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Contemporary Drift in Emerging Micro(nano)plastics Removal and Upcycling Technologies from Municipal Wastewater Sludge: Strategic Innovations and Prospects

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Abstract

Purpose of Review Annually, huge amounts of microplastics (MPs) are added to farmlands through sewage sludge (SS)/biosolid applications as a fertilizer. Most research emphasizes the enormity of the problem and demonstrates the fate, impacts, and toxicity of MPs during SS treatment processes and land applications. None has addressed the management strategies. To address the gaps, the current review evaluates the performance analysis of conventional and advanced sludge treatment methods in eliminating MPs from sludge.

Recent Findings The review uncovers that the occurrence and characteristics of MPs in SS are highly governed by factors such as population density, speed and level of urbanization, citizens' daily habits, and treatment units in wastewater treatment plants (WWTPs). Furthermore, conventional sludge treatment processes are ineffective in eliminating MPs from SS and are accountable for the increased small-sized MPs or micro(nano)plastics (MNPs) along with altered surface morphology facilitating more co-contaminant adsorption. Simultaneously, MPs can influence the operation of these treatment processes depending on their size, type, shape, and concentration. The review reveals that research to develop advanced technology to remove MPs efficiently from SS is still at a nascent stage.

Summary This review provides a comprehensive analysis of MPs in the SS, by corroborating state-of-the-knowledge, on different aspects, including the global occurrence of MPs in WWTP sludge, impacts of different conventional sludge treatment processes on MPs and vice versa, and efficiency of advanced sludge treatment and upcycling technologies to eliminate MPs, which will facilitate the development of mitigation measures from the systematic and holistic level.

Keywords Microplastics · Microplastics management · Sewage sludge · Sludge treatment processes · Toxicity

Abbreviations

A^2O	Anaerobic-anoxic–oxic technology
AD	Anaerobic digestion
ARB	Antibiotic-resistant bacteria
ARGs	Antibiotic resistant genes
BPA	Bisphenol-A
CAS	Conventional activated sludge
CAST	Cyclic activated sludge technology
CPL	Caprolactone
DBP	Dibutyl phthalate
DMP	Dimethyl phthalate
EDCs	Endocrine disrupting compounds
EPDM	Ethylene propylene diene monomer
EPS	Extracellular polymeric substance
EU	European Union
FTIR	Fourier transform infrared
HDPE	High-density polyethylene
HRT	Hydraulic retention time
HTC	Hydrothermal carbonization
HTL	Hydrothermal liquefaction
LOD	Limit of detection
MBR	Membrane bioreactor
MF	Melamine formaldehyde

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MFs	Microfibers
MGEs	Mobile genetic elements
MNPs	Micro(nano)plastics
MPs	Microplastics
NPs	Nanoplastics
PA	Polyamide
PA6	Polyamide-6
PAHs	Polyaromatic hydrocarbons
PC	Polycarbonates
PCB	Polychlorinated biphenyls
PCL	Polycaprolactone
PE	Polyethylene
PES	Polyesters
PET	Polyethylene terephthalate
PHB	Polyhydroxy butyrate
PMMA	Poly(methyl methacrylate)
PP	Polypropylene
PPCPs	Pharmaceuticals and personal care products
PPE	Personal protective equipment
PS	Polystyrene
PTFE	Polytetrafluoroethylene
PU	Polyurethane
PVAc	Polyvinyl acetate
PVC	Polyvinyl chloride
ROS	Reactive oxygen species
SDS	Sodium dodecyl sulfate
SRT	Sludge retention time
SS	Sewage sludge
SUPs	Single-use plastics
UASB	Upflow anaerobic sludge blanket reactor
UF	Urea formaldehyde
WWTPs	Wastewater treatment plants

Introduction

Mass production of plastic products since the 1950s gained popularity worldwide due to their low cost, ease of use, and durability. It has been estimated that global plastic production was around 1.7 million tons in 1950, which has increased to 367 million tons in 2020 [1, 2]. Moreover, if current plastic production and use trends persist, annual global plastic production will reach 590 million tons by 2050 [1]. However, its widespread applications across different sectors, its long-lasting nature, low degradability, and improper disposal have caused massive plastics pollution in the different environmental compartments [3•]. Furthermore, these plastics undergo fragmentation and weathering by mechanical (abrasion, erosion), chemical (hydrolysis, photo-oxidation), and biological (microbial degradation) means, which lead to generation of micro-or nano-sized plastic particles that are defined as micro(nano)plastics (MNPs) [4•, 5]. Microplastics (MPs) are the plastic particles having diameter less than 5 mm; however, nanoplastics (NPs) are nano-sized plastic particles with diameter between 1 and either 100 or 1000 nm [5].

Furthermore, the COVID-19 pandemic has caused an unprecedented surge of the production, consumption, and disposal of personal protective equipment (PPE) and single-use plastics (SUPs), and their improper disposal and management have led to enormous MNPs pollution [6]. However, a decrease of 2.2% in plastic use was observed in 2020 due to economic slowdowns. However, as economic activities resumed in 2021, plastic consumption has also rebounded, leading to a further surge in MNPs pollution [7]. MPs are considered a contaminant of emerging concern due to their persistent nature and hazardous impact on the ecosystem and human health [8, 9]. It can intentionally be manufactured by the industries for various applications such as microbeads for cosmetics, industrial feedstock, and synthetic fibers in textile (primary MPs), or can be generated from plastics deterioration by various mechanisms such as chemical processes, ultra-violet radiations, weathering, and/or microbial actions (secondary MPs) [10••].

Sewage sludge (SS) is the solid, semi-solid, or liquid residue produced during municipal wastewater treatment [11]. The annual SS production in the European Union (EU) was 5.5 million tons per year in 1992, which has increased to more than 9.0–9.5 million tons per year in 2017. Furthermore, in developing countries like China, 36 million tons of SS were produced in 2019, whereas it was estimated that the complete treatment of SS in India would generate 3955 thousand metric tons of dry SS annually [12].

Recently, several studies reported that modern WWTPs could efficiently remove MPs (88–98%) from the wastewater [13, 14]. However, the removed MPs during WWTPs are likely to be concentrated in SS through adhesion, precipitation, and filtration processes [15, 16]. Furthermore, the main disposal routes of SS are landfilling, incineration, and land-based applications such as soil buffer, soil improvement, and soil amendment [17]. However, its application as a soil amendment to agricultural land is preferable, owing to its high content of organic matter, macro- and micro-nutrients, and economic viability [18]. For instance, about 50% of SS is processed for agricultural usage in Europe and North America [19]. However, land application is a significant way of SS management in western members of the EU, whereas it is banned in some EU countries such as Germany and the Netherlands [20]. Moreover, Ireland utilized 80% of municipal sludge for agricultural purposes in 2017 [21].

It is estimated that annually 1241, 1518, 13,660, 26,042, and 21,249 tons of MPs are added to farmlands in Australia, Canada, China, EU, and the USA, respectively, through biosolid application [22]. Therefore, soil amendment is an important pathway of MPs transportation to the agro-ecosystem, where it gets accumulated and undergoes a series of transformations propelled by physical (mechanical abrasion through agricultural activities), chemical (thermal oxidation, photooxidation, and photodegradation), and biological (biodegradation) means under long-term application, where particle size of MPs continues to decrease towards NPs, which allows vertical distribution of MPs with alteration in morphology encouraging interaction with co-contaminants [23, 24].

Raw SS is treated before its land application through different biological, chemical, and heat treatment processes such as dewatering, thickening, mesophilic and thermophilic AD, lime/alkali stabilization, composting, and thermal treatments to reduce sludge volume, weight, and potential health risk [25, 26]. For instance, 73% of SS is treated through anaerobic digestion (AD) in the UK, followed by lime stabilization accounting for 22% [10••]. Researchers suggested that conventional sludge treatment technologies are ineffectual in MPs removal. Additionally, limited studies reported that these treatment processes can influence the abundance and morphology of MPs which are accountable for the increased small-sized MPs or MNPs and changed surface morphology which facilitates more adsorption of co-contaminant, adversely influencing the terrestrial ecosystem functioning [27, 28]. Furthermore, studies have reported that MPs can adversely influence the efficiency of the different sludge treatment processes [29•, 30]. Therefore, a better understanding of the impacts of MPs on different sludge treatment processes and vice versa is needed for sustainable management of sludge and to critically determine the magnitude of MPs pollution.

As aforementioned, soil amendment with SS is an important pathway of MNPs transportation from sludge to agroecosystem where it gets accumulated in the soil, influence the soil properties and soil flora and fauna, and migrate to deep soils and aquifers [31-33]. Furthermore, due to the high surface area to volume ratio and strong hydrophobicity of MPs, various harmful inorganic and organic pollutants present in the SS can interact with MPs through various chemical bonding [34]. As a result, MPs behave as a vector of toxic chemicals contaminants [35] and provide a surface for biofilm formation for pathogens carriers and antibiotic resistant genes (ARGs) [36, 37]. However, despite severe impacts associated with the application of SS embedded with MPs in agricultural lands, limited studies investigated the adverse impact of MPs on vegetation [38, 39] and soil organisms [3•, 40]. Furthermore, research to develop a technology to remove MPs efficiently from SS is at a nascent stage and has progressed gradually; very few articles reported effective MPs mitigation strategies from SS such as through pyrolysis [41•], hyperthermophilic composting, biodegradation [29•], hydrothermal carbonization (HTC) [16], hydrothermal liquefaction (HTL) [42], vermi-wetland [43], and enzymatic degradation [44]. Most present reviews highlighted the immensity of the problem, relate the occurrence of MPs in SS with the toxicity to the terrestrial ecosystem, and intensifying risk through the interaction with co-contaminants present in SS. However, none has discussed the management and upcycling strategies.

