



# Microplastic Contamination of Composts and Liquid Fertilizers from Municipal Biowaste Treatment Plants: Effects of the Operating Conditions

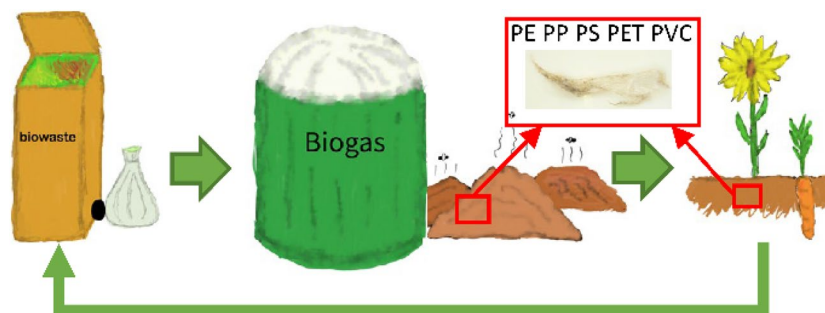
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## Abstract

High-caloric-value household biowaste is an attractive substrate for the production of biogas and fertilizer. Most household biowaste is contaminated by plastics, typically in the form of bags and foils from packaging. Operators of municipal biowaste treatment plants take great care to remove these contaminants, often at the cost of reducing the organic material entering the process. This study compares the residual plastic contamination of fertilizer (composts, digestates) from biowaste treatment plants with compost produced from greenery and digestates produced by agricultural biogas plants processing manure and energy crops. While the fertilizers from the agricultural biogas plants and greenery composts were minimally contaminated by plastic, we found considerable numbers of plastic fragments in the composts/fertilizers from the biowaste treatment plants. Moreover, while certainly being influenced by the quality of the incoming biowaste, this residual contamination appeared to depend largely on the operating conditions. In particular, shredding of the incoming material increased the degree of contamination. Sieving was an efficient method for the removal of fragments > 5 mm but was less efficient for the removal of smaller fragments. In view of the number of the recovered fragments in that size range, it is likely that still none of the finished composts surpassed the current dry weight limits imposed for the plastic contamination of high-quality composts with fragments > 1 mm in Europe (0.3% of dry weight) nor even in Germany (0.1% of dry weight). The contamination of the liquid fertilizer produced via anaerobic digestion by three of the investigated biowaste treatment plants (up to 10,000 particles with a size between 10 and 1000  $\mu\text{m}^{-1}$ ) may pose a more serious concern.

## Graphical Abstract



**Keywords** Biogas · Biowaste · Compost · Fertilizer · Microplastic · Nanoplastic

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## Statement of Novelty

The topic of our manuscript is the residual contamination by (micro-)plastic fragments of quality composts and liquid fertilizer produced by state-of-the-art biowaste treatment plants.

Composts from such plants have been suspected to release significant numbers of plastic fragments into the environment, yet this has never been systematically studied and correlated with plant operation and process conditions. Concomitantly, high-caloric household biowaste is an attractive substrate for biogas and fertilizer. Most household biowaste is contaminated by plastics, typically in the form of bags and foils from packaging. Operators of municipal biowaste treatment plants take great care to remove these contaminants, often at the cost of reducing the organic material entering the process. This study compares the residual plastic contamination of fertilizer (composts, digestates) from biowaste treatment plants with compost produced from greenery and digestates produced by agricultural biogas plants processing manure and energy crops. Our results can contribute to the ongoing discussion of the significance of plastic in the environment and are expected to be of interest to readers from a wider range of communities.

## Introduction

The increasing contamination of the environment by macro- and microplastics, the latter defined as particles  $< 5$  mm, has been the subject of intensive study in recent years [1–3]. Most studies to date have focused on aquatic systems, but the contamination of terrestrial compartments may also be significant [4]. Possible entry pathways for plastics into terrestrial systems are thus of interest and also have implications in the development of future containment strategies. Various entry pathways have been identified, including illegal waste deposits, common agricultural practices such as the use of foils for mulching, the transfer of airborne particles (e.g., rubber particles from tires) to the ground by natural precipitation (snow, rain) [5–7] and the utilization of organic fertilizer [8].

Organic fertilizer is produced at a large scale in technical biogas and composting plants from various organic substrates, including household biowaste. These plants play an important role in recycling organic material (biomass). Plants that deal with high-caloric household biowastes tend to process incoming material with a two-step combination of (1) anaerobic fermentation (biogas production) followed by (2) composting of the solid digestate. The production of biogas, which can be transformed into electricity and heat, improves the economic balance of such waste treatment plants [9]. In addition, biogas represents a possible contribution to the ongoing transition from fossil fuels to renewable energies.

Quality composts, including those from biowaste treatment plants, are strictly regulated in regard to allowable residual contaminants, including plastics. For the European Union regulation is found in EU document 2019/1009 [10]

specifying limits for metals, glass and plastic in composts and digestates from biowaste (0.3% dry weight for each impurity type with a particles size  $> 2$  mm, 0.5% dry weight for the total sum of these impurities). Regulations for Germany are somewhat stricter (limit of 0.1% dry weight for particles  $> 1$  mm) and can be found in the DüMV (Düngemittelverordnung). For details see, e.g., § 3, 4b, DüMV and § 3, 4c, DüMV. A positive identification of the chemical nature of recovered plastic particles is typically not required, even though such identification, e.g., by IR spectroscopy, is possible [11].

Collected household biowaste nearly always contains plastics, mainly bags and foils, but increasingly also coffee and tea capsules. Consequently, stringent removal steps, typically involving sieving, are implemented to reduce such contamination, a practice that incidentally also reduces the amount of organic material entering the digester/composter. Despite these measures, even quality biowaste composts may still contain a significant number of microplastic (MP) fragments, as recently shown by Weithmann et al. [8]. However, in their paper, Weithman et al. included only one biowaste composting plant and one biowaste digesting plant and thus could not establish general conclusions. As far as we could ascertain, no one has to date studied whether MP contamination of fertilizers from biowaste treatment plants is typical and to what extent the operating conditions contribute or not to the final contamination. Moreover, two-stage digester–composter plants often produce liquid fertilizer (LF) in addition to compost. This fertilizer is directly applied to the soil and has, to the best of our knowledge, never been studied regarding possible contamination by MP.

Given the acknowledged need to recycle organic waste in a suitable manner together with the relevance of organic fertilizer as an attractive substitute for artificial fertilizer, the present study systematically studies operational conditions and their influence on the production of MP fragments during biowaste treatment. To the best of our knowledge, this is the first study of its kind.

## Materials and Methods

### Materials

If not otherwise indicated, the suppliers for chemicals were Th. Geyer (Renningen, Germany) and SigmaAldrich (Taufkirchen, Germany). Ultrapure water was produced with an Elga-Veolia-Purelab (Flex2) unit, while ‘Millipore water’ came from a Millipore-Synergy-UV-system (Type 1). Protease A-01 (activity:  $> 1100$  U mL<sup>-1</sup>), Pektinase L-40 (exo-PGA activity:  $> 900$  U mL<sup>-1</sup>, endo-PGA activity,  $> 3000$  U mL<sup>-1</sup>, pectinesterase activity:  $> 300$  U mL<sup>-1</sup>), and Cellulase TXL (activity:  $> 30$  U mL<sup>-1</sup>) were from ASA

Spezialenzyme GmbH (Wolfenbüttel, Germany). Viscozyme L (activity: > 100 FBG U g<sup>-1</sup>) was obtained from Novozymes A/S (Bagvaerd, Denmark).

