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Microplastics in sewage sludge and municipal solid waste

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Abstract

Sewage sludge comes from wastewater treatment, whereas municipal solid waste comprises a broad range of non-liquid waste materials from households, businesses, and institutions. Both share similarities in composition, origins as waste products, and in their final use. Biosolids are the stable form of sewage sludge, while the organic fraction of municipal waste can be useful after being composted. The agricultural application of biosolids and composted organic wastes offers an appealing approach to sustainable agriculture, enriching soil fertility, enhancing water management, and reducing wastes. However, although soil amendment with wastes offers numerous benefits, their usage must be carefully managed to prevent any potential risks to the environment and human health. Regulations and guidelines are in place to ensure the safe handling of biosolids and composted fractions, but none of them is still considering the presence in them of microplastics (MP). The incorporation of MPs into soil matrixes may induce alterations in physical, chemical, and biological soil properties. It has also been shown that MPs can release a wide range of additives and may interfere with the fractionation and distribution of pollutants including trace metals. There is an urgent need to limit the dispersion of MPs due to the agricultural use of biosolids and the composted fraction of organic municipal waste. Especially the latter, due to their high production volume and higher plastic content compared to biosolids. Only by harnessing the potential of organic wastes, can agricultural systems move closer to achieving resource efficiency, and long-term productivity while contributing to sustainability goals.

1. Introduction

Sewage sludge and municipal solid waste are both waste materials generated by human activities that contain substances potentially constituting valuable resources. However, proper waste management practices are required to ensure the safety for human health and the environment of products obtained from sewage sludge or municipal solid waste. The generation of municipal solid waste and sewage sludge is globally increasing due to several factors. The first is population growth because more people mean more consumption and, consequently, more waste generation. Rapid urbanization also leads to increased waste generation due to the concentration of population in cities and towns because cities tend to produce higher volumes of waste compared to rural areas. Another important factor is economic growth and increased industrialization, that historically result in higher levels of waste production due to the introduction of new lifestyles and consump-

tion patterns characterized by increased consumption of goods, single-use items, and extensive use of packaging materials. While in developed countries a considerable effort is being paid to recycling and waste reduction, the situation is worse in developing countries, that often possess inadequate infrastructures and insufficient waste management practices. Finally, climate change and extreme weather events contribute to increase waste generation, for example, through the discharge of untreated sewage into water bodies.

The total generation of municipal solid waste in the EU in 2022 ranged from 301 kg per person per year in Romania to 835 kg in Austria (source: Eurostat), of which the organic fraction of municipal solid waste (OFMSW) represents 40–50 wt% (Kaza et al., 2018). While waste production increases at a slower rate during economic development, organic waste still constitutes a significant portion of a country's waste stream in developed countries (Chen et al., 2020). The generation of sewage sludge is lower than that of municipal waste, with a total annual production of 8.2 Mt (dry weight, source: Eurostat), representing about one-tenth of the total generation of OFMSW in the EU, which amounts to approximately 100 Mt. Fig. 1 illustrates the annual generation of sewage

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sludge and total municipal solid waste in the EU-27 countries in 2021 (or other years depending on data availability as indicated in the caption). Both sewage sludge and municipal solid waste follow the same trend as they are essentially linear with population in countries with similar stage of development.

Both sewage sludge and municipal solid waste can be potentially recycled and processed for energy recovery, and both contain organic matter and nutrients beneficial for agricultural and soil applications. Sewage sludge has been traditionally used as soil amendment due to its content in nitrogen, phosphorous, potassium, and organic matter. Sewage sludge can improve soil fertility and structure, enhancing water retention and aeration, thereby increasing crop yields. With the same rationale, the OFMSW is widely used as soil amendment due to their content in stabilized organic matter and nutrients (Carabassa et al., 2020). Using sewage sludge and composted wastes as soil amendments promotes sustainable waste management by recycling and reusing valuable resources. Besides, their use reduces the need of chemical fertilizers, and help close the nutrient loop in agriculture (Singh and Agrawal, 2008).

The reuse of waste streams constitutes a key element in the context of the circular economy, which has been progressively incorporated into most legal systems. In the European Union, Directive 91/271/EEC on urban wastewater treatment establishes that sludge shall be reused, and Member States shall ensure the progressive reduction of toxic, persistent, or bioaccumulable materials potentially contained in it. Additionally, Directive 86/278/EEC lays down specific rules and standards for the safe use of sewage sludge in agriculture. These provisions include maximum application rates on agricultural lands and define quality criteria for sewage sludge, specifying limits for heavy metals, pathogenic microorganisms, and other harmful substances to avoid potential food contamination risks.

Historically, the focus has predominantly centred on heavy metals and specific pathogens as the primary contaminants of interest in sewage sludge. Accordingly, regulations have been primarily geared towards limiting the presence of metals and pathogens. However, recent studies have assessed the presence of priority and emerging pollutants in treated sludges. These pollutants include pharmaceuticals and personal care products, perfluorinated chemicals, pesticides, and a wide variety of organic chemicals that partition into sludge due to their adsorption affinity (Fijalkowski, 2019). More recently, sewage sludge has been shown to act as a source of microplastics (MPs) in the environment because it concentrates the MPs present in raw wastewater (Petroody et al., 2021).

For the sake of clarity it is important to stress that plastic particles are considered MPs if their larger dimension is <5 mm with a lower boundary of $1 \mu\text{m}$ below which, they are referred to as nanoplastics (NP) (GESAMP, 2019; Gigault et al., 2018).

Regarding municipal solid waste, the Directive 199/31/EC on Waste Landfills has already established limits on the amount of biodegradable waste that can be disposed of in landfills. Additionally, the Waste Framework Directive (Directive 2008/98/EC amended by Directive 2018/851) mandates minimum percentages for the reuse and recycling of municipal waste, with the aim of reaching at least 65 % by 2035. Currently, this Directive is undergoing an additional amendment process with a focus on textiles. Directive 2018/851 also makes it compulsory for all member states to collect the OFMSW starting in 2023. The OFMSW typically includes food leftovers and various vegetal-based substances, providing a balanced mix of carbon, nitrogen, and phosphorous, which makes it suitable for agricultural use as fertilizer and soil amendment. However, to ensure good product quality, treatment systems must remove non-biodegradable materials such as plastics, metals, and glass.

