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Research Paper

Plastic input and dynamics in industrial composting

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ABSTRACT

Green and biowaste, processed within large facilities into compost, is a key fertilizer for agricultural and horticultural soils. However, due to improper waste disposal of plastic, its residues often remain or even lead to the formation of microplastics (1 µm – 5 mm, MiPs) in the final compost product. To better understand the processes, we first quantified 'macroplastics' (> 20 mm, MaPs) input via biowaste collection into an industrial composting plant, and, then determined MiP concentrations at five stages during the composting process (before and after shredding and screening processes), and in the water used for irrigation. The total concentrations of MaPs in the biowaste collected from four different German districts ranged from 0.36 to 1.95 kg ton^{-1} biowaste, with polyethylene (PE) and polypropylene (PP) representing the most abundant types. The "non-foil" and "foil" plastics occurred in similar amounts (0.51 ± 0.1 kg ton^{-1} biowaste), with an average load of 0.08 ± 0.01 items kg^{-1} and 0.05 ± 0.01 items kg^{-1}, respectively. Only 0.3 ± 0.1 kg MaP t^{-1} biowaste was biodegradable plastic. Compost treatment by shredding tripled the total number of MaPs and MiPs to 33 items kg^{-1}, indicating an enrichment of particles during the process and potential fragmentation. Noticeably, a substantial amount of small MiPs (up to 22,714 ± 2,975 particles L^{-1}) were found in the rainwater used for compost moistening, being thus an additional, generally overlooked plastic source for compost. Our results highlight that reducing plastic input via biowaste is key for minimizing MiP contamination of compost.

1. Introduction

Plastic production increased by 4 % from 2020 to 2021, resulting in 390 Mt produced globally, nevertheless, proper recycling and disposal of plastics are often insufficient (PlasticsEurope, report 2022). Consequently, plastic items are found also in biowaste, and, if not removed during composting – in compost (Van den Zee and Molenveld, 2020; Edo et al., 2022; Braun et al., 2021; Laforsch et al., 2023). However, the dynamics of plastics within a compost facility, i.e., the plastic input by biowaste, as well as their removal or enrichment during different steps of composting, are still unclear.

Biowaste, including green waste, is often sold as compost after respective treatment in large composting companies. In agriculture, as well as in gardening horticulture, compost is used as an important organic fertilizer and soil amendment, as it improves soil quality and fertility, and additionally supports a circular economy (Siebert and Kehres, 2008; Viaene et al., 2016; Colombini et al., 2022). The EU compost production equals 17.3 million tons per year, with Germany being the largest compost producer. Around 14 million tons of this compost are derived from green and biowaste, and about 85 % of the final compost is reused as a fertilizer (European Compost Network (ECN) report, 2019). However, the biowaste used for compost production may contain macroplastics (MaPs, >20 mm), mesoplastics (MePs, 5 mm – 20 mm), and microplastics (MiPs, 1 µm – 5 mm). Especially kitchen waste tends to be the most polluted (Ricci and Centemero, 2018, Friege and Eger, 2022). If biowaste is not separated by type at the source or plastic

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is not properly removed before composting, these plastic particles might enrich the final product or break down to smaller sizes, i.e., forming further MiPs or even submicron plastic particles (<1 μm). Hence, compost application was identified as a principal input pathway of plastics into agricultural soils (Kumar et al., 2020; Sajjad et al., 2022; Porterfield et al., 2023; Braun et al., 2021; 2023).

Municipalities and operators of composting facilities pursue different strategies for collecting unmixed biowaste and removing foreign matter, including plastics. Typically, after collecting, green and biowaste undergo sorting, shredding, and sieving stages, preparing it for subsequent decomposition (Edo et al., 2022). This decomposition occurs under certain humidity and temperature conditions for a predetermined period through the activity of microorganisms (Diaz et al., 2007). Simultaneously, European regulations govern the acceptable presence of foreign materials in the final compost product, such as metals, glass and plastics. The compost cannot be commercialized if it contains more than 0.5 % foreign matter (dry weight, dw), larger than 2 mm. The regulations aim to reduce the plastic content to 0.25 g kg^{-1} dw from July 2026 onwards (*Bundesgütegemeinschaft Kompost* (BGK), Federal Quality Association for Compost, 2021; ECN, 2021; ECN, Germany, 2023).

Steiner et al. (2023) screened 14 waste treatment plants with anaerobic and aerobic treatments for the presence of plastics. The authors highlighted that final sieving efficiently reduced the number of MaPs and MePs but lacked efficiency for smaller MiPs. In aerobic facilities, the final product contained plastic fragments (mainly polyethylene (PE)) over 5 mm and between 1–5 mm, i.e., 101 and 21 items kg^{-1} dw, respectively. Conversely, anaerobic digestion produced liquid fertilizer with up to 10,000 particles of 10–1000 μm, raising more environmental concerns. In another study, placed 5x5 cm plastic pieces (PE, polypropylene (PP), polylactic acid (PLA), and polystyrene (PS)) in compost piles generated17 to 52 MiP particles during composting (>50 μm, Sholokhova et al., 2023). Although recent studies underscore the possibility of plastic fragmentation in composting facilities (e.g., Groß et al., 2024), there is little information available on the true plastic inputs; i.e., potential correlations of plastic input and plastic load of biowaste and composted material during the different steps of compost production. Especially, the contamination of the irrigation water during the composting process with MiP has not been evaluated, yet.

Here, we aimed to better understand the input and dynamics of plastics during composting. We hypothesized that i) most large plastic items entering the compost facility would be effectively removed after composting. Further, we assumed that ii) there will be a relative enrichment of small MiPs during the composting process, i.e., every treatment step during composting will reduce the size of plastic particles but increase the number of such small particles that the sieves cannot separate. Finally, we hypothesized that iii) the composting process will lead to an additional MiP contamination in the water used to irrigate the compost. To test these hypotheses, we monitored the plastic content, starting with the fresh waste throughout the whole composting process in a large industrial composting facility in Germany, processing biowaste. In detail, we determined the content of large plastic items in the incoming biowaste and analyzed the plastic content after each treatment step and in finished compost (i.e., at six sampling points) as well as in the irrigation water before and after composting.