### Selection of Biowaste Treatment Plants and Sample Denomination

Plants representing three basic types were included in this study, namely, simple composting plants (aerobic treatment, six plants), simple anaerobic digesters (“biogas plants”, three plants) and two-stage plants comprising (1) anaerobic digestion and (2) aerobic composting (five plants). The selection included plants treating biowaste, green cuttings and/or energy crops; for details, see Table 1. Plants in category 1 included both plants that convert household biowaste and some that convert other organic materials, including greenery. Plants in category 2 were all agricultural biogas plants, converting mainly agricultural waste (manure) and/or energy crops. Household biowaste was not used. However, one of these plants, plant #2.1, processed organic waste from local markets and landscape conservation material alongside the typical mix of agricultural waste and energy crops. The plants from category 3 were all current state-of-the-art biowaste treatment plants, processing high-caloric household biowaste and recovering part of the energy in the form of biogas via anaerobic digestion during the first.

treatment stage. Three of these plants separated the digestate obtained during the fermentation step by press filtration into a solid digestate going into the composting stage and into LF intended for direct application to agricultural soil. Most of the category 3 plants added a certain percentage of green cuttings to their solid digestate prior to composting; for details, see Table 1. Plants processing biowaste are indicated by bold print in Table 1.

Depending on the plant type, samples were taken of the precomposts (before final sieving), the finished composts (after final sieving), and the solid and liquid fertilizers obtained after anaerobic digestion. Samples are coded according to plant number and source (P, F, and L for precompost, finished compost, and liquid fertilizer, respectively). In the case of the agricultural biogas plants, the indicator “S” is used for the solid digestate. When several samples were taken from a given plant/source, each sample is indicated by a number following the plant number. Sample P\_3.3-2 would thus correspond to the second sample of precompost taken from plant #3.3.

### Sampling of Composts and Liquid Fertilizers

Bulk samples were taken from the composts according to the guidelines of the German Association for Quality Compost [12]. A slight modification to the standard procedure was introduced to avoid additional contamination of the compost

samples with plastics, particularly via the plastic foil recommended in the standard protocol for sample mixing. Instead, the individual aliquots obtained from a given compost heap were pooled, mixed and stratified directly on the concrete floor (after a ‘washing’ step with compost from the same heap). To obtain a representative sample, the interior of the heap was made accessible using a wheel loader. Then, individual samples were taken at evenly dispersed points. The number and volume of individual samples depended on the volume and grain size of the compost pile; the current procedural guidelines were followed. For example, in the case of 100 m<sup>3</sup> of compost with grain sizes of 2–20 mm (typical finished compost), 16 individual samples (1 L each) were taken, and a minimum of 4 mixed samples (2 L each) were created. For the coarser precomposts, the number of samples taken was identical to the number of samples taken for the corresponding finished composts.

In most cases, the precomposts and finished composts for a given plant were sampled at the same visit and consequently stemmed from different processing batches. In one case (plant 3.3), one processing batch was sampled before (precompost) and again several days after (finished compost) the final sieving step. In all cases, sample aliquots were transferred to 3 L Fido jars (Bormioli Rocco, Fidenza, Italy) for transport. If immediate analysis was not possible, samples were stored at 4 °C in the glass vessels. Solid digestates from the agricultural biogas plants were sampled analogously to the compost. Solid digestates from the biowaste digester composters were not accessible to sampling for technical reasons. Samples of the liquid digestates (liquid fertilizer, LF) (~6 L) were collected from the outlet of the storage tanks into glass vessels. The first few liters of LF were discarded to rinse the outlet pipe before the sample was taken. If necessary, LF samples were also stored at 4 °C. Backup samples of approximately 1 L were taken for all samples and stored at –20 °C. Glass vessels intended for transport, for storage or for backup samples were washed in advance with Millipore water.

### Analysis of Plastic Fragments in Solid Digestates and composts

A significant concern during the analysis of MP in environmental samples is possible contamination with MP from the ambient air, clothing, laboratory tools, or reagents used during sample processing. To avoid any such contamination, precautionary measures were taken. Cotton lab coats were worn throughout. Unless direct handling was necessary, samples were covered with a glass or aluminum foil lid. Sample processing took place in a laminar-flow box to prevent airborne particles from falling into the samples. All laboratory tools used were made of glass, metal or polytetrafluorethylene (PTFE), a polymer that is rarely found in

**Table 1** Technical details of the investigated plants (plants handling biowaste are indicated by bold print)

|                       | Plant 1.1_I                        | Plant 1.1_II   | Plant 1.2   | Plant 1.3   | Plant 1.4  | Plant 1.5  | Plant 1.6   | Plant 2.1  | Plant 2.2                        | Plant 2.3  | Plant 3.1  | Plant 3.2                                   | Plant 3.3   | Plant 3.4   | Plant 3.5  |
|-----------------------|------------------------------------|--|---|---|--|--|---|--|----------------------------------|--|--|---|---|---|--|
| Substrate preparation | Cuttings                           | <b>Biowaste</b>  | Cuttings  | Cuttings: 25,000 t year <sup>-1</sup>                 | <b>Biowaste, 32,000 t year<sup>-1</sup>, cuttings 3000 t year<sup>-1</sup></b> | Cuttings: 9500 t year <sup>-1</sup>                | <b>Biowaste 60,000 t year<sup>-1</sup></b>  | Agricultural waste; energy crops; grapes, organic waste from local markets; landscape conservation material; 18,000 t year <sup>-1</sup> | Agricultural waste; energy crops | Agricultural waste; energy crops; 15,000 t year <sup>-1</sup>          | <b>Biowaste (88%) and cuttings (12%); 44,000 t year<sup>-1</sup></b> | <b>Biowaste; 18,000 t year<sup>-1</sup></b> | <b>Biowaste with a small fraction of added cuttings</b> | <b>Biowaste (75%) and cuttings (25%); 35,000 t year<sup>-1</sup></b>  | <b>Biowaste (85%) and cuttings (15%); 20,000 t year<sup>-1</sup></b> |
| Substrate preparation | Shredder, magnetic separator       | <b>Bag slicer, sieving (80 mm); magnetic separator; mixing with shredded cuttings; sieving (80 mm)</b> | Shredder  | Shredder  | <b>Biowaste sieving (80 mm); mixing with shredded cuttings</b>                 | Shredder   | <b>Bag slicer, sieving; mixing with up to 5% shredded cuttings, sieving (80 mm)</b> | Shredding of solid material; mixing with liquid substrate  | No data available                | Shredding of solid material; mixing with liquid manure                 | <b>Shredder, sieving (80 mm)</b>                                     | None  | <b>Shredder</b>   | <b>Biowaste: cross-flow shredder, sieving (60 mm), cuttings: sieving &gt; 80 mm, shredder, sieving, fraction is used for fermentation and &lt; 15 mm for composting</b> | <b>Shredder; sieving (100 mm)</b>                                    |
| Fermentation          | -                                  | -  | -   | -   | -  | -  | -   | Main fermenter: 50 days, 53 °C<br>Secondary fermenter: 25 days, 54 °C;   | No data available                | Main fermenter: 150–160 days<br>Secondary fermenter: no data available | <b>Plug flow; 55 °C, ca. 17 days</b>                                 | <b>Box fermenter, 40 °C, ca. 40 days</b>    | <b>Plug flow, 55 °C, ca. 21 days</b>                    | <b>Plug flow, ca. 21 days<sup>a</sup></b>   | <b>Plug flow (20 days, 52–55 °C)</b>                                 |
| Composting            | 10–12 month movement every 2 weeks | <b>12 weeks; movement every 2 weeks</b>  | 3–4 months, plate-like stacks, movement every 4 weeks | 3–4 months, plate-like stacks, movement every 4 weeks | <b>1 week intensive rotting; weekly movement (10 weeks)</b>                    | 4 weeks covered floor aeration; 2 weeks maturation | <b>9 movements every 5 days, then drying</b>  | -  | -                                | -  | <b>Up to 5 weeks</b>   | <b>&gt; 5.5 weeks</b>                       | <b>Up to 9 weeks</b>                                    | <b>6 weeks</b>  | <b>5 weeks</b>   |
| Product preparation   | -                                  | -  | -   | -   | -  | -  | -   | Solid-liquid separation  | No data available                | Solid-liquid separation  | <b>Solid-liquid separation</b>                                       | -   | <b>Solid-liquid separation</b>                          | -   | <b>Solid-liquid separation</b>                                       |

