

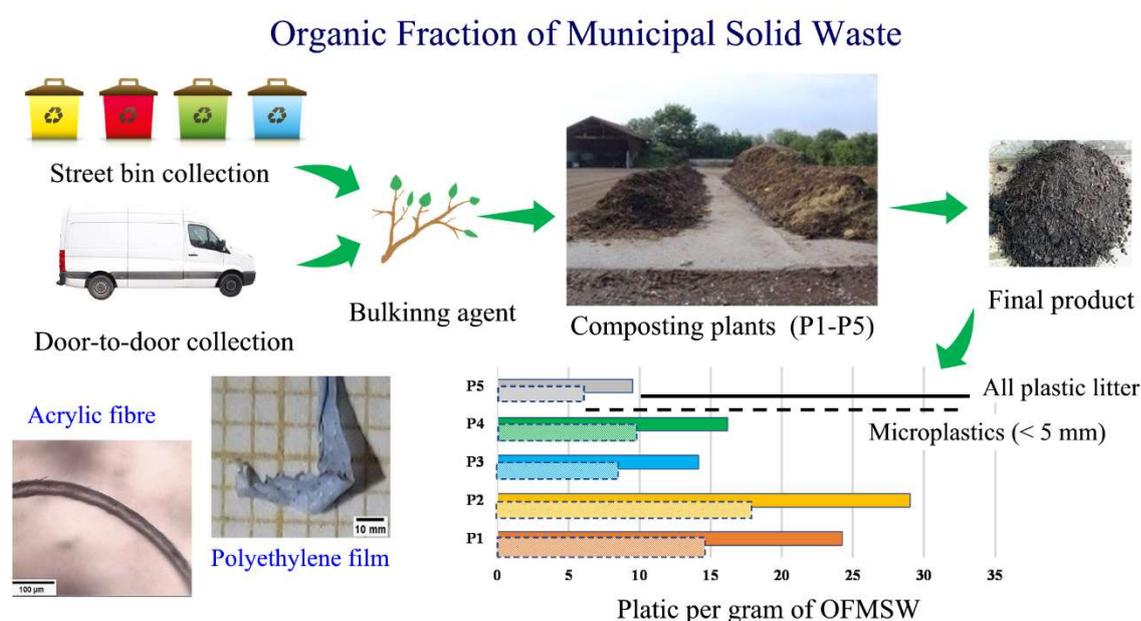
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# Microplastics identification and quantification in the composted organic fraction of municipal solid waste

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## Abstract

Composted Organic Fraction of Municipal Solid Waste (OFMSW) is used in agricultural soils as a source of organic matter and nutrients. Besides, its use avoids landfilling or incineration following the principles of circular economy. It is well established that source separated OFMSW is suitable for compost production, but its quality depends on their content in non-compostable materials. In this work, we selected and studied the final refined compost from five OFMSW facilities over a five-month period. The plants displayed differences in collection systems, concentration on non-desired materials, treatment technology and density of served population. The presence of plastic was studied using a separation and identification process that consisted of oxidation and flotation followed by spectroscopic identification. The results showed a concentration of plastic impurities in the 10-30 items/g of dry compost range. The concentration of small fragments and fibres (equivalent diameter < 5 mm) was in the 5-20 items/g of dry weight range and were dominated by fibres (25 % of all particles < 500 µm). Five polymers represented 94 % of the plastic items: polyethylene, polystyrene, polyester, polypropylene, polyvinyl chloride, and acrylic polymers in order of abundance. Polyethylene was more abundant in films, polystyrene in fragments, polypropylene in filaments, and fibres were dominated by polyester. Our results showed that smaller plants, with OFMSW door-to-door collection systems produced compost with less plastic of all sizes. Compost from big facilities fed by OFMSW from street bin collection displayed the highest contents of plastics. No debris from compostable bioplastics were found in any of the samples, meaning that if correctly composted their current use does not contribute to the spreading of anthropogenic pollution. Our results suggested that the use of compostable polymers and the implementation of door-to-door collection systems could reduce the concentration of plastic impurities in compost from OFMSW.

Keywords: Organic Fraction of Municipal Solid Waste; Plastic impurities; Microplastics; Bioplastics; Compost

## 1. Introduction

Plastics were introduced by the middle of the 20th century and progressively gained a privileged position in our lives due to their outstanding properties, which allowed the rapid substitution of other materials and a plethora of new uses associated to the demands of our modern society. The current worldwide production of plastic is estimated in 368 million tonnes with main uses in packaging and building & construction, which represent together 60 % of the plastic demand in the EU plus United Kingdom, Norway and Switzerland (PlasticsEurope, 2020). For the same geographical unit, the amount of plastic waste collected roughly represents half of the total amount produced and still 7.2 million tonnes of plastics are sent to landfills (PlasticsEurope, 2020). Improper waste management and the wearing of plastic goods during use lead to the accumulation of plastic debris in all environmental compartments. The problem is far from new. The early findings of plastic debris in the ocean can be tracked back to the 1970s, but during the last decade the concern about the dissemination of small and very small plastic fragments

attracted substantial attention from researchers and even from the general public (Ryan, 2015). Small plastics are termed microplastics (MPs) if their larger dimension is < 5 mm with a lower boundary of 1 µm below which, plastic particles produced from the fragmentation of larger debris are usually classified as nanoplastics (NPs) (GESAMP, 2019; Gigault et al., 2018). Although a considerable body of evidence exists on the occurrence of large plastic debris, the data on the smaller fractions of MPs are still scarce due to the difficulties associated to their separation and analysis (Xu et al., 2020).

Plastic debris enter the environment through different ways. Atmospheric dissemination has recently received attention as small airborne debris, mainly consisting of synthetic fibres, may travel long distances from their source (González-Pleiter et al., 2021). The role of wastewater treatment plants is also well-known. Domestic and industrial wastewaters contain a large number of fibres, MPs used in personal care products, tyre wear debris and other MPs that are not completely removed in treatment plants (Ali et al., 2021; Liu et al.,

2021). The typical removal efficiency of current wastewater treatment process is > 90 % meaning that most MPs contained in raw wastewater accumulate in sludge (Xu et al., 2021). In fact, the use of wastewater sludge as soil amendment could be a contributor to MPs dissemination into soil, and from it to other environmental compartments (Edo et al., 2020a; Gao et al., 2020). Besides, agricultural practices like mulching may result in involuntary dissemination of plastic debris to the environment. However, the information available on the sources, fate and effects of MPs in soil are scarce (Yuanqiao et al., 2020). Only limited data are available due to the reduced number of studies accomplished so far and because of the lack of acute effects on biota, which complicates risk assessment (Cheng et al., 2021; Wang et al., 2019; Zhu et al., 2018). For the case of fibres, the information is even more scarce (Selonen et al., 2020). Besides, MPs release dangerous additives, sorb other pollutants and their fragmentation is known to produce smaller and more concerning fragments including NPs (Bueno-Ferrer et al., 2010; Guo et al., 2020; Tagg and Labrenz, 2018)

Apart from sludge and concerning agricultural practices, digestates and compost produced from the organic fraction of municipal solid wastes (OFMSW) are used throughout the world as soil amendment due to their content in stabilized organic matter and nutrients (Carabassa et al., 2020). The current legal scenario for the waste management sector forces improvements in selective collection, total amount of collected OFMSW and recycling procedures (Laso et al., 2019). The Directive 2018/851 makes the collection of OFMSW mandatory for all member states from 2023. Together with organic wastes, there is the possibility of collecting other types of wastes with similar biodegradability and compostability, which includes compostable bioplastic materials certified according to the EN13432. The implementation of adequate waste management policies aimed to promote the efficient separate collection of OFMSW, diverting it out of landfills, constitutes a key element in this new framework. The total generation of municipal solid waste ranges from 0.11 to 4.54 (average 0.74) kg per person per day (Kaza et al., 2018), from which the OFMSW represents approximately 40 % by weight. Therefore, the efficiency of separate collection systems, the biological treatment technology applied, and the requirements for the quality of compost are key drivers for sustainable waste management. In terms of OFMSW collection, different strategies can be implemented, ranging from the street bin containers to personalized door-to-door collection systems with different impacts on product quality. The collected OFMSW can be treated in composting or in combined anaerobic/aerobic treatment facilities with a variety of different layouts and technologies.

In this work, the presence of plastic debris in samples of composted OFMSW recovered and treated in five different industrial facilities (four composting plants and one using anaerobic digestion followed by composting) was studied. The purpose was to identify whether the collection and treatment systems affect the concentration of MPs in final refined compost. In this framework the efforts were mainly focused to quantify the number of plastic particles contained in the final OFMSW compost, their typology and polymer composition, with emphasis on the fraction < 1000  $\mu\text{m}$ . Special attention was paid to compostable biopolymers due to their role as a tool to promote a high quality collection of OFMSW especially in door-to-door collection systems.

