximately 21–42% is deposited in landfills [12,24]. In 2018, landfills in the US received 27 million tons of plastic waste [25]. Though some plastic products are marked as "biodegradable" nowadays, their complete breakage is only possible when composted in industrial units specifically designed for polymer's molecular breakdown, the biodegradation of these products in landfills is limited [26]. All the waste undergoes numerous treatment stages in landfills: initial aerobic biodegradation, a transition from aerobic to anaerobic condition, acid formation and hydrolysis, methanogenesis to form methane, and final maturation and stabilization [27]. Each stage accelerates the rate of plastic breakage and produces secondary MPs [28]. Furthermore, different anthropogenic activities associated with mass-produced MPs/NPs, such as microbeads from pharmaceuticals and personal care products (PPCPs), shampoos, shower gels, lipsticks, sunscreens, masks, eye shadows, or other intentionally produced microparticles for particular purposes, often end up in landfills. Waste generated from dedicated industries and facilities that handle these products can also be a source of primary microplastics in the landfill leachate.

2.1. Microplastics from the wastewater treatment plant

Microplastic existence ranges up to 3160 particles L^{-1}, 125 particles L^{-1}, and 170.9 × 10^3 particles kg^{-1} of total solid dry weight (TS dw) in untreated wastewater treated wastewater and sludge, correspondingly [29]. Most MPs got trapped in the FOG flocs during treatment stages and/or settle downs in sludge [30–33]. While low-density MPs get trapped in FOG during skimming, the high-density MPs settle down with sludge [28]. 60–99% of MPs from wastewater sources are detained in the sludge from wastewater treatment plants (WWTPs) [30–33]. From management and materials handling standpoint, landfilling is one of the most straightforward solutions for sludge disposal [34]. Based on the biosolid program report of 2019, around 22% of the sewage sludge is disposed of in landfills [35]. According to the same report, approximately 4.75 million dry metric tons of biosolids were

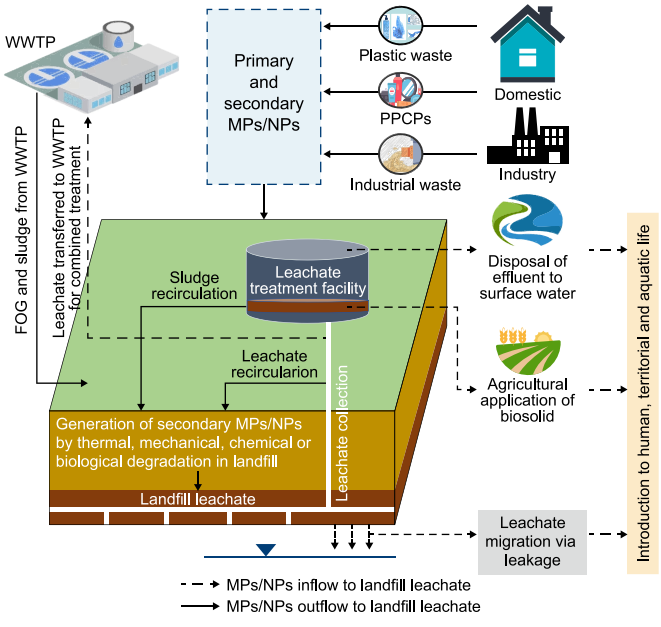

Fig. 1. Source and environmental pathways of microplastics via landfill leachate.

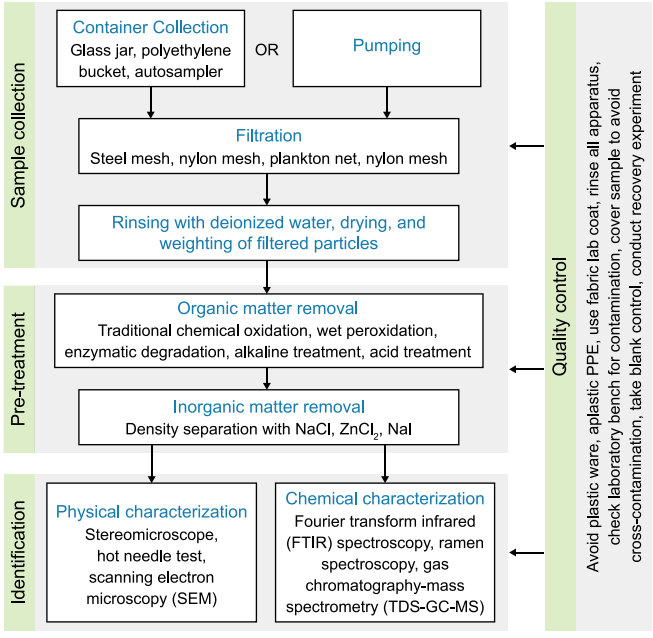

Fig. 2. Flow chart summarizing the analysis steps of microplastics in landfill leachate.

Table 1
Current extraction, quantification, and characterization method of MPs in landfill leachate.