2. Materials and methods

2.1. Compost facility

The experiment was carried out at a facility using tunnel composting

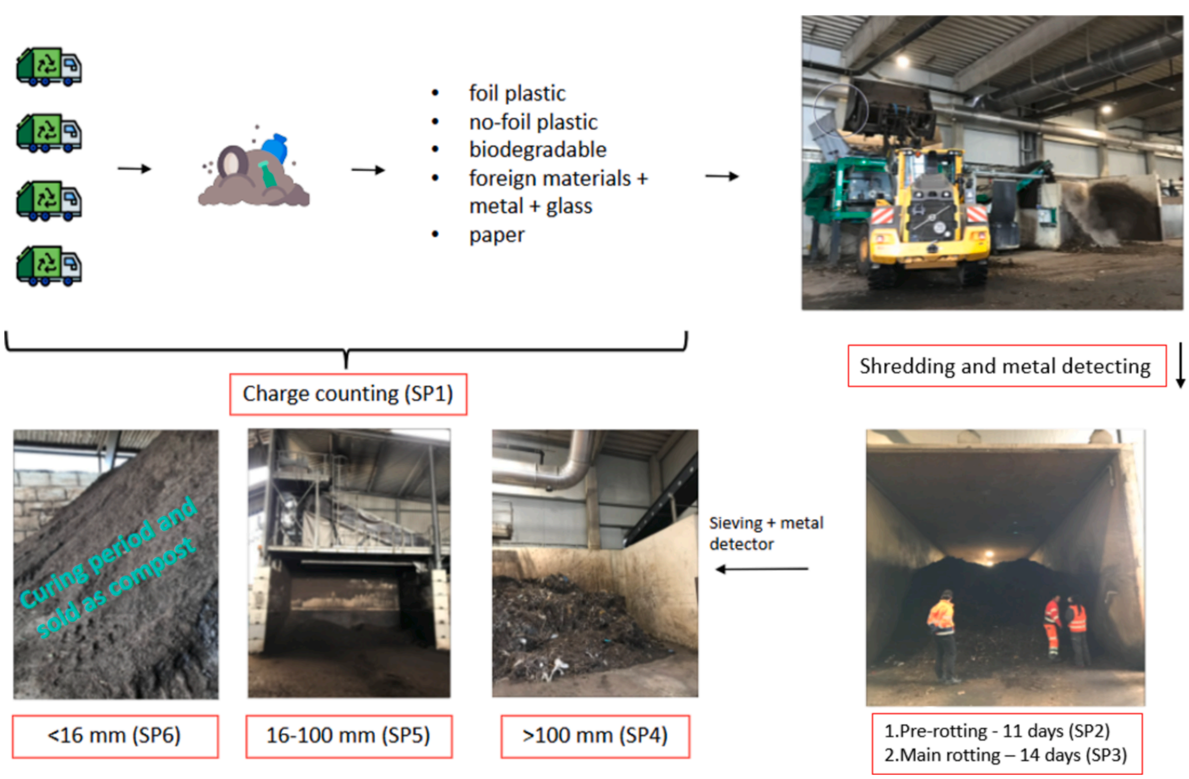

Fig. 1. Schematic representation of the composting process, with each sampling point (SP) distinctly underlined and squared in red, corresponding to the fractions collected for macroplastics, mesoplastics and microplastics. Following the arrows from left to right, charge counting (SP1) is done for quality control by carefully searching for foreign materials within the initial biowaste. Subsequently, the waste undergoes shredding and metal detection before entering a tunnel for pre-rotting (11 days, SP2). This is followed by a round of decompacting and main composting (14 days, SP3). At the end of the main rotting, the compost is sieved into three fractions (SP4-SP6) and ferrous metals are removed again. The fraction > 100 mm (SP4) is disposed of in most cases. The final fraction, <16 mm (SP6), is marketed and serves as a valuable soil amendment and organic fertilizer (the icons for the image are taken from www.flaticon.com). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

(annual input quantity: 30.000 tons of organic waste, consisting of around 98 % biowaste and around 2 % of green waste), producing around 7.000 tons of compost per year.

The organic and green waste was first shredded and subjected to ferrous metal separation via a conveyor belt (Fig. 1, sampling point 1 (SP1), Figure S1). A wheel loader then transported it into a tunnel, for pre-rotting under controlled pressurized ventilation (spigot system) and irrigation via a sprinkler system for 11 days (Fig. 1, SP2, Figure S1). The temperature development is measured directly within the rotting material and indirectly within the exhaust air. In addition, the direct temperature measurement (core temperature) serves as proof of hygiene (Figure S2). Excess water that escapes from the organic material during the rotting process (process water) is drained into a collecting tank via a channel system on the tunnel floor. After pre-rotting, the rotting material is transported out of the tunnel with a wheel loader, loosened, and mixed in a decompactor. A wheel loader then conveys the material into another tunnel for main rotting (duration: 14 days, Fig. 1, SP3; temperature reaching 70 °C after the second day, decreasing to 30 °C at the end of the process; Figure S2). The waste is then de-compacted again using wheel loaders and roughly fractionated using star sieves. Here the fraction > 100 mm is separated (Fig. 1, SP4), which is either fed back into the composting cycle as a feedstock or disposed of (incineration), depending on the foreign matter content.

A conveyor carries the fraction < 100 mm to the fine processing hall, where a second metal removal is conducted before the composted material is divided into two fractions through a flip-flow screen: 16 to 100 mm (Fig. 1, SP5) and < 16 mm (Fig. 1, SP6).

2.2. Sampling

2.2.1. Charge counting (incoming biowaste) for plastics > 20 mm and other foreign matter (SP1)

To identify the income of plastic items > 20 mm and other foreign matter via the collected biowaste, we conducted a so-called batch analysis (*Chargenanalyse*) for 4 different trucks from four districts ("District 1–4", Table 1), with three of them feeding the plant for later sampling (SP1). A "batch" corresponds to a sample quantity of 2x250 kg, representing the contents of a collection vehicle and helps to allocate possible waste contamination to specific collection areas (Figure S3). This procedure follows the BGK (2021) method for "Batch analysis", mirroring the company's process as well as guidelines for foreign materials treatment indicated in ECN, 2021. In addition, we analyzed a fifth sample (1 × 500 kg) as an additional reference after initial shredding.

When the collection truck arrived the biowaste was unloaded and carefully mixed using an excavator. Then, two sampling units, each 250 kg of biowaste, were taken and sorted manually (Figure S3) by careful examination and separation to identify foreign matter larger than 20 mm (foreign materials < 20 mm are disregarded at this stage). The foreign matter > 20 mm was categorized by visual inspection into seven main categories: 1) foil plastics, 2) no foil plastics, 3) biodegradable plastics, 4) glass, 5) metal, 6) other foreign materials and 7) paper (Figure S4). For simplification, we combined categories 4–6 (glass, metal, and other foreign materials) into one category (Table 1). The first differentiation of plastics was based on physical appearance, i.e., plastics were categorized as foil (as plastic bags) or non-foil (like pieces from plastic boxes). Biodegradable plastics were identified by their green color and labeling that indicated biodegradability (Figure S4).