Despite stringent efforts in collection strategies and plant engineering, obtaining clean organic fractions from municipal wastes remains a significant challenge. The key factor to achieve good results lies in performing separate collection of biowastes, as mandated by the EUs Waste Framework Directive, with a deadline of December 31st, 2023. Various approaches can be employed, from placing specific containers in public spaces to implementing personalized door-to-door collection methods, each having different impacts on product quality (Gallardo et al., 2010). Good quality OFMSW undergoes immediate composting or can be treated in combined anaerobic/aerobic treatment facilities. Nevertheless, achieving the full separation of the degradable organic fraction from impurities such as plastics or textile remnants remains a challenging task. The presence of these elements, even in small quantities, poses a threat to endeavors aimed at sustainable waste management (Edo et al., 2022).

2. Sewage sludge and biosolids

Biosolids are organic materials derived from wastewater treatment, after the liquid components are separated from solid residues. These solids undergo a combination of physical and chemical treatments to remove harmful pathogens and reduce potential environmental risks, resulting in the formation of a nutrient-rich, semisolid product known as biosolids.

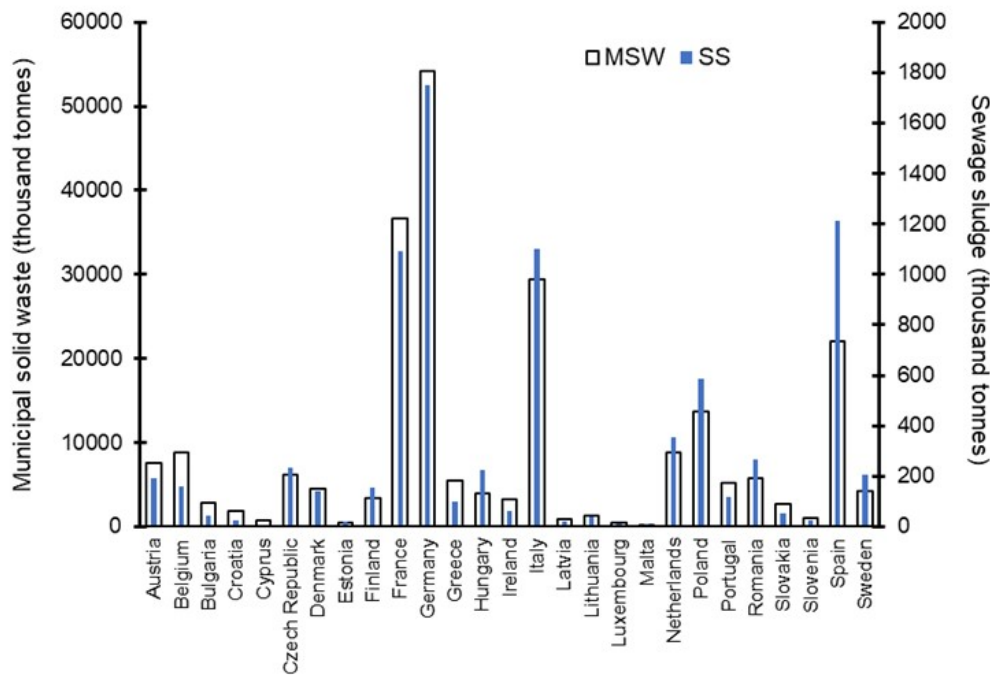


Figure 1: Generation of municipal solid waste (MSW) and sewage sludge (SS, dry weight) in the EU. Data from 2021 except Belgium, Denmark, and Italy (SS, 2010), Portugal (SS, 2016), Spain (SS, 2018), Bulgaria and Germany (SS 2019), Finland, Netherlands, and France (SS, 2020), and Austria, Italy, and Ireland (MSW, 2020). textitSource: Eurostat.

The terms “biosolids” and “sewage sludge” are often used interchangeably. However, to be more precise, while sludge refers to a mud-like or semisolid material, the term biosolids specifically refers to sludge that has undergone treatments such as thermal stabilization, aerobic or anaerobic digestion, drying, or disinfection, among others (Collivignarelli et al., 2019). As an operative definition, biosolids are stabilized organic solids derived from sewage treatment processes, mostly resulting from the biological treatment of wastewater, which can be safely managed and used beneficially for their nutrient, soil conditioning, energy, or other values (Shammas and Wang, 2008). Safe means meeting legal pollutant and pathogen requirements for land application or any other disposal.

When wastewater is treated at a sewage treatment plant, it goes through several stages to remove contaminants. The initial steps are the physical removal of large debris and solids: grit removal and primary settling. Then, wastewater undergoes biological treatments, where microorganisms break down organic matter. As a result, two types of sludge are produced from both primary and secondary settlers. The latter mainly comprises the biomass generated during biological treatment. The resulting sludge must be processed to reduce the load of pathogens, remove water, and stabilize the material. The steps involved in this process are thickening, dewatering, and digestion.

The first process is thickening, which reduces the volume of sludge. This is usually achieved through gravity thickening, dissolved air flotation (DAF), and rotary drums. Gravity thickening employs conventional sedimentation tanks. DAF uses pressurized air to remove solids that do not naturally float or sink, which is the case of activated sludge (hence, DAF is more effective for treating secondary sludges than primary ones). Air bubbles sweep sludge particles to the surface, where they are removed by a scraper. Rotary drum thickeners consist of slowly rotating cylinders with a porous medium located in their wall. Remnants of water can be removed using several dewatering processes, such as centrifugation, vacuum filtration, and filter presses. Typically, thickening results in a range of 4–8 % dry solids, while dewatering generally allows reaching >30 % dry solids. Finally, sludge can be dried in rotary dryers to produce a pelleted material with >90 % dry solids suitable for safe storage before use as fertilizer or fuel for incineration plants.

