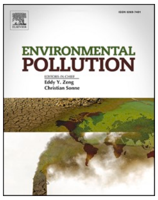
Environmental Pollution 349 (2024) 123986

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol



Plastics in biogenic matrices intended for reuse in agriculture and the potential contribution to soil accumulation☆

Stefano Magni[a], Marco Fossati[b], Roberta Pedrazzani[c], Alessandro Abbà[d], Marta Domini[d], Michele Menghini[c], Sara Castiglioni[b], Giorgio Bertanza[d], Andrea Binelli[a], Camilla Della Torre[a,*]

[a] Dipartimento di Bioscienze, Università degli Studi di Milano, Italy
[b] Istituto di Ricerche Farmacologiche Mario Negri IRCSS, Italy
[c] Dipartimento di Ingegneria Meccanica e Industriale, Università degli Studi di Brescia, Italy
[d] Dipartimento di Ingegneria Civile, Architettura, Territorio, Ambiente e di Matematica, Università degli Studi di Brescia, Italy

ARTICLE INFO

Keywords:
Microplastics
Sewage sludge
Compost
Manure
Soil pollution

ABSTRACT

The spread of biogenic matrices for agricultural purposes can lead to plastic input into soils, raising a question on possible consequences for the environment. Nonetheless, the current knowledge concerning the presence of plastics in biogenic matrices is very poor. Therefore, the objective of the present study was a quali-quantitative characterization of plastics in different matrices reused in agriculture as manures, digestate, compost and sewage sludges. Plastics were quantified and characterized using a Fourier Transform Infrared Spectroscopy coupled with an optical microscope (μFT-IR) in Attenuated Total Reflectance mode. Our study showed the presence of plastics in all the investigated samples, albeit with differences in the content among the matrices. We measured a lower presence in animal matrices (0.06–0.08 plastics/g wet weight w.w.), while 3.14–5.07 plastics/g w.w. were measured in sewage sludges. Fibres were the prevalent shape and plastic debris were mostly in the micrometric size. The most abundant polymers were polyester (PEST), polypropylene (PP) and polyethylene (PE). The worst case was observed in the compost sample, where 986 plastics/g w.w. were detected. The majority of these plastics were compostable and biodegradable, with only 8% consisting of fragments of PEST and PE. Our results highlighted the need to thoroughly evaluate the contribution of reused matrices in agriculture to the plastic accumulation in the soil system.

1. Introduction

Plastic pollution of natural environments is one of the most worrisome problems of the last decade. The soil system in particular is threatened by this kind of contamination, being considered one of the largest environmental reservoirs of plastics (Hurley and Nizzetto, 2018). In this matrix, plastics of smaller size such as microplastics (MPs)-plastics in size from 1 μm to 1000 μm- and nanoplastics (NPs) -plastics below 1 μm-according to International Organization for Standardization (ISO) (2020), can migrate vertically reaching deep soils and leaching into groundwater (Guo et al., 2020; Sajjad et al., 2022). Erosion by wind and water can transfer plastics from soil to air and aquatic systems thus contributing to the further contamination of rivers, lakes and, ultimately, sea (Magni et al., 2021; Sajjad et al., 2022). Indeed, MPs can be transported across long distances, both in the air and in groundwater, thus becoming ubiquitous. In addition, they undergo fragmentation and weathering due to anthropogenic activities and environmental factors (e.g., photochemical reactions) (Zhu et al., 2023). Plastics can be transferred from soil to organisms such as plants, collembola, mites, isopods, nematodes, earthworms, and birds also reaching the human food chain (He et al., 2018; Wang et al., 2020; Sajjad et al., 2022; Su et al., 2023). As far as concerns the adverse impacts of plastics on the soil ecosystems, plastics can change the chemical-physical structure of the soil, can modify soil microbial composition, affect plant growth, and induce negative effects on organisms (reviewed in Guo et al., 2020; Chae and An, 2018; Sajjad et al., 2022), threatening the soil biodiversity and ecosystem functioning (Rilling and Lehman, 2020). Nevertheless, the monitoring of plastics in soil is still limited compared to what would be

---

☆ This paper has been recommended for acceptance by Eddy Y. Zeng.

* Corresponding author. Camilla Della Torre, via Celoria 26, 20133 Milano, Italy.

E-mail address: camilla.dellatorre@unimi.it (C. Della Torre).

https://doi.org/10.1016/j.envpol.2024.123986
Received 4 January 2024; Received in revised form 11 April 2024; Accepted 12 April 2024

needed for assessing the effective risk for soil communities related to the input of these contaminants in the environment.

The amount of plastics detected in soils is highly variable, ranging from a few plastics/kg up to thousands of plastics/kg (reviewed in Buks and Kaupenjohann, 2020; Saadu and Farsang, 2023). In particular, the agricultural soil is contaminated by high levels of plastics, and such contamination can have negative consequences on food production and safety (UNEP, 2022).

The main routes of plastic input into the agroecosystems are mulching, irrigation, deterioration of farm equipment, runoff water, tillage and atmospheric deposition (Bläsing and Amelung, 2018). Besides, there is evidence that the spread of biogenic matrices used as soil conditioners and fertilizers represents a relevant input of plastics on soil, thus raising a question on possible adverse consequences for the environment (Ng et al., 2018; Watteau et al., 2018; Zhang et al., 2020; Braun et al., 2021; Colombini et al., 2022; Bandini et al., 2022; Sivarajah et al., 2023). In this view, the European Union Regulation on the quality of fertilizers (EU, 2019/1009) sets a threshold of 3 g/kg of plastic impurities in digestate and compost. However, the regulation considers only the fraction of plastics >2 mm, thus overlooking the fraction of MPs and NPs, which are more likely to be taken up by organisms and crops (Jiang et al., 2022; Wang et al., 2023) and therefore critical for food safety. Besides, no regulation is applied to sewage sludge (in fact, in Europe, the Council Directive 86/278EEC on the reuse of sewage sludge in agriculture was established in 1986 and is currently undergoing a protracted and contentious process of revision). Furthermore, so far there are few studies regarding the presence of plastics in biogenic matrices which still limits our full understanding of the risk of these contaminants associated with the application of such matrices in the agroecosystems.

Therefore, our study aims to fill this gap of knowledge by measuring the amount of plastics present in different biogenic matrices intended for reuse in agriculture, either from animal and human origin, namely: manure, digestate, compost and sewage sludge and to characterize their size, shape and polymer composition through a Fourier Transform Infrared Microscope System (μFTIR). Our goal was to understand how the spread of these matrices, according to the common agricultural practice, can contribute to plastic accumulation in the soil ecosystem, thus providing useful information for the optimum management of these substrates whose agricultural value (in terms of content of organic carbon and nutrients) is, on the other side, widely recognized and appreciated.