The foreign material content was calculated for each category after BGK (2021) manual (Table 1, BGK, 2021); plastic types were assessed by Attenuated Total Reflectance Fourier Transform Infrared (ATR-FTIR, Supplementary Materials).

Following the composting facility's practice, we wanted to account for the initial amount of plastics in representative amounts of biowaste, as those are considered major contributor for the creation of MiPs throughout the composting process (Van Wijnen et al., 2019). Additionally, as homogenization of the incoming waste was not possible and hence also no reliable MiP analysis, MiP monitoring started directly after the first homogenization (SP2, after shredding).

2.2.2. Pre-rotting for 11 days (SP2-3)

We collected each 3×10 L of the waste material after shredding and pre-rotting (Fig. 1, SP2) and of the material after pre-rotting and de-compacting (SP3).

2.2.3. Main rotting for 14 days, sieving and curing (SP4-6)

We collected 3x10 L of each of the three different final fractions (>100 mm, 16–100 mm, and < 16 mm) after main rotting (Fig. 1, SP4, 5 and 6).

2.2.4. Irrigation and processing water

Rainwater, collected after draining from the rooftop of the composting facility, was stored in a large concrete tank (Figure S5, A). The roof, as well as all connecting pipes, from the roof to the collection container and from the container to the nozzles in the composting tunnels, consist of plastic.

The excess water that escapes during the decomposition process is drained into a collection shaft through channels located at the bottom of the tunnels. The excess water from the biofilter is also collected in this shaft. From there, the process water is pumped into a storage tank (Figure S5, B and C) made of concrete.

To elucidate if the irrigation water is contaminated during the composting process, we collected each 2x1 L of rainwater (used for the main rotting process), and processed water (more information in Supplementary Materials, Water sample collection).

2.3. Plastic analyses

The plastics > 20 mm collected during the batch analysis were

Table 1
Mass distribution of paper and foreign matter categories, including plastic, extrapolated from 2x250 kg, of four districts (delivered by four trucks) were examined. The last sample is a composite sample of the entire tunnel input of 1x500 kg.

	Foil Plastics	No-foil Plastics	Biodegradable Plastics	Other foreign Materials	Paper	Location
kg plastic ton^{-1} biowaste						
Truck 1	0.36	0.48	0.16	6.88	37.28	District 1
Truck 2	0.24	0.12	0	5.44	n.a.	District 2
Truck 3	0.48	0.44	0.32	3.12	n.a.	District 3
Truck 4	0.94	1.01	0.50	16.02	47.00	District 4
Mean (±SE)	**0.5 ± 0.15**	**0.51 ± 0.18**	**0.25 ± 0.1**	**7.8 ± 2.8**	**42.14 ± 4.86**	
Number of items ton^{-1} waste (mean)	84	50	2	n.a.	n.a.	
kg plastic ton^{-1} biowaste						
Shredded composite	2.1	2.1	0.52	24.44	n.a.	Tunnel entry
Number of items ton^{-1} waste	296	152	6	n.a.	n.a.	Tunnel entry

n.a.* – non analysed

cleaned using water and ethanol, cut into pieces and positioned on adhesive tape for ATR-FTIR analysis. We identified 9 groups of plastics types (Fig. 2A), based on frequency in their appearance: 1) PE, 2) PP, 3) polyamide (PA, mostly nylon), 4) polyester (PES, mostly polyethylene terephthalate (PET)), 5) acrylates, 6) copolymers, used often to improve performance compared to conventional plastics (those that could have been identified and including ethylene methyl methacrylate (EMMA), polyester:nylon 75:25, polyester:elastane 82:18, nylon:elastane 83:17, rayon:pet 80:20, cotton:PEPP, etc., 7) others (including plastics with only a few representatives, such as varnish, PS, PVC or additives), and 8) unidentified (with either lower than 50 % hit quality (HQ) index or too dirty surfaces to confirm the finding from the ATR-FTIR characterization) and 9) biodegradable (more information in Supplementary materials, "ATR-FTIR and Raman Microscopy analysis").

The five fractions of waste material and compost (SP2-6) were dried, sieved according to MeP and MiP size limits (5 mm, 1 mm, and 500 μm, Fig. 2B, Table 2) and analyzed via ATR-FTIR (Figure S6).

For the compost fraction, <16 mm, we conducted wet sieving and comprehensive analysis, crucial as this fraction is marketed as compost (Supplementary Materials, Final compost fraction analysis).

The water samples were also analyzed with μ-Raman (each 1 L was processed and placed on a whole filter with particles > 150 μm and 20 % of the filter for particles 6 – 150 μm), following an extraction protocol with density separation via NaCl (density of 1.2 g cm^{-3}), enzymatic steps (including enzymes cellulase/amylase solution for 3 days at 50 °C, lipase for 1 day at 45 °C and protease for 1 day at 45 °C) and Fenton reagent reaction (Peneva et al. unpublished data).

Table 2
Number of items found per kg of green and biowaste/compost dry weight after visual inspection and extrapolation, divided into size groups of macroplastics (MaPs), mesoplastics (MePs) and microplastics (MiPs). For the final compost fraction"<16 mm" both dry and wet sieving were applied. For the other fractions, only dry sieving was applied. "-" indicates that no plastics within this size range were found.