Another commonly used sludge treatment process is anaerobic digestion, which not only concentrates solids but also allows for the production of biogas (a mixture comprising about 60 % methane and 40 % carbon dioxide). The process can operate in three possible temperature ranges, namely psychrophilic (<20 °C), mesophilic (35–40 °C) and thermophilic (50–55 °C). The system’s configuration can be single-stage or double-stage running sequentially in different ar-

rangements. The interplay of this phases determines the yield of biogas. Biogas can be used in the water treatment plant to produce energy, or can be injected into national gas grids, similar to the biomethane produced from municipal solid wastes (De Mes et al., 2003). Another increasingly used process is the recovery of phosphorus from digested sludge as struvite (Saerens et al., 2021). The recovery of excess phosphorus is beneficial for the circular economy due to the limited phosphate rock reserves and the presence of cadmium in all phosphate rocks (Suciu et al., 2022).

The end product are biosolids, which are stable, nutrient-rich materials that comprise organic matter, nitrogen, phosphorus, and other plant nutrients. By applying biosolids to farmland, soil fertility, water retention, and crop productivity can be improved, thereby reducing the reliance on synthetic fertilizers, and promoting sustainable nutrient cycling. Additionally, biosolids find application in various other areas, such as land reclamation projects, mine site remediation, and even as a component in landscaping and horticultural products.

As stated before, and depending on wastewater characteristics and treatments, it may contain heavy metals or organic contaminants. To tackle this issue, monitoring measures are to be implemented in compliance with Directive 86/278/EEC. However, some countries like Germany and the Netherlands are more restrictive concerning land use and have adopted incineration as the common practice to process sludge. Conversely, Ireland, Spain, Bulgaria, and Baltic and Scandinavian countries make extensive use of sludge and biosolids as fertilizers. The use of wastewater sludge in the EU countries is shown in Fig. 2

Besides anaerobic digestion, sludge can be composted. This process stabilizes organic materials before using them as a soil amendment or fertilizer. Composting involves the controlled aerobic degradation of organic materials and can be applied to a variety of substrates, including sludge and the OFMSW. Biosolids can also be co-composted with OFMSW with the rationale of improving compost because that produced from OFMSW contains relatively fewer amounts of organic matter and nutrients (Tognetti et al., 2007). In addition to organic matter, composted biosolids provide nutrients such as nitrogen, phosphorus, and potassium, as explained below

3. Municipal solid waste

The proper treatment of the organic fraction of municipal solid waste (OFMSW) is a central issue in the current shift towards a circular economy. In the

EU, the Waste Framework Directive (2008/98/EC, amended by 2018/851) encourages the composting of biowaste (including home composting) and promotes the use of composted materials. Composting is a natural biological process that transforms the degradable portion of waste into a stable material suitable for soil application

Composting starts with a series of pretreatments required due to the high heterogeneity of municipal solid waste, necessary even if the organic fraction has been recovered under separate collection schemes. Impurities of non-biodegradable materials like glass or plastics that may affect the composting process and the quality of the final product are separated using sieves, magnets, air classifiers, or ballistic separators. Before composting, the raw waste is mixed with other compostable materials (such as manure) to achieve the proper carbon-to-nitrogen ratio. Additionally, a bulking agent, a material with low decomposition rate, is added to provide mechanical structure and porosity during the decomposition phase.

During composting itself, a wide range of organisms, from bacteria to worms, are involved in the decomposition of the biodegradable materials present in raw waste. This is a complex multi-stage process, with several factors influencing the roles of different organisms, such as temperature, moisture, and nutrient content and makes composting a challenging process difficult to accomplish satisfactorily outside industrial facilities. Throughout the composting process, various parameters such as temperature, pH, moisture, and carbon-to-nitrogen ratio must be monitored to ensure efficient digestion. Waste materials to be composted are placed in piles, windrows, or enclosed containers for the digestion that takes place in three phases:

1. **Mesophilic Phase:** After a lag of 12–24 h, in the initial phase of composting, the temperature gradually increases due to the activity of mesophilic microorganisms, typically ranging from around 20 °C to 40 °C. During this phase, easily degradable organic materials decompose, releasing heat.
2. **Thermophilic Phase:** In this phase, the temperature in the compost pile rises significantly, reaching 70 °C or even more and thermophilic microorganisms become dominant. The high temperatures accelerate the breakdown of complex organic materials, including tougher compounds, pathogens, and weed seeds. The intense heat also helps to sterilize the compost.
3. **Maturation or Curing Phase:** During this phase, the compost tends to stabilize. The temperature decreases to ambient levels, and the product

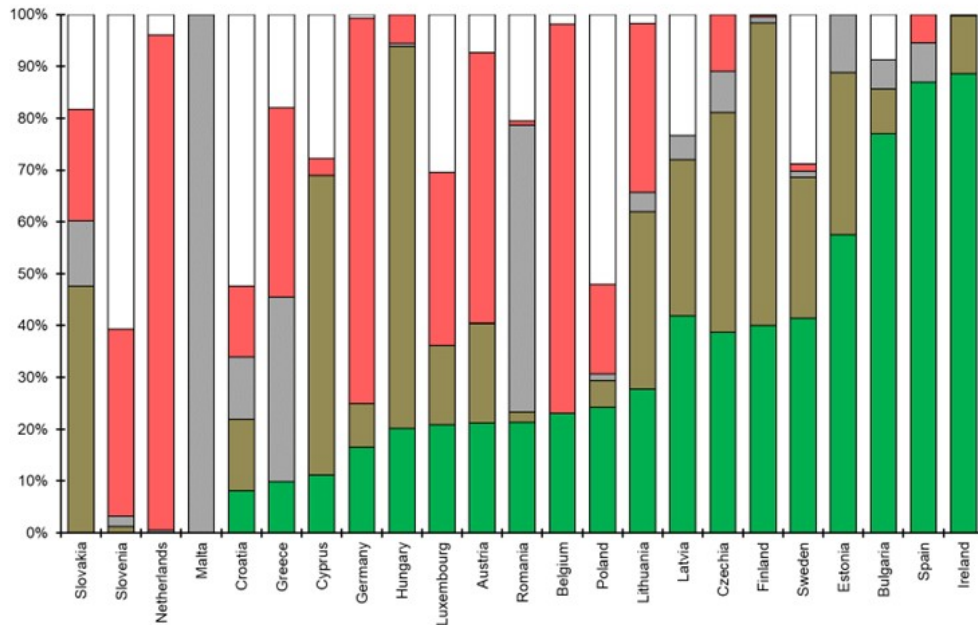


Figure 2: Sewage sludge disposal in EU countries (2020 or the latest year with data; no data available for Denmark, France, Italy, and Portugal). Green: direct agricultural use; brown: composting; gray: landfilling; red: incineration; white: other uses. textitSource: Eurostat.

develops a stable structure. After this phase, the compost resembles mature humus, ready to be used as a valuable soil conditioner and fertilizer.