## 2. Experimental section

### 2.1. Materials

Compost samples were obtained from five different composting facilities located in the northeast of Spain. The samples were taken in five consecutive months in 2021 (from February to June) and consisted of two replicates of about 200 g each per selected plant. All samples were collected after the refining operation and following the procedure to avoid contamination described below. Samples were stored in sealed aluminium bags for their transportation to the laboratory.

### 2.2. OFMSW facilities and collection systems

The facilities were selected based on the different technologies used and the diverse strategies followed to collect organic wastes. Their main characteristics are listed in Table 1. The OFMSW collection systems taken into consideration in this study were based on different combinations of street bin and door-to-door collection. In street bin collection, containers for organic waste are located at curbside and are periodically washed out and the collected organic waste transferred to the composting plant. This system does not permit any control concerning the disposal quality and does not guarantee that citizens use compostable bags. In door-to-door collection systems, citizens place twice a week their organic waste (small volume, usually 7-10 L) in specific places from where it is collected by dedicated trucks. In this collection system, the use of compostable bags is encouraged or mandatory. In all cases, the waste collected is a mixture of domestic and commercial activities (including restaurants), with higher intensity for the later in more densely populated areas. Once in the plant, the OFMSW undergo a sequence of pre-treatment steps with the purpose of removing all non-compostable or oversized materials. The intensity of pre-treatment systems is directly linked to the need of removing non-compostable materials, which is more or less stringent depending on the biological technology adopted to treat the OFMSW. Usually, a pre-treatment step is optional in composting, but it is a requirement in

**Table 1.** Summary of the main characteristics of composting plants and input materials (Source: information from the Agència de Residus de Catalunya).

Plant	P1	P2	P3	P4	P5
Capacity (tonnes/yr.)	20000	45000	12500	8000	750
Collecting systems	15 % door-to-door; 85 % compost containers	25 % door-to-door; 75 % compost containers	30 % door-to-door; 70 % compost containers	35 % door-to-door (including a wholesale market); 65 % compost containers	100 % door-to-door, compostable bags mandatory
Contamination of non-compostable materials (%)*	14.4	4.4	8.3	10.0	1.6
Pretreatment	Sieve (80 mm) and magnetic separator	Sieve (80 mm), magnetic separator, pulper and grit removal	Sieve (80 mm) and magnetic separator	Sieve (80 mm) and magnetic separator	No pre-treatment
Composting technology	Windrows	Anaerobic digestion + tunnel composting	Aerated static pile	Tunnel + windrows	Aerated static pile
Bulking agent	Local sources (65 %) and other origins, mostly agricultural wastes (35 %)	Private wood processing companies (100%)	Local recycling centres (60 %), other local public bodies (20 %), and private companies (20 %)	Local public bodies (75 %), and private companies (25 %)	Clean grinded wood from local recycling centres
Population density (inhab./km <sup>2</sup> )	90	4200	1900	2000	20

\* Averages for the 4th term 2020 and 1st term 2021; details in Table S1 (Supplementary Material)

anaerobic treatment followed by composting (Plant P2 in our study). In plants P1-P4 the OFMSW are mixed and sieved (80 mm) and a magnetic separator is used to remove ferromagnetic metals. P5 is somewhat special because it serves a small community with high citizenship commitment, which includes the exclusive use of compostable bags besides door-to-door collection. The composting technologies used in the different plants are conventional and consist of open windrows, aerated static piles, and in-vessel tunnels.

Table 1 shows the amount of non-compostable materials as determined by periodic inspection of 200-250 kg of OFMSW at the entrance of each plant (6-74 independent samples depending on plant size). The (mass) percentage of non-compostable materials listed in Table 1 corresponds to the 4th term of 2020 and the first term of 2021, which are the reference periods for the final compost sampled in this work. A detailed characterization of the non-compostable fraction is included as Supplementary Material in Table S1. Another important factor is the addition of a vegetal fraction as bulking agent to facilitate aeration, and balancing carbon-to-nitrogen ratio (Adhikari et al., 2008). As shown in Table 1, bulking agents have

different origins, from local wood processing companies to pruning and garden wastes.

### 2.3. Microplastics separation

A total amount of 60 g (30 g/sample in two replicates) was processed for each composting plant and sampling month (a total of 300 g of evaluated material per plant at the end of the study). Compost samples were first processed using size separation with stainless-steel sieves into three different fractions: > 3.55 mm (large), 1.00-3.55 mm (medium) and < 1.00 mm (small). All samples > 1.00 mm were separated in batches of about 10 g each, immersed in ultrapure water and sonicated using an ultrasound disperser (BioBlock Scientific, France) operating at 500 W for 30 s with the aim of separating aggregates of plastics with other particles. Subsequently the samples were filtered through 375 µm stainless steel filters and dried at 60 °C.

The fraction containing particles < 1 mm was separated in batches of about 5 g, put in contact with 30 mL of hydrogen peroxide (30 % w/v) and kept covered in oven at 60°C overnight to remove the organic matter that could interfere with visual examination and spectroscopic characterization. Samples were then

filtered through 25  $\mu\text{m}$  stainless steel meshes and washed with deionized water to remove residual hydrogen peroxide. The resultant material was put in contact with  $\text{ZnCl}_2$  solution (concentration 700 g/L, density  $1.70 \pm 0.05 \text{ g/cm}^3$ ) to perform a density separation. The suspensions were allowed to settle at least for 1 h and the supernatant filtered again through 25  $\mu\text{m}$  stainless steel filters. The sediment was discarded, and the samples dried at  $60^\circ\text{C}$  before visual inspection.

The methodology used, based in a combination of sieving, oxidative treatment, and density separation used a protocol adapted from similar ones reported elsewhere for water and soil samples (Möller et al., 2021; Rodrigues et al., 2020b). The method as described here yielded high recovery rates ( $> 95\%$  for plastics  $> 500 \mu\text{m}$ ) in line with previously published results (Kang et al., 2020). We did not observe degradation of plastic fragments that could be attributed to oxidative treatment or corrosion due to  $\text{ZnCl}_2$  and that could impair polymer identification. Besides, as stated below, a quality check performed for bioplastics also showed recovery rates  $> 98\%$  for fragments  $> 500 \mu\text{m}$ .

#### 2.4. Microplastics analysis and classification

All suspected plastic particles were picked up with metal tweezers or a needle, depending on their size, and stored in closed glass containers until spectroscopic characterization. Then they were identified, photographed, and measured using a Euromex-Edubluu stereomicroscope equipped with Image Focus software. Additionally, potential plastics were classified into four typologies: fragments, films, fibres, and filaments. Fragments were particles with irregular shape, while films corresponded to particles with one dimension significantly lower than the other. Fibres and filaments had one dimension (length) considerably higher than the other two (Rosal, 2021). In what follows, we considered fibres or filaments particles with length/width  $> 4$  and differentiated filaments from fibres because filaments displayed the same thickness along their length and presented sharp ends (Magni et al., 2019). Fig. S1 (Supplementary Material, SM) shows a scheme of the steps followed for the separation and identification of plastic particles. The quantitative results were expressed as number of plastic particles per unite mass of compost (dry weight, DW basis).

The chemical identification of polymer type was performed using Fourier Transformed Infrared Spectroscopy (FTIR). In detail, for particles  $> 1 \text{ mm}$  Attenuated Total Reflectance (ATR-FTIR) spectra were recorded in a Thermo-Scientific Nicolet iS10 equipped with Smart iTR-Diamond. Particles  $< 1 \text{ mm}$  were put on KBr discs and analysed using a Perkin-Elmer Spotlight 200 Spectrum Two micro-FTIR apparatus equipped with an MCT detector. In both cases 32 scans were used with  $8 \text{ cm}^{-1}$  spectral resolution and  $4000\text{--}550$

$\text{cm}^{-1}$  spectral range. The obtained spectra were compared with the databases existing in software Omnic 9 (Thermo Scientific) and with our own databases. Pearson correlation was used with a minimum of 65% matching for positive identification as stated elsewhere, except in some specific cases for which a case-by-case study was performed (González-Pleiter et al., 2021; Liu et al., 2019).

#### 2.5. Bioplastics determination

The applicability of the procedure reported above to biodegradable and compostable polymers was assessed as follows. Four materials made of compostable bioplastics certified by the European Standard EN-13432 were selected: two commercial bags from local markets, one wrapping film from a commercial food package, and specimens from the material Mater-Bi, supplied by Novamont S.p.A. The biopolymers were cut into small pieces  $< 1 \text{ mm}$ , carefully mixed with compost at concentrations of 10 particles/g and separated using the procedure outlined before for the smaller fraction (oxidation with  $\text{H}_2\text{O}_2$  at  $60^\circ\text{C}$ , filtration and flotation using  $\text{ZnCl}_2$ ). The fragments previously added were recovered with efficiency  $> 98\%$ , and correctly identified using FTIR, thereby showing that (micro)bioplastics were resistant to the chemicals used for the separation and that the method of  $\text{ZnCl}_2$  flotation displayed efficiencies comparable with those reported elsewhere for conventional plastics (Rodrigues et al., 2020b). This validation procedure was repeated three times. The infrared spectra of the commercial biodegradable materials used in this work are included in Fig. S2 (SM) together with the identification of the main peaks. The absorptions corresponding to starch-based materials are clearly observed and attributed to thermoplastic starch, which is widely used for the manufacture of commercial biodegradable plastics in compounds with different copolymers and fillers.