		MaP/ MeP> 5 mm	Large MiP 1 – 5 mm	Small MiP 0.5 – 1 mm	Total
Pre-rotting	Before pre-rotting (SP2)	2 ± 2	10 ± 5	–	12 ± 5
	After pre-rotting (SP3)	9 ± 10	0 ± 0	–	9 ± 6
After main rotting	> 100 mm (SP4)	7 ± 3	10 ± 4	–	17 ± 3
	16–100 mm (SP5)	8 ± 6	33 ± 38	–	41 ± 18
	< 16 mm, dry (SP6)	26 ± 7	7 ± 4	–	33 ± 13
	< 16 mm, wet (SP6)	2 ± 3	11 ± 2	2 ± 3	15 ± 6

3. Results

3.1. Charge counting – in search for > 20 mm plastics and foreign materials (SP1)

Paper was the most frequently found material, followed by other foreign materials and conventional plastics. The lowest amounts of foreign matter could be attributed to biodegradable plastics (Table 1). Although paper is not considered foreign material according to the

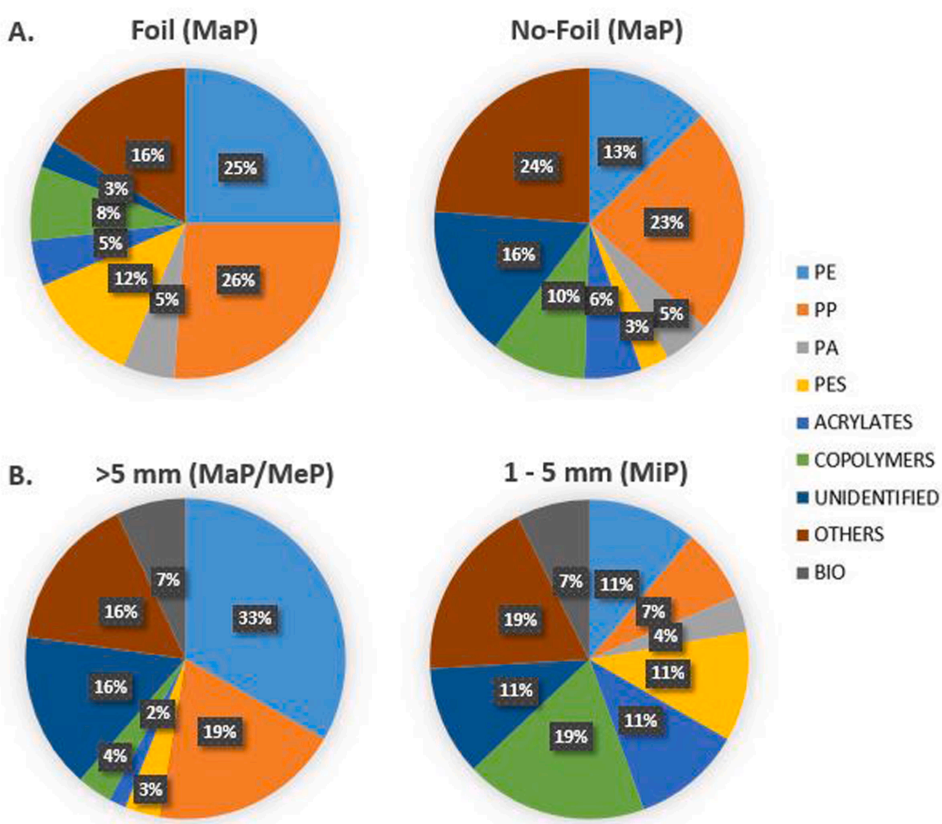

Fig. 2. .Pie chart, representing the composition of the plastics found in the biowaste during charge counting of biowaste from 4 districts (A), charge counting fraction > 20 mm, for both categories "foil" (on the left) and "no foil" and (B) all characterized macro and mesoplastics (MaPs/MePs) and microplastics (MiPs) found throughout the composting process.

"Batch analysis" method (BGK, 2021), we decided to examine it in two batches and found 42.14 ± 4.86 kg paper ton^{-1} biowaste (Table 1).

The plastic content in the "Truck 4″ exceeded that of the other three, underscoring the heterogeneity and varying amounts of foreign materials entering the composting plant, particularly plastic. On average, the plastic foils amounted to 0.5 ± 0.15 kg ton^{-1} biowaste, while "no-foil" plastics contributed to 0.51 ± 0.18 kg ton^{-1} biowaste, indicating a similar presence of foil and no-foil plastics in a 1:1 ratio (Table 1). The lowest plastic content was found for truck 2 (0.36 kg t^{-1} biowaste, equaling 0.04 % of plastic in the biowaste). The tunnel entry carried the largest share of overall foreign materials with 4.2 kg plastic t^{-1} biowaste (sum of foil and no foil plastics), equaling 0.42 % of plastic in the biowaste.

The charge analysis of 2.5 tons of biowaste revealed a total of 500 items of both "foil" and "no-foil" plastics, but only 7 items from plastics were classified as biodegradable. Of those 500 items, 269 were found in the four trucks, with 168 "foil" and 101 "non-foil" items, whereas the rest 224 was found in the shredded composite from the tunnel (with 148 "foil" and 76 "no-foil" items). Approximately half of the plastic items were PE and PP, 122 and 134 items, respectively (Table S1). For "foil"-type plastics, PP constituted 26 %, followed by PE (25 %), others (16 %), and PES (12 %, Fig. 2A), with unidentified plastic items weighing only 3 % of the total plastic. In contrast, for the "no-foil" category, others (24 %) and PP (23 %) had the largest percentage (% of plastic weight), followed by acrylates (16 %), PE (13 %), and copolymers (11 %, Fig. 2A), while PES made up the smallest percentage (3 %).

PP corresponds to almost the same percentage, 26 % and 23 %, in the "foil" and "no-foil" plastic categories, respectively. Interestingly, there were no plastic bottles in the total 2.5 tons checked. Bags and snack food wrappers were some of the most common representatives in the "foil" category of plastics.

In the section copolymers, biodegradable plastics were included, as ATR-FTIR analysis always resulted in a mixture of plastics. These biodegradable plastics were identified either as polyethylene: acrylate copolymer, or polyester. For this plastic category, the hit quality was always under 60 %, which restricted the accurate conclusion of their composition. Noteworthy, all foreign materials had elevated contents in the shredded composite, though it remains uncertain whether this indicated the formation of smaller, more abundant foreign items by the shredding, or simply a remaining sample heterogeneity when taking aliquots for analyses.

3.2. Pre-rotting and main rotting (SP2-6)

In this part, we focused on assessing MaPs, MePs, and MiPs in the green and biowaste, during and after composting (Table 2). The five fractions yielded wet weight between 2.57 and 4.14 kg; the moisture content varied from 15 % to 45 % (Table S2).

The total plastic load increased after the main rotting (SP4-6) and was largest in the fraction 16–100 mm (SP5), followed by < 16 mm (SP6) sold as compost (Table 2). Plastics found in different fractions comprised mainly MaP/MeP (>5 mm) and large MiPs (1–5 mm), while small MiP (0.5–1 mm) were only found at SP6 (Table 2). The largest number of plastics within the category > 5 mm was observed for SP3 and SP6, with 9 ± 10 and 26 ± 7 items kg^{-1} dw, respectively. SP5 showed the largest number of large MiP (33 ± 38 items kg^{-1} dw), i.e., at least about 3 times larger than in any other fraction. Yet, an elevated number of large MiPs kg^{-1} dw was also found in the waste fractions at SP2 and SP4. A slow but steady 3-fold concentration increase of total plastics is visible from the pre-rotting to the final fraction, starting with total of 12 ± 5 items kg^{-1} dw to 33 ± 13 items kg^{-1} dw, respectively (Table 2). The majority of large MiP items was thus also found at the final SP5 and SP6 fractions (Table 2 and Table S3).