Therefore, the objective of the current review article is to provide a comprehensive analysis on conventional and advanced sludge treatment processes to eliminate MPs from the SS by amassing state-of-the-knowledge through (1) the global occurrence of MPs in SS with sources, fate, and transport mechanism; (2) the impacts of conventional sludge treatment processes on MPs, and vice versa; and (3) analysis of advanced sludge treatment and upcycling technologies for MPs removal and management from SS. This comprehensive view will facilitate the development of MNPs mitigations measures from the systematic and holistic level. Furthermore, the recommendations are provided to develop a sustainable treatment process to efficiently remove MNPs from sludge and highlight the knowledge gaps.

Methodology

This literature survey has been conducted systematically comprising of three steps broadly, i.e., (i) literature search and screening, (ii) extraction of data and relevant body of knowledge, and (iii) qualitative and quantitative analysis of findings. Potential literature was selected after searching peer-reviewed research articles, reviews, reports, and book chapters available on well-known scientific databases such as ScienceDirect, Google Scholar, Web of Science, and PubMed. The keywords used for this literature survey, exclusively or in combination, include microplastics, micro(nano)plastics, nanoplastics, sewage sludge, toxicity, sludge treatment methods, occurrence, impacts, management, and co-contaminants. Then, the literature collected from scientific databases and websites was carefully sorted, scrutinized to remove repetitive results, and screened regarding the objectives of the review. This was followed by extraction of recent data and evidence on the various subjects, including the global occurrence of MPs in SS, impacts of MPs on SS and vice versa, MPs toxicity through SS application, and development in MPs management from SS. After extracting the relevant knowledge, the authors synthesized and analyzed the quantitative and qualitative findings, which are summarized in tables and figures of this review.

Sources and Prevalence of Microplastics in Sewage Sludge

Based on the generation, MPs in the SS also can be classified into two categories, i.e., primary and secondary MPs [45]. Primary MPs encompass the MPs deliberately produced by the industries because of their commercial viability, such as plastic granules in industrial feedstock, microfibers in textile industries, and microbeads in cosmetic industries [46]. These MPs are used in broader applications in our daily life. For example, manufacturing of small-sized microbeads to utilize in PCPs (toothpaste, nail polish, shampoo, shaving soaps, skin and hair care, insect repellents, creams and lotions, and face cleaners), cosmetics, pharmaceuticals (nanocapsules), detergents, paints, polymeric cement, dermal exfoliators, polymeric flocculants, sandblasting shots, coatings, polishing agents, and fluid absorbents in horticulture [47–49]. In addition, the small-colored pellets of plastic, known as industrial feedstock, are produced by the plastics industry to be melted down and molded to form larger plastic artifacts—the unintentional release of such particles is considered to contribute significantly to the occurrence of MPs in the sewage. Additionally, the manufacturing of microfibers (MFs) for garment production also contributes significantly. Therefore, the production and utilization of primary MPs heavily contribute to domestic and industrial sludge.

In contrast, secondary MPs are smaller-sized uneven plastic pieces that have been produced inadvertently due to the degradation of larger plastic particles such as plastic bags, bottles, crates, pipes, synthetic cloths, packaging covers, ropes, and nets [48, 50]. Over a period, these large pieces of plastic litter will degrade through the biotic or abiotic process. In abiotic degradation, the breakdown of larger particles occurs through various means such as photo-oxidation (UV light), thermal (temperature), and mechanical forces (such as compression, tension, abrasion). In contrast, biotics includes the breakdown of biodegradable plastic material through microbial actions [50]. Furthermore, fragmentation and weathering can alter the physicochemical properties of MPs and cause the leaching of additives. The other substantial sources of secondary MPs in domestic sewage are the laundering of synthetic textiles in washing machines, flushable wet wipes, and sanitary towels [50, 51]. Domestic laundry of synthetic textiles is considered a significant source of secondary MPs, specifically shredded MFs [52]. It is estimated that North America contributes about 880 t/yr MFs emission, followed by Finland (150 t/yr) and Norway (100–600 t/yr), through laundries and household washing [53]. These shredded MFs travel from a washing machine to domestic wastewater and then enter a municipal WWTPs. Furthermore, automobile rubber tires generate a considerable amount of MPs due to mechanical wear entering the sewerage streams through the rainwater run-off. Moreover, accumulated plastics in landfills also generate a significant amount of secondary MPs, which can be transported to sewage through landfill leachates [54, 55].

Abundance and Fate of Microplastics in Sewage Sludge from Municipal Wastewater Treatment Plants

Occurrence of Microplastics in Sewage Sludge

It is difficult to compare the results of different studies effectively because of the lack of standard procedures for SS sample collection, sorting, pretreatment, and characterization of MPs. Apparently, the Table 1 suggests that the concentration of MPs in SS largely varies from 1000 to 314,000 MPs particles/kg in dry weight of SS independent of any region or country. This considerable variation in the quantification of MPs is caused by different factors such as population density of the region, level of economic development, type of sludge [33], plastic consumption, daily habits of citizens, diversity of urbanization [49], type of treatment units in WWTP [46], seasonality, diurnally, pretreatment method used, sludge treatment methods [56, 57], and waste management activities [58]. Moreover, it is challenging to compare different studies due to the limitation of the exact quantification of MPs from the complex matrix of SS [45, 46]. Furthermore, the lack of uniform terminologies for identifying and classifying MPs makes it challenging to compare studies based on shape, size, color, and type [57].

Factors Affecting Physical and Chemical Characteristics of Microplastics in Sewage Sludge

Most of the studies reported that MFs are the dominant shape, followed by fragments. The possible sources of MFs are the laundering of synthetic textile and industrial products [51]. In contrast, fragments are generated from plastic products used in everyday activities such as packaging bags and from resin-type plastics used in industrial activities, including adhesion agents, foam boards, and insulation boards [68–70]. Additionally, the sources of granular MPs are microbeads in pharmaceuticals and personal care products (PPCPs), and automobile and electronic manufacturing industries [45]. The films are produced from packaging products (including ready-to-eat boxes, preservative films, disposal, and drinking water bottles) and industrial-grade films used in magnetic tapes, photographic films, and X-ray plates [49]. MFs of polyesters (PES), acrylics, and polypropylene (PP) are significantly observed in SS, which are extensively used in the textile industry [58, 59].

Furthermore, several studies reported a wide variety of polymer types in sludge, including PE, PP, polystyrene (PS), PES, nylons, polyacrylamide, PA, polyvinyl acetate (PVAc), polyethylene terephthalate (PET), polyvinyl chloride (PVC), PC, polytetrafluoroethylene (PTFE), ethylene

Table 1 Global occurrence of microplastics in sewage sludge with particle shape, size, and chemical characterization

Country	Population equivalence/served	Level of treatment	MP concentration in sludge (MP/kg d.w. of sludge)	Type of particle	Chemical identification	Size	References
Australia	0.23 to 0.7 million	Primary, secondary	• Primary sludge: $(15.90–45.70)\times10^3$; • Secondary sludge: $(37.80–46.10)\times10^3$; • Digested sludge: $(48.50–56.50)\times10^3$	Fibrous > fragments > granular	PET, PE, PP, Nylon	>25 µm	[55]
Canada	1.3 million	Primary, secondary	• Secondary sludge: $(4.4\pm2.9)\times10^3$; • Primary sludge: $(14.9\pm6.3)\times10^3$	**Primary:** fiber > fragment > foam > pellet; **Secondary:** fiber > fragment	NR	>1.0 µm	[46]
China	0.25 to 0.8 million	Primary, secondary, and tertiary (CAST, A^2O, MBR)	• Primary sludge: $(6.74–16.62)\times10^3$; • Secondary sludge: CAST sludge- $(17.91–19.47)\times10^3$; A^2O sludge-$(11.04–13.79)\times10^3$; • Tertiary sludge: MBR- $(5.88–13.07)\times10^3$; • Sludge cake: $(6.32–13.04)\times10^3$; • Total MPs: $(13.06–29.66)\times10^3$	Fragments > fibrous > granular > film	19 chemical species including PE, PP, PET, epoxy resin, MF resin, PVC, PC	• Fragments- L: 837–1494 µm and W: 527–981 µm; • Film- L: 828–1197 µm and W: 499–833 µm • Granules- L: 217–258 µm and W: 147–230 µm • Fibers- L: 1280–2804 µm	[49]
China	1 million	Primary, secondary, and tertiary (A^2O, UV)	• Secondary sludge: 890.68 ± 436.50 n/L; • Dry sludge: $(16.166\pm1.088)\times10^3$	Fibrous > fragments > microbeads	PET (20.22%), PP (21.48%), PVC (2.06%), nylon (4.44%), olefin (1.55%), acrylic (0.21%), PS (2.51%), and rayon (13.88%)	>25 µm	[59]
Finland	NR	Primary and secondary with MBR	• Activated sludge: $(23\pm4.20)\times10^3$; • Digested sludge: $(170\pm28.70)\times10^3$; • MBR sludge: $(27.3\pm4.70)\times10^3$	Fibrous > particle	Polyester (PES), PE, PA, and PP	>250 µm	[60]
Germany	0.007–0.21 million	Secondary (8), tertiary level (4); sludge sampling (6 locations)	• Dewatered sludge: $(1.00–24.00)\times10^3$	NR	PE, PP, PA, PS	<500 µm	[61]
Iran	0.10 million	Primary and secondary	• Primary sludge: $(214\pm16)\times10^3$; • Secondary sludge: $(206\pm34)\times10^3$; • Thickened sludge: $(200\pm13)\times10^3$; • Digested sludge: $(238\pm31)\times10^3$; • Dewatered sludge: $(129\pm17)\times10^3$	Fibrous > particle	(PES/PET), PP, acrylic, PA, PC, PS, PE	>37 µm	[62]
Italy	1.2 million	Primary, secondary, and tertiary (sedimentation, sand filter treatment and disinfection)	• Recycled activated sludge: $(113\pm57)\times10^3$ (microplastic particles: $(59.5\pm21.6)\times10^3$; microplastic fibers: $(53.3\pm48.9)\times10^3$)	Fibrous > films > fragments > Lines	13 polymeric species: Co-polymers of acrylonitrile-butadiene, PE, PES, PA, PU, polyacrylate, PTFE, PP, ethylene-acrylate, EP diene, styrene–isoprene	>0.01 µm	[63]