#### 2.6. Contamination control

Several measures were taken to avoid the potential contamination of samples with plastic materials during sampling and laboratory handling. Sample collection was performed by a single person, which used non plastic tools. The samples were quickly introduced in close aluminium bags, labelled, and shipped to the laboratory. All material used were previously cleaned carefully with pure water. During laboratory handling only glass and steel material was used which was previously cleaned with ultrapure water at least three times. All solutions used for filtering and density separation were prepared with ultrapure water and filtered through  $0.45 \mu\text{m}$ . Glassware was also cleaned with ultrapure water and then heated at  $450^\circ\text{C}$  for 4 h. Clean materials were always covered with aluminium foil also heated to  $450^\circ\text{C}$  for 4 h to remove all possible contamination with potentially interfering materials. In addition, laboratory clothes were made of cotton. During laboratory manipulation, contamination controls

consisted of 47 mm Petri dishes containing glass fibre filters, which were kept open near the workplace during all manipulation procedures. Most of the particles (30) that appeared in control filters were white cellulose fibres. The particles identified as plastics in procedural controls were one red polyamide fibre, one yellow acrylic fibre, three polyester fibres (two black and one transparent), and one red polysiloxane filament. The fibres or filaments with similar colour and typology found in the samples that corresponded to the controls with plastic were discarded and not included in the total number of plastic particles given below.

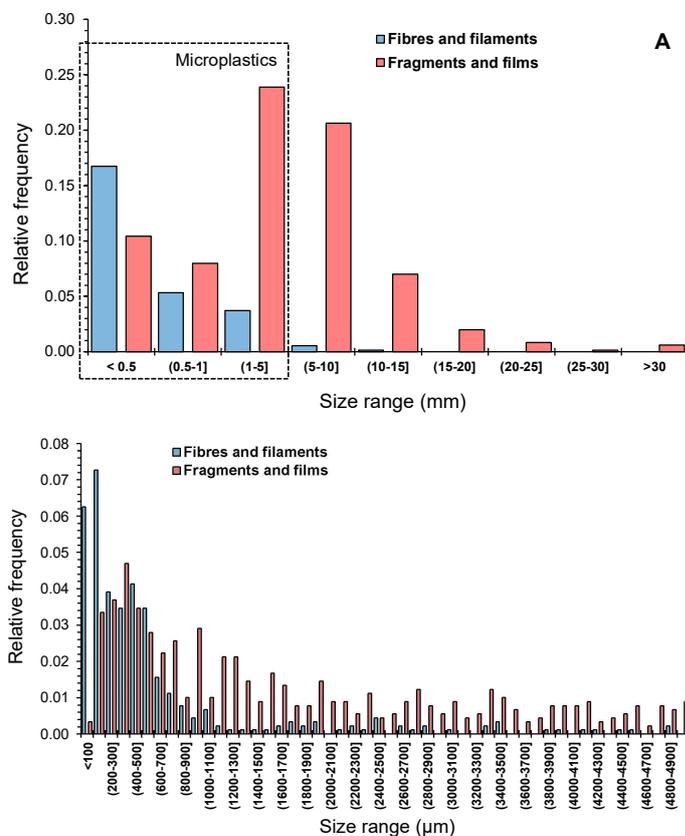
### 3. Results and discussion

#### 3.1. Abundance of plastic litter

The average total amount of compost processed per each plant was  $297.9 \pm 10.1$  g during the five-month study with an average of  $59.6 \pm 1.3$  g of compost per plant per month. The visual inspection of samples led to a total number of 10563 particles classified as potential impurities from which a sub-fraction of 1532 particles was analysed by ATR-FTIR or micro-FTIR depending on their size. The subsample size (15 %) was calculated to allow a maximum error  $< 2.5$  % in the estimation of composition with 95% confidence intervals, as shown elsewhere (Kedzierski et al., 2019). FTIR analyses identified 1357 particles consisting of synthetic polymers distributed among all typologies, which represented  $> 85$  % of the particles analysed. Overall, fibres represented the dominant shape (42.7 %) followed by fragments (31.2 %), films (22.1 %) and filaments (3.9 %). Fragments and films were treated together as well as fibres and filaments because of the methodology used to calculate equivalent diameters from projected images. For fragments and films, the equivalent diameter was that of the circle with the same projected area. For fibres and filaments, the equivalent diameter used was the aerodynamic diameter calculated for the average density of the most common polymers (Rosal, 2021). Fig. 1 shows the relative frequency of all plastic litter detected (Fig. 1A) and for the lower size fraction (Fig. 1B,  $< 5000$   $\mu\text{m}$ ). The same data disaggregated for the different plants are presented in Fig. S3 (SM).

The size distribution of plastic impurities was markedly dependent on their typology. While almost all fibres and filaments (97.4 %) were below 5 mm, only 57.6 % of particles and films were below that threshold. Plastic particles with equivalent diameter  $< 2$  mm represented 89.6 % and 37.7 % of the total amount of fibres-filaments and fragments-films respectively. These cutoffs are relevant because impurities in compost, which include metals, glass, and plastics, with size from 2 mm to 5 mm are legally limited in a number of standard methods for the assessment of compost quality. Sometimes this is done by establishing a maximum concentration for the sum of impurities with a given particle size, while some regulations specify

individual limits for films and other plastic materials (Saveyn and Eder, 2014). Fig. 1 shows that most plastic particles corresponded to the smaller size ranges with clear prevalence of MPs (equivalent diameters  $< 5$  mm). For the case of fibres, the most abundant sizes were in the range of several hundreds of microns or below.



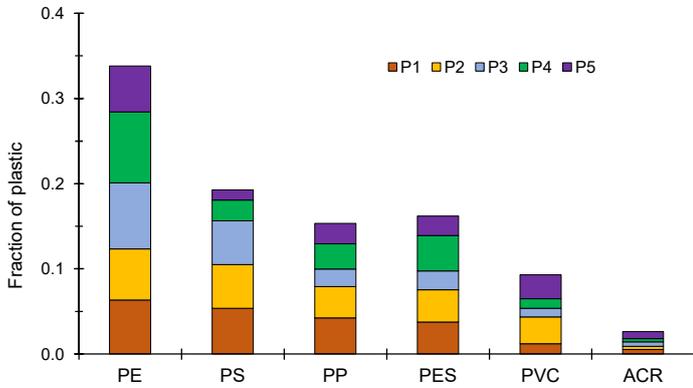
**Figure 1.** (A) Size distribution for the plastics sampled in this work (all plants, all samples) and (B) for the MPs (all plants, all samples,  $< 5$  mm). Size was calculated as equivalent diameter as indicated in the text.

#### 3.2. Chemical composition of plastics in compost

Chemical analyses (ATR-FTIR and micro-FTIR) allowed the identification of 16 different synthetic polymers or groups of polymers in 1322 particles. From the other 210 inspected particles that were not identified as plastics, 62 corresponded to cellulose-cotton and vegetal debris, and the rest to materials whose composition could not be spectroscopically assessed. About 40% of the cellulose-based materials displayed non-natural colours (black, red, blue, orange) evidencing some type of anthropogenic processing. This type of materials might be classified as pollutants as the industrial origin behind their non-natural colours suppose the presence of dyes and other industrial additives (Edo et al., 2020b).

The most abundant polymer was polyethylene (PE) followed by polystyrene (PS), polyester (PES), polypropylene (PP), polyvinyl chloride (PVC), and acrylic polymers (ACR, which include polymethylmethacrylate and acrylic fibers and films). These six polymers account for 96.7 % of the plastic

materials found and were the only ones exceeding 1 % of the total number of 1322 particles identified as plastics. PE was predominant in all plants and throughout the sampling campaign. Besides, samples from plants P1, P2, and P3 displayed a considerable amount of PS, and also PP and PES. Fig. 2 shows the fraction of the different polymers identified in all compost samples throughout the period under study.