2. Methods

2.1. Sample origin and collection

The biogenic matrices that were examined were selected with great care for the following reasons: first, we chose swine and cow manure as a point of reference, situating their application on agricultural land as their “natural” endpoint. Then, we concentrated on a typical End-of-Waste product (compost), which is manufactured for agricultural organic waste recovery. Finally, we examined sewage sludges derived from various sources, as specified below. The use of the three types of matrices is regulated by different legislations, though the destination is the same (agricultural soil), thus making this comparison particularly interesting. In addition, one of the selected sewage sludges does not fit the limits of the local regulation in force for reuse in agriculture.

In short, the following substrates were investigated:

- One swine manure (M1), in the form of slurry, was collected from a swine farming facility. As reported in the Regional Action Program (2020–2023) for water protection from nitrate pollution according to the Council Directive 91/676/CEE, it has been stored long-term (at least 180 days).
- Two substrates were collected from a cow farming facility: bovine manure (M2) and bovine manure digestate (D) derived from the anaerobic digestion of M2 and other agricultural residues. Both matrices were in the form of slurry.
- Sewage Sludge (S1) was collected in a wastewater treatment plant (WWTP) with a nominal capacity of 50,000 PE (Population Equivalent). The sewage treated is mainly domestic, with a small contribution of industrial discharges (4% in terms of volume). The S1 complies with national regulation for reuse in agriculture.
- Sewage Sludge (S2) was collected in a second WWTP with a nominal capacity of 96,000 PE that treats both domestic and industrial sewages. The S2 does not comply with national regulation for reuse in agriculture: total chromium is significantly above the limit value.
- A further sample (S3) was collected consisting of a mixture of sewage sludges after additional treatment in a composting plant, in order to obtain a fertilizer for agricultural use.
- Compost (C) was collected in a composting plant that treats the organic fraction of municipal solid waste (50,000 ton/year), separately collected in the surrounding municipalities (62 municipalities in 3 different provinces and 10 commercial activities such as supermarkets and restaurants). Received wastes are mostly biodegradable kitchen and canteen waste (EWC, 200108) mainly collected by means of door-to-door waste collection system. The residual fraction consists of biodegradable waste (EWC, 200201) and wood not containing hazardous substances (EWC, 191207).

The main characteristics of the substrates are reported in Table S1.

The sampling operations, which occurred in September 2021, are described as follows: M1, M2 and D samples were collected from their stabilization ponds, just before they were spread on land. For each substrate, in order to obtain a more representative sample, three subsamples were collected by a telescopic rod with a beaker. These subsamples (with the same volume) were used to make the final sample. A similar procedure was followed for the collection of sludge samples (S1, S2 and S3). The C sample was collected from the compost pile temporarily stocked in the platform before the commercialization. The compost pile was overturned with a mechanical shovel in order to reach the bulk of the pile. Then, the composite sample was obtained by collecting aliquots from different areas of the pile. The final sample was obtained by means of the quartering method.

One kg of each sample was collected in glass mason jars equipped with vacuum sealing lids and kept at −20 °C prior to processing for plastics characterization.

2.2. Plastic quantification and characterization

Plastic measurement was carried out on three replicates from each matrix, using about 5 g of sample for each replicate, according to a procedure already established for sewage sludges (Magni et al., 2019). First, plastics were isolated from the collected matrices using sodium chloride (NaCl) hypersaline solution (1.2 g/cm^3), which allows the separation of plastics from the particulate matter, by exploiting the generated density gradient. The suspensions were left for three to 4 day at 4 °C, to facilitate the separation of particles and plastics. The supernatants were then filtered through 8 μm cellulose nitrate membrane filters (Sartorius™ 50 mm) using a vacuum pump. The filters obtained were then further treated with 15% hydrogen peroxide (H_2O_2) to complete the digestion of the organic residues left on the filters, keeping the filters under a laminar flow hood to avoid the potential contamination of the samples by fibres. Differently with respect to the other matrices, the compost samples (approximately 0.14 g each replicate) were not digested and filtered, but were directly analysed to select the potential plastic particles, as reported by Mazzoleni et al. (2023) in a study on former food products.

The digested filters were visually analysed using a stereomicroscope to select the suspected plastic particles from the remaining mineral and natural particles still present on the filters after the digestion process. These particles were transferred to a clean filter in order to carry out a chemical characterization that confirmed their plastic nature. All the collected particles were characterized using a Fourier Transform Infrared Microscope System (μFTIR; Spotlight 200i equipped with Spectrum Two). The FTIR infrared spectrum of each particle was acquired in Attenuated Total Reflectance (ATR) mode with 32 scans and wavelengths between 600 and 4000 cm^{-1}, analysed using the Spectrum 10 Software and compared with standard spectra libraries. The similarity between the spectra of the samples and the standard ones was accepted only if they had a matching score ≥0.70. Each plastic particle was also classified according to its shape (fragments – fibres – pellets), colour and size. The size of the plastic particles was determined using the ImageJ Software and the dimensional classification proposed by the ISO (2020), classifying them into MPs (from 1 μm to 1000 μm), large MPs (from 1 mm to 5 mm) and macroplastics (≥5 mm).

During the sampling phase, to evaluate any possible contamination by plastic, a Petri dish was kept open, with a cellulose nitrate filter inside. To monitor the eventual contamination by plastics during sample processing, nitrate cellulose membrane filters (n = 17) were also processed in parallel to samples during the homogenization and filtration process. A total of 5 fibres of polyester (PEST) were detected in blank filters, corresponding to 0.41 ± 0.80 plastics/sample. This quantity is well below the 10% of the average plastics detected in all samples, indicating a poor interference. Plastic debris detected in these filters were subtracted from the final count when the same shape, colour and polymer were also detected in the samples.

2.3. *Normalization procedure and statistical analysis*

The normalizing procedure aimed to estimate the plastic load generated by spreading these substrates across the agricultural area.

A fundamental step was to establish the average rate of application for each substrate (refer to Table 1), taking into account (i) the maximum amount of nutrients available for the culture, (ii) the current regulations and (iii) the dry matter and content of nutrients. The obtained values are in accordance with the current agricultural practices.

Then, the plastic load released in the agricultural field was estimated by combining the plastic measurement and the average rate of application.