Table 1 (continued)

Country	Population equivalence/served	Level of treatment	MP concentration in sludge (MP/kg d.w. of sludge)	Type of particle	Chemical identification	Size	References
Mauritius	NA	Advanced primary, tertiary	• Wet sludge: $(2.25–11.30) \times 10^3$ particles/kg	Fibrous > fragments > foam > sphere	PE spheres, EVA foams, and cotton-PA (polyamide) fibers	• Fibers: 0.25–4 mm length, • Fragments: 0.50–0.25 mm	[57]
Spain	0.3 million	Primary, secondary (A^2O)	• Primary + secondary sludge: $(314 \pm 145) \times 10^3$; • Heat dried sludge: $(302 \pm 83) \times 10^3$	Fibrous > fragments	PES fibers > acrylic fibers > PE > dyed cotton > PP Other polymers identified: PMMA, PCL, PU, PS	**Wet sludge:** • Fragments-L: 36–377 µm and W: 22–36 µm; • Fibers- L: 213–4716 µm W: 5–34 µm **Dry sludge:** • Fragments-L: 29–533 µm and W: 11–369 µm; • Fibers- L: 71–2224 µm W: 7–58 µm	[33]
Sweden	0.012 million	Primary, secondary	• Slightly dewatered sludge: $(16.70 \pm 1.96) \times 10^3$	Fiber > fragment > flakes	NR	>300 µm	[64]
Thailand	0.22 million	Primary, secondary, tertiary	• Secondary sludge: $(2.63 \pm 1.26) \times 10^4$	Fibrous > films > fragments	9 types of polymers: PET, PE, PP, PS, acrylics, EPDM, PU, PA, Alkyd	Largest group of MPs in the sewage sludge was of size fraction 0.05–0.5 mm	[65]
Turkey	2 million	Primary, secondary	• Dried sludge: 32×10^3	Fibrous > fragments > pellets	PC, PET, PES	1000–2000 µm (dominant fibers size in sludge)	[66]
UK	0.15 million	Primary, secondary	• Reception tank: 107.5×10^3; • Thickened sludge: 50.2×10^3; • Digestate sludge: 180.7×10^3; • Centrifugation feed tank sludge: 286.5×10^3; • Sludge cake: 97.2×10^3; • Pre lime cake: 74.7×10^3; • Lime cake: 37.7×10^3	Particle > fibers	21 different polymers dominated by PES, PVAc, and PE, polyacrylate, ABS, PVC	>50 µm (average length of fibers: 969.9 µm, and average particle size: 170.6 µm)	[10••]
USA	NR	Primary, secondary, tertiary treatment	• Sewage sludge: 1×10^3	NR	NR	45–400 µm	[67]

NR not reported

propylene diene monomer (EPDM), alkyds, polycaprolactone (PCL), urea formaldehyde (UF), melamine formaldehyde (MF), and epoxy resins [10••, 71]. MPs chemical composition in SS depends on the daily habits of plastic consumption, different population, commercial patterns, and industrial activities [49, 57]. Notably, PE (polyethylene) and PP are substantially used in the production of daily products such as PPCPs, bags, containers, and packaging films [72]. However, PVC is widely used in industries to make pipes, whereas polycarbonate (PC) has industrial applications in glass, packaging, and industries of medical apparatus. In addition, epoxy resins are widely used in the electronics and civil engineering industry, while MF resins are extensively applied to furniture and vehicles [49].

Factors Influencing Abundance of Microplastics in Sewage Sludge

The level of WWTPs also influences the abundance of MPs in sludge. In primary treatment, MPs removal depends on their characteristics such as size, density, and shape. MPs with higher density may get settled due to gravity, whereas lower density may float on the surface and later be separated by skimming followed by settling [50]. Alavian Petroody et al. [62] found microparticles with a size > 500 µm and lower density than water is efficiently separated from wastewater in the primary settling tank, while clarifier played an important role for separation of < 500 µm. In the secondary treatment, the amount of small-sized MPs increased because of the efficient removal of large-sized MPs in the preliminary and primary stages. However, higher MPs per gram of dry weight can be in secondary treatment than primary due to return activated sludge that carries a large number of MPs into the aeration tank leading to deposition in sludge [59, 62]. Besides, MPs abundance in SS can be influenced by the difference in the secondary treatment process to a certain degree. For instance, Yuan et al. [49] reported 1.76 times higher MPs dry abundance in the sludge of CAST (cyclic activated sludge technology) than improved anaerobic-anoxic–oxic technology (A^2O) owing to longer hydraulic retention time (HRT) and sludge retention time (SRT), provided sufficient contact between MPs and SS, which facilitated MPs accumulation and final sedimentation. Furthermore, Lares et al. [60] found membrane bioreactor (MBR) process had better MP removal efficiency compared to conventional activated sludge (CAS), ultimately leading to a slightly higher MPs concentration in MBR sludge than CAS sludge. In addition, Table 1 also suggests that the sludge treatment processes affect the abundance of MPs in SS [10••, 62], which is discussed in the "Effect of Sludge Treatment Processes on Abundance and Characteristics of Microplastics" section. In conclusion, municipal WWTPs are considered a source and recipient of MPs since they receive a substantial number of MPs but are unable to hold such particles and releases into SS and effluent [59, 66]. More than 90% of MPs are concentrated in SS, and the rest is discharged in the effluent. Therefore, it is imperative to have a detailed discussion on the sources and fate of MPs in sewage to identify the potential environmental risks (Fig. 1).

Fate of Microplastics in Sewage Sludge from Wastewater Treatment Plants to Its Application

As mentioned, MPs are transported to the WWTPs through various sources, including domestic and industrial sewage, surface run-off, and landfills. However, WWTPs are efficient in removing MPs from the wastewater. For instance, Iyare et al. [13] reviewed the efficiency of WWTPs in removing MPs and reported a 72% average removal from preliminary and primary treatment. Furthermore, on average, 88% and 94% of MPs were removed from secondary and tertiary WWTPs, respectively. Though the removal efficiency is high from wastewater, the majority of MPs are probably to concentrate in SS (> 98%) due to the high hydrophobicity of MPs which promotes adherence to the organic portion of SS [73, 74].

Moreover, wastewater contains a considerable amount of other chemical contaminants along with MPs, which include toxic heavy metals [75, 76], polychlorinated biphenyls (PCB), PPCPs, polyaromatic hydrocarbons (PAHs), perfluorinated surfactants, and endocrine disrupting compounds (EDCs) [77–79], which are not efficiently biodegraded or volatilized in WWTPs. Furthermore, wastewater also includes human pathogens including virus, ARGs, mobile genetic elements (MGEs), and antibiotic-resistant bacteria (ARB) [80, 81]. All these chemical and biological contaminants were found to be concentrated in SS [82–84]. Therefore, raw SS is treated before its application with different sludge treatment processes such as lime stabilization, anaerobic/aerobic digestion, composting, dewatering/thickening, and pasteurization [85].

Notably, conventional sludge treatment processes are ineffective in eliminating MPs [10••, 74]. Furthermore, these sludge treatment processes can influence the abundance and morphology of MPs, and their presence can impede the operation of sludge treatment processes, which is discussed in detail in the "Microplastics Presence in Different Conventional Sludge Treatment Processes: Mutual Impacts" section. Moreover, land application of MPs embedded sludge leads to long-term accumulation of MPs in soil; adversely affect the soil biophysical properties, flora, fauna, and microbial community, ultimately affecting the food web and soil ecological services [3•, 86]. Furthermore, MPs present in SS can interact with other chemical and biological contaminants

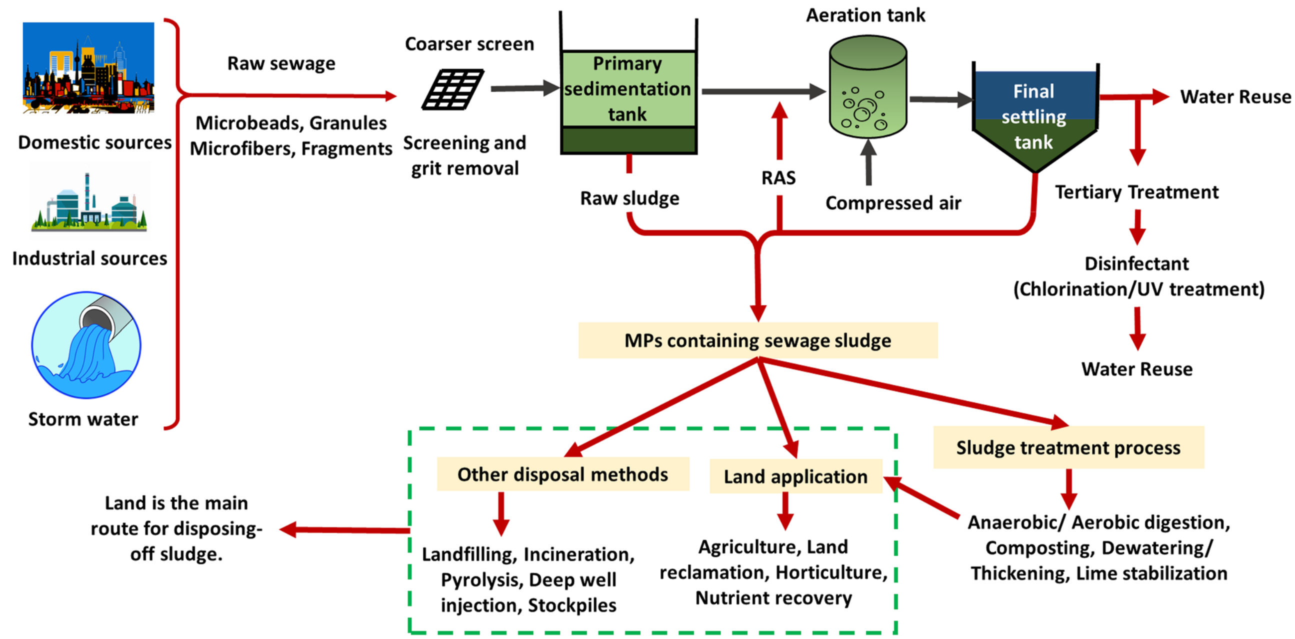

Fig. 1 Sources and fate of microplastics in sewage sludge

through the adsorption–desorption process and escalate the risk of human exposure, summarized in Fig. 2.