**Table 1** (continued)

|               | Plant 1.1_I | Plant 1.1_II | Plant 1.2 | Plant 1.3                           | Plant 1.4                           | Plant 1.5                         | Plant 1.6                           | Plant 2.1                          | Plant 2.2                          | Plant 2.3                          | Plant 3.1                                | Plant 3.2    | Plant 3.3                                | Plant 3.4    | Plant 3.5                                |
|---------------|-------------|--------------|-----------|-------------------------------------|-------------------------------------|-----------------------------------|-------------------------------------|------------------------------------|------------------------------------|------------------------------------|--|--------------|--|--------------|--|
| Final sieving | 12 mm       | 10–15 mm     | 10 mm     | 15 mm                               | 12 mm                               | 20 mm                             | 25 mm, 12 mm and 10 mm              | -                                  | -                                  | -                                  | 12 mm                                    | 20 mm        | 10 or 12 mm                              | 12 mm        | 10 mm                                    |
| Products      | Compost     | Compost      | Compost   | 15,000 t year <sup>-1</sup> compost | 12,000 t year <sup>-1</sup> compost | 3000 t year <sup>-1</sup> compost | 14,000 t year <sup>-1</sup> compost | Solid digestate, liquid fertilizer | Solid digestate, liquid fertilizer | Solid digestate, liquid fertilizer | Compost/liquid fertilizer <sup>a,b</sup> | Only compost | Compost/liquid fertilizer <sup>a,b</sup> | Only compost | Compost/liquid fertilizer <sup>a,b</sup> |

<sup>a</sup>One part of the digestate was dried and composted, and another part was mixed with fresh substrate and returned to the fermenter

<sup>b</sup>Part of the liquid fraction was returned to the fermenter to mash the substrate; separation was performed with dewatering press

environmental samples and was excluded here from the analysis. All required solutions and the deionized water used to prepare them were filtered through 0.2 μm pore membranes (mixed cellulose ester membrane, diameter 47 mm, Whatman ME 24, Merck KGaA) before use. Enzyme solutions were filtered through 0.45 μm pore membranes (regenerated cellulose membrane, diameter 100 mm, Whatman RC 55, Merck KGaA) and stored in glass bottles with glass caps before use. All laboratory equipment was thoroughly rinsed with filtered deionized water, 35% ethanol, and again with filtered water before use and between steps to avoid cross contamination. Blanks subjected to the same treatment as the environmental samples were used to detect possible contamination in the laboratory.

In preparation for analysis, the digestate or compost samples were filled into a rectangular metal form (790 mm × 510 mm × 150), thoroughly mixed with a metal shovel and quartered. Sample aliquots for analysis of the plastic content were taken from two quarters (bottom right and top left). Sample aliquots for the determination of the dry weight (DW) were taken from the bottom left quarter, while sample backups (1 L) were taken from the top right quarter. For the determination of the DW, 100 mL sample aliquots were weighed into 250 mL Schott-Duran beakers and dried at 105 °C (oven: Memmert UM 500, Memmert, Schwabach, Germany) for at least 24 h. Afterward, the beakers were allowed to cool to room temperature in a desiccator, and the DW was determined by reweighing the beakers.

For the recovery of individual plastic fragments, approximately 3 L of material were used. The wet weight was measured and correlated to the dry weight determined from another aliquot of the sample, see above. Then the material was evenly distributed into six glass vessels (capacity 3 L each). The material was suspended in 2.5 L of water and first sieved with a mesh size of 5 mm. All retained particles (fraction > 5 mm) were collected with tweezers, while the material passing the sieve was sieved again at 1 mm, followed by collection of the retained particles (fraction 1–5 mm). The sieves were obtained from Retsch GmbH (Haan, Germany; test sieve, IS 3310-1; body/mesh, S-steel; body, 200 mm × 50 mm).

For the analysis of the chemical nature of the particles, attenuated total reflection-Fourier transform infrared (ATR-FTIR) spectrometry (spectrometer: Alpha ATR unit, Bruker Optik GmbH, Ettlingen, Germany; equipped with a diamond crystal for measurements) was used. Spectra were taken from 4000 to 400 cm<sup>-1</sup> (resolution 8 cm<sup>-1</sup>, 16 accumulated scans, OPUS 7.5 software) and compared with entries from an in-house database described previously [11] or the database provided by the manufacturer of the instrument (Bruker Optik GmbH). An incident light microscope (microscope, Nikon SMZ 754T; digital camera, DS-Fi2; camera control unit, DS-U3; software, NIS Elements D) was used for visual



documentation and dimensional analysis of all particles identified by ATR-FTIR as synthetic plastics.

### Analysis of Plastic Fragments in the Liquid Fertilizers

The LF samples were also sieved with 5 mm and 1 mm sieves to obtain all fragments > 1 mm. In addition, fragments with sizes ranging between 10 and 1000  $\mu\text{m}$  were treated and analyzed as described previously [13, 14]. Briefly, the LF sample was mixed well with a metal rod, and 50 mL were quickly poured into a 300 mL glass beaker (Schott-Duran). The metal rod and the glass beakers were washed in advance with Millipore water. Subsequently, the samples were purified using an enzymatic-oxidative digestion sequence, as summarized in Table 2 below. Blank samples were processed in parallel in the same way.

Between steps, the sample was filtered through a 10  $\mu\text{m}$  stainless steel mesh filter (47 mm diameter, Rolf Körner GmbH, Niederzier, Germany) with a vacuum filtration unit (3-branch stainless steel vacuum manifold with 500 mL funnels and lids, Sartorius AG, Göttingen, Germany) and rinsed with filtered deionized water to remove residues from the reagents of the previous step. Then, the sample was rinsed back into the reaction vessel with either 20 mL of filtered deionized water or the amount of buffer specified in Table 2, and the filter was placed in the reaction jar to be used again in the subsequent step. All filtrations were conducted under a laminar flow hood to minimize contamination with MP from the surrounding air.

For the final density separation step, the retained matter was transferred from the filter into a clean glass beaker using a metal spatula, and an aqueous  $\text{ZnCl}_2$  solution (50 mL;  $\rho = 1.8 \text{ g cm}^{-3}$ ) was added. The mixture was stirred with a magnetic stir bar until all aggregates had been dispersed. Then, the mixture was transferred into a straight-walled separation funnel (400 mL). The mixture was stirred for several minutes with a glass rod and left to settle overnight (at least 12 h). The plastic fragments separated from any mineral matter by rising to the top. After separating the sediment, this less-dense fraction was filtered onto a new 10  $\mu\text{m}$  stainless steel mesh filter, which was then rinsed with 98% filtered ethanol and filtered deionized water to remove residual  $\text{ZnCl}_2$ .