Data is presented by mean ± standard deviation. To evaluate the significant differences between the plastic content in the different matrices, data were statistically analysed using GraphPad Prism 8.0.2 software package. The one-way analysis of variance (one-way ANOVA), followed by the Fisher LSD *post-hoc* test, was applied out taking $p \leq 0.05$ as a significant cut-off.

3. Results

3.1. *Plastic quantification and input into soil*

A total of 1156 particles were detected in all the investigated samples. Among them, 832 were identified as plastics, while 324 were cellulose-based debris (28%). Plastics represented a different fraction of analysed particles depending on the considered substrate, namely 12% in M1, 20% in M2, 6% in D, 61% in S1, 82% in S2, 59% in S3, and 89% in C, respectively. The size, shape, polymer and the library matching score of each plastic item are reported in Table S2.

Table 1
Average rate of application of each substrate to fields for maize production.

MATRIX	Average rate of application [t w.w./ha]
M1	40
M2	40
D	80
S1	26
S2	28
S3	31
C	10

The average number of plastics detected in each sample was 1.67 ± 1.16 in M1, 1.00 ± 0.00 in M2, 0.67 ± 0.58 in D, 42.33 ± 15.82 in S1, 57.67 ± 62.64 in S2, 34.33 ± 11.93 in S3 and 139.67 ± 6.11 in C. The concentration of plastics in g of wet weight and dry weight of each matrix is reported in Table 2 and Table S3, respectively. The content of plastics was significantly different in the matrices considering both the content measured on wet weight ($p < 0.0001$, $F_{6,14} = 199.6$) as well as on dry weight ($p < 0.0001$, $F_{6,14} = 142.2$): the higher abundance of plastics was detected in C, followed by the three S, while a much lower presence of plastic debris was measured in M1, M2 and D.

Considering the rate of application of each matrix to fields for maize production (Table 1), we estimated the possible yearly input of plastics to arable fields by using our data. Manures and D can introduce the same order of magnitude of plastics: 3.20×10^6 plastics/ha each year for M1 and M2 and 4.80×10^6 plastics/ha for D, respectively. A higher amount of plastic input is calculated for the three sewage sludges, ranging from 8.96×10^7 plastics/ha introduced by S2, to 9.73×10^7 plastics/ha introduced by S3, up to 1.32×10^8 plastics/ha introduced by S1. Lastly, C without compostable and biodegradable plastics (CBP) can enrich soil by 8.44×10^8 plastics/ha, while including also CBP the amount of plastics increases to 9.86×10^9 plastics/ha.

3.2. *Plastic characterization*

Concerning the quality characterization, the three matrices of animal origin showed a very similar plastic composition (Fig. 1): in M1 and D only fibres were detected, while in M2 67% of plastics were fibres and 33% were fragments. As for the size, MPs were more abundant than large MPs in both M1 and M2, and no macroplastics were detected. In D, MPs and large MPs occurred at the same percentage and again no macroplastics were detected. The polymeric composition was very homogeneous, showing mostly the presence of PEST particles, and a few items of polyacrylate (PAK) and polyethylene (PE).

Also in the three sewage sludges, fibres represented the majority of plastics, while fragments represented about 20% of the total. MPs accounted for 59–72% of the total, and some macroplastics were also detected in S2 and S3. The polymer composition was more heterogeneous with respect to M and D. In S1 PEST represented 52% of the total, polypropylene (PP) and PE were detected at 11–13%, while polyurethane (PU), PAK and other polymers were identified at lower percentage. PP was the most common polymer (35%) in S2 followed by PEST (20%). Polyamide (PA), PE, PAK and PU were detected at a frequency of about 10%, and other polymers occurred at a very low percentage. In S3 PEST was again the most common polymer (37%) followed by PP (26%), PAK and PE were detected at a frequency of 13% and 12% respectively, while PA and PU were the least abundant polymers (Fig. 2).

Table 2
Quantity of plastic items measured in the different matrices expressed as plastics/g in wet weight (N = 3 replicates). Different letters indicate statistically significant differences among samples ($p \leq 0.05$). C-no CBP = compost excluding compostable and biodegradable plastics.

matrix	plastics/g w.w.
M1	0.08 ± 0.02^a
M2	0.08 ± 0.00^a
D	0.06 ± 0.05^a
S1	5.07 ± 1.90^a
S2	3.20 ± 0.37^a
S3	3.14 ± 1.09^a
C	986 ± 87.83^b
C-no CBP	84.44 ± 80.91^c

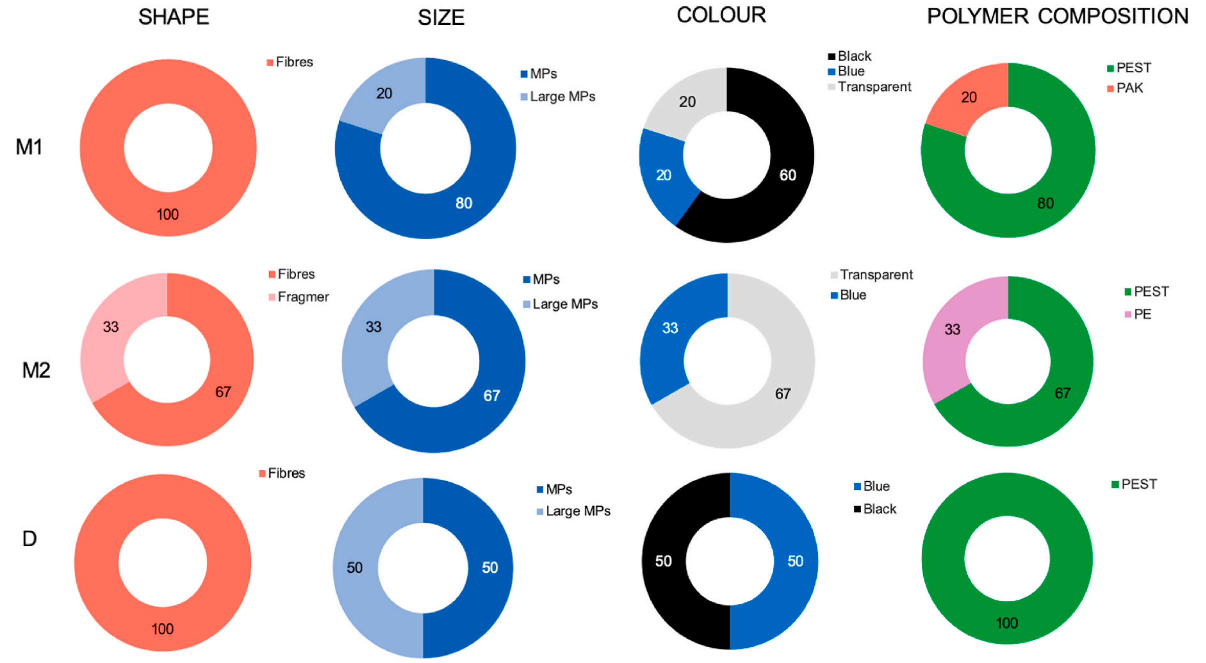


Fig. 1. Characterization of plastics in terms of size, shape, colour and polymer composition in swine manure (M1), bovine manure (M2) and digestate (D). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