Microplastics Presence in Different Conventional Sludge Treatment Processes: Mutual Impacts

As aforementioned, several studies reported that raw SS contains many toxic substances. However, it also contains a substantial amount of organic matter and beneficial nutrients [87], promoting its land utilization instead of landfilling and incineration, particularly from an environmental and economic perspective. For that, SS is treated prior to its land application to reduce sludge volume, weight, and potential health risk while restoring its agronomic value and promoting a circular bioeconomy. The common sludge treatment processes include dewatering, thickening, mesophilic and thermophilic AD, lime/alkali stabilization, composting, and thermal treatments [25, 88, 89]. So far, limited studies have demonstrated the variation in the amount and morphology of MPs during the conventional sludge treatment process. Therefore, the current review collates state-of-the-art theoretical, experimental, and numerical evidence. Table 2 summarizes the effect of different sludge treatment processes on MPs.

Effect of Sludge Treatment Processes on Abundance and Characteristics of Microplastics

Limited studies investigated the effect of different conventional sludge treatment processes on MPs concentration and characterization. In order to assess the impact, Mahon et al. [90••] analyzed the impact of different sludge treatment processes (lime stabilization processes, full-scale AD, and thermal drying) on MPs with a minimum limit of detection (LOD) of 250 µm. They reported the occurrence of smaller size MPs in final sludge product from the lime stabilization process, which was ascribed to flaking and shredding of MPs resultant from elevated temperature, mechanical mixing, and pH. In addition, the authors reported the lowest number of MPs from anaerobically digested sludge compared to lime stabilized sludge, suggesting AD might reduce the MPs abundance. In contrast, Harley-Nyang et al. [10••] reported more MPs in digested sludge cake than the lime stabilized cake through visual identification because they adopted 50 µm minimum LOD; furthermore, samples were collected across the whole treatment streams, and the different extraction process was performed. But Horton et al. [27] also reported less MPs abundance in AD when compared with lime stabilization despite adopting a minimum LOD of 25–178 µm, which was attributed to the automated analytical method, eliminated human bias of counting, and enhanced sensitivity. Therefore,

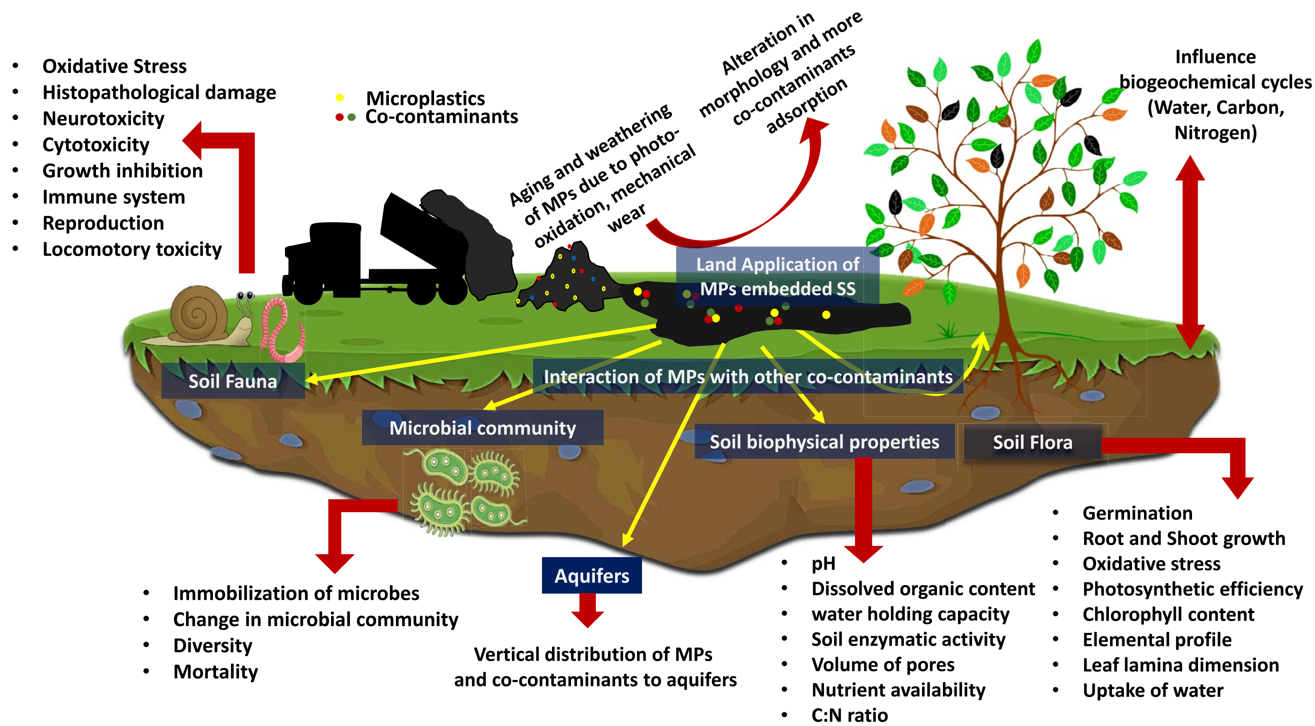

Fig. 2 Impacts of microplastics on different components of terrestrial ecosystem

more research is required with a uniform extraction process and characterization method to provide a clear scenario of whether MP concentrations are elevated in the AD process.

Some studies reported that dewatering could reduce the MPs abundance. For instance, Alavian Petroody et al. [62] reported a more than 50% reduction in MPs numbers after dewatering. They explained that attached MPs were released during the probable destruction of flocs in the digestive process that returned to WWTP with released water. Moreover, dewatering and pretreatment of sludge can cause under-reporting of MPs abundance. For instance, Li et al. [73] reported that sludge from different dewatering units had different average MPs concentrations (belt-type group > filter pressure > centrifuged group > plate-frame group). However, centrifugation and plate-frame filter pressing may under-estimate the MPs abundance. Like, in the centrifugation process, low-density MPs remain in the water phase and may be released back into the water. Lime is often added to reduce moisture content during plate-frame filter pressing, complicating the MPs analysis. Application of different pretreatments of sludge before MP analysis can influence MPs leading to underestimation of abundance. Like, Li et al. [91] reported that pretreatment can influence the size, surface morphology, and adsorption potential, highest influence observed with pretreatment with alkali (NaOH), followed by the high concentration of acids, low concentration of acids, then H_2O_2 and Fenton.

Additionally, different sludge treatment processes alter the surface morphology of MPs. In this context, Mahon et al. [90••] demonstrated variation in surface morphologies which varied with the treatment types. The authors reported a more shredded and flaked appearance of MPs in lime stabilized sludge resulting from elevated temperature and mechanical mixing. In addition, they reported the presence of deep cleavage in MPs in anaerobically digested sludge, whereas more melding, wrinkling, and fracturing in MPs in thermal dried sludge. Similarly, in a recent study, Li et al. [28] compared the impact of sludge treatment processes, i.e., AD, thermal hydrolysis, thermal drying, and aerobic composting on MPs. Their results showed an increase in abundance of MPs after thermal hydrolysis in the sludge, indicating breaking into smaller particles, alteration in micro-morphology of MPs after AD treatment, and cracking in MPs after thermal heating and drying. Moreover, more distorted edges after thermal drying and aerobic composting damaged the surface of MPs and led to erosion. This uneven, rough surface and more fragmentation of MPs by sludge treatment processes will increase the surface area-to-volume ratio, further allowing more adhering of organic pollutants on MPs surface; smaller size increases the bioavailability of

Table 2 Impacts of different sludge treatment processes on microplastics

Process	Impact on MPs	References
Lime stabilization	• Smaller size MPs due to elevated temperature, mechanical mixing, and pH • Shredded and flaked appearance	[90••]
AD	• Less MPs abundance in AD sludge when compared with lime stabilized sludge • Presence of deep cleavage in MPs • Alteration in micro-morphology of MPs	[27, 28, 90••]
Dewatering	• More than 50% reduction in MPs numbers after dewatering might be due to probable destruction of flocs in the digestive process that returned to WWTP with rejected water • Different average MPs concentrations (belt-type group > filter pressure > centrifuged group > plate-frame group)	[62, 73]
Pretreatment	• Pretreatment can influence the size, surface morphology, and adsorption potential • NaOH > high concentration of acids > low concentration of acids > H_2O_2 and Fenton	[91]
Thermal drying	• More melding, wrinkling, and fracturing in MPs in thermal dried sludge • Cracking in MPs after thermal heating and drying	[28, 90••]
Thermal hydrolysis	• An increase in MPs number	[28]

such particles [90••]. Similarly, Zhang et al. [92] reported an increase in extracellular ARGs after MFs exposure during AD owing to an increase in host numbers with increased exposure to MFs suggested enhanced horizontal transformation. This can accelerate the transportation of ARGs in the environment during sludge utilization and disposal.

Effect of Microplastics on Different Sludge Treatment Processes

Apart from the studies that investigated the impact of sludge treatment processes on MPs, the operation of sludge treatment processes is observed to be influenced by the abundance of MPs, which is summarized in Table 3.