The composition of MaPs/MePs and MiPs (identified by ATR-FTIR; Table S4, Fig. 2B, Fig. 3A and 3B) was categorized according to categories of "Batch analysis". Since most plastic items were found in size groups of ">5 mm" and "1–5 mm", only those two were illustrated in Fig. 2B and Table S3. The data showed significant variation in polymer composition between the two main size categories MaP/MeP (>5 mm), and large MiP (1–5 mm). For MaP/MeP, PE emerged as the most abundant polymer type across the five fractions, followed by PP, whereas biodegradable polymers exhibited the lowest abundance (Fig. 2B, Fig. 3A and 3B). Conversely, PE and PP were less frequently detected within MiPs, with "acrylates'' and "others" emerging as the predominant polymer types (Fig. 2B and Fig. 3B). Notably, PA was the least abundant polymer and was nonexistent in size of MaP/MeP. Other items with minor abundance were either not identified (appearing as PS foam, 2 items) or Aluminium oxalate (likely as part of cellophane), whereas the biodegradable plastics comprised poly-2-hydroxyethyl methacrylate at SP2. At SP3, the copolymer was identified as PE-co-vinyl acetate, but with a minute HQ of 29 %.

The composition of the plastics changed at SP4: for the ">100 mm" fraction all four items categorized as PES visually looked like biodegradable MiPs, one plastic was recognized as styrene-allyl, and the rest three plastics as vinyl chloride, as parexyl, and as butyl hydroxide. For the fraction "16–100 mm" (SP5), the unidentified plastic visually looked like plush or plaster and was not identified according to the library spectra. The two biodegradable ones were identified as PES; however, due to the obvious physical alterations we sorted them as biodegradable. The copolymer there was identified as styrene-acrylonitrile resin (SAN).

At all sampling points, in the size of MaP/MeP, PE was the most abundant polymer type representing 25 % of plastics in the initial biowaste and 29 % in the last processing step at SP6 (Tables S1 and S4). Other plastic types like PP, PA, copolymers, and Acrylates had a higher

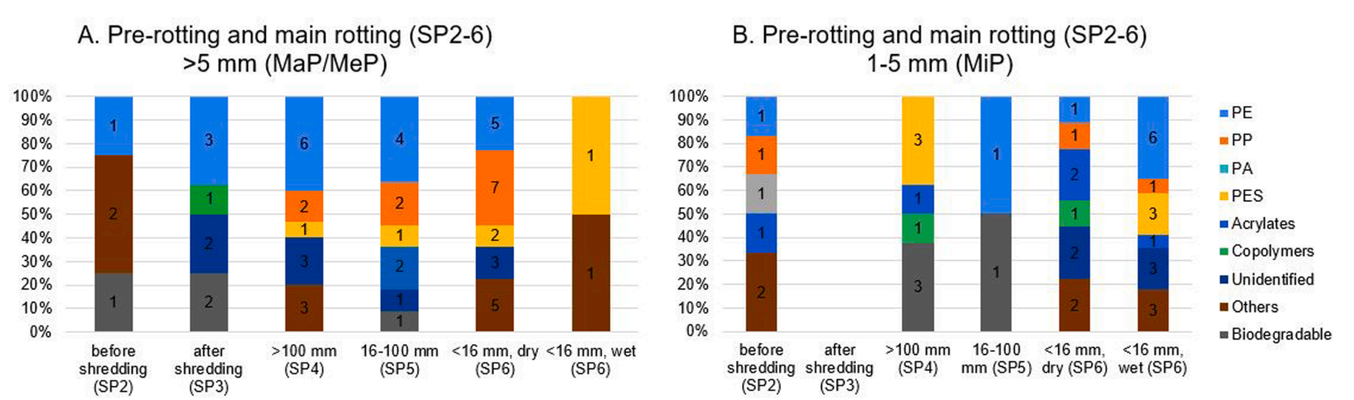

Fig. 3. Stacked bar chart representing the number of plastics (inside of the stacks), their type (as a legend with colors) and the representative percentage (on the y-axis) found in the five fractions of bio and green waste and compost analyzed, for macroplastics/mesoplastics (MaP/MeP), >5mm (A.) and microplastics (MiP, 1–5 mm (B.)). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

percentage at SP1 than in the final analyzed waste fractions. On the other hand, PES, unidentified and biodegradable plastics showed the opposite trend (Table S1 and S4). Noteworthy, none of the factions created by the composting plant for plastic sizes of > 5 mm contained PA (Fig. 2B and Fig. 3A).

3.3. Additional extraction and detection of small MiPs of 20–500 μm and > 500 – 1000 μm from compost fraction < 16 mm

From the 500—1000 μm MiPs, only 3 to 50 % of particles selected under the stereomicroscope could be identified via Raman spectroscopy. Likely due to some remaining organic material, identification of smaller-sized MiPs was difficult, despite strong signals at 1600 cm^{-1}, 2884 to 2904 cm^{-1}, and 3070 cm^{-1} observed for many of the particles, typically associated with plastic spectra. From the total number of 12 identified plastic particles in the 15 g dw of compost, polybutylene adipate terephthalate (PBAT), represented 42 %, followed by PE and PS with 16 % and finally PP, PVC and Nylon with 8 %. Thus, after extrapolation, for the 500–1000 μm MiPs this resulted in 800 ± 529 items kg^{-1} dw compost. For the smaller size-fraction, 20–500 μm, from the 10 on average items selected in 5 g of dw compost under the stereomicroscope, only 1 appeared as biodegradable. However, this particle could not be identified via Raman spectroscopy, preventing any definitive conclusion.

3.4. Water samples analysis

Three main polymer types were identified in high quantities for the rainwater: PE was most abundant with 22,714 ± 2,976 items L^{-1}, followed by PA and PP with 3,108 ± 748 and 686 ± 398 items L^{-1}, respectively (Fig. 4). The same pattern was observed for the process water, though the numbers of plastic items were lower with 13,864 ± 3,866 items L^{-1} for PE; 617 ± 28 items L^{-1} for PA; 497 ± 197 items L^{-1} for PP, respectively (Fig. 4). The size range for those MiPs had been set from 6 to 150 μm, yet, all particles found were in the size of 6–70 μm. Additionally, we found 35 ± 0.4 and 100 ± 0.6 larger MiP items (150 – 600 μm) per L of rain and processed water, respectively.