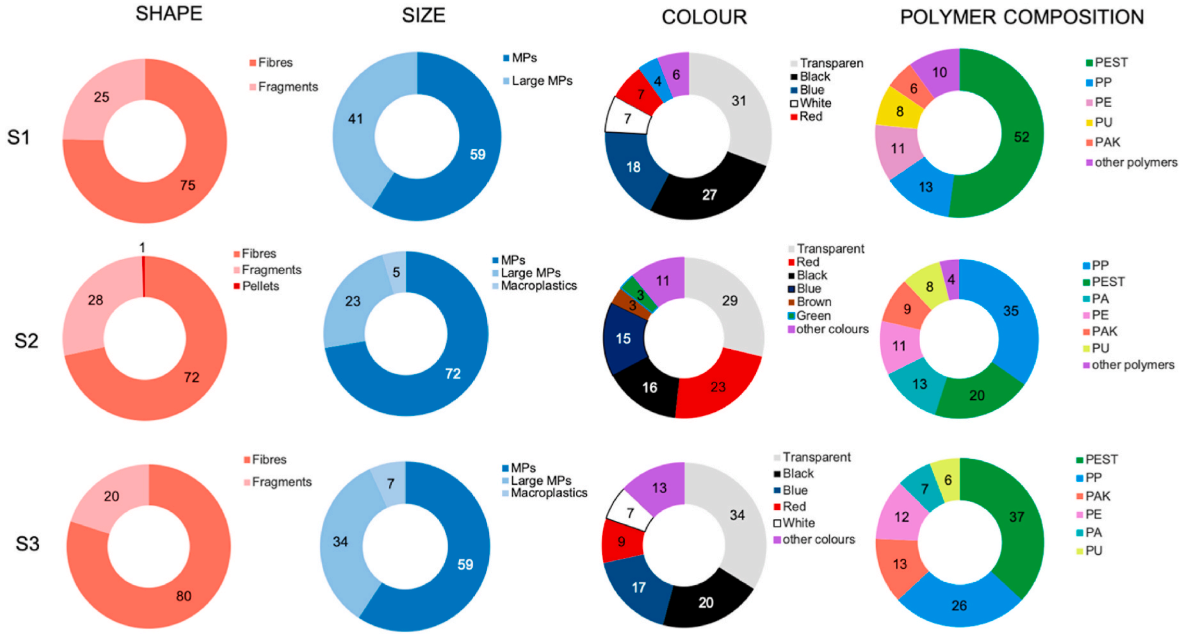


Fig. 2. Characterization of plastics in terms of size, shape, colour and polymer composition in the three sewage sludges collected in different WWTPs. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

The plastic items identified in C were almost fragments of MPs made for the 92% of compostable and biodegradable plastics (CBP). The remaining 8% was represented by plastic particles made of PEST (54%), PE (26%), PA (9%) and PP (6%), and other polymers were detected at low percentages (Fig. 3).

4. Discussion

The presence of plastics in biogenic matrices intended for reuse in agriculture can lead to a further plastic input into agricultural soils, thus rising an issue on possible negative consequences for the environment. Notwithstanding, the current knowledge concerning the presence of plastics in these residues is very poor. Therefore, the objective of the present study was a quali-quantitative characterization of plastics in different matrices reused in agriculture as manures, digestate, compost and sewage sludges. Our goal was to understand how the spread of these matrices, according to the common agricultural practice, can contribute to plastic accumulation in the soil ecosystem, thus providing useful information for the optimum management of these substrates.

Even though it is difficult to make comparisons among studies, due to the different sampling and detection methods, the plastic content measured in our samples is in line with levels reported in other studies

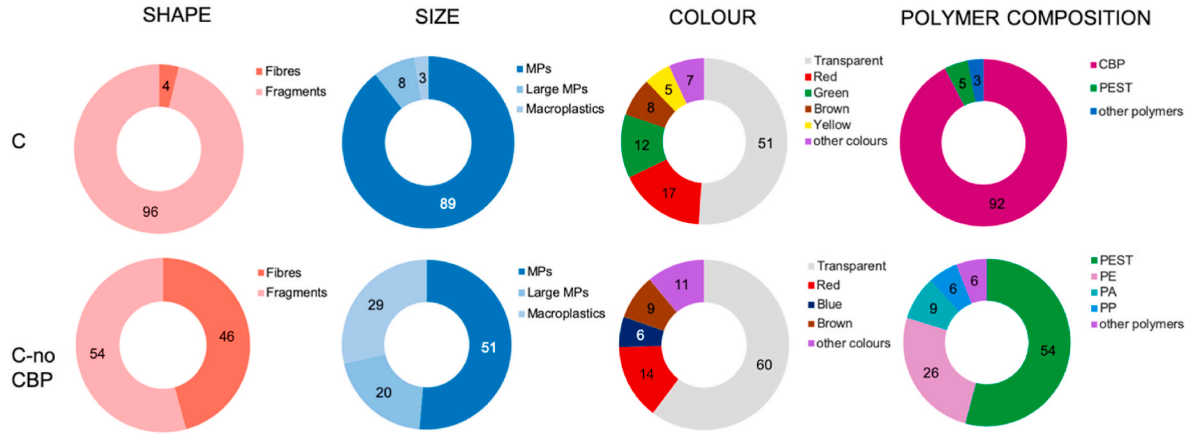


Fig. 3. Characterization of plastics in terms of size, shape, colour and polymer composition in compost (C) and compost excluding compostable and biodegradable plastics (C-no CBP). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

(Table S3), except for C, in which we detected a greater number of plastics than what is reported in the previous studies.