Most studies investigated the impacts of MP types, sizes, and concentrations on methane and hydrogen production through AD. Different MNPs exhibit different impacts on AD based on their physical and chemical characteristics. Some MPs can generate reactive oxygen species (ROS), such as OH• and H_2O_2, which induces cytotoxicity and reduces cell viability. For instance, Wei et al. [96] reported a significant reduction of 12.4–27.5% in methane production when AD was amended with PE-MPs (concentration: 100 and 200 particles/g-TS). This reduction was attributed to ROS generation despite the leaching of additive ATBC, which reduced cell viability by 7.6–15.6%, inhibiting sludge acidification, hydrolysis, and methanogenesis with shifting of the microbial community against AD. In addition, Zhang et al. [101] reported a population decline of key methanogens and acidogens such as *Longilinea* sp., *Methanobacterium* sp., *Methanosaeta* sp., and *Levilinea* sp. on long-term exposure to PET MPs which suppressed both methanization and acidification due to dibutyl phthalate (DBP) leachate and induction of excessive oxidative stress. However, the mechanism of ROS generation in anaerobic conditions is still ambiguous; thus, further research is required to understand ROS production under anaerobic conditions.

Furthermore, additives leached may inhibit or enhance hydrogen and methane production from AD through different mechanisms. For instance, Li et al. [93] reported a reduction in methane production in AD on exposure to PES-MPs ascribing incomplete digestion, which led to inhibition of methane production potential and observed no significant impact on the microbial community. Additionally, the leaching of toxic chemicals affects digestion efficiency. For instance, leaching of bisphenol-A (BPA) from PVC-MPs facilitated rupturing of the microbial cell wall. It caused an inhibitory effect on the hydrolysis-acidification process, leading to decreased methane production when AD was amended with PVC (20–60 particles/gTS) [95].

In contrast, Chen et al. [94] stated increase in production of methane during AD of WAS on exposure of polyamide-6 (PA6) MPs due to leachate caprolactone (CPL), which binds with enzyme molecules to alter the active site, thereby enhancing catalytic enzyme activity (α-glucosidase, F_{420}, protease, butyrate kinase, and acetate kinase), affinity with the substrate, and promoting methanogenesis and acidification processes. Furthermore, Tang et al. [102] demonstrated that the high concentration of additives such as dimethyl phthalate (DMP) fostered disintegration of sludge in AD by expediting cell lysis and improved methanogens development by providing more biological substance for methanogenesis. Moreover, the authors reported that a high concentration of DMP could contribute spreading of ARGs in the antibiotic stress such as erythromycin, tetracycline, and sulphadimidine. In the same way, Pittura et al. [99] reported a reduction in methanogenesis by 4–58% in pilot-scale upflow anaerobic sludge blanket reactor (UASB) treating municipal wastewater and stated the strong association of MPs toxicity with their leachates mostly additives, particularly antioxidants in case of PP and PS, for the reduction in the process.

Moreover, MPs negatively influence methane and hydrogen production by changing the protein structures, affecting

Table 3 Impacts of microplastics on different sludge treatment processes

Process	MPs	Operating conditions (C: Concentration and T:Time)	Major findings	References
AD	PES	C: 0–200,000 PES-particle/kg activated sludge; T: 59 days	Methane production was reduced by 88.53 ± 0.5 to 95.08 ± 0.5% with slight inhibition in AD with improvement in dewaterability	[93]
AD	PA6	C: 5–50 particles/g-TS; T: 45 days	PA6 enhanced methane production by 39.5%	[94]
AD	PVC	C: 20–60 particles/g-TS; T: 45 days	PVC inhibited methane production by 75.8 ± 0.2 to 90.6 ± 0.3%	[95]
AD	PE	C: 100 and 200 particles/g-TS; T: 44 days	12.4–27.5% reduction in methane production during short-term exposure	[96]
Aerobic digestion	PET	C: 15 particles/L of PET MPs; T: 95 days	Inhibition of WAS aerobic digestion by 10.9 ± 0.1% when MPs spiked into wastewater; suppressing WAS aerobic digestion by 28.9 ± 0.1% when MPs were directly added to the digester	[30]
Dewaterability	PS, PE, PVC	C: 1–300 mg/L of MPs; T: 60 days	29.6–47.7% reduction in the sludge dewaterability was observed	[97]
Anaerobic fermentation	PS	C: 30–90 particles/g-TS; T: 28 days	Low PS concentrations (30 particles/g total solid) significantly increased the production of volatile fatty acids by 112.8 ± 2.4%. In contrast, high concentrations of PS (90 particles/g total solid) significantly decreased volatile fatty acids production by 83.01 ± 0.76%	[98]
Anaerobic digestion (UASB)	PP	C: 5 PP-MPs/g-TS to 50 PP-MPs/g-TS	4–58% decrease in methanogenic activity at 18–50 PP-MPs/g-TS	[99]
Alkaline anaerobic fermentation	PET	C: 10, 30, and 60 particles/g-TS of PET MPs; T: 21 days	11.6 ± 0.1 to 29.3 ± 0.1% reduction in hydrogen production from WAS throughout the 21 days test period	[100]

the key enzymes and functional genes. In particular, Feng et al. [103] demonstrated that 20 μg/L of cationic PS could induce higher inhibition in methane production when compared with 100 μg/L of anionic PS and reported that nanoscale PS could penetrate granular sludge and alter the protein secondary structure of extracellular polymeric substance (EPS). Moreover, the authors reported a decrease in the abundance of functional genes for methanogenesis (*ACAS* and *mcrA*) after exposure to PS-NPs. Zhang et al. [104] reported a significant reduction of 19.3% in methane production with NPs. In contrast, only a reduction of 17.9% was observed with MPs under a concentration of 0.25 g/L, concluding that NPs have slightly more inhibition capacity in methane production than the MPs. Venkiteshwaran et al. [105] reported an increase in methane production when waste polyhydroxy butyrate (PHB) was fed to an anaerobic digester with no change in digester function. However, a shift in bacterial communities was observed with an initial reduction in lag time before the methane production, which then increased when PHB co-digestion began when bioplastic PHB was pretreated at high temperature and pH.

In summary, various inhibition/enhancement mechanisms involved in the deterioration of AD performance include (a) producing ROS, (b) penetrating/damaging microbial cells, (c) releasing toxic additives, (d) changing protein structure in granular sludge, and (e) affecting key enzymes activities and functional genes [106••]. However, the effect of different MPs on AD is uncommon and cannot be interpreted with a similar mechanism; more studies are required to understand the mechanisms crucial for sustainable sludge management. Similarly, limited studies investigated the impact of MPs on the aerobic digestion process. Wei et al. [30] demonstrated that MPs influence the aerobic digestion of WAS through two different entry routes on exposure to PET MPs. The authors reported a decrease in hydrolysis and an increase in WAS solubilization when MPs spiked into wastewater, owing to the alteration in WAS characteristics. In contrast, when MPs were directly added to the digester, severe inhibition on hydrolysis was observed along with fewer impacts on solubilization, causing a lot more severe impact on aerobic digestion. In addition, Wei et al. [30] reported a reduction in key bacteria populations involved in the aerobic digestion process (e.g., *Chitinophagaceae*, *Saprospiraceae*, and *Xanthomonadaceae*) through releasing toxic chemicals or/and inducing oxidative stress. Furthermore, the occurrence of MPs in the aerobic digestion process decreases ARGs removal efficiency owing to an increased abundance of bacterial hosts of ARGs with the addition of MPs, which emphasizes the potential threat caused by entering more ARGs into the local environment during sludge utilization [107].

Likewise, the effect of chronic exposure of MPs on sludge dewaterability was investigated by [97]. The authors observed size and polymer-dependent effects on the dewaterability of sludge. Dramatic reduction in the sludge dewaterability was observed due to the physical crushing of flocs by large MPs, whereas in the case of PVC-MPs, plastic additives lead to a certain level of cytotoxicity. In contrast, the authors reported reduced dewatering performance on exposure to small size PS due to alteration in the spatial distribution and composition of EPS by the decreased population of key organisms, impeding microbial activity, affecting the final sludge disposal [97]. Furthermore, anaerobic fermentation, which is crucial for sustainable utilization of WAS, was affected with MPs based on the types and concentrations. Compared to the control, Zheng et al. [98] reported an increase in the production of volatile fatty acids (VFAs) to $112.8 \pm 2.4\%$ on exposure to low PS concentration due to enzymatic activity and enhancement in solubilization, whereas higher concentration of PS led to a decrease in the production of volatile fatty acids due to inactive microbial actions because of ROS production, synergistic toxicity of aged MPs with external contaminants, and excess sodium dodecyl sulfate (SDS). Wei et al. [100] reported repression of hydrogen production under exposure to PET MPs in alkaline anaerobic fermentation due to suppression of sludge hydrolysis, acetogenesis, and acidogenesis with inhibition of methanogenesis and homoacetogenesis under alkaline conditions. Furthermore, their results showed a reduction in the abundance of hydrogen producers, i.e., *Fonticella* sp., *Bacteroides sp.*, and *Tissierella sp.*, while the presence of hydrogen consumers, i.e., *Acetoanaerobium* sp., *Methanosaeta* sp., and *Methanobacterium* sp., was hardly observed, suggesting leaching of DBP and induction of ROS were the reasons for loss of activity of anaerobic microbes.

Strategic Drift in Sludge Treatment Processes to Reduce Microplastics Contamination from Sewage Sludge

As mentioned in the previous section, that conventional sludge treatment technologies (including dewatering, drying, thickening, lime stabilization, anaerobic and aerobic digestion, and composting) are ineffectual in MPs removal. Moreover, the conventional processes are accountable for increased small-sized MPs and changed surface morphology of MPs in sludge which facilitates more co-contaminant adsorption, eventually adversely influencing the soil biota and its ecosystem functioning. However, research to develop advanced technologies to remove MPs from SS efficiently is at a nascent stage. It has progressed gradually, with few articles reported effective MPs mitigation strategies from SS. This section collates all such articles, compares the performance of the state-of-the-art technologies, and discusses their mechanisms for MP removal from SS, which is also summarized in Table 4. Furthermore, Fig. 3 provides a synopsis regarding strategic drift in sludge treatment processes from conventional to advanced treatment processes in reducing MPs concentration from sewage sludge.