Country	Sampling location	Volume (L)	Sampling technique	Reported MP size range	Filter type and size	Organic matter removal	Inorganic matter removal	Quantification and characterization method	References
China	Collecting well or/and equalization basin	24	PE bucket	100–5000 μm	SS sieves with mesh sizes of 150, 75, 45, and 25 μm 0.45 μm filter paper	WOP: 0.05 M Fe (II) + 30% H_2O_2	DS: NaI	ATR-FTIR,μ-FTIR	[37]
China	-	3	-	-	47 mm and 20 μm Nylon membrane filter	CO: 30% H_2O_2	-	ATR-FTIR,μ-FTIR, stereomicroscope with AxioCam digital camera	[38]
China	Equalization basin, bioreactor, membrane tank, final effluent, rejected water of RO system	1 (raw leachate),40 (treated leachate)	Glass jar	20–100 μm	SS sieves with mesh sizes of 150 μm and 10 μm	WOP: 0.05 M Fe (II) + 30% H_2O_2	DS: $ZnCl_2$	μ-Raman spectrometer, optical microscope	[39]
China	Equalization basin, adjustment tank, membrane bioreactor, ultrafiltration,nanofiltration, and reverse osmosis effluent	5		0.5–5 mm	Mesh sieve of 48 μm,20 μm filter paper	CO: 30% H_2O_2		ATR-FTIR, SEM, stereomicroscope with AxioCam digital camera	[40]
Indonesia	Leachate pond, leachate drain	-	HDPE bottle	80–5000 μm	SS sieves with mesh sizes of 5 mm, 200 μm, and 20 μm 0.45 μm cellulose nitrate filter paper	WOP: 0.05 M Fe (II) + 30% H_2O_2	DS: NaCl	μ-FTIR, microscope with camera	[41]
India	Groundwater	1	Glass bottle	1000–5000 μm	0.45 μm cellulose nitrate filter paper	-	-	ATR-FTIR,SEM, stereomicroscope with camera	[42]
Thailand	Leachate pond			-	SS sieves with mesh sizes of 330 μm	WOP: 0.05 M Fe (II) + 30% H_2O_2	DS: NaCl	FTIR, stereomicroscope	[43]
France	Coast near landfill	1000–2000	Pumping	20–5000 μm	SS sieves with mesh sizes of 500 μm, 200 μm, 80 μm and 20 μm		DS: $ZnCl_2$	μ-Raman spectrometer, stereomicroscope with camera	[15]
Finland	Settlement pond, pump station	10, 80, 120	Pumping	50–500 μm	SS sieves with mesh sizes of 5000, 411, and 47 μm	CO: H_2O		ATR-FTIR	[44]
Finland	Pump station	70, 109	Pumping, PE bucket						
Finland	Leachate pond, outflow pipe	70, 120	Pumping, PE bucket						
Norway	Pump station	20, 5	Pumping						
Norway	Well, pump station	10, 40	Pumping						
Ice land	Borehole, outflow pipe	295, 55	Pumping						
Iceland	Leachate pond outflow pipe	44, 307	Pumping						
Bosnia and Herzegovina	Receiving basin	2.5	Pumping, glass bottle	-	Plankton net of nylon mesh material with pore size 23 μm	WOP: 0.05 M Fe (II) + 30% H_2O_2	DS: $ZnCl_2$	Stereomicroscope	[45]
Serbia	Sedimentation and aeration lagoon	2.5	Pumping, glass bottle						
Serbia	leachate lagoon	2.5	Pumping glass bottle						

Abbreviations: SS, stainless steel; CO, chemical oxidation; WOP, wet peroxidation; DS, density separation.

produced in the US in 2019 [35]. During wastewater treatment, a significant quantity of MPs is retained in sludge. Therefore, landfilling sludge from WWTPs can deliver a substantial number of MPs in landfill leachate. According to the average reported MP concentration in sludge from the US, approximately 2.5×10^{12} items per gram of MPs can be delivered to landfills via WWTP sludge each year (assuming the MP counts from Rolsky et al. [36]). Thus, even though sludge is a sink of MPs for WWTPs, it is an enormous source of MPs to landfills leachate.

3. Detection of microplastics in landfill leachate

The detection of MPs in landfill leachate generally contains three steps, i.e., sample collection, sample pretreatment, and MP characterization/quantification, as summarized in Fig. 2. However, the method applied in each step for the leachate sample is not standardized yet. Studies used techniques according to sample characteristics, available resources, and research goals. Most studies maintained quality control to avoid potential leachate sample contamination and sample loss bias.

3.1. Sample collection

Container collection (polyethylene bucket or glass bottle) is the most reported method in landfill leachate studies (Table 1) due to its straightforwardness. However, collection capacity is limited in container collection practice. Generally, only a few litters per collection event are possible with containers. If the leachate contains high organic matter content and solids, such as the untreated leachate sample, given the ease of the following filtration process, container collection is more appropriate [20]. Autosampler collection is another straightforward method reported in wastewater sample collection for MP analysis and could also be suitable for leachate sample collection. However, it poses the same limitations as container collection [20,33,46]. Depending on the mesh size and leachate characteristics, pumping can effectively increase the sampling volume. With pumping, up to 2000 L of leachate sample collection has been reported [44].

Separate or simultaneous filtration with steel, nylon, or plankton nets is conducted while sampling [46,47]. For leachate filtration, steel meshes are commonly reported (Table 1). Filtration using the different sieves of mesh size between 150 mm and 10 μm has been reported. Plankton nets are also mentioned in some literature [45]. Sample collection with custom-made filtering houses with mashes of varying sizes employing concurrent sample collection and filtration is also a popular technique [8,44]. Applying different series of sieves ensures MP characterization based on their sizes. However, the mesh-based size categorization can be highly biased. Some MP particles might not pass through the sieves even if sufficiently small due to their irregular shapes [46]. Moreover, unique morphology, like fibers, can pass vertically through a small sieve and promote inaccuracy in the outcome [48].

The volume of the collected leachate sample varies between milliliters and liters [38]. The sample volume collected for raw and treated leachate is generally different. A large volume of raw leachate typically contains a high organic load content which may clog filters faster. On the other hand, a higher volume can be collected for treated leachate as it contains fewer pollutants. Moreover, treated leachate is expected to have fewer microplastics. Hence, it is wise to sample larger volumes, especially if a large particle is targeted to be identified [47,49].

A sample might have uneven temporal and spatial distributions of MPs in leachate [20]. A sample's representativeness can be increased by collecting large volumes, taking a 24-h composite sample, adjusting the sampling frequency, and sampling mode corresponding to the goal of the study [33,46,50]. As landfill leachate is a complex liquid, a microplastics sampling guide is highly recommended to be developed for prescribing appropriate sampling mode and frequency depending on the sample characteristics to reduce sampling inaccuracies and increase data quality.

3.2. Pretreatment of sample

3.2.1. Organic matter removal

Landfill leachate is rich in particles or organic suspended matter, interfering with the detection and characterization process. To count and characterize MPs properly, non-plastic material needs to be removed by chemical pretreatment. The organic matter from the sample are mostly removed by conventional chemical oxidation, novel wet peroxidation, enzymic degradation, and acid or alcohol hydrolysis [39,51–53]. Inorganic matter is mostly removed using the density separation technique. Fig. 2 summarizes the conventional pretreatment methods available for leachate samples.

Conventional oxidative treatment is the standard digestion process to remove organic matter. Chemicals, including H_2O_2, NaClO, etc., are usually utilized as oxidizing reagents [54–56]. However, chemicals that can dissolve the biological substance while maintaining the integrity of microplastics are limited for conventional oxidative treatments. Some oxidizing chemicals might react with some particular polymers. So far, H_2O_2 has shown the best performance degrading organic matter without denting considerable microplastics [57]. The traditional procedure is to apply 30% H_2O_2 for the pretreatment to isolate MPs from the sample [38,44]. However, the reaction rate of this H_2O_2 pretreatment process is slow. Therefore, this method might not be practical for samples containing extensive organic matter like landfill leachate due to prolonged treatment time. To accelerate the reaction rate, moderate heat for a short period (30 min) can be applied [58]. Even though utilizing heat for the progression of the conventional chemical oxidization process for leachate study is scarce, the result should be promising for analyzing MPs in leachate. However, applied heat may melt some tiny MPs and alter the findings [58].