Based on our results, the spreading of all the matrices considered could lead to an enrichment of plastics for the soil, even though remarkable differences were observed among them, since human wastes can introduce from 25 to 200 times more plastics in the agricultural soil with respect to animal ones. It should be noted that each sample was collected only once and no replicates were taken over time. Therefore, albeit our samples are surely representative of wastes that are ready to be spread on land, we must consider that the plastics present in the different matrices may be subject to a certain degree of fluctuation over time, and therefore we may have measured the worst-case scenario or the minimum contamination of the different residues. This could have influenced the comparisons carried out between the matrices. However, both the two manures and the three different sewage sludges have similar levels of plastic, both from a qualitative and quantitative point of view, although they come from different plants. Therefore, we do not expect a huge variation in the content of plastics over time for these matrices.

Among animal matrices, the digestate was less contaminated than the M2 from which it is produced, suggesting that the further processing step (especially the separation phase after anaerobic digestion) could contribute to reducing the load of plastic in the manure. Indeed, if we consider that also the separated solid fraction is spread on land, the actual total plastic load is not expected to change.

Our results confirmed sewage sludge as an important reservoir of plastics from household and industrial origin, that could be transferred to natural ecosystems. Indeed, the spread of sewage sludge is considered a main driver of plastic contamination of agricultural soil, together with mulching (Crossman et al., 2020; Brandes et al., 2021) since this matrix represents the sink of plastics that enter WWTPs and sediment following weathering processes, fouling and flocculation (Magni et al., 2019). Estimations of an annual load of plastics spread on soils available in the literature confirm this outcome (Nizzetto et al., 2016). Also, a recent study confirmed that the long-term application of chicken manure, sludge and domestic waste compost significantly enriched the content of MPs of soil in a field with wheat-maize rotation. Moreover, the MP content increased over time, due to the fragmentation and degradation of plastics in smaller pieces (Zhang et al., 2022).

Concerning the qualitative characterization of plastics, the detected shapes, represented by fibres and fragments, highlighted that plastics in all the analysed matrices originated from fragmentation of larger items, while no pellets or beads -which characterize plastics of primary origin- were found. This evidence contributes to identify the origin of the plastic found in the different matrices, which is a key aspect for identifying possible solutions for the reduction of this contamination.

The two manures are different due to the type of animals bred (e.g., the dry content in M1 – swine manure – is significantly lower than M2 ones – bovine manure), the feeding system, the mode of the removal of manures from livestock housings and their stocking conditions. Nevertheless, the quali-quantitative characterization of plastics seems to confirm that regardless of the type of farming, manures and digestate are subject to a fairly limited input of plastics of similar origin. The contamination might have different origins: primarily the ingestion by livestock of feed contaminated with plastics. For instance, fibres and fragments of PP and PE were found in feed samples from swine and bovine farms in China (Wu et al., 2021). Mazzoleni and collaborators (2023) found fragments of PP and PE in former food products included in animal feed. Another important contribution to plastic contamination could derive from equipment and instruments used in the breeding plants (manure scrapers, water supply system and work wears just to mention some). Lastly, since fibres represent the largest fraction of atmospheric MPs (Su et al., 2023), contamination through atmospheric deposition cannot be excluded (manure is stored in open air lagoons at least 120 days, before being spread on land).

As expected, the plastic composition of sewage sludges was more heterogeneous than the animal matrices. Albeit the three sewage sludges considered in this study are different in the nominal capacity of the WWTPs, type of treated wastewater (with different fraction of industrial discharges with respect to domestic ones) and process scheme, the plastic chemical composition was quite similar and showed the presence of polymers in line with what is normally detected within WWTPs (Sun et al., 2019). The largest fraction of plastics identified in all the three sludges are fibres of PEST and to a lower extent PA. Another polymer present in synthetic fibres, often found in all sewage sludges, is PAK. These fibres of textile origin likely reach WWTPs being released from clothes-washing machines (Napper and Thompson, 2016). Indeed, a study by De Falco et al. (2019) measured up to 1,500,000 fibres released from one washing, highlighting that this route heavily contributes to the global MP pollution of natural ecosystems. Furthermore, Zubris and Richards (2005) demonstrated that fibres released in soil upon treatment with sewage sludges could be retained in this matrix for up to 5 years after application. Other polymers detected in high percentage were PP, which is used mainly for packaging and liquid handling and PE, that can be released from several household goods such as single-use items, bottles, bags, food containers and toys, as well as from pipes for water and sewage.

Lastly, the highest level of plastic contamination occurred in C. The plastic content observed in the present study is higher than those reported in the previous studies (Table S3). This is largely related to the presence of a very high number of CBPs, which have not been detected in the other research. The presence of CBPs in compost could be due to the fact that the complete degradation of these plastics is slow and can take days to months depending on composting conditions such as temperature, humidity and the initial size of CBP (Lavagnolo et al., 2020; Cucina et al., 2021a, 2021b). According to international standards, plastics can be labelled as compostable if at least 90% of the weight is reduced into particles smaller than 2 mm within 3 months and mineralizes within 6 months (Gerritse et al., 2020). Moreover, during the biodegradation process the transient formation of fragments of small size (μm range) occurs (Wohlleben et al., 2023). Therefore, a significant amount of CBP fragments of small size could be still present in the matrix and spread in the environment, if the compost cycle and the delivery time for spreading do not match (Agarwal, 2020).

The effects of CBP on soil have been barely investigated, however CBP might modify the soil physico-chemical properties, and the soil microbial community (Zhou et al., 2021), induce negative effects on different organisms, and might affect plant performances (reviewed in Liwarska-Bizukojc, 2021; Fan et al., 2022). This evidence highlights the need to evaluate the actual release of CBP from compost into soil upon treatment, as well as to expand current information concerning the potential hazard of CBP for soil communities under realistic exposure scenarios.

Besides CBP, we also identified a number of plastics in C much higher than in all the other matrices. Indeed, the presence of plastics in this matrix has already been demonstrated (Bandini et al., 2022), since plastics can occur mainly due to incorrect disposal and poor waste separation in composting plants (Gui et al., 2021; Scopetani et al., 2020). Moreover, differently from the other matrices, most of the plastic debris detected in C were fragments, therefore indicating an origin more linked to the fragmentation of plastic items than to the washing of synthetic garments, as previously discussed. Concerning the polymeric composition, PEST and PE were the most abundant polymers, in line with observations from other studies (Weithmann et al., 2018; Gui et al., 2021). In particular, PE plastics likely originate from the fragmentation of plastic bags disposed of incorrectly in organic wastes. Also, contamination after door-to-door collection cannot be excluded. Compost application has emerged as a major pathway for plastic entry into agroecosystems, with plastic loads in compost reaching levels of up to 1.36 g/kg (Gajst, 2016; Bläsing and Amelung, 2018; Braun et al., 2021). A recent study demonstrated that plastic mulching and application of organic compost significantly increased the content of MPs in apple orchards over time (Wang et al., 2023). The application of compost, particularly composted municipal biowaste or green cuttings, has gained popularity in Europe as a means to enhance soil fertility, with an increase in compost production observed from 14 million tons in 1995 to approximately 37 million tons in 2018 (Eurostat, 2022). However, this widespread use of compost is contributing to plastic pollution in agricultural fields. Braun et al. (2023) highlighted that plastic contamination varies widely among composts, with plastic content ranging from 0.002 to 1.36 g/kg dry weight and 22 ± 2 to 2800 ± 616 plastic items/kg dry weight. Factors contributing to these variations include the origins of the compost and different analytical methods employed in various studies. Despite efforts by compost plants to reduce plastic content through sorting and sieving, plastic persists in the final compost product.