4. Discussion

4.1. Charge counting – searching for > 20 mm plastics and foreign materials (SP1)

Paper dominated in the biowaste inputs, likely reflecting that paper, being compostable, is organic and considered biodegradable. For hygienic reasons, biowaste in Germany is frequently pre-collected in small paper bags or wrapped in paper before being placed in the organic waste bin for collection. Also paper absorbs much more moisture from biowaste than plastic and, therefore, has a higher wet weight. After composting, and unlike plastics, the paper was largely degraded.

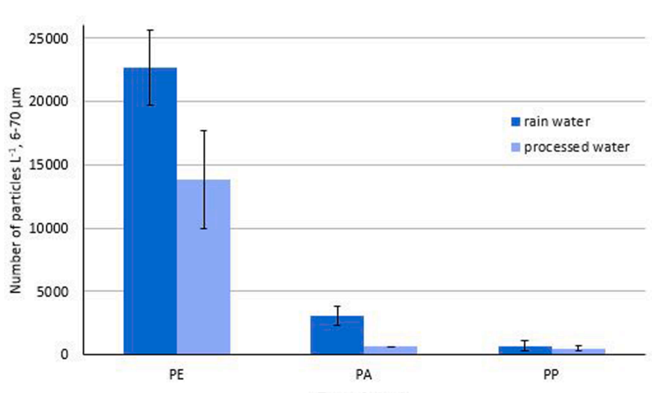

Fig. 4. Number of small microplastics (6 to 70 μm) of polyethylene (PE), polyamide (PA) and polypropylene (PP) found in rainwater (used for irrigation) and processed water (after irrigation). Data is presented as means ± standard deviation.

The biowaste input typically maintained a 1:1 gravimetric ratio of "foil" to "no foil" plastic contaminations, except once where foil dominated. Also Colombini et al. (2022) reported that plastic films constituted up to 80 % of compost contaminations. In line with Van den Zee and Molenveld (2020), half of the plastics can be categorized as "flexible packaging." Despite equal weight distribution in our study, the category "foil" comprised 316 items, while "no-foil" had 177 items in the 2.5 tons, respectively. This discrepancy underscores the importance of considering both weight and quantity metrics when analyzing plastic abundance, as weight-based assessments ignore particle size information and hence may not accurately capture material distribution. The higher item numbers in the "foil" category are attributed to the intrinsic weight differences between foil and non-foil materials, with foil materials being lighter. Our data also suggest that the easier fragmentation of "foil" structures contributes to their larger item numbers compared to "no foil" pieces.

The percentage of PE in the "foil"-type was nearly two times larger than in the "no-foil"-type category. That aligns with the common use of PE in plastic bags and wrappings (López and Serna, 2022). For instance, all collection trucks had paper/plastic bags frequently used in bakeries and baking sections in big supermarkets. Given that this packaging is primarily composed of paper, with only a small portion being plastic, it may often be mistakenly disposed of in biowaste. When a piece consisted of a mixture of different materials, mostly bags from bakery shops where paper and plastic were abundant, we carefully separated the respective components. In those cases, the plastic parts of pieces were categorized as part of the foil group due to their foil-like form. The paper from these bags was not included in the paper group during charge counting, which only accounted for fully paper-based items like boxes made entirely of paper.

Polypropylene, in turn, is equally found in both "foil" and "no foil" plastics, with double amount in "no-foil" in comparison with PE, likely reflecting that PP is commonly used in food packaging, such as boxes for food deliveries. The same trend was observed by Van den Zee and Molenveld (2020), with PET being the second most commonly found rigid plastic type. Additionally, PP is a common component of fruit stickers, another significant plastic contaminant in compost (EPA, 2022; Groß et al., 2024). Besides, we found meat packages still unopened, and, occasionally, also small toys, rubber gloves and plant pots.

PE was the most frequently detected plastic type in compost reported by Edo et al. (2022), Colombini et al. (2022) and Van den Zee and Molenveld (2020), accounting for 35 %, 71 % and 29 % of all plastic items respectively. Interestingly, both Edo et al. (2022) and Colombini et al. (2022) found PS as the second most abundant plastic type, whereas here PS was rather rarely detected (3 items per ton), and, thus, included in the "others" category. Additionally, we found many other plastic items lacking clear identification likely due to a layer of grease on their surface, which could not be completely removed even after meticulous sample cleaning. Also, Van den Zee and Molenveld (2020) could not identify 17 % of the plastic items due to their altered surfaces when investigating plastic loads in a composting facility in the Netherlands. Van den Zee and colleagues reported 21 % of biodegradable bags in green and biowaste samples, in contrary to our results, with less than 1 % of the plastic items being biodegradable bags.

According to the Biowaste Ordinance, cities and districts have the option of restricting the list of authorized input materials in their waste bylaws. In many nationwide statutes, as in the statutes for this composting plant, the use of biodegradable plastic collection bags is expressly prohibited. This is presumably why relatively few such bags were found in the organic waste. A German team of researchers, who initiated in 2019 a project called BabbA (Biologisch abbaubare Beutel in der Bioabfallverwertung) found that composts contain significant amounts of biodegradable MiPs smaller than 1 mm, which can persist in the soil over extended periods (Forberger et al., 2023). Their final report highlights paper bags as a promising alternative to biodegradable bags and calls for a critical revision of current standards for compostable materials. Other research groups summarized that it can take up to 18 weeks until 85–99 % of biodegradable plastics are fully degraded (Le et al., 2023). However, this depends, besides on the composting conditions, strongly on the type of biodegradable polymers used to create the blend, i.e., polylactic acid-poly(hydroxyalkanoate) PLA-PHA based biodegradables can be > 95 % degraded in just 4 weeks (Sintim et al., 2020). Noteworthy, the remaining parts from biodegradable bags were very easily broken once touched and tried to be placed under the ATR-FTIR (Van den Zee and Molenveld, 2020).

Our study neither aimed at reporting typical variations in plastic inputs between different composting plants nor accurately assessing in-situ variations. Instead, we aimed to report realistic contaminations by sampling large volumes. We found 272 plastic items (comprising foil, no foil, and biodegradable plastics) within 2 tons of biowaste, corresponding to 136 items per t^{-1} of biowaste. Some larger plastic items were found after the sieving, shredding, and rotting stages (SP2-5), underscoring difficulties in their effective removal throughout the process and/or their transformation into smaller sizes during shredding. That highlights the need to reduce plastic inputs by optimizing top-down policymaking.