National Oceanic and Atmospheric Administration (NOAA) employed a new technique called wet peroxidation (WPO) to increase the efficiency and reduce the pretreatment time of the traditional oxidation process. This method is also widely used to isolate microplastic from samples collected from freshwater, seawater, wastewater, landfill leachate, sediments, and organisms [37,57,59–61]. In WPO, a ferrous catalyst is used along with H_2O_2 for oxidation. NOAA laboratory applied equal amounts (20 mL) of 30% H_2O_2 and 0.05 M Fe^{2+} (Fenton reagent) for the water sample. Simultaneous mixing and heating are then conducted to complete the digestion [61]. The majority of microplastics are considered to remain unaffected with WPO while reducing oxidation time from days to hours or minutes. For extraction and identification of microplastics in leachate, in some cases, a modification of NOAA laboratory methods is applied by adjusting the amount of the chemical documented in the original method [37]. To prevent the chance of violent boiling at the beginning of the reaction, WPO should be started in a cold bath for safety [39]. In some studies, wet peroxidation is conducted multiple times for better results [39].

Other potential digestion methods for MP analysis from landfill leachate are enzymatic degradation, acid, and alkaline treatment. An enzymatic degradation is an emerging approach for removing organic matter from the aqueous sample. Oxidative enzymatic degradation is conducted using enzymes, such as lipase, proteinase, cellulase, chitinase, and amylase. Enzymatic degradation is evidenced to remove a high amount of organic matter [51,62] with minimum to no degradation MPs in the wastewater sample matter

[51,62]. However, no research employed enzymatic degradation to isolate MPs from landfill leachate samples. The reason could be the high cost of enzymes, the complexity of the process (multi-step process), and the prolonged treatment time (13 days for wastewater sample) of the method, which could be further delayed for an increased amount of organic matter in the sample like landfill leachate [48,63].

3.2.2. Inorganic matter removal

Following the oxidation process, density separation is conducted to remove the inorganic substance. The mechanism is to float MPs in a solution of higher density than the targeted polymers. The density of common polymers mainly varies from 0.90 to 1.6 g cm^{-3}. The common salts used for density separation are sodium chloride (NaCl), sodium iodide (NaI), sodium polytungstate (SPT), and zinc chloride ($ZnCl_2$). The salt is selected based on the density of the targeted polymer for extraction. For example, NaCl (density: 1.2 kg L^{-1}) is used to separate low-density polymers, such as polyethylene, polystyrene, and polypropylene, where NaI (density: 1.6–1.8 kg L^{-1}) or ZnCl (density: 1.5–1.7 kg L^{-1}) extract heavy polymer, such as polyethylene terephthalate (PET) and polyvinyl chloride (PVC).

Most importantly, separating organic matter and inorganic matter from the leachate sample to isolate MPs should be conducted without causing any damage to MPs. Currently, there is no existing pretreatment guideline for leachate pretreatment. Studies conducted on the pretreatment of leachate samples are based on experience from other water and wastewater samples [39,43,44]. However, the chemical composition of landfill leachate is very different from other sources. Unlike municipal wastewater, landfill leachate generally contains a higher level of ammonia, chemical oxygen demand, and metals, which can interfere with the traditional pretreatment process [37]. Thus, a standardized methodology designed for the pretreatment process of landfill leachate is necessary.

3.3. Microplastic characterization and identification

A complete characterization of MPs from landfill leachate should define the physical (shape, size, and color) and chemical (polymer composition) properties of MPs. It is difficult to conduct a complete characterization for MP complex matrices such as landfill leachate using a single identification technique. Thus, a combination of analytical methods has often been used.

For physical characterization, visual identification with the naked eye is the simplest approach. The microplastic size range of 2–5 mm can be characterized by the naked eye [64]. However, the detection method is size-limited and error-prone, thus challenging for identifying MPs from the complex environmental matrix as landfill leachate. In leachate analysis, the stereomicroscope is the most used instrument for counting and classifying physical characteristics like microplastic size, shape, color, and surface morphology (Table 1). Nevertheless, stereoscopic microscopes are also susceptible, especially with samples burdened with a high level of organic matter. Hidalgo-Ruz et al. [65] estimated that up to a 70% error ratio could be observed if used only stereomicroscope for identification. Furthermore, the rate of error increased with the decreasing particle size. Transparent tiny particles (<100 μm) or fibrous MPs are hard to identify with stereoscopic microscopes [66]. Therefore, using a stereomicroscope as the only identification instrument to study leachate is unreliable, as landfills have an abundance of such particles. For complex environmental samples, associating scanning electron microscopy (SEM) can facilitate analysis by providing high-resolution images of the microplastic particles [67]. Transparent and high-magnification of images characterize microplastics' surface morphology and differentiate microplastics from organic particles. Nevertheless, the color of microplastic cannot be identified by SEM tests [68]. Moreover, SEM-based procedures are expensive, time-consuming, and require substantial effort for sample preparation and examination, limiting the number of samples tested [69].

For chemical characterization, Fourier transforms infrared (FTIR) spectroscopy is the most used instrument for analyzing microplastic in landfill leachate. By utilizing an encoded polymer spectrum library, FTIR spectroscopy can confirm the presence of plastic particles and identify specific polymer types. Depending on the targeted size, a different mode of FTIR has been used. He et al. [36]. applied attenuated total reflectance (ATR) mode (ATR-FTIR) for evaluating MPs with a size greater than 1 mm and Micro-FTIR (μ-FTIR) for assessing MPs with a size less than 1 mm. For tinier MPs (<1 mm), another means is Raman spectroscopy [70]. It detects the polymer composition of a sample by analyzing the chemical bond polarity of the particle. The projection of a laser beam on a particle in different frequencies of back-scattered light produces a unique spectrum for each polymer depending on the molecular structure and atoms present [71,72]. However, possible interference resulting from foreign bands and fluorescence from dyes and pigments can interfere with the accuracy of the readings [73]. Micro-Raman spectroscopy can detect particles smaller than 20 μm [72]. Kazour et al. [15] used Raman spectroscopy and detected MPs in the range of 20–500 μm from the surface water sample adjacent to a coastal landfill.