Digestion may take place using different arrangements and practices, depending on the characteristics of the compostable material and the size of the plant. In small plants, the most common method involves passive aerated piles or windrows, which are periodically turned around (every 3–4 days for at least 2 months) to homogenize and allow air passage. Piles and windrows only differ in shape. Large-scale composting can also take place in aerated static piles or windrows, where air flows upwards through blowers distributed at the bottom. Alternatively, in-vessel composting confines the organic materials into drums or silos, allowing a precise control of the environmental composting conditions. This method efficiently processes a wide variety of wastes in less space. The entire composting process may last 6–12 weeks, depending on the composition of raw materials and the choice of technology.

As with wastewater sludge, waste minimization and energy recovery can be achieved through anaerobic digestion (Paritosh et al., 2018). At the end of anaerobic digestion, a significant proportion of the raw material entering the digester can be further processed through composting. Therefore, anaerobic digestion and aerobic composting are not mutually exclusive (Qi et al., 2022). Lignocellulosic materials have low yields of biogas, while food wastes from residential sources have much higher yields and di-

gest at a faster rate. Consequently, the use of both technologies concatenated depends on logistic variables, such as the availability of source separation for organics separately from vegetal fractions.

The availability and efficiency of separate collection schemes are the key drivers for the sustainable management of municipal solid waste, especially its organic fraction. The EU's Waste Framework Directive mandates that all Member States collect biowaste separately and not mixed with other types of waste before 31st December 2023. Different strategies can be implemented for it, ranging from street bin containers to personalized door-to-door collection systems, each with different impacts on product quality. As indicated before, the collected OFMSW can be treated in composting or in combined anaerobic/aerobic treatment facilities with various layouts and technologies. However, the quality of the final product strongly depends on how well the source separation is done (Rodrigues et al., 2020).

4. Plastics in biosolids and in the organic fraction of municipal solid waste

Recent studies have revealed the occurrence of microplastics (MPs) in sludge/biosolids, composts sourced from various origins, agricultural soils amended with biosolids, and, to a lesser degree, un-

treated soils. The literature shows a higher abundance of the most commonly produced polymer types: primarily polyester (PES), polyethylene (PE), polypropylene (PP), polystyrene (PS), polyamide (PA), and polyvinyl chloride (PVC), as well as industrially processed natural substances like regenerated cellulose and other textiles fibers. Their analysis is not an easy task as sludge and composted wastes are intricate matrices, that require difficult procedures to digest organic matter and separate plastics.

Table 1 presents a selection of recent results reporting the concentration of MPs in sludge and biosolids from different origins and treatments. Petroody et al. (2021) encountered 214 ± 16 MPs/g (dry weight, insofar in all cases), in sludge from a primary settler, 206 ± 34 MPs/g in the sludge from a secondary clarifier, 200 ± 13 MPs/g after thickening, 238 ± 31 MPs/g after aerobic digestion, and 129 ± 17 MPs/g after dewatering. This low variability is remarkable and rather unusual, considering simultaneous samples correspond to different residence times. The same authors found that most of the MPs sampled (85%) were fibers, primarily attributed to textiles such as carpets and curtains. Among the fragments, the majority were identified as PE, suggestive of cosmetic microbeads (Petroody et al., 2021). Horton et al. (2021) collected sludge samples from eight different wastewater treatment plants (WWTPs) in the UK and reported MP concentrations as high as 1.04×10^4 MPs/g. This value is very high compared to other studies but may be attributed to a locally high concentration in the influent. In fact, in the same research, the authors recorded a high concentration of MPs in the influent dominated by an unusually high concentration of polyethylene terephthalate (PET). Harley-Nyang et al. (2023) summarized the results of sixty-five studies across twenty-five countries. Most of the reported results ranged in the tens to hundreds of MPs/g, with an average (after excluding one outlier) of 208 MPs/g and a maximum of 3.6×10^3 MPs/g.

The intrinsic variability of wastewater due to local specificities, the existence of industrial activities or seasonal factors are aspects that must be considered to correctly interpret the data concerning the occurrence of MPs in WWTPs and biosolids. In most cases, samples are simultaneously collected from the influent, effluent, and sludge; however, it is important to note that these samples do not represent the same waste stream due to the different retention times. Furthermore, the analysis of biosolids is complex due to the substantial organic matter content, requiring a sequence of separation and purification methods. Typically, this involves the oxidation of organic matter followed by flotation and filtration be-

fore identification through spectroscopic techniques. Increased processing can lead to loss of information and to a higher probability of contamination thereby adding uncertainty in the reported results. Additionally, the analyses employ small amounts of sample, typically of a few grams or less, which can contribute to the variability (Horton et al., 2021). Nonetheless, the dispersion of data observed in the studies documented in the literature is not high enough to render estimation of actual microplastic emissions through wastewater biosolids unfeasible.

Koutnink et al. (2021) analyzed a collection of 76 studies and calculated the median concentrations of MPs in influent, effluent, and sludge. They concluded that only 4% of the MPs entering WWTPs were effectively detected in the effluent and sludge. The seeming paradox was resolved by Ziajahromi et al. (2021), who demonstrated that the majority of MPs (69–79%) were eliminated during preliminary treatments like screening and grit removal. The MPs recovered with grit are typically disposed of in landfills, thus transforming landfills into significant repositories for MPs. In fact, landfilling grit prevents the dispersion of the majority of MPs entering WWTPs (Ziajahromi et al., 2021). When considering the grit stream, the balance of MPs in a WWTP reasonably closes. Utilizing our own data and removal efficiencies sourced from the literature (Fig. 3), we estimated that the MPs detected in sludge accounted for 13% of the MPs removed within the plant (Edo et al., 2020; Iyare et al., 2020). This is a rough approximation using data from a single plant, but it falls within the range of 7.5–16% estimated elsewhere (Ziajahromi et al., 2021).