Depending on the initial amount and the quality of the matrix, the amount of plastic recovered by the outlined purification procedure can vary. To avoid matrix interference, which would make FTIR analysis difficult, overloading of the aluminum oxide sample carrier filters (0.2  $\mu\text{m}$ , Anodisc, Whatman GE Healthcare) must be avoided. Therefore, samples with a high amount of material were suspended in filtered deionized water, evenly filtered through a 5  $\mu\text{m}$  stainless steel mesh filter (diameter: 47 mm), and then halved

using custom-made pliers. One half was washed into a clean 100 mL beaker, while the other was kept as a backup sample. This process was repeated as often as necessary to achieve a subsample that could be transferred onto 3–5 aluminum oxide filters for spectroscopic measurement. The filters were analyzed with focal plane array-based  $\mu\text{-FTIR}$  spectroscopy (10), which allows the determination of the fragment shape, size, color and polymer type, using a Bruker Hyperion 3000 FTIR microscope (Bruker Optik GmbH) equipped with a  $64 \times 64$  pixel FPA detector in conjunction with a Tensor 27 spectrometer. The samples were measured in transmission mode with a  $3.5 \times$  IR objective (spatial resolution 11.05  $\mu\text{m}$  per pixel) and a wavelength range of  $3600\text{--}1250 \text{ cm}^{-1}$  (resolution  $8 \text{ cm}^{-1}$ , 6 accumulated scans). Data processing was conducted using Bruker OPUS software version 7.5 (Bruker Optik GmbH), and automated spectral analysis was performed with the “BayreuthParticleFinder” module in ImageLab version 4.1 (EPINA GmbH, Retz, Austria) based on random forest decision classifiers [15, 16] for 22 different polymer types. The results of the automated spectral analysis were checked by experienced personnel for quality assurance and finally corrected with the blank values.

## Results

### Plastic Contamination of the Sampled Composts and Solid Digestates

When choosing the biowaste treatment plants for our study, we attempted to cover the current technical range of such plants, i.e. both simple composting plants and two stage digester–composters. Simple composting plants processing greenery and agricultural biomass digesters (biogas plants) processing mostly manure and energy crops were included for comparison. Table 3 summarizes the number of plastic fragments found per kilogram dry weight (DW) in the investigated composts and solid digestates. Composts sampled from plants processing biowaste are indicated by bold print. Note that solid digestates could only be sampled from the biogas plants, for technical reasons they were not available from the biowaste treatment plants.

Plastic fragment were collected in two size categories, > 5 mm and 1–5 mm. Consequently, we were able to distinguish between larger particles (> 5 mm) and those constituting MP (< 5 mm). The lower limit of 1 mm in the 1–5 mm category is of interest, since the current German regulations of compost quality in regard to contamination by plastic fragments consider only particles > 1 mm, the EU is expected to soon follow (current limit > 2 mm). Fragment numbers were normalized to the DW of the samples since the water content of the samples varied significantly; for details, see Table 3.

**Table 2** Enzymatic-oxidative digestion sequence for analysis of microplastics (10–1000 µm) from liquid fertilizer samples

| Step | Volume                     | Reagents  | Function of reagents  | Incubation time | Incubation temperature                         |
|------|----------------------------|---|---|-----------------|--|
| 1    | 50 mL                      | 10% sodium dodecyl sulfate*                     | Anionic detergent to solubilize lipids and proteins                             | 72 h            | 50 °C  |
| 2    | 50 mL in 2 × 25 mL batches | 30% H <sub>2</sub> O <sub>2</sub> **            | Oxidizing agent to degrade organic material                                     | 2 h             | 35–40 °C temperature controlled in an ice bath |
| 3    | 50 mL & 10 mL              | 0.1 M Tris HCl buffer (pH 9)*** & protease A-01 | Hydrolyzation of proteins into soluble peptides                                 | 12 h            | 50 °C  |
| 4    | 25 mL & 5 mL               | 0.1 M NaAc buffer**** (pH 5) & Pektinase L-40   | Degradation of any pectin in the primary cell wall and middle lamella of plants | 72 h            | 50 °C  |
| 5    | 25 mL & 1 mL               | 0.1 M NaAc buffer (pH 5) & Viscozyme L          | Splitting of the β(1,3) linkages of cellulose                                   | 48 h            | 50 °C  |
| 6    | 25 mL & 5 mL               | 0.1 M NaAc buffer (pH 5) & Cellulase TXL        | Splitting of the β(1,3) linkages of cellulose                                   | 24 h            | 40 °C  |
| 7    | 40 mL                      | Fenton's reagent**                              | Oxidation of degradation products and remaining organic material                | 2 h             | 40 °C  |

\*≥ 95% SDS; Karl Roth; \*\*30% H<sub>2</sub>O<sub>2</sub>; Fischer Scientific; \*\*\*≥ 99.3%, buffer grade; Karl Roth; \*\*\*\*≥ 98.5%, sodium acetate; Karl Roth

In three of the plants (#1.6, #3.1, and #3.3), we were also able to sample the precomposts. However, only in the case of plant #3.3 did we have access to material from a particular batch of compost before and after the final sieving step. In all other cases, precomposts and finished composts were sampled at the same time; i.e., the samples originated from different batches/initial loadings and thus cannot be compared directly. In plant #1.6, samples were taken of finished compost, material after an intermediate sieving step, and precompost. In the case of biogas plant #2.1, we had access to digestate pellets made from dried and pressed digestate, as well as to fresh digestate.

A comparison of the precomposts and finished composts shows that the final sieving step (typically using a 10 or 12 mm mesh size, Table 1) reduces the contamination of fragments > 5 mm, while sieving is less efficient in regard to the removal of fragments in the 1–5 mm range. In case of plant #3.3 were pre- and final composts were available for a given compost batch and hence directly comparable, the final sieving reduced the number of particles > 5 mm from 194 in the precompost to 53 in the finished compost while having little to no effect on particles in the range of 1–5 mm (46 and 48 particles, respectively).

With values between 10 and 15 particles kg<sub>DW</sub><sup>-1</sup>, finished biowaste composts from simple composting plants (category 1) showed only slightly higher plastic contamination than did finished greenery composts, which was not anticipated, given that incoming biowaste is much more contaminated by plastics than is greenery (Fig. 1). The data for plant # 1.6, i.e., the only simple biowaste composter where precompost data were also available, particularly demonstrates the

efficiency of the final sieving step in terms of reducing the number of fragments.

In comparison, the finished composts from the digester–composters contained a significantly higher number of particles in both the > 5 mm and 1–5 mm categories despite the use of similar final sieving mesh sizes. The digester–composters included in our study tended to use shredders to process incoming biowaste, whereas the simple biowaste composters mainly used bag slicers. While shredding is effective in making material accessible for digestion and biogas production, it presumably increases plastic and MP contamination. By comparison, digester–composter #3.2, which used an initial box fermentation step (no initial substrate preparation/shredding), had significantly lower plastic contamination in its finished compost, particularly from the difficult-to-remove 1–5 mm size fraction. Moreover, the content of particles in the > 5 mm fraction could presumably be further reduced by using a 10/12 mm mesh for final sieving instead of the currently used 20 mm mesh.

In general, the final sieving had a significant effect on the particle dimensions. Most retained particles had a longish shape. Whereas the range of particle length was nearly identical for particles sieved with a 10, 12, or 15 mm mesh size, the average width of the particles passing the final sieve nearly doubled from 10/12 to 15 mm. Particles capable of passing the 20 mm mesh were considerably larger in both length and width, for details see Fig. S1. Average values and standard deviations for the fragment sizes (length × width) are given in Table S1.