3.4. Quality assurance

Due to the omnipresence of MPs in the surroundings, quality control is mandatory to minimize cross-contamination and acquire reliable results. To minimize contamination, all apparatuses used in the sampling and extraction processes (such as sampling containers, glassware, stainless sieves, Petri dishes, tubes, and vacuum filters) should be rinsed thoroughly with deionized water [37,39] or Mili-Q water and Ethanol [45] for several times before and in between sampling. The apparatus should be immediately covered with aluminum foil after cleaning and during the procedure [37,39,45]. After sampling, the filters are safer positioned in glass Petri dishes to protect from atmospheric MPs [39]. Measurements can be taken to quantify the airborne MPs as a precaution. Moreover, to avoid the contamination of airborne microplastics, sorting suspicious particles can be conducted in a vertical flow cabinet (P [37]. The liquid chemicals used in the study should be filtered before use. Nitrile gloves and clean lab coats of natural fabrics should be worn throughout the sampling, pretreatment, and identification process [37,39,45]. Sampling equipment, either a plastic container or pumping equipment (PVC hose or power cable), can cause cross-contamination. Therefore, multiple blank samples should be taken to detect possible sources of contamination during sampling, and the same identification procedure should be conducted as the leachate sample. For future reference, the presence of plastic materials used at or in the locality of the sampling point should be documented (photographed and identified).

4. Occurrence of micro-/nanoplastics in landfill leachate

4.1. The concentration of MPs in landfill leachate

MPs were detected in both raw and treated landfill leachate. The detected MPs in untreated and treated landfill leachate varied between 0–382 and 0–2.7 items L^{-1}, respectively (Table 2). This high variation of concentration might be contributed to the difference in sampling strategy, analytical technique, or leachate treatment

Table 2
Concentration and treatment of MPs in landfill leachate on a global scale.

Landfill description					Treatment process	MP concentration			References
Country	Landfill location	Number of landfills	Landfill type	Landfill status		Untreated leachate (items L^{-1})	Treated leachate	% Removal	
China	Shanghai, Wuxi, Suzhou, Changzhou	6	MSW	Non-haz, active, closed	-	0.42-24.58	-	-	[37]
China	Shanghai	1	MSW	Non-Haz	-	4-13	-	-	[38]
China	Suzhou	1	MSW	Non-haz	Membrane bioreactor with activated sludge, nanofiltration, reverse osmosis	235.4 ± 17.1	0.4 ± 0.1	99.8%	[39]
China	Shanghai	1	MSW	Non-haz	Membrane bioreactor (Anoxic/Oxic), ultrafiltration, nanofiltration, reverse osmosis	1.2 ± 0.57	0.6	50%	[40]
Indonesia	Bogor City	1	MSW	Non-haz	-		-	-	[41]
India	Chennai	2	MSW	-	-	2-80	-	-	[42]
France	Dollemard Coast	1	MSW, IW, SP	-	-	6	-	-	[15]
Finland	Turku, Salo, Lahti	3	MSW, IW	Non-haz	-	0.16-1.10	-	-	[44]
Finland	South-east	1	IW	Haz	Filtration and activated carbon	0.30	0.32	3%	
Finland	Lahti	1	MSW, IW	Non-haz	Artificial soil filtration	1.97	0.03	99%	
Norway	Skedsmokorset	1	MSW, IW	Non-haz	Sequence batch reactor	1.3	0	100%	
Norway	Ask, Anonymous	2	MSW, IW, mixed	Non-haz		1-4			
Ice land	Fiflholt	1	MSW, IW	Non-haz	Sand bed filtration	0.2	0.06	76%	
Iceland	Alfsnes Fiflholt	2	MSW, IW	Non-haz	-	0-4.51	-	-	
Sweden	-	-	-	-	-	-	0-2.7	-	[74]

Abbreviations: MSW, municipal solid waste; IW, industrial waste; SP, special waste.

process. Moreover, the complexity of the waste composition in landfill highly influences MP concentration and compositions in leachate. High variation of MP concentration is also reported in wastewater. For wastewater, the reported MPs varied between 1–3169 and 0.0007–125 item L^{-1} for raw and treated wastewater, respectively [29]. The MP abundance in untreated and treated landfill leachate is lower than in untreated and treated wastewater. One possible reason is that the fragmentation of plastics and MPs in landfill strata influences the higher abundance of smaller MPs/NPs than sewage, which is not detected by the currently used methods and is not reflected in the number-based concentration reports.

Landfill age or status may influence the concentration of MPs in leachate. For example, Su et al. [38] found that the average MPs abundances in younger landfills were higher (8–10 items L^{-1}) than the number of MPs (4 items L^{-1}) in leachate from older landfills. The growing trend of plastic use could be a possible reason behind this observation. The global production of plastic waste increased by 26% from 2010 to 2016, and the occurrence of plastic in solid waste raised to 12% globally, rocketing 242 metric tons in 2016 [75]. Plastic waste comprises 2.95–21.76% of solid waste in landfill. Consequently, higher plastic waste is reported in active landfills than in older or closed landfills [76]. Received plastic waste undergoes fragmentation in landfills and produces secondary MPs with time, which percolate in leachate. However, the microbial breakdown of polymer with time in landfills might be another possible reason for the lower concentration of MPs in the leachate of older landfills [77]. Meanwhile, the scant detection of tinier particles in currently available studies might partially account for this observation. Due to analytical complexity, most leachate studies conducted the MP analysis with sizes ranging 50–5000 μm (Table 1), with tinier MPs or NPs largely ignored and undetected in old landfills.

Studies indicated geographical variation in MP concentration in landfill leachate, which could be related to regional plastic generation. A survey conducted in 2010 showed that the highest plastic waste-generating country in Europe is Germany, which produces 14.48 million tons of plastic waste annually. In China, the plastic waste generation was 59.08 million tons per year [78], and the measured microplastics concentration from China varied between 0.4 and 235.4 items L^{-1} [37–39]. In comparison, the concentration range of MPs in the leachates from Nordic countries was 0–4.5 items L^{-1} (Iceland, Norway, and Finland), while the concentration range of MPs in the leachates from different regions of China varied between 0.42 and 235.4 items L^{-1} [44] (Table 2). The higher generation of plastic waste in China is one of the main contributors to higher MP abundance in leachate compared to European countries.