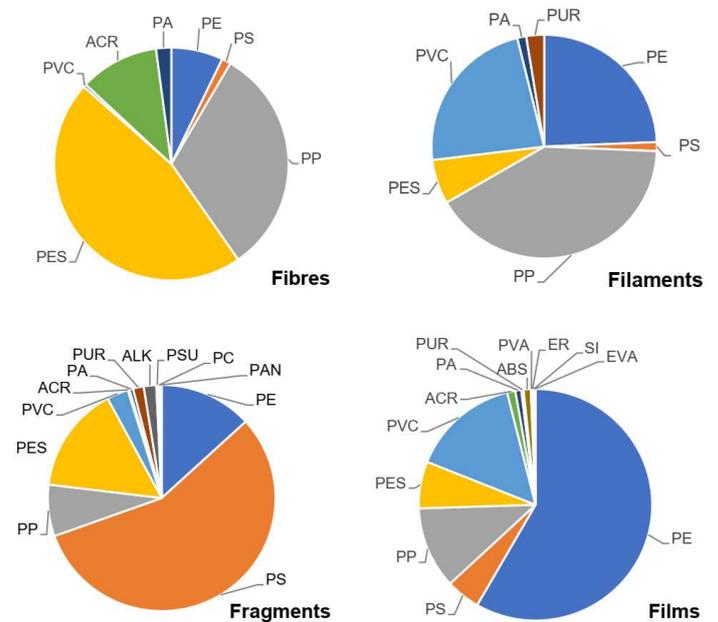


**Figure 2.** Main synthetic polymers found and their relative frequency (in unit fraction) for the different plants studied. PE: polyethylene; PS: polystyrene; (PES) polyester; (PP) polypropylene; (PVC) polyvinyl chloride; (ACR) acrylic polymers.

Apart from the most abundant plastics, other synthetic polymers were occasionally found. These were polyamide, polyurethane, alkyd resins, acrylonitrile butadiene styrene, polysulfone, polycarbonate, polyvinyl acetate, epoxy resins, silicone, ethylene-vinyl acetate and polyacrylonitrile, which, altogether accounted for the remaining 3.3 % of the specimens identified as synthetic polymers. Among the minor polymers, polyamide and polyurethane and alkyd resins, exceeded 0.5 %, which corresponded to less than six particles for type and per sample. No particles that could be attributed to biopolymers were found in any of the samples.

Polymer composition differed considerably among typologies. Fig. 3 shows the different materials found in the form of fragments, films, fibres, and filaments. Fragments were dominated by PS and films by PE, which corresponds with the major uses of those polymers in packaging. Most fibres were PES, PP, and acrylic, while filaments displayed a variety of polymers dominated by PP, PE, and PVC. PVC appeared occasionally as green colour filaments, probably due to its use as industrial yarns. PVC was also found as transparent films, which was attributed to its massive use for food wrapping. In the present study, PS was barely found in its expanded foam form. On the contrary, most of the fragments found corresponded to the material widely used in the food industry as yogurt container, fruit carrier, or other similar containers as a consequence of their proven food safety (Gelbke et al., 2019). Most fibres were PES, PP, and acrylic, typical

polymers used in textiles for clothing and other industries. Overall, the composition of the plastic debris found in this work corresponded to the usual polymer found in packaging materials like bags or food containers and were consistent with data reported elsewhere (Gui et al., 2021). The correspondence between typology and polymer type can be visualized in the PCA plot given in Fig. 4 (SM). Besides, some images of the plastic particles samples in this work are shown in Fig. S5 (SM).

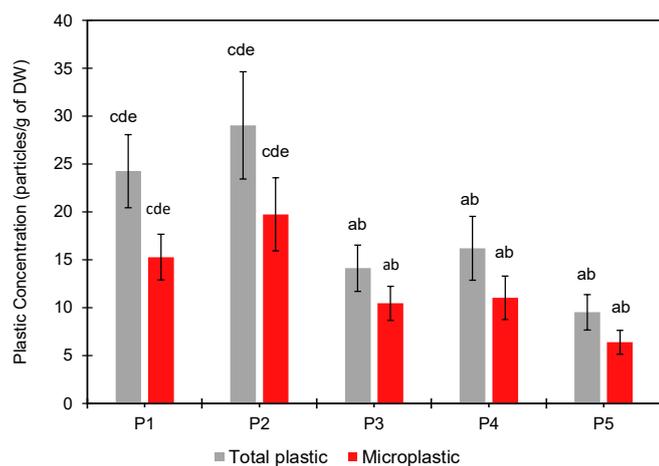


**Figure 3.** Composition of plastic particles by typology. PA: polyamide; PUR: polyurethane; ALK: alkyd resins; ABS: acrylonitrile butadiene styrene; PSU: polysulfone; PC: polycarbonate; PVA: polyvinyl acetate; ER: epoxy resin; SI: silicone; EVA: ethylene-vinyl acetate; and PAN: polyacrylonitrile.

Fig. 4 shows the total concentration of plastic particles and MPs are expressed in items per mass of dry compost. The results showed a considerable reproducibility with limited changes along the sampling period as evidenced by the relatively small differences between maximum and minimum values for plastic concentration along the five month sampling period. Plants P1 and P2 consistently displayed a concentration of plastics in the range of 20-35 particles/g of DW; P3 and P4 in the range of 10-20 particles/g of DW, while P5 was the one with the lowest plastic content, as low as 8-12 particles/g of DW (average  $9.5 \pm 1.9$  particles/g of DW). The overall fraction of MPs (with respect to the total number or plastic particles) was in the 63-74 % range, representing 6.4-19.7 MPs/g DW. The results showed that the majority of plastic impurities consisted of small particles, below the 5 mm threshold. The concentration data disaggregated per month and facility are shown in Fig. S6 (SM).

As indicated before, the results showed fibres were the main typology followed by fragments and films (Fig. S3). The data showed that fibres and filaments were

predominant in plants P2 and P5 (> 50 %), while fragments and films slightly outnumbered fibres and filaments in plants P1 and P3. The data on the different typologies showed a consistent tendency to increase in the number of fibres (from 6.3% in M1, to 10.4% in M6) along with a reduction in the number of fragments during the same period (8.0% to 5.0%). The data, split into months, plants and for these two main typologies are shown in Fig. S7 (SM). This tendency was not clearly accompanied by a change in the fraction of the different polymers as shown by Fig. S8 (SM), which represents the fraction of PES in fibres and of PS in fragments and might be due to seasonal variations.



**Figure 4.** Concentration of plastic particles per g of compost (DW, dry weight) representing total concentration (grey) and microplastics (red, < 5 mm). Different letters indicate significant differences ( $p < 0.001$ , ANOVA, Bonferroni test).

Different studies have reported the presence of MPs in soil environments because of the use of wastewater sludge as fertilizer (Edo et al., 2020a; Li et al., 2018). The data available suggest that wastewater sludge could release tens of thousands of MPs per kg of dry soil when dispersed in agricultural lands (Zhang and Liu, 2018). The use of compost obtained from biowaste composting processes for soil amendment is another way of entry for MPs to the environment, which deserved much less attention. MPs have been reported in compost samples from several countries up to thousands of items per kg (van Schothorst et al., 2021; Vithanage et al., 2021). Weithmann et al. (2018) demonstrated that fertilizers from composting facilities contained 70-122 MPs/kg of dry product in plants using aerobic and anaerobic treatment of OFMSW respectively. The study, however, was limited to particles > 1mm (Weithmann et al., 2018). Another recent study on the presence of MPs in compost from organic rural domestic wastes found an average abundance (0.05–5 mm) of  $2.4 \pm 0.4$  MPs/g of dry weight compost (Gui et al., 2021). Our results showed a concentration of plastic particles in the 10-30 particles/g of dry weight compost, mostly < 5 mm (about 5-20 MPs/g), and 30 % of them < 1 mm, which

is a reasonably good agreement with Gui's findings in spite of the different origin of the raw biowaste.

Even if the presence of plastic in compost is well documented, the risk posed to agricultural soil and interconnected environments is still poorly known. MPs in compost are suspected to act as a carrier of toxic chemicals including metals, to influence the carbon cycle, to alter microbial distribution and to produce negative effects to soil biota. Besides, as strange materials in soil, they will possibly induce changes in soil physical, chemical, and biological properties. However, most of the existing studies on the environmental impact of plastic debris performed so far showed no risk or very limited risk for short term exposure (Selonen et al., 2020). Judy et al. (2019) demonstrated that municipal wastes containing plastic had no effect to wheat seedling and biomass production as well as to earthworm growth, reproduction, and mortality even after up to 9 months of exposure (Judy et al., 2019). However, other researchers found significant effects, particularly when studying smaller plastic particles. Kim et al., (2020) exposed the nematode *Caenorhabditis elegans* to submicron PS particles and found significant offspring decrease for concentrations of 10 mg/kg of soil, which intensified in clay-rich soils (Kim et al., 2020). The higher plant *Vicia faba* suffered changes in enzymatic (catalase, superoxide dismutase, peroxidase) activity when exposed to PS MPs (5  $\mu$ m or smaller) and growth reduction when treated with 100 nm PS-NPs (Jiang et al., 2019). In sum, the long-term impact of plastics is essentially unknown, especially referring to the effect and potential accumulation in tissues of small size MPs and NPs.