An additional concern is that the conservation of mass does not necessarily imply the conservation of particle number. The potential fragmentation of MPs during wastewater processing can result in either an increase in the number of particles or an apparent decrease if the resulting fragments fall outside the range of measurement (the lower limit is in the tens of microns using micro-FTIR). The existing research showed that the size range of MPs depends on the treatment undergone by the sludge, with digested and lime-stabilized sludge exhibiting a smaller average MP size compared to raw mixed sludge. This suggests that some treatments might indeed break MP particles, although the available findings are not conclusive (Harley-Nyang et al., 2022; Mahon et al., 2017).

Regarding OFMSW, while the utilization of compost derived from biowaste has been acknowledged as a substantial contributor of MPs within agricultural domains, its quantification has been less studied, with the available findings reflecting a broad

Table 1. MPs found in sludge and biosolids in selected studies. Results are given per gram of sludge (dry weight in all cases).

Location	Analytical procedures	Concentration, size, and composition	Reference
12 WWTPs in the north-western part of Germany	NaOH for 24 h at 60 °C; NaCl flotation 1.14 kg/L; filtration using 500 µm PA nets with aliquots filtered onto 0.2 µm Al ₂ O ₃ filters; ATR-FTIR and micro-FTIR	1-2.4 MPs/g; No MPs > 500 µm; PE, PP, PA, and PS; fibres were not studied	(Mintenig et al., 2017)
28 WWTPs from 11 provinces in China	NaCl 1.2 kg/L; filtration using 37 µm filters; H ₂ O ₂ 30 % overnight; identification using micro-FTIR	1.60-56.4 MPs/g; average 22.7 ± 12.1 MPs/g; MPs were mainly polyolefin and acrylic fibers, PE and PA films, alkyd resin, and PS spheres; no size details given	(Li et al., 2018)
WWTP near Madrid, Spain	H ₂ O ₂ (33 % w/v) at 50 °C for 20-24 hours; flotation with NaCl, 1.2 kg/L; filtration using 25 µm stainless steel filters; identification using micro-FTIR	183 ± 84 MPs/g (mixed sludge) and 165 ± 37 MPs/g (dried sludge); Fragments in mixed sludge 36-377 µm; fragments in dried sludge 29-533 µm; fibres in mixed sludge 213-4716 µm length and 5-34 µm width; fibres in dried sludge in 71-2224 µm length and 7-58 µm width; fibres 62 % in mixed sludge and 84 % in dried sludge; PES, PP and acrylic fibres and PE fragments	(Edo et al., 2020)
3 WWTPs in Australia	Digestion with H ₂ O ₂ ; flotation with NaI; filtration using 25 µm; oxidation post-filtration (30 % H ₂ O ₂ + FeSO ₄); identification using micro-FTIR	15.9-45.7 MPs/g (primary sludge), 37.8-46.1 MPs/g (secondary sludge), 48.5-56.5 MPs/g (digested sludge); 63.5 % fibres; PET (> 60 %), PE, PP and PA	(Ziajahromi et al., 2021)
WWTP in the city of Sari, Iran	H ₂ O ₂ 30 % at 70 °C for 5 h; density separation with NaI followed by filtration using 37 µm stainless steel filters; identification using micro-Raman spectroscopy	From 214 ± 16 MPs/g (primary settler) to 129 ± 17 MPs/g (after aerobic digestion and dewatering); 85 % of MPs were fibres, half of which were PES; size not given	(Petroody et al., 2021)
8 WWTPs in the UK (six activated sludge, two trickling filters and one biological aerated flooded filter)	25 µm stainless steel filters; Fenton's reagent (70 mL of 30 % H ₂ O ₂ + 30 mL Fe (II) 0.05 M), < 50 °C, overnight; ZnCl ₂ flotation followed by enzymatic digestion and refiltered to remove particles > 178 µm; semiautomated micro-FTIR	301-10,380 MPs/g; almost all particles < 100 µm (due to the filtration removal of larger particles); PE, PP and PET (especially in samples with higher MPs load); morphology was not studied	(Horton et al., 2021)
1 WWTP in Devon, UK	Oxidation of organic matter with Fenton's reagent (30 % H ₂ O ₂ +0.05 M FeSO ₄) < 50 °C overnight; flotation with ZnCl ₂ , 1.5 kg/L; 1.2 µm GF/C filter; identification with micro-FTIR before anaerobic digestion	37.7-107.5 MPs/g after anaerobic digestion 97.2-286.5 MPs/g; 42.5 % fibres, 57.5 % particles; most (57.1-82.1 %) MPs < 500 µm	(Harley-Nyang et al., 2022)

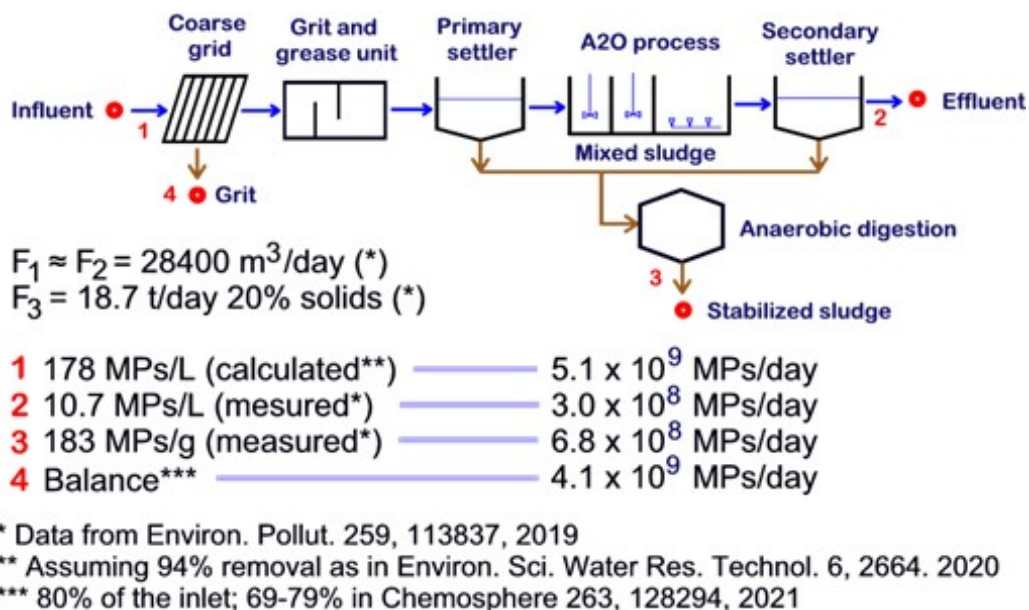


Figure 3: Number balance for MPs in a wastewater treatment plant.