The local waste management approaches could be another crucial factor in MP concentration in leachate. The average MP concentration from Southeast European countries (Serbia, Bosnia, and Herzegovina) is three orders of magnitude higher than in Nordic countries (Finland, Iceland, and Norway) (Table 2). This could be the consequence of systematic waste management practices in developed countries, while proper sorting during waste collection is not entirely applied in developing countries such as Serbia, Bosnia, and Herzegovina [45]. Cenk and Fikret [79] investigated the municipal waste management of 35 European countries regarding four treatment approaches — recycling, composting, landfilling, and incineration (2012 database). The result revealed that the primary municipal solid waste treatment process for Serbia, Bosnia, and Herzegovina is landfilling. In contrast, waste recycling is much higher in Iceland, Norway, and Finland than in Serbia, Bosnia, and Herzegovina. In addition to recycling, the rate of waste incineration in Norway is the highest among all countries. The compost preparation is also higher in Nordic countries (Finland, Iceland, and Norway) than in Southeast European countries (Serbia, Bosnia, and Herzegovina). Another report mentioned that Serbia is experiencing the sharpest decline in municipal waste recycling in Europe. The current recycling rate of the county is only 0.4%. All these scenarios are attributed to the abundance of MPs between developed Nordic and developing Southeast European countries.

4.2. Polymer composition

More than 28 kinds of polymers were identified in the leachates of different landfills (Fig. 3). Among all types of polymers, studies indicate that low-density polyethylene (LDPE), high-density polyethylene (HDPE), polystyrene (PS), polypropylene (PP), PVC, and

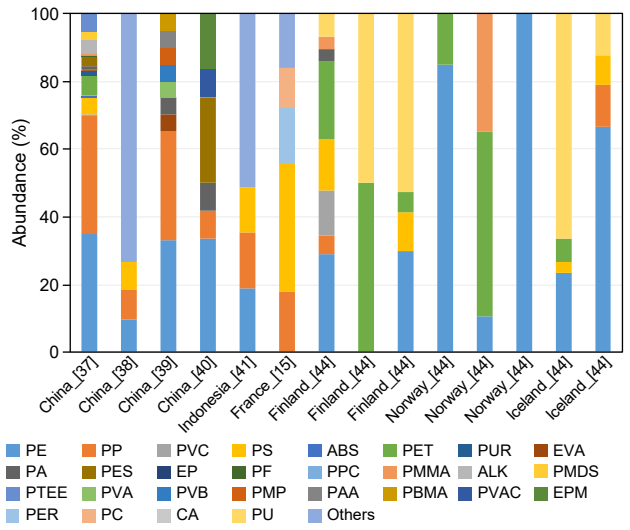

Fig. 3. Polymer composition of microplastics in landfill leachate worldwide. The number after each country's name is the reference number.

PET are the most abundant plastic polymers in the landfill leachate worldwide [8,37–39].

The polymer composition is directly related to the contemporary application of polymeric materials in anthropogenic uses. Due to their unique property and cost-effectiveness, the polymers mentioned above are extensively used for various short-term use products like shopping bags (PE), water bottles (PET), disposable drinking cups (PS), etc. Due to its high flexibility, PVC is widely used in different sectors like construction, waterproofing, medical equipment, clothing, toys, and other sports supplies [80,81]. The polymer composition in landfill leachate is similar to that in wastewater. The most occurring MPs in wastewater are polyester (PES) (60–96%), polyamide (PA) (3–20%), PE (64–78%), PP (20–100%), and PS (12–80%) and other polymers, such as alkyd and acrylic [11]. Wearing synthetic clothes during washing is one of the significant sources of PES and PA. Facial and body wash contains PE and PP. Packaging films and water bottles also include PE. Car wash and cosmetic products can contribute to PP [28].

Polymer types in landfill leachate may depend on the regional difference of Municipal Solid Waste (MSW) composition and condition of the landfill (young/old, active/closed). The polymer composition of leachate directly reflects the pattern of the current consumption scenario of plastic products. Moreover, the forthcoming MPs contamination in leachate can also be projected from the everyday use of the plastic polymer. Su et al. [38] compared MPs in leachate generated from landfills of different ages. Unlike PE and PP, Polyether Urethane (PEUR) in the younger landfill was more abundant in the older ones. In contrast, in samples from older landfill leachate, PEUR was undetected. This finding might be attributed to the change in application fields and the lifetime of various plastic products. Due to good mechanical and fiber-producing ability, the use of PEUR is increasing every year. PEUR is primarily used in the transportation and construction sector and has a longer lifespan (up to 35 years) compared to traditional packaging polymers like PE and PET (0.5 years) [82].

The polymer composition can influence the fate of MPs along the leachate treatment processes. For instance, commonly, higher-density polymers are more likely to accumulate in sludge, while polymers with lower density are more prone to pass with treated effluent. For example, as the density of PES (1.37 gm cm^{-3}) is higher than PE (1 gm cm^{-3}), more PES can be expected to settle down in the leachate sludge compared to PE. Moreover, the polymer composition is also crucial to evaluate the effects of different treatment methods on the removal efficiency of MPs. For instance, granular activated carbon (GAC) utilizes the polarity of solids, and therefore, non-polar MPs, like PE and PP, might exhibit a successful removal when applied in leachate treatment [83]. However, the complex composition of leachate might alter the anticipated outcome. The relationship between the fate, transport, and removal efficiency of MPs in the leachate treatment process regarding polymer composition is not well documented. Studies analyzing MPs' fate in the leachate treatment system are needed to confirm their correlation with polymer composition.

4.3. Shape

The morphologies of MPs identified in leachate include lines, flakes, films, fragments, pellets, beads, foam fiber, and granules (Fig. 4a). Fiber and fragments are the most abundant shapes of MPs in leachate worldwide (Fig. 4b). Due to the form, fiber and fragments might be more favorable to infiltrate into the leachate from landfills with rainwater. On the other hand, the dominant existence of fibrous MPs in the leachate was primarily caused by the fact that fibers were easier to pass through the garbage and enter the leachate due to their small sizes [40].

The parent plastic products can be traced from the shape of MPs. For example, films are usually generated from plastic bags and packages. Plastic bags are thin and transparent, so they can easily get broken through the exposure to the sun. Granules and spheres are mostly from plastic containers, water bottles, microbeads, or food storage containers [43]. The shape can also indicate the source (primary/secondary) of plastics. For example, most of the MPs found in the leachate are in irregular shape and hackly in structure with rough edges [37,38], which indicates the production of