Rodrigues et al., (2020) demonstrated that separated OFMSW collection systems could play a key role to control the contamination with non-compostable materials in biowaste processing plants (Rodrigues et al., 2020a). The contamination of OFMSW with non-compostable materials results in a high impact on compost quality in terms of the concentration of plastics and other debris, and this is true despite the plant engineering efforts adopted to remove them. It is desirable that the organic materials generated at home are completely free of non-biodegradable and compostable materials (i.e., conventional plastic, glass, textiles, metals). It is a well-known fact that door-to-door collection, rather than publicly available compost containers reduces the amount of impurities in compost and rejection rates in composting plants (Malamis et al., 2017). Our study showed lower rejection rates in plants with higher rate of door-to-door collection compared with combined collection systems that make use of street bin dedicated containers as shown in Table 1.

Our results allowed some insight into the actions that may help to control the presence of plastic impurities in OFMSW compost. Table S2 shows the covariance

matrix relating the concentration of plastics with other quantitative variables that represent the differences among plants. Most of the variance was explained by plant capacity, which in turn displayed significant autocorrelation with population density, but the fraction of door-to-door collection and the fraction of impurities were also significant ( $p$ -value < 0.05) to explain variability. Instead, the sampling month was not significantly explaining variance and was removed from the analysis. The PCA plot (Fig. S9, SM) expresses this information as a set of new variables, the principal components (PC). PC1 explains 73.7 % of variance (68.7 % considering only fibres) and is mainly influenced by plant capacity and door-to-door collection fraction. Besides Plant capacity and door-to-door collection are negatively correlated. PC2 explains 8.2 % (12.8 % for fibres) of variance and is mainly influenced by the presence of impurities and population density, which are uncorrelated. The effect of plant size and population density are probably explained by the sociodemographic characteristics of the population served, where more OFMSW are expected to be accompanied by higher plastic waste due to different lifestyle (Shittu, 2020).

It is widely accepted that more careful collection schemes and more stringent selection at the plant entrance, lead to lower plastic contents in the final product. However, our results showed that even for plants in less populated places with rigorous collection policies, plastic impurities, in particular small MPs, were still present. Plant P5 would disseminate  $4.8 \times 10^9$  MPs/yr and the five plants studied in this work would contribute to MP pollution with a total load of  $1.4 \times 10^{12}$  MPs/yr altogether. Besides, it is important to note that composting conditions combine temperature, humidity, and an aerobic environment, which are known to trigger plastic ageing and fragmentation as a consequence of mechanical stress, oxidation and abiotic disintegration (Gui et al., 2021). As it has been shown, the plastic fragments spread into the environment will continue to undergo ageing and fragmentation process to produce smaller fragments including NPs (Sorasan et al., 2021).

The analyses performed in this work demonstrated that bioplastics were completely absent from all compost samples analysed. In the specific case of P5, all bags accepted in door-to-door collection should be compostable (consideration supported also by the very low amount of impurities rejected by the process; see Table 1). The data provided by the analyses at the entrance of all plants (Table S1, SM) indicated that compostable bags were found in most plants, reaching ~90 % of all bags in plant P5. However, we could not find any fragment of biodegradable bags or other biodegradable polymers in spite the efforts made in that direction. The case of P2 was special as it represented the only plant with an anaerobic digestion process followed by post-digestate composting. Fig. 4 shows

that P2 compost was the one with highest concentration of plastic debris, which could be explained by the pretreatment technology applied in this plant before OFMSW digestion. Anaerobic digesters are sensitive to improper materials so a high performance pretreatment is required to avoid failures in the digestion phase. These systems could be responsible for the mechanical fragmentation of plastics entering with OFMSW, which would appear later as contaminants in the final product. This explanation would require additional studies to be confirmed.

Our results highlighted the presence of plastic impurities in compost from OFMSW. The results from non-compostable waste shown in Table S1 has been made publicly available by the Agència de Residus de Catalunya (<https://sdr.arc.cat/>). The data in the additional information to each characterization file showed that most non-classified waste corresponded to a few types of waste. The most common are facemasks, coffee capsules, kitchen scourers and cleaning cloths, drug blisters, rests of food packaging and a variety of different plastic-containing objects including electronic devices. The presence of such macrowastes gives a clue on the origin of the small plastic debris obtained in this work, which can be mostly attributed to an improper waste management and separate collection, although certain contribution of in-plant contamination could exist (for example, due to atmospheric deposition) or cross-contamination through the use of bulking agents containing plastics. Measures should be taken to avoid the presence of MPs into the final compost in order to limit as much as possible their dispersion to soils. This goal could be achieved by limiting the use of non-compostable plastics in domestic uses like food packaging in favour of bioplastics. Another strategy could be the design of environmentally friendly plastic goods, easy to sort and classify at home by the end-users.

#### 4. Conclusions

The presence of plastic debris in compost obtained from OFMSW was studied by analysing samples of final compost taken over a five-month period from five different composting facilities representative of different collection systems, rates of impurities, and technology. From our study we concluded that: (1) The total concentration of plastic particles was in the 10-30 items/g of DW range; (2) The concentration of MPs was in the 5-20 items/g of DW; (3) Fibres were predominant and were mostly in the lower size range ( $25 \% < 500 \mu\text{m}$ ); (4) The plastic materials found were polyethylene, polystyrene, polyester, polypropylene, polyvinyl chloride, and acrylic polymers in that order of abundance; (5) Smaller plants, with door-to-door collection schemes produced compost with less plastic impurities; and (6) Compostable bioplastics are completely absent from compost even if there was

evidence that they arrived to the composting plants at least as biodegradable plastic bags.

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# Supplementary Material

## Microplastics identification and quantification in the composted organic fraction of municipal solid waste

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### Contents:

**Table S1.** Detailed characterization (in wt.%) of non-compostable materials identified at the entrance of all plants (Source: Agència de Residus de Catalunya).

**Figure S1.** Scheme of the steps followed for the separation and identification of plastic particles.

**Figure S2.** ATR-FTIR spectra of the bioplastics used as references in this work. MaterBi was supplied by Novamont and the remaining #1, #2 and #3 were certified materials obtained from local markets.

**Figure S3.** Size distribution for the plastics recovered from the different plants studied (P1 to P5, details in Table 2). Size was calculated as equivalent diameter as indicated in the text.

**Figure S4.** PCA plot for the relationship between typology and polymer composition.

**Figure S5.** Micrographs of some plastic particles sampled in this work.

**Figure S6.** Concentration of plastic particles per g of compost (DW, dry weight). Error bars represent plus/minus one standard deviation for the five samples taken from each plant. M1 to M5 represents months during the sampling period (February-June 2021).

**Figure S7.** Concentration of fibres (A) and fragments (B) for the different plants during the sampling period. M1 to M5 represent months during the period February-June 2021.

**Figure S8.** Unit fraction of PS in fragments (A) and PES in fibres (B) for the different plants during the sampling period. M1 to M5 represent months during the period February-June 2021.

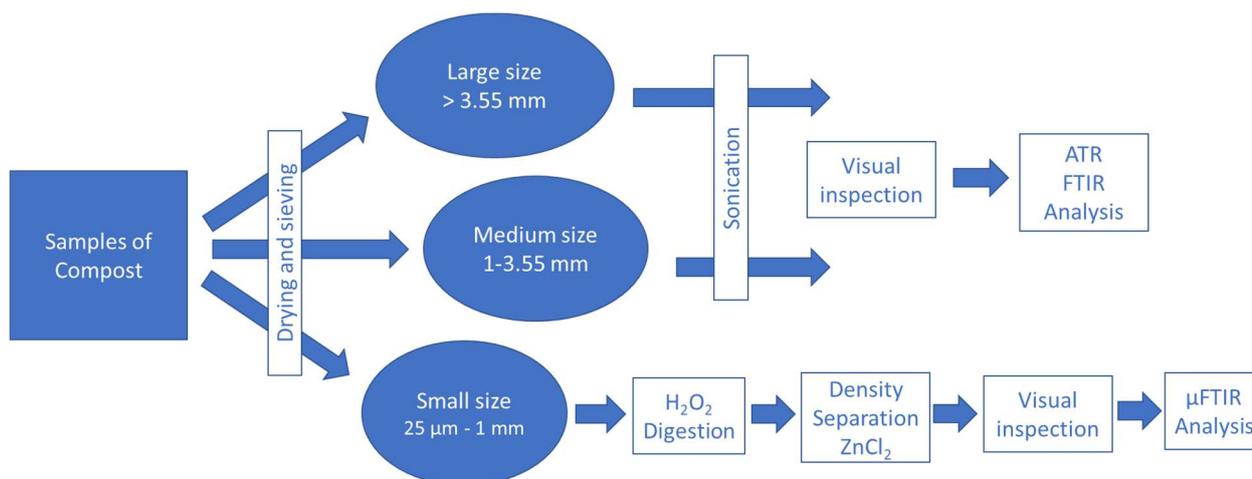
**Table S2.** Covariance matrix for the quantitative variables included in this study.