range of concentration (Vithanage et al., 2021). Compost samples sourced from organic municipal waste processed in The Netherlands exhibited an MPs content of $2.80 \pm 0.66 \text{ MPs/g}$, higher than that of compost from garden and greenhouse waste, which exhibited $1.25 \pm 0.56 \text{ MPs/g}$ (van Schothorst et al., 2021). In another investigation focusing on MPs in compost derived from rural domestic waste, an average abundance of $2.4 \pm 0.4 \text{ MPs/g}$ of dry weight compost was obtained for the size range 0.05–5 mm (Gui et al., 2021). Sholokhova et al. (2022) examined the presence of MPs in compost sourced from separate collections of OFMSW and reported concentrations of 3.78–6.43 MPs/g, varying based on waste type and season, with particles mostly measuring <1 mm (83.8–94.9%).

In a recent study, we investigated the presence of plastic particles (>25 μm) in the compost produced from the OFMSW taken from five composting facilities, representative of different collection systems and technologies. The outcome revealed an overall plastic concentration ranging from 10 to 30 items/g, mostly consisting of particles <5 mm (MPs), which were in the 5–20 MPs/g range, with fibers predominantly present in the lower size fraction (25% <500 μm) (Edo et al., 2022). Weithmann et al. (2018) reported 20–24 MPs/kg in compost from municipal organic biowaste subjected to aerobic treatment, 70–146 MPs/kg for compost from anaerobic digestion, and a notably higher 895 MPs/kg for compost produced by a biowaste digester processing commercial waste. The huge discrepancy could be attributed to a limitation of the latter study, focused on particles >1 mm.

The primary distinction among different studies lies in the range of sizes sampled, with studies investigating plastic particles in the tens or hundreds of microns range reporting higher counts. Overall, the small size of MPs discovered in the majority of compost samples highlights the necessity of limiting these minute MPs in organic waste fertilizers to avoid their dispersion into soils. Additionally, it has been demonstrated that compost originating from smaller plants and employing door-to-door collection practices exhibits fewer plastic impurities compared to compost generated from organic fraction of municipal solid waste collected from street bins. This correlation can be attributed to the direct link between inadequate waste management and citizen involvement.

In general, compost facilities exclusively processing agricultural residues yield cleaner compost, albeit not entirely free of plastic materials due to the widespread nature of plastic pollution. In a case of pure vegetal compost produced at a facility in northern Spain, we measured $2.4 \pm 0.9 \text{ MPs/g}$, which is at least one fourth the number concentration found in OFMSW-derived compost, yet still considerable. The most abundant synthetic polymers observed in compost samples were those more commonly used for consumer products, namely PE, PS, PES, PP, PVC as well as acrylic fibers, rubbers, and cellulose-based polymers (Edo et al., 2022; Weithmann et al., 2018). Up to date, no compostable bioplastics were found in compost from OFMSW (Edo et al., 2022).

The limited information on the size distribution of MPs provided in most environmental samples is a barrier to compare results. The utilization of differ-

ent size classes poses a substantial challenge when attempting to compare data and draw meaningful conclusions on the occurrence of MPs, but even the application of standard binning with specific size cutoffs would restrict the insight that can be gained from employing a comprehensive size description for all MP particles. Fig. 4 presents the cumulative distribution function for two distinct datasets: mixed and heat-dried biosolids (red), and plastic particles sourced from compost derived from municipal waste (green) (Edo et al., 2022; Edo et al., 2020). All MP particles, including fragments, fibers, and films, were individually characterized based on their two orthogonal projected dimensions. Specifically, for fragments and films, these dimensions correspond to length and width, while for fibers, they represent length and diameter. For nearly isometric particles, the diameter of the sphere with an equivalent volume to that of the particle, d_v , can be approximately computed, as demonstrated elsewhere (Rosal, 2021). In the case of films, the third dimension, which is the smallest and not recorded, was assumed to be one-tenth of the smallest dimension among the other two. For fibers d_v was calculated assuming a cylindrical shape. The individual particle mass can be then readily estimated using the tabulated average density corresponding to each polymer (or an average for all of them).

A clear difference is evident between the two datasets. The median size of MPs in biosolids was 66.3 μm , whereas that of MPs extracted from compost was 1100 μm . Employing straightforward calculations, the mass concentration of MPs can be accurately determined, thereby circumventing significant errors such as the credit card per week issue (Pletz, 2022). The reason behind discrepancies is that the weight of the average particle and the total weight divided by the number of particles may be very different. For the data shown in Fig. 4, the mass concentration of MPs measured in biosolids (mixed and heat-dried sludge) was 135 μg MPs/g (or 135 g MPs/t biosolids) and in compost 15–60 mg MPs/g compost (15–60 kg MPs/t compost).

Considering the legal limit existing in the region of Madrid (where the studies were carried out) to the application of biosolids, established in 5 t/ha (dry weight) per year, the maximum emission of plastics, could reach 1.7 kg/ha every year. In Madrid, the production of sludge pellets accounts for roughly 100,000 t/year containing 13 t of MPs. No limits exist on the use of compost from organic wastes, that can potentially be a much greater source. The biowaste generation in the region of Madrid (6.6 million inhabitants) could yield 400,000 t/year of compost (in dry weight) containing 20,000 t of MPs in a rough

estimation.