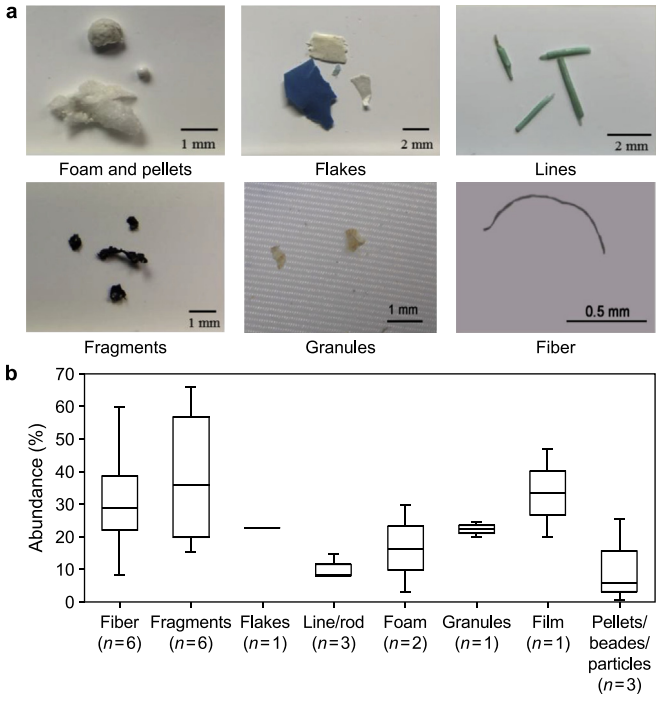

Fig. 4. **a**, Example of different shapes of microplastic detected in landfill leachate. **b**, Relative abundance of different shales of microplastics in the landfill leachate. The median, 10th, 25th, 75th, and 90th percentiles were plotted as vertical boxes with error bars. [15,37–41,43].

secondary MPs from plastic debris by the fragmentation process. Moreover, shapes can also indicate the location from where the microplastic originated. For example, resin pellets might be the predominant shape of microplastic near industrial areas, while fragments and foam might be in high concentration near the fishing port [84].

Studies from almost all regions indicated that MPs found in landfill leachate had irregular shapes and rough surface texture caused by the fragmentation of plastic products in the landfill environment [39,45]. The surface texture of the MP surface is an important parameter to presume the possible threat to the environment. The husky surface might enhance contaminant adsorption, such as heavy metals and organic pollutants, enhancing the environmental risk of leachate disposal. Again, the surface roughness influences the removal rate through the different treatment processes. For instance, with smooth textures, fibers and pellets were comparatively less prone to being trapped by mechanical methods [85]. Angular and twisted morphologies with curved-surface textures make fragments and granules easily captured [86].

4.4. Size

The size of microplastic refers to the largest length of plastic particles. Size is one of the most critical characteristics of microplastics, which determines its potential damage to humans and the environment. MPs in leachate displayed a wide size range from <20 to 5000 μm. The wide variation in size could be highly influenced by the detection method. For example, most tiny particles will get lost if a larger sieve is employed during sampling. To get a whole scenario about particle distribution, studies should consider extracting MPs for a more extensive size range (1–5000 μm). To properly compare results among numerous research, clear criteria for defining the size of MPs should be followed, and standardized sampling and extraction protocols should be established [87].

The number of microplastics in landfill leachate increases with the decrease in particle size. For example, He et al. [37] detected that 75% of microplastics in the sample were in between the size of 100–1000 μm and 20% of the count were in the range of 1000–5000 μm while only about 5% of particles were with the size of >5000 μm. A similar result was found by Ref. [42] while evaluating the microplastic abundance in groundwater near municipal solid waste dumpsites in South India. Microplastic produced by fragmentation during landfilling process causes travelers to leachate with rainwater. It could be inferred that smaller particles were more readily accumulated in the leachate, while larger MPs would retain more in the solid phase of landfills. Su et al. [38] confirmed this fact by comparing the MP occurrence in landfill refuse and leachate and identified that the size of microplastic in the leachate is much smaller (0.83 mm) than the refuse (4.97 mm).

Size is an influential factor that can affect the removal efficiency of MPs by different treatment units. Fragmentation during the treatment process generates multiple smaller MPs or NPs from one larger MP particle; therefore, a negative removal efficiency of that treatment steps could be reported. For example, Sun et al. [39] reported the concentration of microplastics in untreated leachate as 235.4 ± 17.1 items L^{-1}, which increased almost 150 times after going through the membrane bioreactor. Similar findings were achieved for WWTPs. The wastewater influent, MPs between 20 and 100 μm comprises around 45%, and after preliminary treatment, the concentration of MPs in that size range was observed to be 70%. Therefore, following the size distribution in each leachate treatment step is necessary to judge the success of a particular treatment procedure.

4.5. Color

The colors of MPs depend on the colors of their parent plastic and life span. For example, transparent fibers might originate from the fragmentation of fishing lines or nets, while colored particles are more likely derived from the breakdown of commonly used plastic commodities, such as textile and packing products [88]. However, they can change by the weathering effect. Color is an overlooked property that has been well-defined in very few research. However, the color of the MPs can provide critical hints about the solid waste composition and the duration of the fragmentation process. For example, the dominant, white-colored plastics indirectly indicate the degradation process that takes place on-site for a long time, transforming other color contents into white [42]. The high abundance of transparent and yellowish color suggested that most particles were aged and presented in the landfill system for a long time [39]. The yellowish color also may indicate a higher quantity of organic material in the samples [8]. Sun et al. [39] also identified that over 90% of their detected MPs were transparent or yellowish, while some (<10%) were in other colors. Kilponen [8] also detected an abundance of transparent and yellowish color particles from the leachate sample of Finland. In contrast to landfill leachate, in flowing water, colored MPs (white, yellow, green, red, orange, blue, black, and grey) are the most dominant ones accounting for 50.4–86.9% of the total MPs [89]. This difference is possibly due to the difference in the retention time of plastic in landfill leachate and flowing water. As MPs stay and fragment in the landfill for a longer time, the original color of the polymer alters due to weathering effect. The level of threat to the biota by MPs is related to their color. For example, due to the likelihood of their food, white microplastics are likely to be ingested more by plankton, fish, and other species. In that way, microplastics might enter food webs and cause several physiological toxic effects with intake [90]. Hence, to realize the potential threat of MPs by landfill leachate, their color should be characterized in more analysis.

5. Removal of MPs in the landfill leachate treatment facilities

If not properly managed, MPs from landfill leachate can pollute the nearby environment. For instance, Cordova and Riani [41] measured the MP concentration in the receiving river before and after the discharge of leachate effluent. They identified that the MP count increased around three times after the leachate discharge in the river. In another report, Silva et al. [91] estimated a yearly release of 0.03 billion or 291 items L^{-1} of MPs via European landfills leachate. Therefore, careful MP removal management is necessary to mitigate MPs migration from landfill leachate into the environment. Currently, the leachate treatment facilities are not specially designed for addressing MPs contamination, though several studies indicated a high removal of MPs by traditional leachate treatment process [39,44].