4.2. Plastic loads (> 500 μm) throughout the composting process and in final compost (SP2-6)

The threefold increase of cumulative MaPs/MePs and MiPs from the initial stage throughout the composting process supports the hypothesis that plastics enrich with weight loss of other organics during composting. Furthermore, large plastic items likely underwent shredding (SP2), facilitating fragmentation and hence the formation of secondary MiPs (Gui et al., 2021; Sholokhova et al., 2023). Reported numbers of large MiPs in the end-product are lower than reported by Weithman et al. (2018) and Braun et al. (2020). The authors found 24 items kg^{-1} dw and an average of 21 ± 31 items kg^{-1} dw, respectively. Other studies found 310 to 410 items kg^{-1} dw to 2,533 ± 457 items kg^{-1} dw, with sizes of 1–5 mm and 0.05–5 mm, respectively (Premasiri, 2021; Gui et al., 2021), thus underscoring the complexity and variability in MiP distribution within composting facilities. In contrast, reported MeP/MaP numbers were higher than the numbers of MaPs routinely collected by the facility, with mean values of 2–4 items kg^{-1} (unpublished data of the composting facility), which were confirmed by an earlier study (Braun et al., 2021). Varying results point to a high heterogeneity of plastic loads within the compost, letting us assume that the found 26 ± 7 items reflect rather maximal loads.

Here, we focused on quantifying plastics and MaPs loads from the incoming organic waste, examining 2 tons across four trucks and a shredded composite from the tunnel, however, this represents only 1.1 % of the total 180 tons in the tunnel. Additionally, we observed a high heterogeneity, especially for large plastics. Different numbers may also be due to differences in the methodologies used: when using wet sieving for the final biowaste fraction, we found about 50 % more plastic items in the < 16 mm fraction.

However, for obtaining representative measurements an adequate sampling strategy is key. For future studies, increasing replicates (including the number of subsamples used for one composite sample), also from different seasons across the year, could reduce the uncertainties. Particularly since plastic contamination is likely higher during the colder months compared to spring and summer when unpackaged fruits and vegetables are more readily available and consumed in larger quantities (Adam et al., 2024). Although there are similarities between studies from different countries, such as the prevalence of foil-like plastics and the dominance of PE (Colombini et al., 2022, Edo et al., 2022), national regulations vary, which can add to differences in findings among different countries or even continents.

A key challenge remaining is the lack of a standardized methodology for MiP extraction from complex organic matrices such as compost, digests, and food waste (Porterfield et al., 2023). We are confident that the detected MiP load was not affected by sampling or sample processing, because strict contamination precautions were taken, using, e.g., only non-plastic materials, and because the processed lab blanks did not point at significant MiP loads.

Overall the composition of the plastic types found was diverse, with PE dominating MaPs/MePs (Fig. 2B and Fig. 3A), while no specific plastic type dominated within MiPs.

On the other hand, PS, for example, was hardly detected and hence categorized under "others" in the fresh biowaste (SP1). However, after pre- and main-rotting, we identified a total of 9 PS items within the total 11.9 kg dw (SP2-6, Table S2), with 0.3 items kg^{-1} and 0.4 items kg^{-1} falling within the size range of MaP/MeP and MiP, respectively, pointing to a slight increase throughout the composting process. Notably, expanded PS has a very low density (0.01–0.05 g cm^{-3}) and a large potential to generate numerous MiPs (British Plastic Federation). In this case, the paradox between particle number and mass is evident, as PS can produce a significant number of MiPs with minimal weight. Gui et al. (2021) highlight the rapid fragmentation of expanded PS, generating 53 MiP particles from a single item within 30 days of composting, surpassing the formation rates of PE and PP, which only produce 9 and 5 MiP items, respectively. As we observed a low abundance of PS in our experiment, we can relate this to the changing trends in plastic usage. With regulations like the EU's ban on single-use plastics (DIRECTIVE (EU) 2019/904), the decrease in single-use PS, commonly used in food and beverage containers, may explain the observed trend. We believe that this shift in consumer behavior aligns with the ongoing efforts to reduce plastic pollution.

Overall, the total count of MaPs/MePs and MiPs in the final fractions "16–100 mm" (SP5) and "<16 mm" (SP6) surpasses that in the fractions taken after pre-rotting (SP 2–4) by two to three times, which may be attributed to the steps of shredding and composting (Gui et al.,2021; Sholokhova et al., 2023).

In principle, there might be an additional source of MiP: atmospheric deposition (Kernchen et al., 2022; O'Brien et al., 2023). To account for such processes, we analyzed MiP loads in rainwater from the rooftop. It likely contained both atmospheric deposition from rather long-distance transport, as well as from the blown-out material of the composting facility itself.

4.3. Small MiPs (500–1000 μm) in final compost and water samples

In general, the selection of small MiPs under a microscope, a widely employed method for plastic particle recognition, can introduce a potential source of error (Mariano et al., 2021), especially when organic matter is still present in the sample. This was evidenced by the relatively low number of identified items and by a notable occurrence of false positives as only between 3 and 50 % of selected items could be identified as plastic. Also, extracting MiPs from high-organic matter content soils or compost is challenging, with varying success reported in the literature (Hurley et al., 2018; Braun et al., 2023). To address challenges in extracting MiPs smaller than 1 mm from organic matrices like compost, thermoanalytical methods, such as TED-GC-MS, can support the analysis, providing mass-based concentration for single polymer types (Wiesner et al., 2023).

In the final compost, we found 800 ± 529 items kg^{-1} dw compost, of small MiPs (500–1000 μm). In comparison, Van Schothorst et al. (2021) analyzed MiPs in the size range of 0.03 to 2 mm by applying 30 mL of distilled water to 5 g of compost, followed by centrifugation. This method allowed them to identify 1,253 ± 561 items per kg of compost made of green cuttings, using a microscope and a hot plate at 130 °C. Further, Edo et al. (2021) employed a two-step protocol and identified 1,357 out of 1,532 particles as synthetic polymers, with the most common sizes ranging from 150 to 700 μm. In contrast, Steiner et al. (2022) utilized a seven-step enzymatic-oxidative digestion protocol for MiPs (10–1000 μm) in liquid fertilizer, ending with density separation using $ZnCl_2$ and subsequent dilution in filtered water to avoid matrix interference during FTIR analysis. They found between 6,000 and 12,000 particles L^{-1} of liquid fertilizer. The high variation in MiP loads reported by different studies might result from differences in methodology, including the number of steps, reagents used, and particle size ranges analyzed. However, the results might also point to the high heterogeneity of MiP loads within the compost, as already observed by other studies (Braun et al., 2021).