Finally, the production of LF (by press filtration) seems to increase the contamination of composts prepared from the

**Table 3** Number of plastic fragments in two size categories (> 5 mm and 1–5 mm) found in compost and solid fertilizer samples

| Sample                     | Number of plastic fragments $\text{kg}_{\text{DW}}^{-1}$ |              | DW content [%] |
|----------------------------|--|--------------|----------------|
|                            | > 5 mm   | 1–5 mm       |                |
| F_1.1-I                    | 1.58   | 1.58         | 64.46          |
| <b>F_1.1-II</b>            | <b>11.07</b>   | <b>2.46</b>  | <b>74.9</b>    |
| F_1.2-1                    | 13.27  | 4.82         | 80.15          |
| F_1.2-2                    | 7.55   | –            | 76.62          |
| F_1.2-3                    | 3.44   | 3.44         | 62.46          |
| F_1.3-1                    | 6.35   | 1.27         | 58.25          |
| F_1.3-2                    | 5.13   | –            | 72.53          |
| <b>F_1.4-1</b>             | <b>10.69</b>   | –            | <b>60.52</b>   |
| F_1.5-1                    | 42.14  | 7.80         | 61.83          |
| <b>P_1.6-1</b>             | <b>34.78</b>   | –            | <b>86.50</b>   |
| <b>P_1.6-2<sup>a</sup></b> | <b>90.00</b>   | <b>6.67</b>  | <b>73.31</b>   |
| <b>F_1.6-1</b>             | <b>11.53</b>   | <b>4.60</b>  | <b>68.71</b>   |
| S_2.1-1 <sup>b</sup>       | 13.04  | 15.66        | 86.13          |
| S_2.1-2 <sup>b</sup>       | 4.90   | 1.84         | 88.27          |
| S_2.1-3                    | 18.99  | 22.79        | 31.73          |
| S_2.2-1                    | 13.60  | –            | 18.57          |
| S_2.2-2                    | 7.79   | –            | 19.9           |
| S_2.3-1                    | 0.00   | –            | 21.93          |
| <b>F_3.1-1</b>             | <b>67.41</b>   | <b>23.68</b> | <b>45.86</b>   |
| <b>F_3.2-1</b>             | <b>28.65</b>   | <b>4.77</b>  | <b>64.88</b>   |
| <b>P_3.3-1</b>             | <b>193.95</b>  | <b>46.31</b> | <b>57.49</b>   |
| <b>F_3.3-1</b>             | <b>53.14</b>   | <b>48.53</b> | <b>57.41</b>   |
| <b>P_3.4-1</b>             | <b>53.62</b>   | <b>7.94</b>  | <b>39.66</b>   |
| <b>F_3.4-1</b>             | <b>15.41</b>   | <b>5.60</b>  | <b>51.57</b>   |
| <b>F_3.5-1</b>             | <b>97.88</b>   | <b>16.04</b> | <b>70.22</b>   |

Samples from plants processing biowaste are indicated by bold print

<sup>a</sup>Sample taken after an intermediate sieving step

<sup>b</sup>Sample of digestate pellets

corresponding pressed solid digestate, since the composts from plants #3.1, #3.3, and #3.5 were the most contaminated ones found in this investigation. Incidentally, the addition of cuttings as structuring material to the digestate for the composting step, which presumably would dilute the particle contamination (such added cuttings contain little to no plastic) is not able to compensate for this effect. All three plants in question, #3.1, #3.3, and #3.5, add cuttings in varied amounts, yet there is no correlation between the added amount and the residual plastic contamination of the finished composts. Moreover, composts from plant #3.2 (no press filtration, *no addition* of cuttings) were less contaminated than composts from plant #3.4 (no press filtration, *addition* of cuttings). Of course, other process conditions also contribute to the final results, while the number of sampled plants is at present too small for a statistically significant analysis of this contribution.

### Chemical Signatures of the Plastic Fragments Found in the samples

Figures 2, 3 and 4 summarize the chemical signatures of the plastic fragments found in the investigated compost and solid digestate samples according to relative percentage. In Fig. 2 arrows indicate samples from plants processing biowaste. Only plastic types accounting for > 5% of the total number of fragments are specified. All other types are grouped as “others”.

According to these data, polyethylene (PE)-based fragments tend to dominate in the compost samples prepared from biowastes. This is independent on whether the compost is prepared by simple biowaste composting or via the more intensive two-stage process of digestion–composting. In all cases the domination of PE is more pronounced in the sieving fraction > 5 mm. The polymer type distribution



**Fig. 1** Left: incoming greenery waste, right: incoming biowaste, both at plant #1.1



found in the greenery compost seems to be more diverse, with a stronger tendency towards polypropylene (PP) over PE. However, in particular in case of the simple composts, the samples typically contained only a small total number of fragments, a statistically relevant analysis is therefore not possible.

Of some concern is the category “other plastics”, which tended to accumulate in the 1–5 mm size fraction. Among the “other plastics”, we found mainly polyethylene terephthalate (PET), Platilon T (a thermoplastic adherent polyurethane film) and other polyurethanes, as well as polyvinyl chloride (PVC), acrylonitrile compounds such as acrylonitrile butadiene styrene and styrene–acrylonitrile, and Teflon compounds. Polymers such as poly(lactic acid) (PLA) or poly(butylene adipate-co-terephthalate) (PBAT) were occasionally also found, but were not considered in this study due to their presumed biodegradable nature. Notably, fragments with PBAT and PLA signatures were found only in the composts from the two-stage digester–composters, never in those from the simple biowaste composters.

In the case of the digestates from the agricultural biogas plants (Fig. 3), no sieving took place. Nevertheless, no plastic fragments were found in the case of plant #2.3, which corroborates previous findings (8) that fertilizers (digestates) from such plants are minimally contaminated with plastic. In case of plant #2.2, two samples were investigated. Both contained some plastic fragments > 5 mm, but no smaller fragments. Moreover, the contamination was very uniform. In the case of digestate sample S-2.2-1, 100% of the few fragments found in the size fraction > 5 mm were PP, while in the case of sample S-2.2-2, 50% were PE, and 50% PP (Fig. 3a). It is thus possible that the contamination was caused by a single piece of plastic entering the digester by accident. Both plants #2.2 and #2.3 exclusively processed agricultural waste and energy crops.