Generally, leachate treatment is conducted by biological, physical, or chemical processes. Common treatment steps include soil bed filtration, aeration, sequencing batch reactors (SBR), membrane bioreactors (MBR), oxidation, coagulation/flocculation, activated carbon, stripping, evaporation, and reverse osmosis (RO) [39,44,92]. Leachate recirculation in landfills and leachate transfer to wastewater treatment plants is also applied [45]. Depending on the treatment technique, leachate treatment processes can decrease the concentration of microparticles from 3% to 100% (Table 2). Fig. 5 summarizes the MP removal efficiency and challenges due to the presence of MPs in different leachate treatment processes.

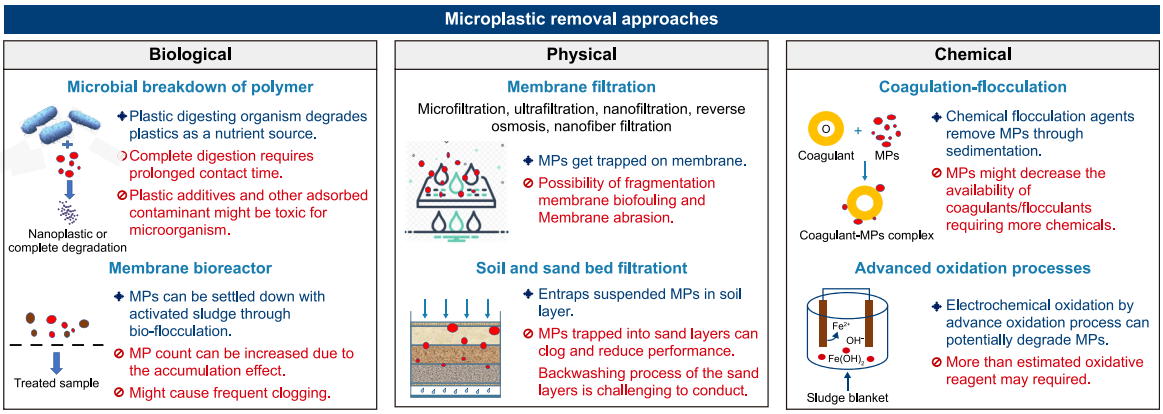

Fig. 5. Efficiency and challenges of microplastic removal by the leachate treatment process.

5.1. Biological treatment

Biological approaches are widely used for leachate treatment for their reliability, cost-effectiveness, and simplicity. Biological treatment methods, such as bio-flocculation, by activated sludge and degradation by plastic-ingesting organisms indicated their potential for microplastic reduction in leachate effluent [44]. analyzed the microplastic in raw leachate and leachate treated with SBR and documented a 100% removal efficiency. In SBR, high-density microplastics can settle down with biomass, reducing the leachate effluent's MP count. MBR is another widely used biological treatment for landfill leachate. Sun et al. [39] analyzed leachate from a leachate treatment unit with MBR. The study reported that membrane treatment effectively reduced microplastics loading to 0.14% for particles and 0.01% for mass concentration, but the average particle density increased. Zhang et al. [40] also examined the fate of MPs in a two-stage treatment with MBR and two-stage Anoxic/Oxic (AO). Zhang et al. [40] reported 50% and 20% removal efficiency after MBR and AO, respectively. In an MBR, the microplastic count can be increased due to the accumulation effect of microplastic in the treatment system. For example, Sun et al. [39] noticed an increased amount (150 times higher) of microplastic in an MBR effluent compared to the raw leachate sample. Consequently, as the MPs got captured by the membrane bioreactor, the microplastic in the sample of membrane tank effluent was significantly reduced, indicating a high removal rate of microplastic in the membrane bioreactor system. It is noteworthy that MPs present in raw leachate might induce frequent clogging in MBR, requiring recurrent backwash to clear the sludge cake layer. Frequent backwashing might accidently release the MPs trapped in the membrane. Therefore, the performance of MP removal by MBR fluctuates depending on the membrane's pore size and the incidence of backwashing [11].

The microbial breakdown of polymer in the landfill leachate is another biological removal process for microplastic. For Example, Different bacteria like *Bacillus cereus, Bacillus gottheilii*, *Alcaligenes faecalis, Bacillus amyloliquefacien, Bacillus brevis, Cyanobacterium, Anabaena spiroides;* Microalgaes like *Scenedesmus dimorphus* (Green microalga), *Anabaena spiroides* (blue-green alga) and *Navicula pupula* (Diatom) and, other microorganisms like *Agios consortium*, *Souda consortium*, *Penicillium Roquefort,* etc. Can successfully degrade polyethylene, polystyrene, polyethylene terephthalate, and polypropylene polymers as their nutrient source [93–97]. Utilizing microbial breakdown for the degradation of MPs/NPs in leachate is less expensive and safe for use to a large extent [93]. However, the efficiency of this method depends on the contact time between the potential microbes and the targeted polymer [75].

5.2. Physical treatment

Different physical treatments, such as filtration and sedimentation. Can potentially remove MPs while employed in a leachate treatment system. Microfiltration (pore size 10–0.1 μm), ultrafiltration (pore size 0.1–10 nm), and nanofiltration (pore size 10–1 nm) are frequently used leachate treatment techniques based on physical separation. All these filtration systems can also separate MPs from the leachate according to their sizes. RO (pore size 1–0.1 nm) is another promising physical separation technique [98], and the competence of RO for removing MPs from landfill leachate has been mentioned in multiple studies [39,45]. Sun et al. [39] analyzed the leachate sample treated with nanofiltration and RO and recognized almost 99% removal of MPs. In contrast, Zhang et al. [40] reported a high efficiency of MP removal by the ultrafiltration technique. Still, they could not find any contribution of NF and RO on MP reduction in leachate effluent. The leachate went through a membrane bioreactor in both studies before advanced separation. However, fragmentation of MPs into nanoparticles can occur during membrane filtration resulting in membrane abrasion and fouling [99].

For leachate treatment, soil and sand bed filtration can successfully remove MPs from leachate. The effectiveness of this treatment is correlated to pore size. Rapid sand filtration can entrap suspended solids within three sand layers of anthracite, silica, and gravel [17]. van Praagh et al. [44] reported a 76% and 99% removal rate with soil and sand bed filtration, respectively. However, MPs trapped in sand layers can clog and reduce performance. Furthermore, the backwashing process of the sand layers is also challenging to conduct [17].