Our data clearly underline the need to consider the presence of plastics in all the biogenic matrices as an important route for their input in soil, and the consequent need to improve the control at source. In fact, removing MPs in WWTP effluent and excess sludge as well as in manures and in the composting facilities is quite challenging (Sadia et al., 2022; Hassan et al., 2023). In this perspective, the replacement of plastic goods used in livestock with bio-based products and the reduction of plastic contamination of feed are suggested measures that could reduce plastic input in animal matrices. The presence of textile microfibres in sludges could be reduced by improving fabric, softeners and detergent types and washing conditions (De Falco et al., 2018). Suggestions for modifying the procedure of collection of the organic fraction of municipal wastes (e.g. paper instead of CBP bags) could also arise from the outcomes of this study.

5. Conclusions

Our study pointed out that the spread of biogenic matrices can contribute to plastic accumulation in the agroecosystems. Nevertheless, in the absence of an adequate environmental risk assessment, it is not possible to establish whether the detected levels of plastics could represent an actual risk for the agricultural systems in which they are applied and the human health. It is therefore mandatory to increase the current knowledge concerning the adverse impacts of plastics on soil biodiversity and ecosystem functions under relevant exposure scenarios. Based on an increased awareness of this relevant issue, it may be necessary to introduce stricter controls and limitations on the acceptable residual concentrations of MPs in organic matrices to be reused in agriculture. For example, most of the plastics found were less than 2 mm. Consequently, it could be important to include also the lower size of particles in the current legislation on compost. Furthermore, in addition to the matrices already subjected to regulation for plastics, it could be useful to set limits for all biogenic matrices intended for agricultural reuse, such as sewage sludge and manure.

In view of the state of art of technology for chemical-physical and biological treatment of wastewater and wastes, which makes the elimination of MPs almost impossible, the reduction of the potential negative impact of this class of contaminants in soil ecosystems, suggests developing control measures and good practices to prevent their release at source.

CRediT authorship contribution statement

Stefano Magni: Writing – original draft, Validation, Methodology. **Marco Fossati:** Writing – review & editing, Investigation. **Roberta Pedrazzani:** Writing – review & editing, Formal analysis, Conceptualization. **Alessandro Abbà:** Writing – review & editing, Resources, Formal analysis. **Marta Domini:** Writing – review & editing, Project administration. **Michele Menghini:** Writing – review & editing, Resources, Formal analysis. **Sara Castiglioni:** Writing – review & editing, Investigation. **Giorgio Bertanza:** Writing – review & editing, Supervision, Funding acquisition. **Andrea Binelli:** Writing – review & editing, Conceptualization. **Camilla Della Torre:** Writing – original draft, Supervision, Funding acquisition, Conceptualization.

Declaration of competing interest

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

Data availability

Data will be made available on request.

Acknowledgements

This study was supported by the Fondazione Cariplo [SLURP - SLUdge Recovery in agriculture: environment and health Protection Project - grant number 2020-1029].

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2024.123986.

References

Agarwal, S., 2020. Biodegradable polymers: present Opportunities and challenges in providing a microplastic-Free environment. Macromol. Chem. Phys. 221, 2000017 https://doi.org/10.1002/macp.202000017.

Bandini, F., Taskin, E., Bellotti, G., Vaccari, F., Misci, C., Guerrieri, M.C., Cocconcelli, P. S., Puglisi, E., 2022. The treatment of the organic fraction of municipal solid waste (OFMSW) as a possible source of micro- and nano-plastics and bioplastics in agroecosystems: a review. Chem. Biol. Technol. Agric. 9, 4. https://doi.org/10.1186/s40538-021-00269-w.

Bläsing, M., Amelung, W., 2018. Plastics in soil: analytical methods and possible sources. Sci. Total Environ. 612, 422–435. https://doi.org/10.1016/j.scitotenv.2017.08.086.

Brandes, E., Henseler, M., Kreins, P., 2021. Identifying hot-spots for microplastic contamination in agricultural soils—a spatial modelling approach for Germany. Environ. Res. Lett. 16, 104041 https://doi.org/10.1088/1748-9326/ac21e6.

Braun, M., Mail, M., Heyse, R., Amelung, W., 2021. Plastic in compost: prevalence and potential input into agricultural and horticultural soils. Sci. Total Environ. 760, 143335 https://doi.org/10.1016/j.scitotenv.2020.143335.

Braun, M., Mail, M., Krupp, A.E., Amelung, W., 2023. Microplastic contamination of soil: are input pathways by compost overridden by littering? Sci. Total Environ. 855, 158889 https://doi.org/10.1016/j.scitotenv.2022.158889.

Buks, F., Kaupenjohann, M., 2020. Global concentrations of microplastic in soils, a review. Soils 6, 649–662. https://doi.org/10.5194/soil-6-649-2020.

Chae, Y., An, Y.J., 2018. Current research trends on plastic pollution and ecological impacts on the soil ecosystem: a review. Environ. Pollut. 240, 387–395. https://doi.org/10.1016/j.envpol.2018.05.008.

Colombini, G., Rumpel, C., Houot, S., Biron, P., Dignac, M.F., 2022. A long-term field experiment confirms the necessity of improving biowaste sorting to decrease coarse microplastic inputs in compost amended soils. Environ. Pollut. 315, 120369 https://doi.org/10.1016/j.envpol.2022.120369.

Council Directive 86/278, 1986 EEC of 12 June 1986 on the protection of the environment, and in particular of the soil, when sewage sludge is used in agriculture (https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX:31986L0278) .

Council Directive 91/676, 1991 EEC of 12 December 1991 concerning the protection of waters against pollution caused by nitrates from agricultural sources. (https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX:31991L0676) .