Thermo-Chemical Methods

Thermochemical treatment of SS is appealing in terms of reducing sludge volume, producing a wide range of by-products, destroying harmful pathogens, efficiently removing micropollutants, and considerable energy recovery. In addition, further utilization of additives and catalysts in these thermochemical processes can increase the quantity and quality of by-products, gaining importance and interest [108]. This subsection collates few key studies which reported the efficient removal of MPs by the thermochemical treatment of SS.

Pyrolysis is one of the popular sludge treatments and valorization method in which SS decomposes at an enhanced temperature under anaerobic conditions, resulting in high calorific value liquid and gaseous products, along with biochar which can be used for soil fertilization [109]. It is reported that pyrolysis can effectively mitigate MPs contamination in sludge. For instance, Ni et al. [41•] reported a reduction of 99.1–99.4% MPs concentration in the residues at 350 °C and 99.8% reduction at 450 °C when MPs embedded SS was pyrolyzed in vacuum tubular furnace at different temperatures (*conditions*-temperatures: 150, 250, 350, 450, 500 °C; heating rate: 10 °C /min; MPs: PE, PP, PS, PA; holding time: 30 min). The pyrolytic mechanism of non-degradable plastics is categorized into four categories, i.e., end-chain scission, random-chain scission, chain stripping, and cross-linking [110]. The process initiates with random or end-chain scission of plastics, leading to depolymerization, generating more chain-terminus radicals, and further producing short-chain liquid products and ashes with increased temperature. In addition, the authors recommended 450 °C for pyrolysis because, at low temperatures, reactions between MPs and organics can generate new MPs and biochar with a high amount of PAHs. In contrast, incinerating plastic-embedded sludge at elevated

Table 4 Selected studies on microplastics removal from sewage sludge through advanced sludge treatment technologies

Technology	Operating conditions	Removal (%)	Mechanism	Advantages	Limitations	References
Pyrolysis	Pyrolysis of MPs embedded SS at different temperatures, i.e., 150–500 °C, with a holding time: 30 min and heating rate: 10 °C/min. (MPs: PE, PP, PS, PA)	99.1–99.4% at 350 °C and 99.8% at 450 °C	Random/end chain scission of plastics followed by generation of chain terminus radicals, and production of short chain liquid products and ashes with increased temperature	No release of PAHs in biochar, dioxins, furans, and PCBs at 450 °C with effective removal	Pretreatment of sludge is required to dry it before utilizing pyrolysis technology	[16, 41•]
Hyperthermophilic composting	SS after 45 days of composting treatment sludge-based MPs at high temperature, which they demonstrated in full scale (200t)	43.70%	In situ biodegradation of sludge-based MPs	Efficient MPs removal with resource recycling, easy operation, short processing time, little residue, and odor emission	Organic mineralization increases with a decrease in organic content leading to less fertilizer efficiency; oily, lipid, or salinity-rich solid waste can cause system instability; human safety of hyperthermophilic composting microbes must be addressed before large-scale application	[29•, 111]
Hydrothermal carbonization	Different temperatures, i.e., 180–260 °C at a heating rate of 10 °C/min for 3 h	79.71% at 260 °C	Depends on the chemical type of MPs: PET-MPs: hydrolysis of PET-MPs dur to presence of hydrophilic group PP-MPs: polymer random chain scission	Efficient degradation of MPs in sludge with reduced environmental risk of sludge as an important carrier of MPs, less harmful emission compared to pyrolysis	Complete degradation of MPs in the sludge cannot be achieved in a single HTC, and the residual MPs (NPs) remain a potential threat to the natural environment	[16]
Hydrothermal liquefaction	Continuous HTL process operated at supercritical conditions (Temp: 400 °C; pressure: 30 MPa)	97% as plastic mass and 76% as particle number	• Dilution of polymer phase while shifting the pyrolysis selectivity from bimolecular hydrogen abstraction and addition to unimolecular β-scission • Initiation of proton transfer reactions between polymer and supercritical water through hydrogen bonding facilitating the formation of carbocations serving as an initiator to activate C–C cleavage in the polymeric chains	Involves the use of wet biomass, no excess energy for drying feedstock; carbon- and cost-efficient way; efficient removal of micropollutants	Decreasing particle size increases the threat of a high concentration of nanoplastics that are more harmful than MPs	[42]

Table 4 (continued)

Technology	Operating conditions	Removal (%)	Mechanism	Advantages	Limitations	References
Vermi-wetlands	The concentrations of 1 µm, 100 µm, and 500 µm PMMA MPs were added to the excess sludge and treated through constructed Vermi-wetland. (Earthworm species: *E. fetida*; wetland plant species of *A. calamus*)	100% (500 µm), 95.44–99.52% (100 µm), and 86.62–95.69% (1 µm MPs)	Synergistic action among earthworms, microorganisms, and plants; an interception effect responsible for MPs elimination in excess sludge in vermi-wetlands	Effective in MPs removal, economical, eco-friendly, and greater ability to stabilize and reduce sludge	High MPs concentration can hinder degradation, and can lead to oxidative stress and neurotoxicity in earthworms	[43, 112]
Enzymatic degradation	Degradation of HDPE beads by three hydrolytic enzymes lipase (Rhizopus oryzae), cellulase (*Trichoderma reesei*), and protease (*Aspergillus oryzae*) at various temperatures	4% initial bead mass was removed with protease (88 mg/L) at 55 °C in a 3-day batch experiment. Moreover, degradation up to 95% of HDPE beads in 20 days of AD retention time (mesophilic conditions) (through simulations)	Adsorption of enzymes on the polymeric surface, followed by hydrolysis/hydroperoxidation of bonds	Efficient MPs removal at lab-scale experiments	Enzymes utilization in large-scale reactions is not economical because the enzymes are hard to be recovered or reused with relatively short lifetimes; further improvement is needed on parameters such as recyclability, efficiency, and robustness	[44, 113]
Biodegradation	Biodegradation of 100 mg of PS-MPs through co-culture of PS-MPs and conventional thermophilic and hyperthermophilic composting inoculum in 50 mL of liquid carbon-free basal medium and 10 mL cell suspension. (Conventional composting: 40 °C; hyperthermophilic composting: 70 °C; incubation period: 8 weeks; shaking at 180 rpm)	Hyperthermophilic composting inoculum 7.3% degradation at 70 °C, which is 6.6 times more than conventional composting	Facilitation of more microbial deposition fostering biodegradation due to changes in operating conditions such as high composting temperature accountable for decreasing the hydrophobicity of MPs by introducing C-O and C=O groups	Low investment, inexpensive, low operating cost, easy operation, no secondary pollutant generation, and flexible to handle	Controlled environmental conditions and nutrient requirement, difficult to scale up batch and pilot scale studies to large scale, time consuming as compared to other techniques, aggregation of microbial assemblage on MPs surface, lack of reproducibility, difficult to find suitable microbial community	[29•, 114–116]

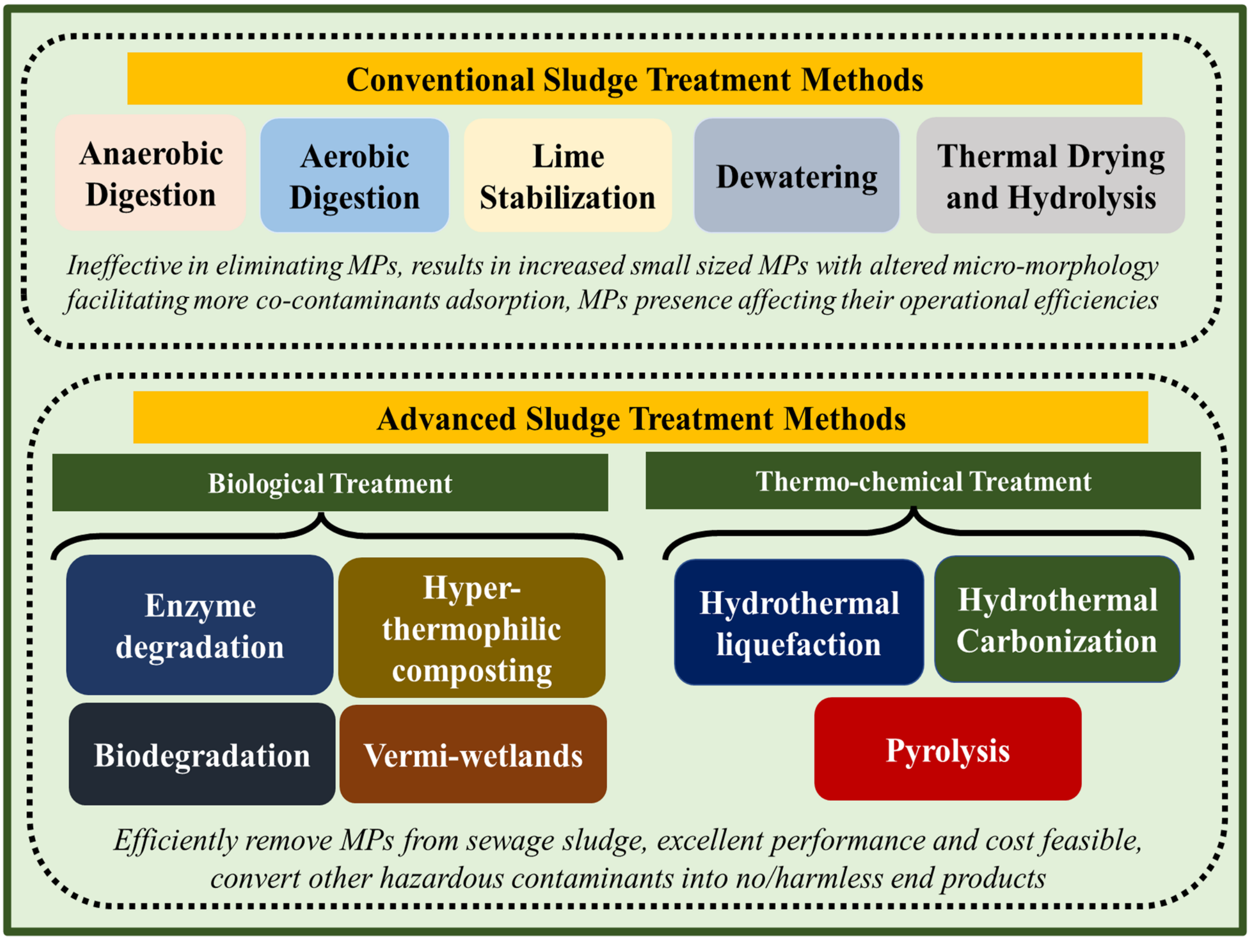

Fig. 3 Strategic drift in sludge treatment processes to reduce MPs from sewage sludge

temperatures can generate more harmful emissions such as furans, dioxins, and polychlorinated biphenyls [41•]. Nevertheless, sludge pretreatment is required before utilizing pyrolysis technology, as sludge needs to be dried, increasing process costs [16].