5. Risks derived from the use of wastes contaminated with plastics

The utilization of biosolids, once treated and processed to adhere to regulatory standards, has been a significant aspect of urban wastewater management in Europe since the entry into force of Directive 86/278. Currently, the processing of organic municipal wastes plays a pivotal role in achieving the EU-27s recycling target of 65% for municipal waste by 2035. This objective forces to find a final use for most of the 75 million tons of municipal biowaste produced annually across Europe (EEA, 2020). However, the application of both biosolids and compost requires careful consideration due to their potential role in the dispersion of contaminants. Over time, the focus has shifted from heavy metals to organic pollutants, such as persistent or emerging micropollutants and, notably to MPs. A major issue is that the current state of knowledge does not allow to establish emission limits for MPs that could be considered exempt from environmental and health risks but the implementation of precautionary approaches could excessively restrict the agricultural use of organic wastes (Gianico et al., 2021).

Several studies allowed quantifying the accumulation of plastics in soil environments due to the use of organic amendments (Table 2). Corradini et al. (2019) performed a study in thirty-one agricultural fields in Chile with different sludge application records during the last ten years with similar application rate (about 40 t/ha). The content of plastic increased with the number of applications from a median of 1.1 MPs/g (1 application) to 3.5 MPs/g (5 applications) while control sites where no sludge had been applied only had 0.2–0.6 MPs/g. The sludge applied had an average of 34 MPs/g. Most MPs (90% in sludge, 97% in soil) were fibers. The authors estimated that the mass concentration of MPs in sludge was 45.5 mg/kg in sludge (particles $>8 \mu\text{m}$), which would represent a dispersion of 1.82 kg/ha of MPs or 0.27 mg/t of soil (25 cm depth, 2.65 g/cm³ soil av. density) per application (Corradini et al., 2019). It has to be noted that this work didn't use spectroscopic analysis of MPs, which were characterized based on visual inspection only. Crossman et al. (2020) studied soils exposed to biosolids in Ontario, Canada. The concentration in biosolids in soil after application (74–140 m³/ha) reached 24–358 MPs/kg. A simple calculation using the data provided by the authors yielded a total input rate in the range of 1.6–19.8 kg MPs/ha. van den Berg et al. (2020) studied 16 agricultural fields

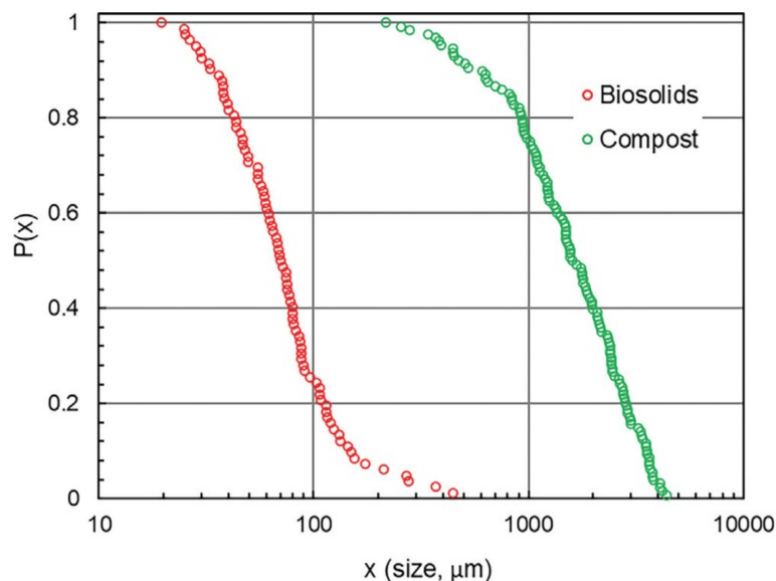


Figure 4: Probability distribution of MPs in samples from biosolids (mixed and heat dried sludge) and compost from OFMSW. $P(x) = \text{textitni}(x)/N$, where $\text{textitni}(x)$ is the number of particles larger than textitx , textitN the total number of particles and $\text{textitx} = d_v$. Data from Edo et al. (2020, 2022).

in Valencia, east of Spain, after 1–8 applications of biosolids with a rate of 20–22 t/ha and demonstrated that every application increased the number concentration of MPs in an average of 0.71 MPs/g of soil. In another study, the authors found no statistically significant differences in the overall number and mass concentration of MPs in treated and untreated soils, which was interpreted as caused by the presence of microplastic from other sources either agricultural or non-agricultural (Radford et al., 2023). Most of the available data provide evidence of the accumulation of MPs in soil following the application of biosolids to land. In fact, the utilization of biosolids is a reasonably well quantified source of MPs although their rates of migration to surface or underground water bodies or their biotic and abiotic degradation remain largely unknown.

In contrast, the studies on the accumulation of MPs in soils treated with urban compost and scarce, but the rationale is the same: compost application is a major pathway for MPs entering agricultural soils. Braun et al. (2023) investigated the effect of the long-term use of compost from municipal biowaste on the plastic content in the upper (<30 cm) soil layer and found that fields undergoing long-term use of compost showed a MP load of 38–171 $\times 10^6$ MPs/ha increasing with more compost application. It is remarkable that the effect was still noticeable after 12 years. Once into the soil, the fate of MPs depends on the biological and physicochemical characteristics of the soil as well as environmental factors, the main difference with MPs from biosolids being their larger size. Once released, MPs can move to other compart-

ments and become fragmented under the action of biological, photooxidative or mechanical processes more easily than smaller plastics (Aoki and Furue, 2021). Watteau et al. (2018) used pyrolysis/gas chromatography/mass spectrometry to track the fate of these plastics in soil regularly amended with household waste compost and found evidence of MPs in amended soil, but not in control soil, although the methodology used did not allow to make quantitative assessments.

There exists a substantial knowledge gap concerning the fate and effects of MPs within soil ecosystems. Soils can experience diverse impacts due to MP pollution. Firstly, plastic materials incorporate various additives such as pigments, plasticizers, antioxidants, and flame retardants, which eventually leach out of the plastic matrix and potentially endanger human health and environmental organisms (Luo et al., 2022). Phthalate plasticizers, like those found in biosolids and compost, are a prominent example of chemicals known to disrupt soil organisms (Zhu et al., 2022). Ma et al. (2017) demonstrated that bis(2-ethylhexyl) phthalate affected the enzymatic activity and induced DNA damage in the earthworm *Eisenia foetida* at concentrations in the low milligrams per kilogram range. The aging of MPs promotes the leaching of additives and results in the loss of antioxidant stabilizers, which in turn facilitates particle fragmentation (Liu et al., 2022). Possible impacts on soil and freshwater organisms include hormonal disruption, potentially causing developmental and reproductive issues; altered behaviour, with cascading effects on ecosystems; and suppres-

Table 2. MPs in soils as a consequence of the use of biosolids in selected studies. Mass units refer to dry weight in all cases.