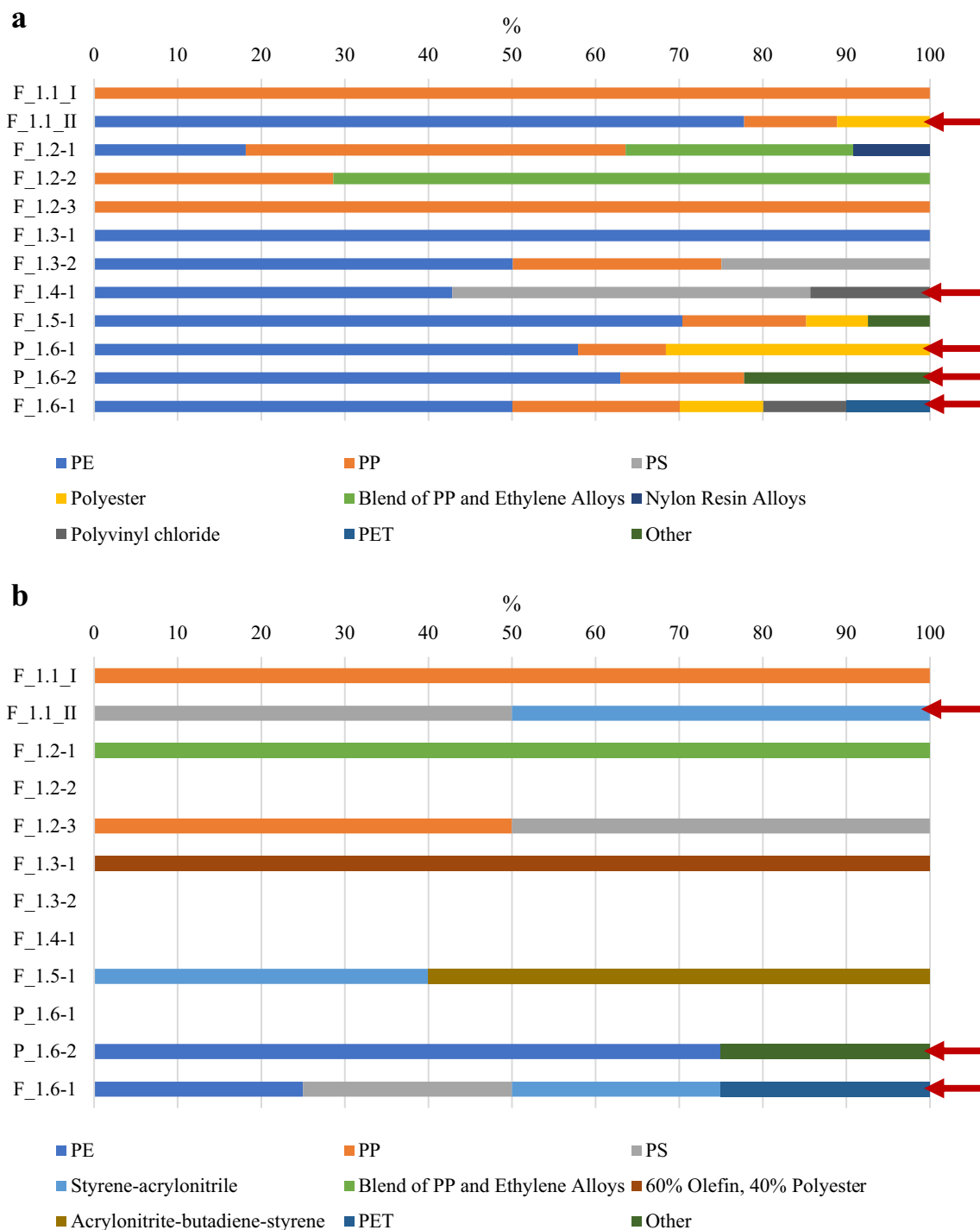
Plant #2.1 processed a wider selection of organic material, including organic waste from local markets and landscape conservation material (see Table 1 for details). In addition to fresh digestate, plant 2.1 produced digestate pellets as fertilizer. Two charges of pellets were sampled in addition to the fresh digestate. The contamination in terms of fragments > 5 mm was similar to that of the digestate from plant 2.2. For the first time, however, we also found significant numbers of fragments in the 1–5 mm size category in a fertilizer from an agricultural biogas plant. The diversity of the particle chemical signatures detected in the fertilizers from plant #2.1 (Fig. 3) is also much broader than that for the other two plants in category 2 and similar to the variability found for the biowaste processing plants.

## Contamination of Liquid Fertilizer by Plastic Particles

Plants #3.1, #3.3, and #3.5 produce LF in addition to biowaste compost. LF has a low solid content and is produced by press filtration of the digestate obtained at the end of the anaerobic step. While the solids from the press filtration step thus enter the composting stage, LF is typically directly distributed on agricultural soil. Since LF is essentially a liquid, plastic particles down to a size of 10 µm could be isolated and identified in the LF samples using environmental techniques developed for water analysis (10), which was not possible in the case of the solid digestates and composts. The LF samples were also analyzed for fragments > 1 mm, but none were found. Presumably, these fragments were retained in the solid digestate during press filtration and went into the composting stage. As discussed before, this effect is most likely also responsible for the fact that the composts from plants #3.1, #3.3 and #3.5 contain a higher number of plastic particles than do the composts of plants #3.2 and #3.4. Table 4 summarizes the particle numbers and plastic types found in the LF samples.

LF from the biowaste treatment plants contained between 6000 and 12,000 MP fragments per liter. Most of these fragments had sizes between 22 and 300 µm. PP, PE, and PET were detected in all samples, while PS, silicone and PVC were found occasionally. In certain cases, e.g., in sample L3.5-1, we again observed large numbers of MP with IR signatures corresponding to biodegradable materials, mainly PBAT; however, these particles were not considered here for reasons already given.

In the case of the agricultural biogas plants (category 2), only the LFs produced by plants #2.1 and #2.3 were sampled since for technical reasons, LF from plant #2.2 was not available. In the case of plant #2.3, which produced a particularly clean solid digestate, the number of contaminating particles found in the LF was also negligible. Moreover, all particles had the chemical signature of silicone. Silicone is used in biogas plants for various purposes, including as an antifoaming agent, for the sealing of concrete floors and walls, and as an antimicrobial barrier layer [17, 18]. Finding silicone residues in LF is therefore not necessarily a surprise. By comparison, plant #2.1, which used a wider range of possibly pre-contaminated substrates and had no pre-sorting and/or sieving system installed, showed a higher contamination level in the case of LF. Most MPs found were in the range of 22–100 µm, and some were also in the range of 100–300 µm. PE, PP, and PS were again observed, but surprisingly large numbers of PET fragments were also observed. This is surprising since PET fragments > 1 mm were not found in the corresponding compost samples.



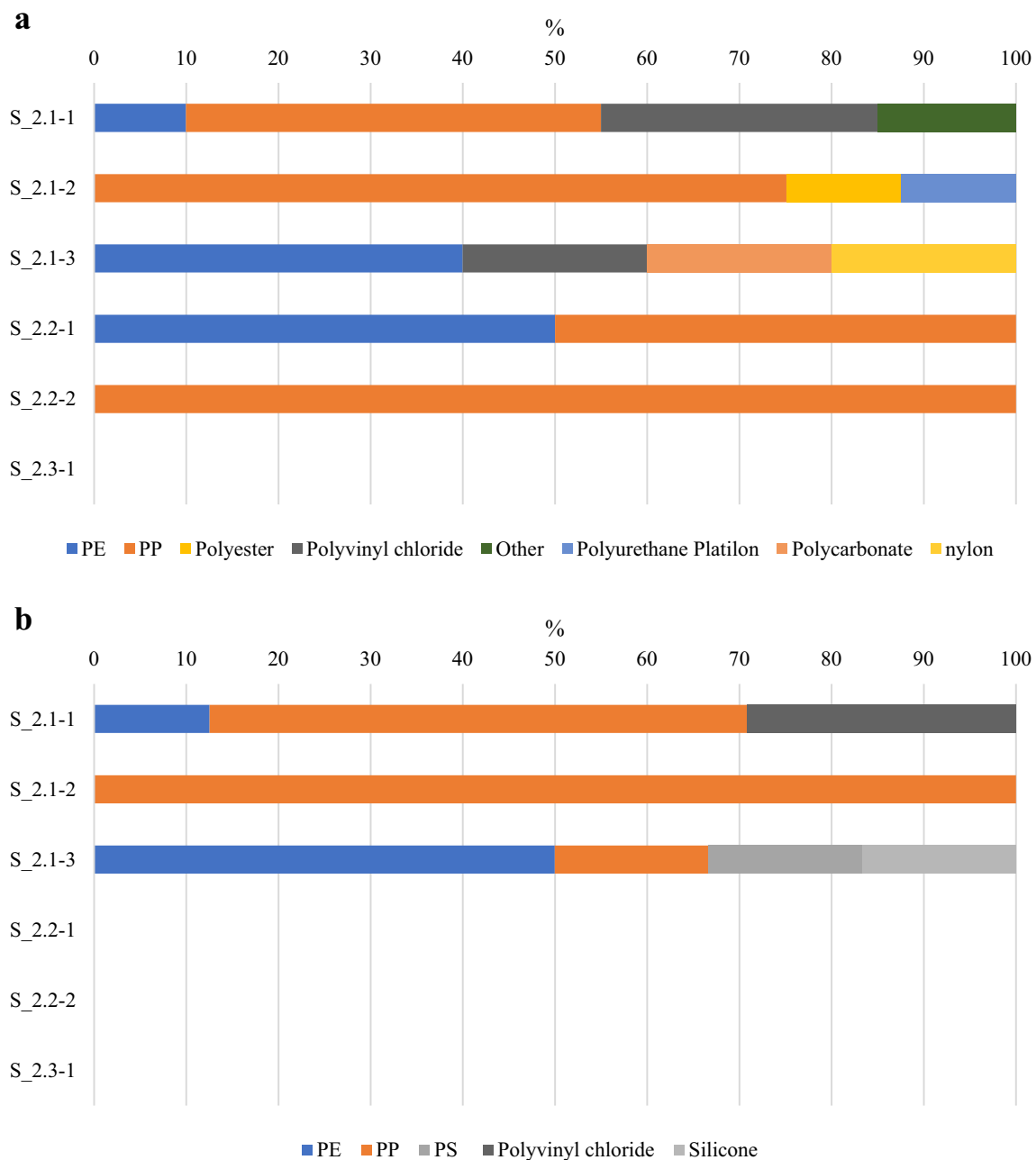
**Fig. 2** Distribution over the material classes for plastic fragments found in plants from category 1 (simple composters). Arrows indicate samples from biowaste treatment plants. For the total number of