**Fig. S9.** PCA plot for the independent variables studied in this work.

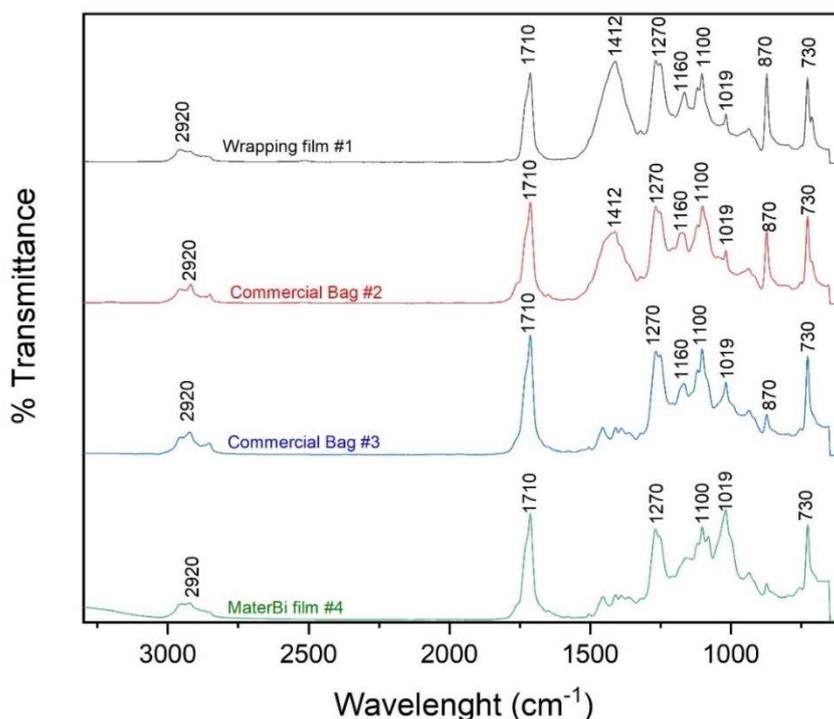
**Table S1.** Detailed characterization (in wt.%) of non-compostable materials identified at the entrance of all plants (Source: Agència de Residus de Catalunya).

	Samples	Glass	Paper & cardboard	Plastic items	Plastic bags*	Metals	Textiles	Total
P1	54	0.66	0.74	5.13	1.70	0.52	2.15	14.43
P2	74	0.23	0.41	1.13	1.07	0.11	0.67	4.43
P3	60	0.60	1.12	1.66	0.83	0.33	1.17	8.32
P4	10	0.99	1.01	2.93	0.91	0.52	1.90	10.03
P5	6	-	0.25	0.25	-	0.06	0.23	1.65

\* Estimation of the presence of compostable bags: P1 10-15%, P2 ~10 %, P3 15-20%, P4 very low, P5 ~90 %



**Figure S1.** Scheme of the steps followed for the separation and identification of plastic particles.

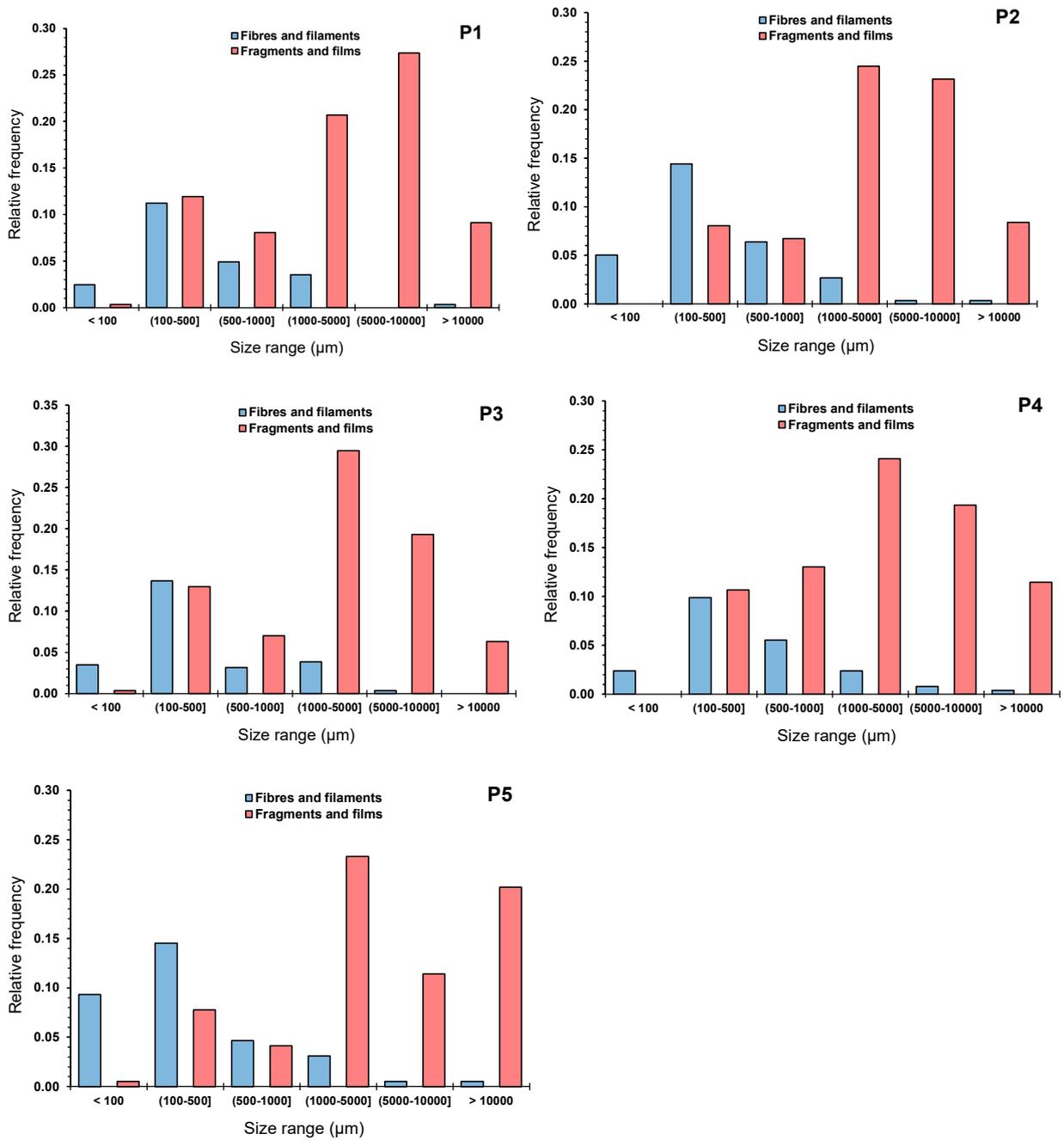


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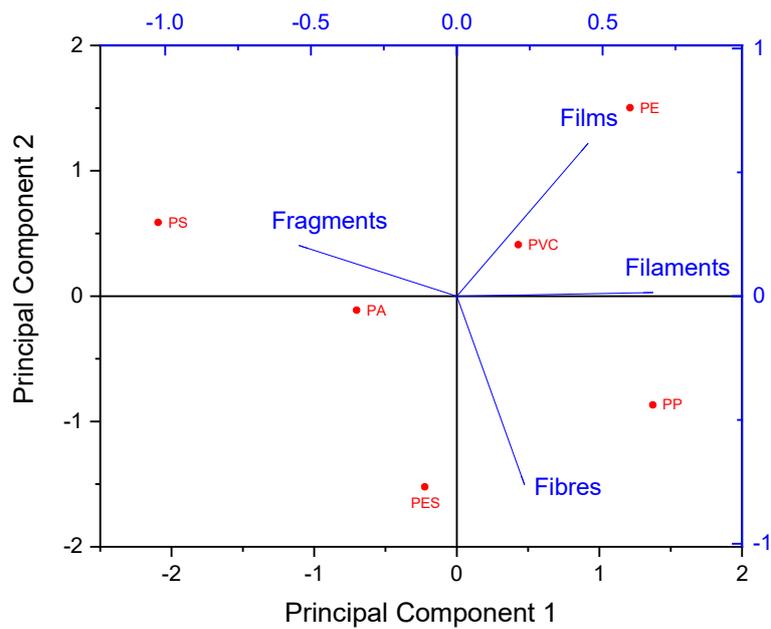
The peaks corresponding to starch-based materials are clearly observed and attributed to thermoplastic starch, which is widely used for the manufacture of commercial biodegradable plastics in compounds with different copolymers and fillers. Accordingly, the spectra of all materials shared some common features. The bands in the 2970–2850  $\text{cm}^{-1}$  region corresponded to the asymmetric and symmetric stretching of C–H aliphatic alkyl chain bonds. The carbonyl peak (C=O stretching) was clearly observed at 1710  $\text{cm}^{-1}$ . The broad band at 1412  $\text{cm}^{-1}$ , predominant in two of the samples (#1 and #2 in Fig. S2), was attributed to the bending and wagging deformation of C–H bonds. The stretching vibration in 1160, 1100, 1019 and 870  $\text{cm}^{-1}$  corresponded to C–O stretching vibration, C–C deformation, and the out-of-plane bending vibrations of C–H bonds. The bands at 1270  $\text{cm}^{-1}$  and 730  $\text{cm}^{-1}$  could be attributed to C–O stretching of ester bonds, and the out of plane deformation of the aromatic ring, probably from PBAT (Nainggolan et al., 2013; Aldas et al., 2020).