5.3. Chemical treatment

Coagulation-flocculation and chemical oxidation are potential chemical approaches to separate and remove suspended solids from water samples. Even though not targeted for MP separation, the coagulation-flocculation process can remove MPs from the leachate sample. Multiple reports have also stated significant MP removal from wastewater samples [100,101]. Coagulants like polyacrylamide (PAM), Fe and Al-based salts, and $FeCl_3 \cdot 6H_2O$ generally remove MPs by forming larger clusters in which MPs get trapped [93]. However, the interaction of MP with the chemical flocculation agents may decrease the efficiency of the leachate

treatment system. Electro-coagulation is another process that can be efficiently removed MPs from influent. In electro-coagulation, metal ions (Fe^{2+} and Al^{3+}) are released from electrodes that react with the hydroxide to form metal hydroxide coagulants and generate sludge blankets for adhering MPs [102]. In wastewater treatment plants, almost 90% of MPs was removed using this method [93]. van Praagh et al. [44] reported a 3% removal of MPs from landfill leachate by flocculation. However, details about the flocculation process were not mentioned in that study.

Besides coagulation-flocculation, several other chemical treatment approaches generally applied in landfill leachate treatment systems can potentially remove MPs from leachate effluent. For instance, electrochemical oxidation (OCl^-, O_3, or H_2O_2) or advanced oxidation processes (combination of O_3/H_2O_2, ultraviolet (UV)/ultrasound, O_3/UV, H_2O_2/UV, H_2O_2/ultrasound, etc.) may degrade MPs breaking the chain of different polymer. Moreover, disinfection processes like chlorination, ozonation, and UV radiation can also break MPs into smaller sizes and even produce nanoplastics [46,100,103]. However, MPs in the leachate might reduce the efficiency of the disinfection processes by devouring disinfectants or protecting bacteria from the attack of disinfectants [17].

6. Perspectives on microplastic removal methods

With all kinds of treatments evaluated for MPs in landfill leachate, most MP removal is driven by separation-based treatment processes rather than degradation, such as bio-flocculation, sedimentation, air flotation, membrane separation, filtration, etc. All separation-based treatments produce MPs concentrated residuals in biosolids, chemical sludge, RO brine, froth in air flotation, etc. More investigation is needed for the final disposal of and/or resource recovery from these MPs-concentrated residuals.

Though multiple treatment technologies were evaluated for MP removal from landfill leachate in literature, in practice, it is unlikely to install new treatment processes only for MPs. It is more realistic to promote monitoring MPs in existing landfill leachate treatment facilities to better understand the fate and removal rate of MPs, based on which to develop improvement strategies for MP management, such as membrane modification and coagulant addition, etc. To facilitate universal MP monitoring in landfill leachate, standardized sampling, detection, and characterization methodologies are necessary but are not in place yet. It is mainly due to the intricacy and variation of landfill leachate matrices, especially high levels of organic matter, which can create significant background noises for the many spectroscopic detection means, such as stereomicroscope, SEM, FTIR, Raman spectroscopy, etc. The development of pertinent pretreatment methods is the key to MPs detection and characterization in landfill leachate matrices.

Discharging landfill leachate to WWTPs and disposing wastewater sludge in MSW landfills create a unique loop for recalcitrant contaminants in landfill leachate to transfer between landfills and WWTPs. Taking the US as an example, 54% of landfill leachate is discharged to WWTPs, while 22% of wastewater sludge is disposed of in MSW landfills. Even if not completely closed, this landfill-WWTP loop provides a niche with extended retention time for MPs and other recalcitrant contaminants. In addition, bioreactor landfills, which recirculate leachate by pumping collected leachate back to the top of waste cells, also provide a longer retention time for leachate. The prolonged retention time for landfill leachate by landfill-WWTP loop and bioreactor landfills can facilitate possible physical fragmentation and bacterial degradation and depolymerization of MPs, which can release plastic degradation by-products and plastic additives as secondary contaminants.

7. Conclusions

- There are primary and secondary MPs in landfill leachate. Primary MPs are manufactured on a micro scale, while secondary MPs are generated by the degradation and/or fragmentation of regular plastic waste in landfills and contribute to the total MPs in landfill leachate. Wastewater treatment residuals are another source of MPs in landfill leachate.
- Methods for sampling and detecting microplastics in landfill leachate vary considerably between studies. Container collection and pumping are the most common methods of sampling. Micro-FTIR or Raman techniques may be the most effective method for characterizing landfill leachate microplastics.
- The high variance of MPs concentration in landfill leachate is reported due to: (i) inconsistent sampling and detection methods and (ii) highly variable solid waste composition. Literature reported less overall MP concentrations in landfill leachate than sewage, possibly because tinier MPs/NPs in landfill leachate are ignored in the number-based concentration reports. Also, fresh landfill leachate contained more MPs than mature landfill leachate due to increased plastic use and plastic waste production. The geographical comparison showed that MP concentration in landfill leachate is directly related to the local plastic waste production and waste management approaches.
- PE, PP, and PS are the most abundant plastic polymers in landfill leachate. Polymer types also determine their physical properties, such as shape, density, etc., influencing their removal efficiencies in landfill leachate treatment processes.
- Fiber and fragments are the most abundant shapes of MPs in landfill leachate as they are easier to pass through the waste layers. The shape can indicate the origin (plastic bags, plastic containers, etc.), source (primary or secondary), and location of the parent plastic waste. The shape of MPs can also change through treatment processes.
- The color of MPs in landfill leachate is mainly determined by their parent plastic waste. The dominance of light-colored (transparent, white, or yellowish) MPs indirectly indicates long-term degradation due to the weathering effect.
- Monitoring of MPs in landfill leachate treatment facilities is limited, and a wide range of removal efficiency was reported. MP removal in engineered biological treatment systems (such as SBR) is believed to be mainly driven by bio-flocculation. High removal efficiency is believed to be an overestimation due to the ignorance of smaller MPs in the detection methods. Microbial breakdown of MPs is reported to take place with extended contact time.
- Membrane filtration processes (MF, NF, UF, RO, etc.) are reported to have high removal efficiency for MPs in landfill leachate with high cost. The fragmentation of MPs into NPs can cause membrane abrasion and fouling. Low-cost physical filtration, such as soil and rapid sand filtration, were reported with moderately high removal efficiencies. Sand layer clogging and backwashing are the main issues with rapid sand filtration.
- Various oxidation and disinfection processes are reported effective in removing MPs/NPs in landfill leachate, while very limited information is available for coagulation-flocculation.

CRediT authorship contribution statement

Mosarrat Samiha Kabir: Conceptualization, Investigation, Writing - Original Draft Preparation. **Hong Wang:** Conceptualization. **Stephanie Luster-Teasley:** Supervision. **Lifeng Zhang:** Supervision, Resources. **Renzun Zhao:** Funding Acquisition,

Conceptualization, Project Administration, Writing, Review & Editing, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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