particles found in each sample, see Table 3. **a** >5 mm size category; **b** 1–5 mm size category. P\_1.6-2: sample taken after the 12 mm sieving step but before the final 10 mm sieving step

### Discussion

Our study focuses on current practices for the recycling of household biowaste into fertilizer using either simple

composting or the more economic two-stage digestion–composting approach. Three agricultural biogas plants (simple digesters) and several composting plants producing greenery compost were included in our study for comparison. As



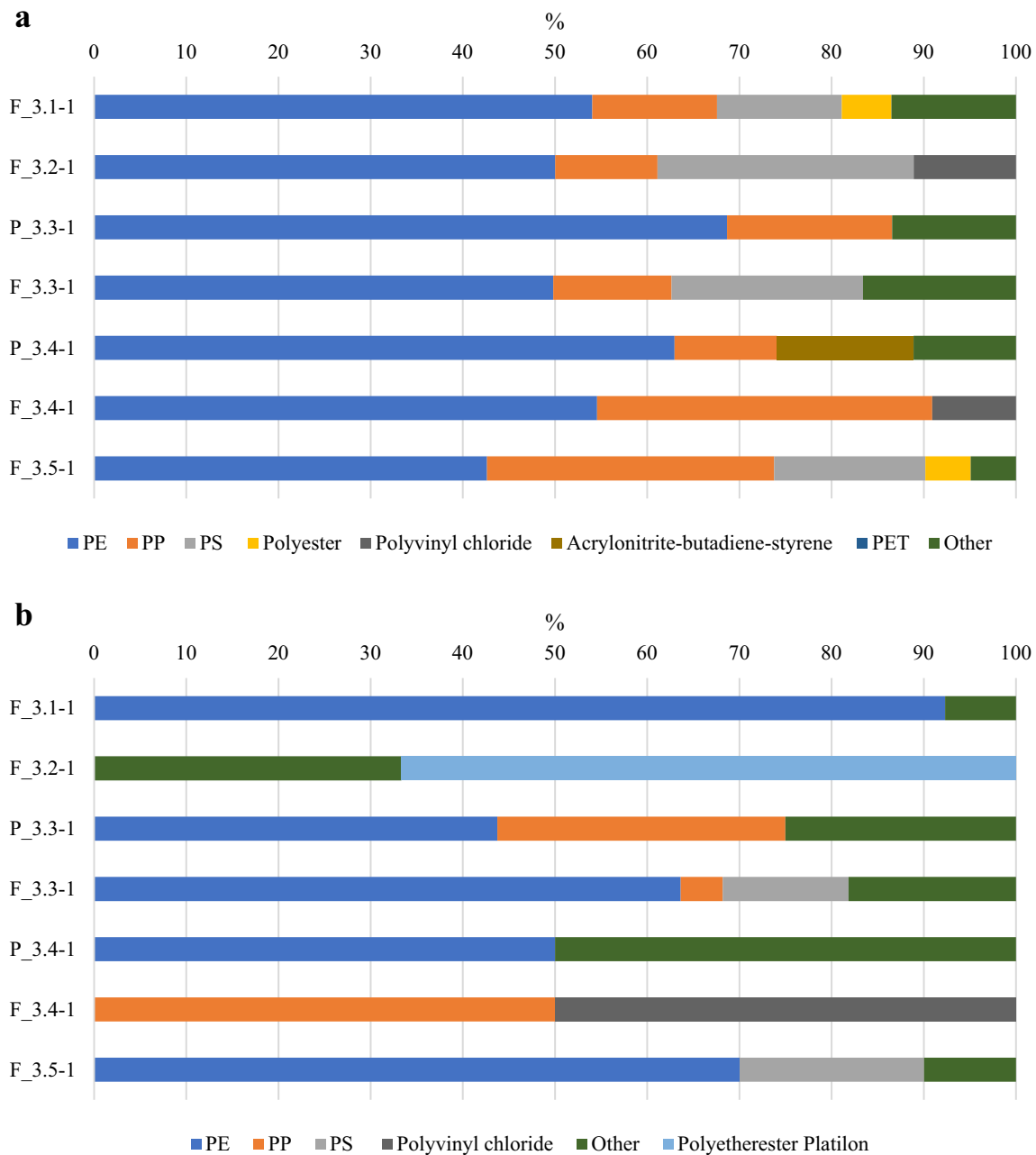
**Fig. 3** Distribution over the material classes for plastic fragments found in plants from category 2 (agricultural biogas plants). For the total number of particles found in each sample, see Table 3. **a** > 5 mm

size category; **b** 1–5 mm size category. S\_2.1-1 and S\_2.1-2: digestate pellets made from dried and pressed digestate.

observed before [8], the liquid and solid fertilizers from the agricultural biogas plants exhibited unremarkable residual plastic contamination. Only the digestate from plant #2.1, i.e., the only agricultural biogas plant in our study that added a wider variety of organic material to its substrate mix, showed increased levels of contamination.

Greenery composts also showed only a low level of plastic contamination compared to the biowaste composts (Table 3). Plant #1.1 is of particular relevance in this regard,

since it comprises two composting lines operated in parallel, one for greenery waste (1.1\_I) and the other for biowaste (1.1\_II), under otherwise similar conditions. One exception in our data set is the greenery compost from plant #1.5, which, despite stemming from cuttings, contained an average of 50 plastic particles  $\text{kg}_{\text{DW}}^{-1}$ . With only 9500 t year<sup>-1</sup>, plant #1.5 was by far the smallest in our study, and more stringent quality control of the input material together with a final sieving using a mesh size below the currently used



**Fig. 4** Distribution over the material classes for plastic fragments found in plants from category 3 (digester–composters of biowaste). For the total number of particles found in each sample, see Table 3. **a** > 5 mm size category; **b** 1–5 mm size category

20 mm might help to improve the quality of the compost from that plant.