Location	Sludge	Controls	Amended Soil	Reference
Mellipilla county, Chile	34 MPs/g	0.2-0.6 MPs/g	1.1-3.5 MPs/g (1 to 5 applications); input 1.82 kg MPs/ha	(Corradini et al., 2019)
Ontario, Canada	8.7-144 MPs/g	6.8 MPs/kg (0.2 mg/kg)	24-358 MPs/kg (2.3-28.5 mg/kg); input 1.6-19.8 kg MPs/ha	(Crossman et al., 2020)
Valencia, Spain	50 ± 35 MPs/g	2.0 ± 1.3 MPs/g	5.2 ± 2.6 MPs/g (increasing 0.71 MPs/g after every application)	(van den Berg et al., 2020)
University farm in Nebraska, USA	9.1 ± 1.7 MPs/g	0.9 ± 0.1 MPs/g	2.6 ± 0.6 MPs/g; input 4.5 x 108 MPs/ha	(Naderi Beni et al., 2023)
Southeast of England	Not given	664 MPs/kg or 86.6 mg/kg	874 MPs/kg (max. 1486 MPs/kg, min. 202 MPs/kg) or 206.8 mg/kg	(Radford et al., 2023)

sion of immune responses, altering the organism's defence mechanisms, among other effects (Ding et al., 2022; Greven et al., 2016).

MPs have the capacity to adhere to seed and root surfaces, hindering water and nutrient uptake, thereby compromising seed germination and plant development (Iqbal et al., 2023). Organisms ingesting MPs might experience blockages in feeding appendages or digestive tracts (Barnes et al., 2009). In specific cases, MPs could accumulate and propagate through food chain through predation. This involves the accumulation and possibly concentration of MPs, additives, and absorbed chemicals in ecosystems. While ongoing research illustrates MP bioaccumulation in certain trophic levels, evidence, of bioaccumulation especially under real-world exposure conditions, remains controversial (Miller et al., 2020). Furthermore, the small fragments of plastic, NPs (<1 µm) can be internalized, inducing oxidative stress, cytotoxicity, and genotoxicity in soil biota, including plants (Wang et al., 2022).

The dispersion of MPs from compost or biosolids affects physicochemical and biogeochemical processes in soil, altering water-holding capacity and potentially interfering with nutrient cycles, thereby modifying soil fertility (Ya et al., 2021). The available results indicate that MPs could impact soil pH, water content, dissolved organic carbon, and biological aspects like soil enzyme activities (Zhang et al., 2023). Besides, the MPs present in compost and biosolids have the capacity to bind harmful chemicals, which might otherwise infiltrate agroecosystems and spread to new environments due to the mobility of plastic particles (Vithanage et al., 2021). Research has demonstrated that MPs alter the cation exchange capacity

and metal speciation, enhancing their organic-bound fractions through adsorption (Yu et al., 2020).

MPs could also alter the soil microbial community by providing adsorption sites for microorganisms. The impact of plastics on soil microbial activity is unclear, but it has been demonstrated that plastics can harbour antibiotic resistance genes and act as vectors for human and animal pathogens (Martínez-Campos et al., 2022). The role of biodegradable plastics, a family of materials increasingly employed in agriculture due to their quicker degradation, seems clearer because of their higher reactivity. Research shows that poly(3-hydroxybutyrate-co-3-hydroxyvalerate) leads to reduced corn growth, altered foliar nitrogen content, and modified microbial diversity, leading to decreased bacterial diversity. The long-term implications of the use of bioplastics for agroecosystem health is completely unknown (Brown et al., 2023).

6. Conclusions and recommendations

The agricultural application of biosolids and composted organic wastes as soil amendments holds substantial promise for sustainable and enhanced crop production, while contributing to effective waste management practices. Biosolids, the nutrient-rich byproducts of wastewater treatment, and composted organic wastes, derived from various organic materials, are both increasingly recognized for their potential to enrich soil quality, improve fertility, and promote sustainable agricultural practices.

As anthropogenic pollutants, the risks associated

with the dispersion of MPs in the environment demand careful assessment. MPs might influence soil biogeochemical cycles, interact with soil microbiota, retain various chemicals including pollutants, and exhibit toxicity towards soil biota. Additionally, their high mobility extends these risks to freshwater and groundwater. Significantly, despite the potential hazards posed by MPs and their relevance to food systems, regulations pertaining to soil environments have yet to be established.

A major problem in the quantification of MPs in complex organic matrixes is the limited methodological standardization of their analyses. However, the available data show a relatively good agreement and in recent years the analytical procedures tended to converge to a set or relatively standard procedures. In spite of different size cutoffs, digestion methods (hydrogen peroxide, Fenton), flotation techniques (NaCl, ZnCl₂, NaI), and spectroscopy methods (FTIR and Raman) methodological differences pose minor challenges compared with the limited use of sound QA/QC practices (Ziajahromi and Leusch, 2022).

Soil acts as a sink—temporary or permanent—for MPs from diverse sources, including biosolids, composted waste, wastewater irrigation, agricultural plastics (mulching, silage, greenhouses, piping), and atmospheric deposition. The available data suggest that the use of biosolids may result in the spreading of several kilograms of MPs per ha and year, predominantly fibers with a size (of the sphere with the same volume) mostly <100 µm. However, considering the huge amount of biowaste generated (173 kg per person every year in the EU according to the European Environment Agency) the potential dispersion of plastic with compost from organic municipal waste may be several orders of magnitude higher (in mass) than that from biosolids.

The rise of bioplastics is noteworthy, but despite their renewable origin, their decomposition could release harmful substances, alter soil characteristics, or impact soil microorganisms, possibly causing environmental consequences. Proper waste management and disposal strategies are vital to prevent bioplastic residues from entering the environment. Notably, nonindustrial composting has shown limited ability to degrade bioplastics. This is somehow in contradiction with the EUs Waste Framework Directive that mandates Member States to foster and encourage home and community composting and an issue that needs urgent attention

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