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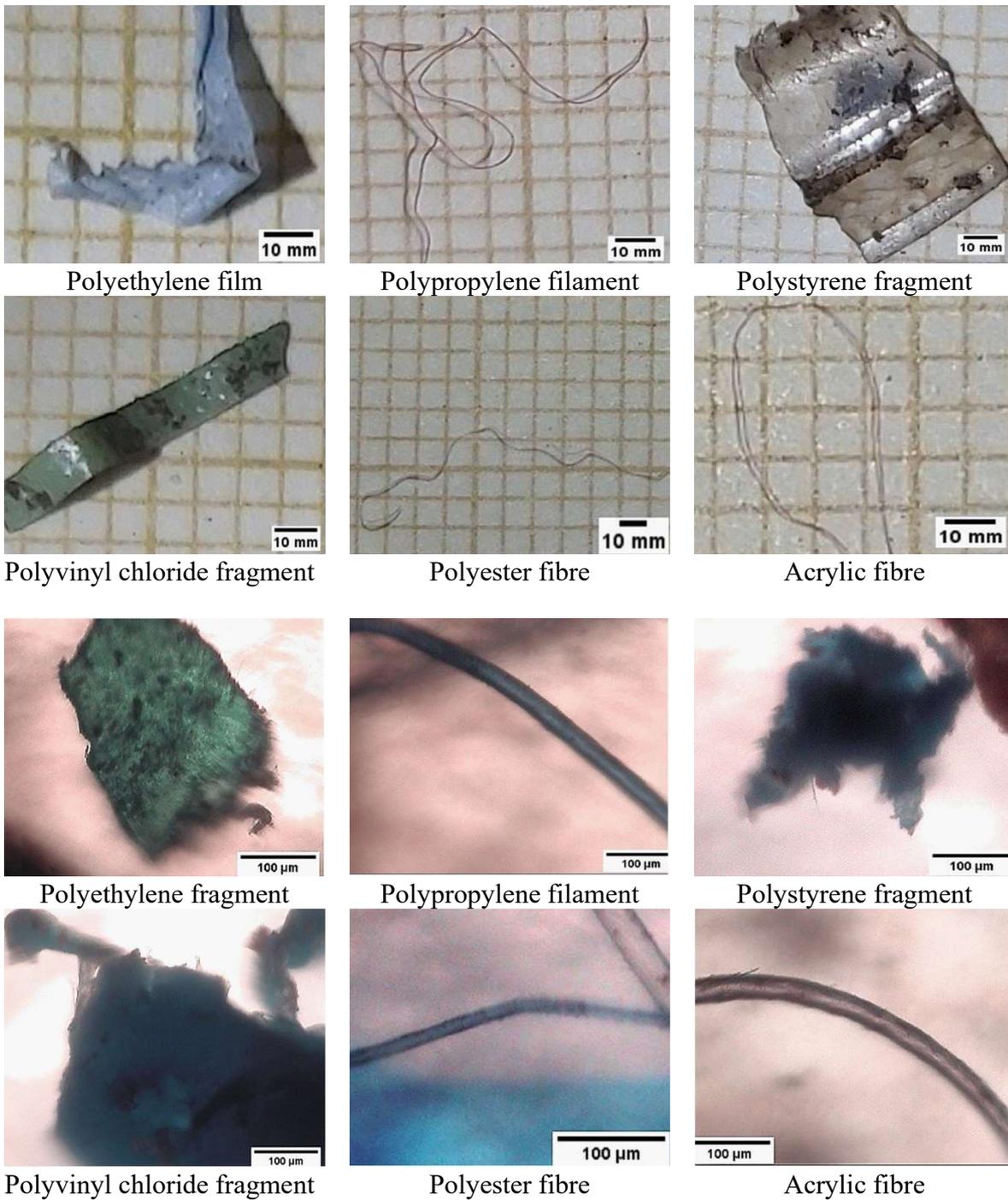
Nainggolan H, Gea S, Bilotti E, Peijs T, Hutagalung SD. Mechanical and thermal properties of bacterial-cellulose-fibre-reinforced Mater-Bi(®) bionanocomposite. *Beilstein J Nanotechnol* 2013; 4: 325-9.