Cucina, M., De Nisi, P., Trombino, L., Tambone, F., Adani, F., 2021a. Degradation of bioplastics in organic waste by mesophilic anaerobic digestion, composting and soil incubation. Waste Manag. 134, 67–77. https://doi.org/10.1016/j.wasman.2021.08.016.

Crossman, J., Hurley, R.R., Futter, M., Nizzetto, L., 2020. Transfer and transport of microplastics from biosolids to agricultural soils and the wider environment. Sci. Total Environ. 724, 138334 https://doi.org/10.1016/j.scitotenv.2020.138334.

Cucina, M., de Nisi, P., Tambone, F., Adani, F., 2021b. The role of waste management in reducing bioplastics' leakage into the environment: a review. Bioresour. Technol. 337, 125459 https://doi.org/10.1016/j.biortech.2021.125459.

De Falco, F., Gullo, M.P., Gentile, G., Di Pace, E., Cocca, M., Gelabert, L., Brouta-Agnésa, M., Rovira, A., Escudero, R., Villalba, R., Mossotti, R., Montarsolo, A., Gavignano, S., Tonin, C., Avella, M., 2018. Evaluation of microplastic release caused by textile washing processes of synthetic fabrics. Environ. Pollut. 236, 916–925. https://doi.org/10.1016/j.envpol.

De Falco, F., Di Pace, E., Cocca, M., Avella, M., 2019. The contribution of washing processes of synthetic clothes to microplastic pollution. Sci. Rep. 9, 6633. https://doi.org/10.1038/s41598-019-43023-x.

EU, 2019/1009. Regulation of the European Parliament and of the Council of 5 June 2019 Laying Down Rules on the Making Available on the Market of EU Fertilizing Products and Amending Regulations (EC) No 1069/2009 and (EC) No 1107/2009 and Repealing Regulation (EC) No 2003/2003.

Eurostat, 2022. Eurostat, 2022. Eurostat: estimated by Eurostat-(online data code: env_wasmun). https://appsso.eurostat.ec.europa.eu/nui/show.do?dataset=env_wasmun&lang=en.

Fan, P., Yu, H., Xi, B., Tan, W., 2022. A review on the occurrence and influence of biodegradable microplastics in soil ecosystems: are biodegradable plastics substitute or threat? Environ. Int. 163, 107244 https://doi.org/10.1016/j.envint.2022.107244.

Gajst, T., 2016. Analysis of Plastic Residues in Commercial Compost. Thesis Univ Nov Goric.

Gerritse, J., Leslie, H.A., de Tender, C.A., Devriese, L.I., Vethaak, A.D., 2020. Fragmentation of plastic objects in a laboratory seawater microcosm. Sci. Rep. 10, 1–16. https://doi.org/10.1038/s41598-020-67927-1.

Gui, J., Sun, Y., Wang, J., Chen, X., Zhang, S., Wu, D., 2021. Microplastics in composting of rural domestic waste: abundance, characteristics, and release from the surface of macroplastics. Environ. Pollut. 274, 116553 https://doi.org/10.1016/j.envpol.2021.116553.

Guo, J.J., Huang, X.P., Xiang, L., Wang, Y.Z., Li, Y.W., Li, H., Cai, Q.Y., Mo, C.H., Wong, M.H., 2020. Source, migration and toxicology of microplastics in soil. Environ. Int. 137, 105263 https://doi.org/10.1016/j.envint.2019.105263.

Hassan, F., Prasetya, K.D., Hanun, J.N., Bui, H.M., Rajendran, S., Kataria, N., Khoo, K.S., Wang, Y.-F., You, S.-J., Jiang, J.-J., 2023. Microplastic contamination in sewage sludge: abundance, characteristics, and impacts on the environment and human health. Environ. Technol. Innovat. 103176 https://doi.org/10.1016/j.eti.2023.103176.

He, D., Luo, Y., Lu, S., Liu, M., Song, Y., Lei, L., 2018. Microplastics in soils: analytical methods, pollution characteristics and ecological risks. Trends Anal. Chem. 109, 163–172. https://doi.org/10.1016/j.trac.2018.10.006.

Hurley, R.R., Nizzetto, L., 2018. Fate and occurrence of micro(nano)plastics in soils: knowledge gaps and possible risks. Curr. Opin. Environ. Sci. Heal. 1, 6–11. https://doi.org/10.1016/j.coesh.2017.10.006.

International Organization for Standardization, 2020. ISO/TR 21960:2020 – Plastics - Environmental Aspects — State of Knowledge and Methodologies, p. 30. First Edition 2020 02.

Jiang, M., Wang, B.Q., Ye, R., Yu, N., Xie, Z.M., Hua, Y.J., Zhou, R.H., Tian, B., Dai, S., 2022. Evidence and impacts of nanoplastic accumulation on crop grains. Adv. Sci. 2202336 https://doi.org/10.1002/advs.202202336.

Lavagnolo, M.C., Ruggero, F., Pivato, A., Boaretti, C., Chiumenti, A., 2020. Composting of Starch-based Bioplastic bags: small Scale test of degradation and size reduction trend. Detritus 12, 57–65. https://doi.org/10.31025/2611-4135/2020.14008.

Liwarska-Bizukojc, E., 2021. Effect of (bio)plastics on soil environment: a review. Sci. Total Environ. 15, 148889 https://doi.org/10.1016/j.scitotenv.2021.148889.

Magni, S., Binelli, A., Pittura, L., Avio, C.G., Della Torre, C., Parenti, C.C., Regoli, F., 2019. The fate of microplastics in an Italian wastewater treatment plant. Sci. Total Environ. 652, 602–610. https://doi.org/10.1016/j.scitotenv.2018.10.269.

Magni, S., Nigro, L., Della Torre, C., Binelli, A., 2021. Characterization of plastics and their ecotoxicological effects in the Lambro River (N. Italy). J. Hazard Mater. 412, 125204 https://doi.org/10.1016/j.jhazmat.2021.125204.

Mazzoleni, S., Magni, S., Tretola, M., Luciano, A., Ferrari, L., Bernardi, C.E.M., Lin, P., Ottoboni, M., Binelli, A., Pinotti, L., 2023. Packaging contaminants in former food products: using Fourier Transform Infrared Spectroscopy to identify the remnants and the associated risks. J. Hazard Mater. 448, 130888 https://doi.org/10.1016/j.jhazmat.2023.130888.

Napper, I.E., Thompson, R.C., 2016. Release of synthetic microplastic plastic fibres from domestic washing machines: effects of fabric type and washing conditions. Mar. Pollut. Bull. 112, 39–45. https://doi.org/10.1016/j.marpolbul.2016.09.025.