While biowaste compost in general tended to contain more plastic fragments than greenery compost, there was a consistent and significant difference in the level of contamination between the composts from the simple biowaste composters and those produced by the two-stage biowaste digester–composters. The composts from the digester–composters were significantly more contaminated by plastic fragments, even when the higher mass reduction achieved by the

two-stage process during biogas production or a possible enrichment of plastic fragments > 1 mm during press filtration in some of these plants is taken into account (Table 3). Simple biowaste composters often use bag slicers to gain access to organic material, whereas two-stage plants tend to use shredders. It seems that the latter approach, as beneficial as it is for efficient biogas production, aids in the formation of plastic fragments. This would explain why the composts produced in digester–composter plant #3.2 were among the least contaminated composts ( $33 \text{ particles kg}_{\text{DW}}^{-1}$ ) found in



**Table 4** Plastic particles per liter of liquid fertilizer and percentages according to size and type; biowaste treatment plants: L\_3.1-1, L\_3.3-1, L\_3.5-1; agricultural biogas plants: L\_2.1-1, L\_2.3-1

| Sample  | Fragment number and type | 11–22 $\mu\text{m}$ | 22–100 $\mu\text{m}$ | 100–300 $\mu\text{m}$ | 300–500 $\mu\text{m}$ | 500–1000 $\mu\text{m}$ |
|---------|--------------------------|---------------------|----------------------|-----------------------|-----------------------|------------------------|
| L_3.1-1 | Number                   | –                   | 5120                 | 2560                  | 1280                  | 1280                   |
|         | PET                      |                     | 25.0%                | 50.0%                 |                       |                        |
|         | PE                       |                     | 25.0%                |                       |                       | 100%                   |
|         | PP                       |                     |                      | 50.0%                 |                       |                        |
|         | PS                       |                     |                      |                       | 100.0%                |                        |
| L_3.3-1 | Number                   | 640                 | 3840                 | 2240                  | –                     | –                      |
|         | PET                      | 50.0%               | 33.3%                | 14.3%                 |                       |                        |
|         | PVC                      |                     |                      | 14.3%                 |                       |                        |
|         | PE                       | 50.0%               | 33.3%                | 28.6%                 |                       |                        |
|         | PP                       |                     | 16.7%                | 14.3%                 |                       |                        |
|         | PS                       |                     | 16.7%                | 28.6%                 |                       |                        |
| L_3.5-1 | Number                   | –                   | 5120                 | 6400                  | –                     | –                      |
|         | PET                      |                     |                      | 40.0%                 |                       |                        |
|         | PE                       |                     | 75.0%                | 40.0%                 |                       |                        |
|         | PP                       |                     | 25.0%                | 20.0%                 |                       |                        |
| L_2.1-1 | Number                   | –                   | 1280                 | 320                   | –                     | –                      |
|         | PET                      | –                   | 25.0%                | 100.0%                | –                     | –                      |
|         | PE                       | –                   | 25.0%                | –                     | –                     | –                      |
|         | PP                       | –                   | 25.0%                | –                     | –                     | –                      |
|         | PS                       | –                   | 25.0%                | –                     | –                     | –                      |
| L_2.3-1 | Number                   | 320                 | 320                  | –                     | –                     | –                      |
|         | Silicone                 | 100.0%              | 100.0%               | –                     | –                     | –                      |

category 3. In contrast to all other plants in that category, plant #3.2 did not use a shredder to process the incoming material. Furthermore, the data from this plant suggest that using an anaerobic digestion step does not necessarily increase the plastic fragment content. Otherwise, one would expect the composts from plant #3.2 to show a higher content of residual plastic fragments.

While the anaerobic treatment and shorter composting times applied in two-stage plants are unlikely to influence the contamination of the finished composts with fragments of conventional plastics, these differences in operating conditions could explain why we found residues of biodegradable plastics in the composts (and LF, see below) from the two-stage plants but not in any of the composts from the single composting plants. The digester–composters included in our study typically only used composting times of 5–6 weeks (after digestion), in contrast to at least 12 weeks in the case of the simple composters. Anaerobic digestion is not expected to contribute to the degradation of biodegradable plastics, and the length of the subsequent composting stage may thus not be sufficient for the full degradation of such biodegradable materials. The pertinent literature suggests that once biodegradable plastics enter the environment, they can persist there for quite some time [19–21].

For all investigated compost samples (both biowaste and greenery), the final sieving step used to prepare the finished composts was effective in reducing the number of contaminating plastic fragments. However, sieving is much more efficient for fragments  $> 5$  mm than for smaller fragments. Any processing step that increases the number of fragments, particularly that of smaller fragments, is therefore problematic. The bias for removing larger fragments in the final sieving step can be seen in Fig. S1. Whereas the lengths of the particles sieved with 10, 12, or 15 mm mesh sizes cover a very similar range, the widths of the retained particles tend to increase with the mesh size used during sieving, see also Table S1. If we presume the fragments pass a given hole “head on”, it makes sense that the fragment width determines the likelihood of passage. In terms of the types of plastics found in the most contaminated ( $> 30$  particles  $\text{kg}_{\text{DW}}^{-1}$ ) and hence most statistically relevant compost samples, PE was the most common type of plastic found in the biowaste composts. PP and PS, as well as some “other polymers” (individually representing  $> 5\%$  of the total), were also found in the composts made from biowaste and/or cuttings. In the greenery composts, PP was the dominant polymer, along with PE.

Due to the restrictions of the analytical methods, mainly the need to isolate and clean fragments for spectroscopy, the

study of the composts was restricted to fragments retained by the 1 mm sieve. Given the especially high load of > 1 mm fragments in the composts from the digester–composters, the additional presence of yet smaller particles in the composts cannot be excluded. Three of the digester–composters produced LFs, recovered via press filtration after anaerobic digestion, which could be analyzed for plastic fragments down to 10 µm using techniques originally developed for water samples. According to the results, the LFs contained up to 10,000 MP particles with sizes of 10–1000 µm<sup>-1</sup>, while no fragments > 1000 µm were found. According to common agricultural practices, LF is applied several times a year at a concentration of 2–3 L m<sup>-2</sup>.

Most fragments/particles found in the LFs were in the range of 22–300 µm and smaller. The mechanical stress exerted on fermented plastics during the press filtration step can presumably lead to fragmentation and in consequence the formation of small MP particles, especially since the materials presumably become more brittle during anaerobic digestion due to the extraction of additives such as plasticizers [22]. It does not take many large fragments to result in a significant number of small ones. For instance, a single 4 mm × 4 mm fragment could break down in more than 100,000 fragments of 100 µm<sup>2</sup>. Recently, it has been shown that PE and PS macroparticles (> 25 mm) can release 4–63 MP particles during the composting process [23]. While we thus still assume that most larger fragments present in a given digestate end up in the solid fraction after press filtration and thus in the composter, some fragmentation under the mechanical stress of press filtration may result in the heavily contaminated liquid fertilizer. The LFs produced by the agricultural biogas plants contained significant numbers of MPs from commodity plastics only in the case of plant #2.1, i.e., the plant where the solid digestate also contained unusually high plastic contamination. While significant, the MP content of the LF from plant #2.1 was still an order of magnitude less than that of the LFs from the biowaste treatment plants.

Chemically speaking, the MPs found in the LFs stemmed mostly from commodity plastics such as PE, PP and PS, all of which have been shown to be toxic or harmful to the environment [24–26]. Moreover, in all investigated LFs, fragments of PET were found in 22–40% of the detected particles (Table 4), even in those cases where PET was not found in larger fragments in the corresponding composts or digestates (Figs. 3, 4). This could be an indication that PET is easily fragmented into MPs under anaerobic/aerobic treatment of organic material. It has been shown that PET in soil can be highly toxic to nematodes [25].

## Conclusions

Plant type and operating conditions have a major influence on the residual contamination of composts and organic fertilizers with plastic fragments. The removal of such fragments would cause a further reduction in process yield and efficiency. Reducing the plastic content in the incoming biowaste is thus still the most important measure for reducing the release of plastics and MPs into the environment via such composts and LF. Whether biodegradable materials may present a solution in this context was not part of our study. The fact that we did find residues of biodegradable material in some of the composts shows that the behavior of such materials during biowaste treatment may have to be reevaluated in the future.

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**Data Availability** All data are available in the main text or the supplementary materials.

## Declarations

**Competing Interests** Authors declare that they have no competing interests.

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