Hydrothermal carbonization is another effective technology for removing MPs from sludge. It is thermochemical technology that converts SS into rich carbonaceous hydrochar in the presence of water, and under moderate conditions (temperature: 180–260 °C; and pressure: autogenous pressure for several hours), application of hydrochar can reduce N_2O emission, promote microbial immobilization of N, and reduce mineral-N content in soil [117, 118]. In this process, temperature plays a significant role in removing MPs from sludge. Xu and Bai [16] reported that HTC reduced MPs abundance by 79.71% at 260 °C, with complete removal of MPs, including PET, PA, polyurethane (PU), and PS, from the sludge, while PP and PE reduced by 79.34% and 55.93%, respectively. An in-depth mechanistic analysis of HTC treatment was performed with two representative polymers such as PET-MPs to represent condensation polymers and PP-MPs to addition polymers. The authors reported that hydrolysis of PET-MPs to monomer due to the occurrence of hydrophilic groups in the molecular chains could be responsible for decomposition. In contrast, PP-MPs demonstrated an increase in molecular weight at low HTC temperature (< 220 °C), whereas polymer random chain scission could be the dominant mechanism in degradation at high pyrolytic temperature. However, the complete degradation of MPs in the sludge cannot be achieved in a single HTC, which is a disadvantage. Moreover, the residual MPs (generally NPs) emerge as a new potential threat to the natural environment.

Hydrothermal liquefaction is another promising route to remove MPs from sludge. In this thermochemical process, wet sludge is treated at 250–400 °C temperature and 4–22 MPa pressure and converted into valuable bio-crude with reduced risk by conversion of micro-pollutants [119]. In addition, the harsh condition utilized by the HTL process can significantly remove MPs from SS [120]. In a recent study, Chand et al. [42] demonstrated that the HTL process operated at supercritical conditions (temperature: 400 °C; pressure: 30 MPa) significantly reduced the amount of MPs

(97% as plastic mass and 76% as particle number). Additionally, the authors reported a decrease in particle size of more resistant polymers such as PE, PP, and polyurethane (PU) [42]. In this process, the molten polymer gets dissolved into supercritical water initially, which causes the polymeric phase to dilute and promotes polymer decomposition while shifting the pyrolysis selectivity from bimolecular hydrogen abstraction and addition to unimolecular β-scission. Further proton transfer reactions between polymer and supercritical water through hydrogen bonding become initiated, facilitating the formation of carbocations which serves as an initiator to activate C–C cleavage in the polymeric chains [121]. HTL involves the utilization of wet biomass; no excess energy is needed to dry the feedstock. Moreover, the process is carbon- and cost-efficient. However, particle size reduction can lead to the generation of NPs which are more harmful than MPs.

Biological Methods

Advanced biological treatment methods can be the ideal solution to eliminate SS-based MPs efficiently. However, different environmental parameters such as temperature, microbial diversity and availability, plastic chemical composition and concentration, and SS composition slow down the process of biological removal. Furthermore, biological methods are still under development, and more evidence is required to implement them on a larger scale [122]. This sub-section collates state-of-the-art evidence to remove MPs through advanced biological treatment of SS.

Hyperthermophilic composting of SS is one of the promising biological technique for effectively removing MPs. In this process, the composting temperature can reach up to 90 °C (due to thermophilic bacteria and without exogenous heating) during the fermentation process (temperature is 20–30 °C high compared to conventional composting) [88]. In addition, the process accelerates the removal of ARGs and MGEs (89% and 49% more than conventional composting) [123, 124]. In a recent study, Chen et al. [29•] reported that technology can reduce 43.7% of MPs from SS after 45 days of treatment through in situ biodegradation of sludge-based MPs; they demonstrated in full scale (200 tons). The authors reported a lower abundance of MPs ranging from 0.3 to 2.0 mm in hyperthermophilic composting with a higher abundance of < 0.3 mm MPs compared to conventional thermophilic composting. Furthermore, the process has some limitations, such as oily, lipid, or salinity-rich solid waste as substrate creates hyperthermophilic composting process unstable. In addition, organic mineralization increases while a decrease in organic content leads to fertilizer efficiency loss. The safety of hyperthermophilic microbes must be addressed concerning plants, animals, and humans before widespread application [123]. In addition, Xing et al. [125] also demonstrated the biochemical mechanism of accelerating the aging of MPs by free radicals during sludge composting and demonstrated that iron oxides adhering led to microbially mediated redox conversion of iron oxide, producing abundant reactive free radicals under anoxic/oxic alteration conditions of sludge composting, thereby damaging PS-MPs through oxidative degradation.

Vermi-wetlands, an environmentally friendly and economical method, use synergistic action among earthworms, microorganisms, and plants to recycle excess sludge [126]. In this process, plant rhizospheres play a substantial role in intercepting sludge flocs and providing enough oxygen for earthworms and microorganisms, thus enhancing sludge degradation. Moreover, the burrowing action of earthworms (involves ingestion, grinding, digestion, and excretion) facilitates microbial growth and maintains the aerobic condition in the system naturally, along with lessening the problem of wetlands clogging [127]. In a recent study, Nie et al. [43] studied the MPs removal performance by vermi-wetlands while recycling excess sludge under laboratory conditions using poly(methyl methacrylate) (PMMA) MPs. The authors demonstrated a reduction of 500 μm, 100 μm, and 1 μm MPs size by 100%, 95.44–99.52%, and 86.62–95.69%, respectively, suggesting an interception effect for MPs elimination in excess sludge in vermi-wetlands. Furthermore, the authors found that all the MPs in earthworm excrements, though detected only 1 μm MPs in digestive organs, indicating an essential role of bioturbation in MPs mobilization in vermi-wetlands. In a similar way, Ragoobur et al. [56] reported a significant reduction in the abundance of high-density polyethylene (HDPE) (146–500 μm), HDPE (1650–2000 μm), PP (1650–2000 μm), and PP (146–500 μm) by 56%, 31%, 22%, and 78% respectively after 14 weeks of vermicomposting. Fourier transform infrared (FTIR) spectra also revealed a 34% and 11% reduction in absorbance for alkane groups for HDPE and PP, respectively. However, the presence of MP was observed to have neither influence on the mortality of earthworms nor on the chemical properties during vermicomposting in terms of pH, C: N ratio, and electrical conductivity. Therefore, the process effectively removes low-concentration MPs from excess sludge and is economical and eco-friendly. Also, vermi-wetlands have the potential to stabilize and reduce sludge. However, higher MPs concentration can adversely affect the earthworms, ultimately reducing the degradation efficiency [43, 128].

Another treatment technology is enzymatic degradation. Plastics undergo enzymatic degradation in two stages. Initially, enzyme adsorption occurs on the polymeric surface, followed by hydrolysis/hydroperoxidation of bonds [129]. Most studies revealed that mainly hydrolytic enzymes are accountable for plastic degradation [130]. In a recent study, Elsayed and Kim [44] studied the enzymatic degradation of HDPE beads by three hydrolytic enzymes lipase

(*Rhizopus oryzae*), cellulase (*Trichoderma reesei*), and protease (*Aspergillus oryzae*) at various temperatures. The authors reported protease was the most effective in bead degradation as 4% initial bead mass was removed at thermophilic condition (55 °C) with the enzyme concentration of 88 mg/L in a 3-day batch experiment. In addition, with increasing enzyme concentration and temperature, HDPE beads degradation was observed to be increased. In addition, the authors stimulated PE beads degradation using the calibrated model by considering interactive decay reaction between two enzymes (protease and cellulase) in AD. The authors reported degradation of up to 95% of HDPE beads in 20 days of AD retention time (mesophilic conditions), portraying the significance of AD in MPs removal. However, utilizing enzymes on a large scale is not economical because enzymes are difficult to reuse or recover with a relatively shorter lifetime. Therefore, further improvement is needed on parameters such as recyclability, efficiency, and robustness [113].