Surprisingly, we discovered MiP items in both process water and rainwater. The high numbers in the rainwater suggest significant contamination by atmospheric deposition (Kernchen et al., 2022), likely due to wind-driven distribution of MiPs, especially as rainwater is stored under the rooftop, even if the containers are closed (Wei et al., 2023). This rooftop consists of a plastic foil roof, and all the pipes connecting the rooftop to the collection container and from there to the nozzles used for compost irrigation are made of plastic, despite the container itself being non-plastic (See Supplementary Materials, Water samples Collection). Hence, any abrasion process from the roof, if there were any, might have contributed to the contamination of rainwater if blown around with the wind. Moreover, in the process water, the MiPs could originate from the compost itself. The big floods in Germany, which may also have transported plastics around, even into the plant, had passed since 9 months. Another input source for the rainwater could be the nearby motorway.

In addition to these potential contamination sources, it is important to consider the possibility of false positives arising from Raman measurements and a potential overestimation of very small-sized MiPs, common in spectroscopic techniques, that could explain the observed high numbers, especially for particles below 10 μm (Xu et al., 2019).

As the process water after being used to irrigate the compost, contains significantly fewer MiPs than the rainwater, we assume that a large part of these MiPs are filtered by the compost. Hence, we must refute our initial hypothesis, that the irrigation water is contaminated by MiPs during composting. Instead, we conclude that the irrigation water is an additional source for small MiPs during composting, which has been overlooked so far.

5. Future implications

Market acceptance of compost and digestion products hinges on users' expectations that these products are free, or nearly free, of foreign substances, particularly plastics, to ensure high-quality organic fertilization. In the future, it remains crucial to develop strategies for reducing the plastic load in compost. This could include more strict regulations, such as limiting plastic contamination in composts. The EU, for instance, permits up to 0.3 % dry weight impurity for plastic, metal, and glass (larger than 2 mm), whereas Germany enforces stricter standards (0.1 % dry weight for particles larger than 1 mm).

However, to reduce the plastic load of compost, the key strategy must be to minimize the plastic input via waste collection into composting facilities, i.e., preventing improper waste disposal in organic bins, and not to leave the burden of purifying the materials to the plant. The contamination of household organic waste, particularly with plastics, has increased in recent years (BGK, 2024). The German Federal Quality Association for Compost (BGK) attributes this to declining public awareness regarding separate waste collection systems and inadequate identification and elimination of foreign material sources. The problem is that during waste collection, single collected bins can pollute a whole batch of waste. To prevent existing plastic contaminations some composting facilities already use a system to recognize foreign matter during the collection of biowaste. Contaminated garbage cans can so directly be excluded. In addition to these measures, ensuring correct use and disposal of biodegradable plastic is crutial. Biodergadable plastic, used as a bin liner for collection of biowaste is becoming more and more popular, although their complete degradation during composting is still under discussion (Flury et al., 2021). An approach to better account for incomplete degradation under various disposal conditions, is the calibration of the actual biodegradability in standards. Further, the provision of biodegradability certificates on biodergadable plastics with clear disposal instructions is recommended to ensure proper end-of-life management (Yu et al., 2024). Encouraging the collection of source-separated green and biowaste, combined with public education and better marketing, can further enhance the purity of compost (Fricke et al., 2017). Expanding the organic waste bin system, as seen in Germany since 2015, could also be a helpful measure (Friege and Eger, 2022). Public education via social media and awareness campaigns as well as regular compliance checks are essential to reduce the plastic contamination of compost (BGK, 2024; Habermann, 2022).

Although our study confirmed that final compost contains plastics, even when applying high MeP/MaP contents, resulting in a total mean amount of 833 items kg^{-1}, this plastic content is substantially lower than plastic values of sludge where 1,000 to 24,000 items kg^{-1} are found (Bläsing and Amelung, 2018). Considering now compost application rates, which vary widely from 7 to 35 t/ha annually (BioAbfV, 2013, WRAP, 2015), the plastic contents found in this study (mean: 833 items kg^{-1}), a plowing depth of 30 cm and a soil bulk density of 1.4, a one-time compost application of 7 and 35 t would result in soil plastic contents of 1.39 to 27.77 plastic items per kg soil, respectively. Twenty years of annual compost application would result in soil plastic contents of 27.8 to 139 plastic items per kg soil, which are considerably lower than reported for soils under plastic mulching or soils under sludge application, most of them having concentrations larger than 1000 items per kg (van den Berg et al., 2020, Büks and Kaupenjohann, 2020, Harms et al., 2021, van Schotthorst et al., 2021, Wrigley and Braun et al., 2024).

6. Conclusions

Here, we investigated the presence and composition of MaPs, MePs and MiPs in fresh biowaste and at different stages throughout the composting process within a German composting facility, following national and state regulations. The analysis included biowaste batch analysis, pre- and main rotting assessments, and collection of irrigating water – before and after composting. The results revealed varying MiP loads in different compost fractions, with PE and PP standing out as the most widespread plastics. We found with up to 1.95 kg per ton large amounts of MaPs in biowaste, highlighting the role of improper waste separation as an entry route for plastics in composting facilities. During the composting process, total plastics became enriched by a factor of three, which might be caused by the fragmentation of large plastic items into smaller ones. Moreover, we could identify irrigation water as an additional source for especially small MiPs. The latter presents, thus, a still under-examined source of a plastic cycle within the industrial compost plant.

CRediT authorship contribution statement

Stoyana Peneva: Writing – original draft, Validation, Methodology, Formal analysis, Data curation. **Quynh Nhu Phan Le:** Writing – review & editing, Formal analysis. **Davi R. Munhoz:** Writing – review & editing, Data curation. **Olivia Wrigley:** Writing – review & editing, Formal analysis, Data curation. **Giovana P.F. Macan:** Writing – review & editing, Data curation. **Heidi Doose:** Writing – review & editing. **Wulf Amelung:** Writing – review & editing, Supervision, Conceptualization. **Melanie Braun:** Writing – review & editing, Supervision, Data curation, 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.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.wasman.2024.11.043.

Data availability

Data will be made available on request.

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