Ng, E.L., Huerta, Lwanga, E., Eldridge, S.M., Johnston, P., Hu, H.W., Geissen, V., Chen, D., 2018. An overview of microplastic and nanoplastic pollution in agroecosystems. Sci. Total Environ. 627, 1377–1388. https://doi.org/10.1016/j.scitotenv.2018.01.341.

Nizzetto, L., Futter, M., Langaas, S., 2016. Are agricultural soils dumps for microplastics of urban origin? Environ. Sci. Technol. 20, 10777–10779. https://doi.org/10.1021/acs.est.6b04140.

Rilling, M., Lehman, A., 2020. Microplastic in terrestrial ecosystems. Science 368, 1430–1431. https://doi.org/10.1126/science.abb5979.

Saadu, I., Farsang, A., 2023. Plastic contamination in agricultural soils: a review. Environ. Sci. Eur. 35, 13. https://doi.org/10.1186/s12302-023-00720-9.

Sadia, M., Mahmood, A., Ibrahim, M., Irshad, M.K., Quddusi, A.H.A., Bokhari, A., Mubashir, M., Chuaff, L.F., Show, P.L., 2022. Microplastics pollution from wastewater treatment plants: a critical review on challenges, detection, sustainable removal techniques and circular economy. Environ. Technol. Innovat. 102946 https://doi.org/10.1016/j.eti.2022.102946.

Sajjad, M., Huang, Q., Khan, S., Khan, M.A., Liu, Y., Wang, J., Lian, F., Wang, Q., Guo, G., 2022. Microplastics in the soil environment: a critical review. Environ. Technol. Innovat. 27, 102408 https://doi.org/10.1016/j.eti.2022.102408.

Scopetani, C., Chelazzi, D., Mikola, J., Leinio, V., Heikkinen, R., Cincinelli, A., Pellinen, J., 2020. Olive oil-based method for the extraction, quantification and identification of microplastics in soil and compost samples. Sci. Total Environ. 733, 139338 https://doi.org/10.1016/j.scitotenv.2020.139338.

Sivarajah, B., Lapen, D.R., Gewurtz, S.B., Smyth, S.A., Provencher, J.F., Vermaire, J.C., 2023. How many microplastic particles are present in Canadian biosolids? J. Environ. Qual. 52, 1037–1048. https://doi.org/10.1002/jeq2.20497.

Su, L., Xiong, X., Zhang, Y., Wu, C., Xu, X., Sun, C., Shi, H., 2023. Global transportation of plastics and microplastics: a critical review of pathways and influences. Sci. Total Environ. 154884 https://doi.org/10.1016/j.scitotenv.2022.154884.

Sun, J., Dai, X., Wang, Q., van Loosdrecht, M.C.M., Ni, B.J., 2019. Microplastics in wastewater treatment plants: detection, occurrence and removal. Water Res. 152, 21–37. https://doi.org/10.1016/j.watres.2018.12.050.

United Nations Environment Programme, 2022. Plastics in Agriculture – an Environmental Challenge, vol. 29. Foresight Brief, Nairobi.

Wang, W., Ge, J., Yu, X., Li, H., 2020. Environmental fate and impacts of microplastics in soil ecosystems: progress and perspective. Sci. Total Environ. 708, 134841 https://doi.org/10.1016/j.scitotenv.2019.134841.

Wang, Y., Liu, L., Cao, S., Yu, J., Li, X., Su, Y., Li, G., Gao, H., Zhao, Z., 2023. Spatio-temporal variation of soil microplastics as emerging pollutant after long-term application of plastic mulching and organic compost in apple orchards. Environ. Pollut. 328, 121571 https://doi.org/10.1016/j.envpol.2023.121571.

Watteau, F., Dignac, M., Bouchard, A., Revallier, A., Houot, S., 2018. Microplastic detection in soil amended with municipal solid waste composts as revealed by transmission electronic microscopy and pyrolysis/gc/ms. Front. Sustain. Food Syst. 2, 81. https://doi.org/10.3389/fsufs.2018.00081.

Weithmann, N., Moller, J.N., Loder, M.G.J., Piehl, S., Laforsch, C., Freitag, R., 2018. Organic fertilizer as a vehicle for the entry of microplastic into the environment. Sci. Adv. 4, 1–8. https://doi.org/10.1126/sciadv.aap8060.

Wohlleben, W., Ruckel, M., Meyer, L., Pfohl, P., Battagliarin, G., Hüffer, T., Zumstein, M., Hofmann, T., 2023. Fragmentation and mineralization of a compostable aromatic–aliphatic polyester during industrial composting. Environ. Sci. Technol. Lett. 10, 698–704. https://doi.org/10.1021/acs.estlett.3c00394.

Wu, R.T., Cai, Y.F., Chen, Y.X., Yang, Y.W., Xing, S.C., Liao, X.D., 2021. Occurrence of microplastic in livestock and poultry manure in South China. Environ. Pollut. 277, 116790 https://doi.org/10.1016/j.envpol.2021.116790.

Zhang, L., Xie, Y., Liu, J., Zhong, S., Qian, Y., Gao, P., 2020. An overlooked entry pathway of microplastics into agricultural soils from application of sludge-based fertilizers. Environ. Sci. Technol. 54, 4248–4255. https://doi.org/10.1021/acs.est.9b07905.

Zhang, J.J., Wang, X.X., Xue, W.T., Xu, L., Ding, W.C., Zhao, M., Chen, Y.H., 2022. Microplastics pollution in soil increases dramatically with long-term application of organic composts in a wheat–maize rotation. J. Clean. Prod. 356, 131889 https://doi.org/10.1016/j.jclepro.2022.131889.

Zhou, J., Gui, H., Banfield, C.C., Wen, Y., Zang, H.D., Dippold, M.A., Charlton, A., Jones, D.L., 2021. The microplastisphere: biodegradable MPs addition alters soil microbial community structure and function. Soil Biol. Biochem. 156, 108211 https://doi.org/10.1016/j.soilbio.2021.108211.

Zhu, J., Dong, G., Feng, F., Ye, J., Liao, C.H., Wu, C.H., Chen, S.C., 2023. Microplastics in the soil environment: focusing on the sources, its transformation and change in morphology. Sci. Total Environ. 165291 https://doi.org/10.1016/j.scitotenv.2023.165291.

Zubris, K.A.V., Richards, B.K., 2005. Synthetic fibers as an indicator of land application of sludge. Environ. Pollut. 138, 201–211. https://doi.org/10.1016/j.envpol.2005.04.013.