Bioremediation also can play a substantial role in removing MPs from sludge by utilizing bacteria, lower eukaryotes, and higher eukaryotes [131]. For instance, Chen et al. [29•] performed PS-MPs biodegradation with inoculum at 70 °C for 56 days to understand the mechanism and reported 7.3% degradation, which was 6.6 times higher than conventional thermophilic composting inoculum at 40 °C. Highly efficient biodegradation in hyperthermophilic composting was accountable to the dominant presence of *Thermus*, *Geobacillus*, and *Bacillus*, which were effective at oxidizing plastic structures. Furthermore, the high composting temperature was accountable for decreasing the hydrophobicity of MPs by introducing $C-O$ and $C=O$ groups, which facilitated more microbial deposition and promoted biodegradation of MPs [132]. Furthermore, some studies revealed the potential of seagrasses to trap MPs. For instance, the common eelgrass *Zostera marina* has the potential to accumulate MPs of size 0.04 to 3.95 mm (mean = 0.95 mm ± 0.05 SE), average adherence of 4.50 ± 0.96 MPs in seagrass-associated biota, and 4.25 ± 0.59 MPs in blades [133]. Even Caribbean angiosperm *Thalassia testudinum* has the potential to encrust MPs on its blades by epibionts (Microfibers (MF): 3.69 ± 0.99 MF/blade; microbeads: 0.75 ± 0.25 beads/blade). In view of the preceding, Masiá et al. [134] mentioned that some seagrasses are susceptible to excessive nitrogen pollution, and some are resistant and could theoretically grow in affected areas such as outfall or sludge of WWTP.

Critical Insights and Future Perspectives

WWTPs are considered a source and recipient of MPs since they receive a substantial number of MPs but cannot hold such particles and release them into SS and effluent. Most of the MPs are likely to be concentrated in SS, along with other chemical and biological pollutants that are not biodegraded or volatilized from SS. Therefore, raw sewage sludge is treated prior to its environmental application through chemical, biological, and heat treatment processes, referred to as conventional sludge treatment technologies, to reduce sludge volume and potential health risks. However, these conventional treatment processes are inefficient in removing MPs from the SS and result in fragmentation of MPs along with the increased abundance of MNPs and altered micro-morphology such as smaller sized MPs in lime stabilized sludge, deep cleavage and altered micro-morphology in anaerobically digested sludge, and damaged surface in aerobically composted sludge. Categorically, more fragmentation of MPs with uneven and rough surface resulting from conventional treatment processes are accountable for increased surface area to volume ratio, increasing the adsorption potential towards toxic co-contaminants, escalating the environmental risk.

Furthermore, MPs presence can affect the operational efficiencies of different conventional sludge treatment processes, such as influencing the methane production from AD, inhibiting waste-activated sludge solubilization in aerobic digestion, and reducing sludge dewaterability in dewatering, affecting the production of VFAs from anaerobic fermentation. Conclusively, these conventional treatment processes are not only ineffective in removing MNPs from SS, but even escalate the environmental risk by influencing MPs abundance and micro-morphology; in addition, MPs presence can impede the operations of different conventional sludge treatment processes. Nowadays, various advanced sludge treatment technologies have been demonstrated to eliminate MPs from SS successfully with outstanding performance and cost feasibility. Although these advanced treatment methods confirmed higher MPs removal efficiencies and an excellent potential to transform other hazardous compounds into no/harmless end products, some of these technologies are still under development. More evidence is required to implement on a larger scale.

Furthermore, to understand the different aspects of MPs in SS and to address the challenges mentioned in the previous sections, the following issues deserve further attention (summarized in Fig. 4):

1. A standard procedure for SS sample collection, pretreatment, sorting, and characterization of MPs should be established to effectively compare the results of different studies. Uniform terminologies for identifying and classifying MPs must be established for better comparison, including polymer shape, size, color, and type. Moreover, results should be recorded in both units, i.e., particle no. and the weight of MPs detected per solid. Further

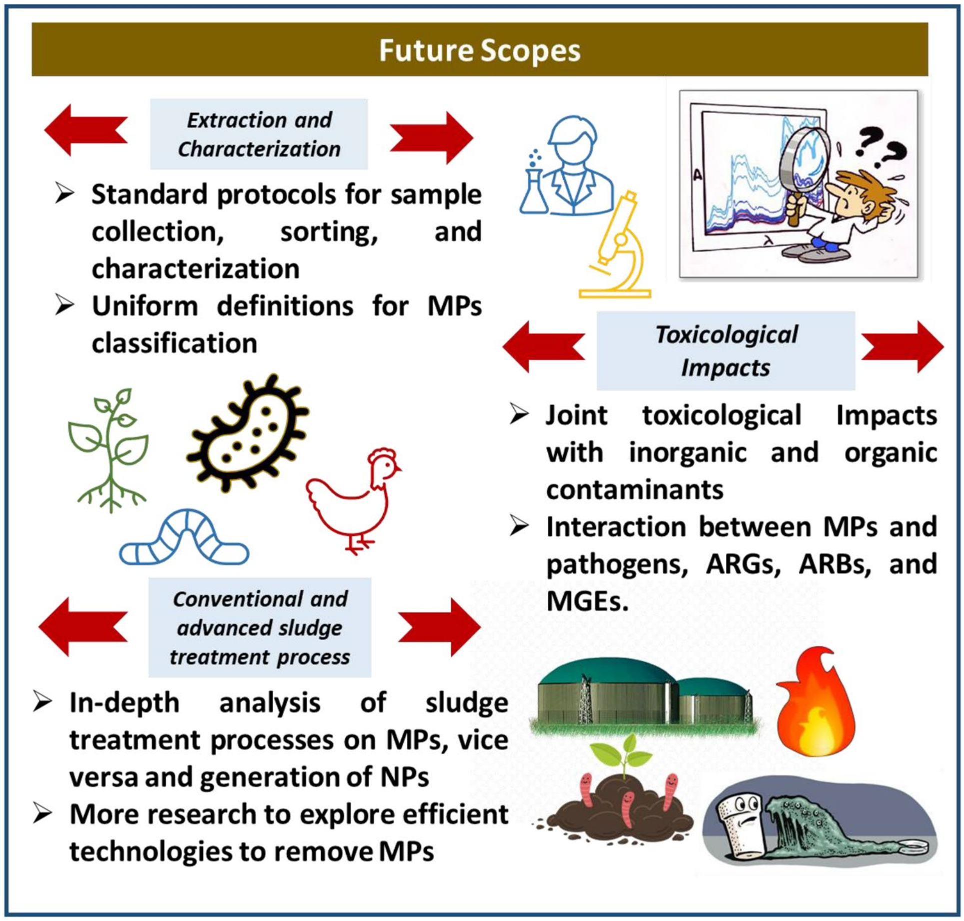

Fig. 4 Future directions for more research on various aspects related to MPs presence in sewage sludge

investigation should focus on extracting MPs from SS with higher separation efficiency without damaging their abundance and morphology.

2. The impacts of sludge treatment processes on MPs abundance and morphological characteristics are still studied superficially. Therefore, in-depth analysis is needed to understand better the impacts of different sludge treatment processes on the characteristics of MPs and comprehensive degradation mechanisms to boost the technology development. Moreover, different MPs' characteristics such as type, shape, and size can affect the efficiency of different sludge treatment processes. Limited studies described the impacts on sludge treatment processes other than anaerobic digestion. Hence, further investigation is required to holistically assess the impacts of different characteristics of MPs on sludge treatment processes for sustainable sludge management.
3. The toxicological impacts of MPs in soils should be urgently addressed concerning soil biophysical properties, plants, animals, microbes, crops, and humans involved via the food web. Additionally, more investigations on the joint environmental toxicity caused by the interaction between MPs and co-contaminants in SS must be prioritized; special emphasis must be given to the interaction of MPs with pathogens, antibiotics, ARGs, and ARBs.
4. Vigorous efforts should be made to explore different cost-effective and sustainable technologies for efficiently removing MPs from SS to prevent further accumulation of MPs on land through biosolid application. Moreover, the application of microbes and microbial consortiums must be considered to eliminate MPs from the SS. More research is needed to investigate about different microbial structures and communities that can degrade the MNPs efficiently. Additionally, studies to set the regulatory standards for discharging MPs pollutants on land through SS application must be prioritized to minimize uncontrolled discharge.

Conclusion

The objective of the review was to comprehensively analyze MPs in SS, by corroborating current state-of-the-knowledge, on different aspects, including the global occurrence of MPs in WWTP sludge, impacts of different conventional sludge treatment processes on MPs and vice versa, and efficiency of advanced treatment and upcycling technologies to eliminate MPs. The conclusions are as follows.

1. MPs abundance and characteristics in SS depend on different factors such as population density, development level of the region, citizen's daily habits, and treatment units in WWTPs. Furthermore, it is found that microfibers are the dominant shape of MPs in SS. Microfibers are (bio)available to wide variety of organisms owing to their small size and length-to-diameter ratio, which makes them more toxic than any other MPs shape.
2. Conventional sludge treatment processes can alter the abundance and morphological characteristics of MPs due to physical, chemical, and biological treatment. Altering micro-morphology can increase the surface area and even generate hydrophilicity, further escalating the risk of interaction with organic and inorganic co-contaminants. Based on limited studies, MPs can influence the efficiency of the sludge treatment process depending on their type, shape, size, and concentration.
3. In real-world scenario, accumulated MPs undergo aging or weathering due to photo-oxidation, abrasion, and biodegradation, which can alter the crystallinity, surface morphology, and oxygen-containing functional groups of MPs, leading to higher adsorption of co-contaminants present in SS. Bioaccumulation and uptake of which, by terrestrial organisms, is a substantial concern for trophic transfer in the agro-ecosystem.
4. Conventional sludge treatment processes are ineffectual in eliminating MPs from SS completely. The review critically discussed the performance analysis and mechanisms of various mitigation options recently published in the context of MP removal. Additionally, more research is needed to develop advanced technologies to remove MPs from SS and impede their accumulation on land. Furthermore, reduction in size during conventional and advanced treatments can lead to the generation of NPs. These NPs have strong fluidity and high specific surface area, which make them bioavailable to soil organism, and even allows absorption into plant cells, posing a substantial threat to the agro-ecosystem. Therefore, the problem should be addressed urgently; otherwise, it can permanently damage the agro-ecosystem.

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Compliance with Ethical Standards

Conflict of Interest The authors declare no competing interests.

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