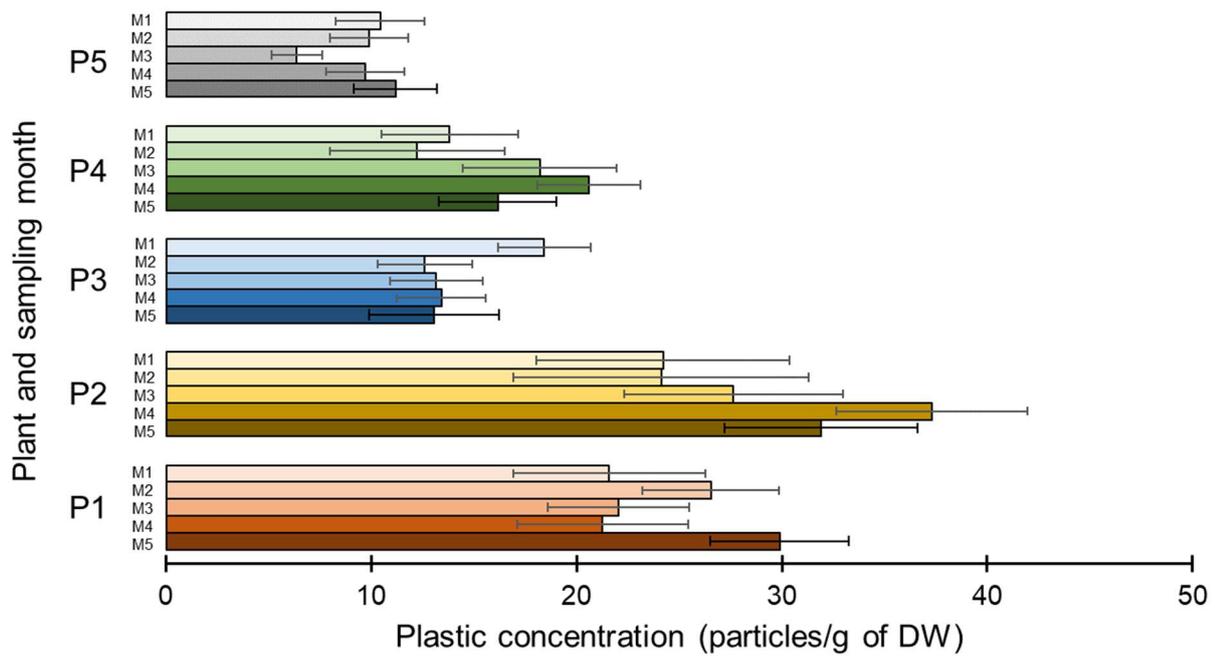
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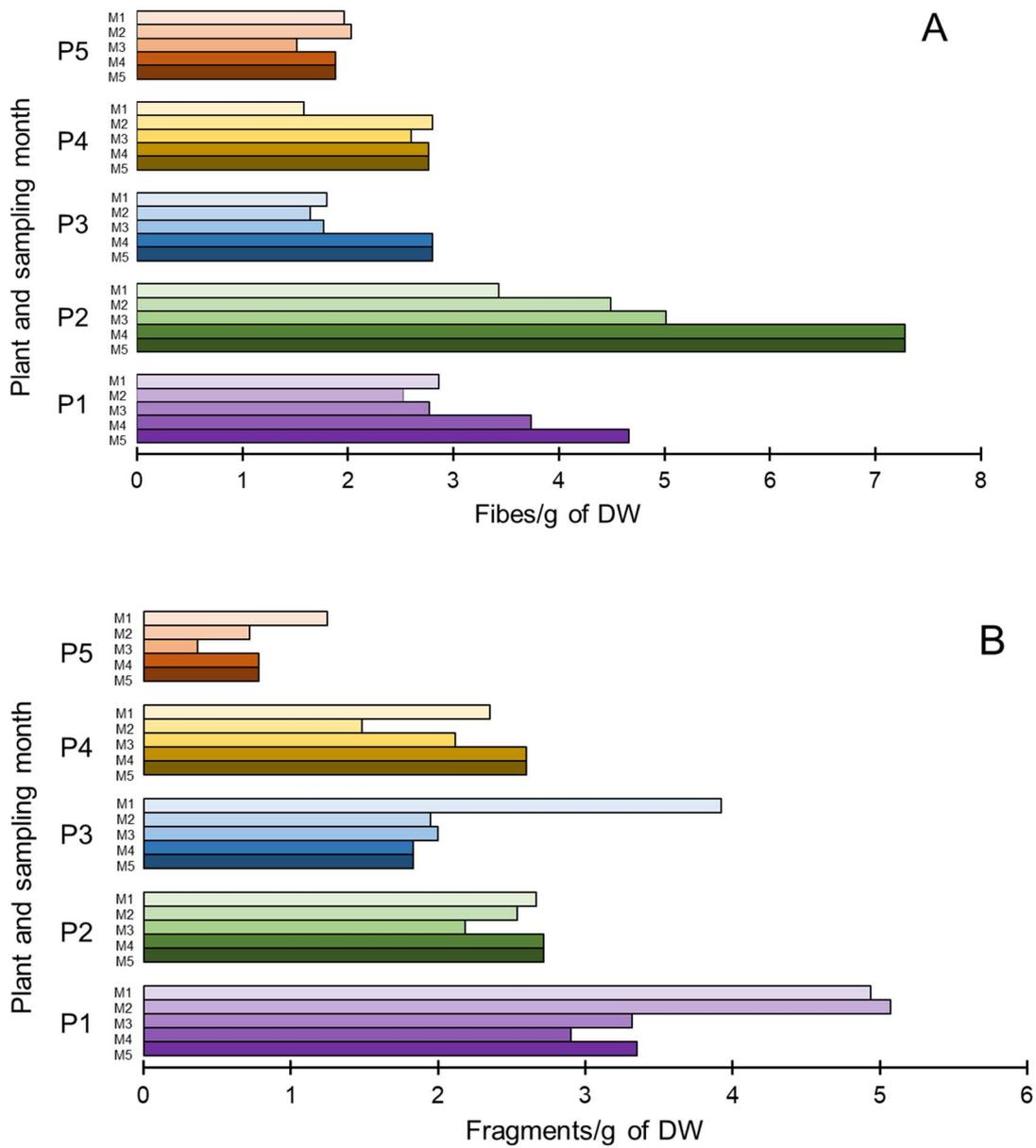
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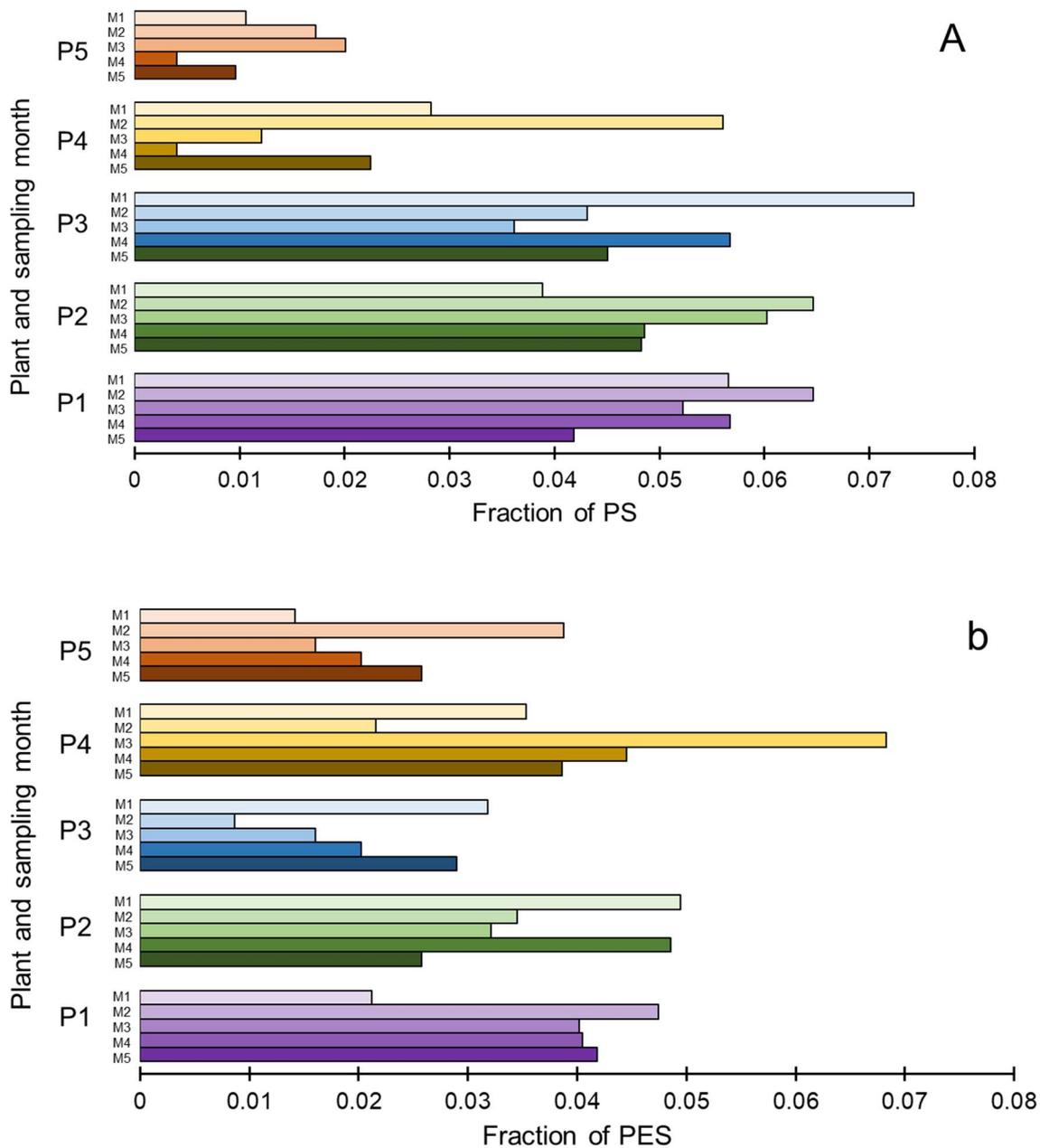
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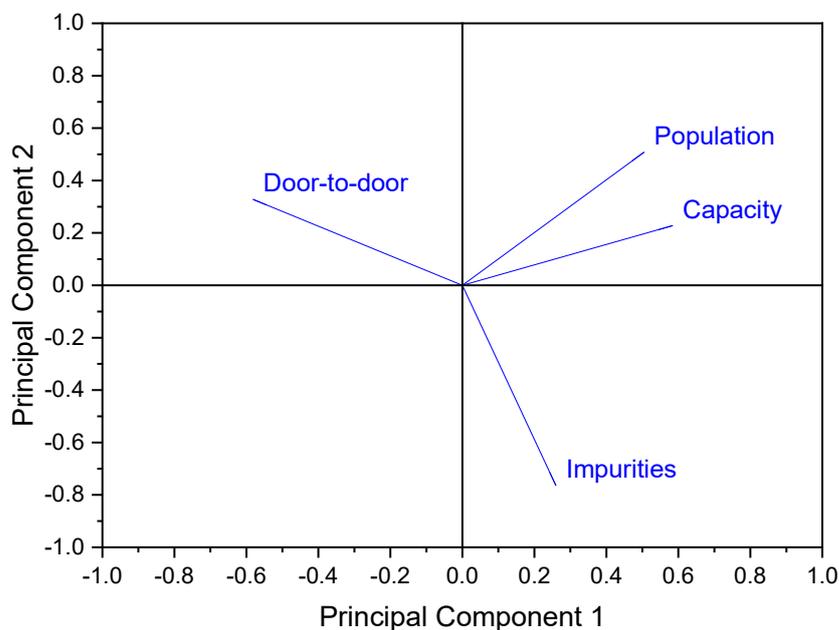
**Table S2.** Correlation matrix for the quantitative variables included in this study.

(all plastics)

	<i>Plastic</i> (particles/g)	<i>Capacity</i> (tonnes/yr)	<i>Door-to-door</i> (fraction)	<i>Rejection</i> (fraction)	<i>Population</i> (inhab./km <sup>2</sup> )
Plastic	1				
Capacity	0.84246	1			
Door-to-door	-0.66939	-0.60043	1		
Rejection	0.37480	0.13939	-0.69715	1	
Population	0.51383	0.76160	-0.43647	-0.31511	1

(only fibres and filaments)

	<i>Plastic</i> (particles/g)	<i>Capacity</i> (tonnes/yr)	<i>Door-to-door</i> (fraction)	<i>Rejection</i> (fraction)	<i>Population</i> (inhab./km <sup>2</sup> )
Plastic	1				
Capacity	0.97445	1			
Door-to-door	-0.52459	-0.60043	1		
Rejection	0.16817	0.13939	-0.69715	1	
Population	0.63732	0.76160	-0.43647	-0.31511	1



**Fig. S9.** PCA plot for the independent variables studied in this work.