Plastic in compost: Prevalence and potential input into agricultural and horticultural soils

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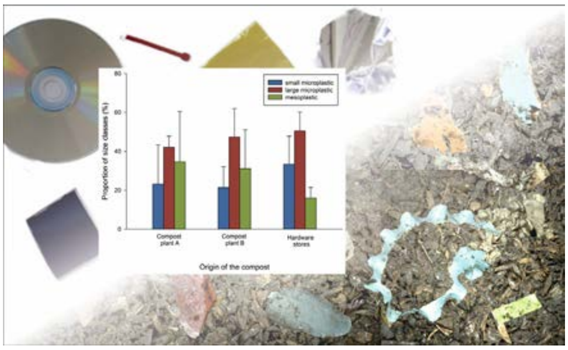
HIGHLIGHTS

- Plastic load of different compost types was analyzed.
- Plastic loads of compost were highly variable.
- Microplastic in the form of fragments was dominant particle type.
- Microplastic contributed only marginally to total plastic weight.
- Composition in compost differed markedly from that reported in sewage sludge.

GRAPHICAL ABSTRACT



ABSTRACT

To maintain and improve soil fertility, compost application is a widely recommended practice. We hypothesized that this practice is, however, also a main entry path for plastic into soil. Hence, we i) quantified the prevalence of plastic in eight composts from different composting plants and hardware stores to derive estimations about related plastic inputs into soil, and ii) characterized the properties of these plastic residues in regard to size and shape for further risk assessment. Plastic remains were analyzed via density separation ($ZnCl_2$) and light microscopy. Testing this method recovered 80 ± 29% of spiked plastic items. Applying this method revealed that all composts contained plastic particles in detectable amounts, with contents ranging from 12 ± 8 to 46 ± 8 particles kg^{-1}, corresponding to calculated plastic weights of 0.05 ± 0.08 to 1.36 ± 0.59 g kg^{-1}. Because of this high variability, an a priori discrimination of plastic loads between compost types cannot be achieved. Upscaling these loads to common recommendations in composting practice, which range from 7 to 35 t compost ha^{-1}, suggest that compost application to agricultural fields goes along with plastic loads of 84,000 to 1,610,000 plastic items ha^{-1} per year (a), respectively, amounting to 0.34 to 47.53 kg plastic ha^{-1} a^{-1}. Large potential inputs should thus also occur for horticultural soils, where application rates of compost usually vary between 6.48 and 19.44 t ha^{-1}, therewith resulting in a minimum plastic contamination of 77,770 plastic items and 0.31 kg plastic ha^{-1} a^{-1}, but a maximum amount of up to 894,240 plastic items and 26.4 kg plastic ha^{-1} a^{-1}. We conclude that compost application must be considered as potential source of plastic for both agricultural and horticultural soils, and technical solutions are needed to minimize these contamination risks while continuing this practice as important option to secure soil health.

Keywords:
Microplastic
Mesoplastic
Green waste
Environmental pollution
Synthetic polymers

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1. Introduction

Plastic is widely recognized as an environmental hazard and has been found in marine and fresh water environments all over the world. In contrast, the prevalence of plastic in agricultural and horticultural soil and especially its entry paths are relatively unknown (Rillig, 2012; Bläsing and Amelung, 2018). As recent studies already determined plastic in compost with up to 1.20 g plastic kg^{-1} (Gajst, 2016; Bläsing and Amelung, 2018), compost must be considered as plausible entry path for plastic into agroecosystems. This was recently confirmed by Watteau et al. (2018) who identified plastic in compost amended fields. To the best of our knowledge, however, up to now the range of plastic loads in different compost types and the contribution of small microplastics to these loads remained unknown.

In circular bioeconomy, the use of organic wastes like compost as fertilizer and soil amendment is an important practice to improve or maintain soil fertility parameters, like soil organic carbon stocks, water holding capacity and soil nutrient contents (summarized in Hargreaves et al., 2008; Diacono and Montemurro, 2010). Accordingly, composts from a variety of source materials have been produced for use in agriculture, including bio waste from households, green cuttings from commercial, garden, and park sites as well as from waste from agriculture and food processing (UBA, 2015). Related to the duration of composting, composts are divided into fresh composts (2 6 weeks, lower degree of rotting) and finished compost (5 12 weeks of composting, higher degree of rotting; VHE (2020). In the European union, for example, about 18 million (Mio.) t of compost were produced in 2008, which were applied to agricultural as well as horticultural soil, and these application rates are expected to increase (ARCADIS, 2010). If compost contains plastic, however, it must be considered as possible entry path of plastic into soils.

Plastic in compost has already been recognized as problematic, but this attention referred mainly to larger items for cosmetic reasons. Plastic mainly enters compost by improper waste disposal and littering, as well as by the usage of conventional plastic bags for biowaste collection, the latter relate to compost produced from biowaste (Bläsing and Amelung, 2018; UBA, 2015). Accordingly, compost plants reduce the plastic amount in compost by several procedures, including manual sorting and sieving before and after composting. Nevertheless, plastic can still be found in the final product and is tolerated by most fertilizer regulations to a certain extent. For example, in Germany, despite having one of the strictest regulations worldwide, plastic is considered as foreign matter in the so called "Düngemittelverordnung" (engl. fertilizer ordinance; DüMV, 2017). Here, up to 0.5 wt% foreign matter of particles >2 mm are allowed; non degradable synthetic materials can make up 0.1 wt%. Smaller items, however, are not considered and accordingly almost nothing is known about the abundance of such plastic items in compost and thus also in soil.

Up to now, as far as we know, only three studies investigated plastic in compost. These studies illustrated that concentrations varied widely, ranging between 2.4 and 120 mg plastic kg^{-1} and 20 to 24 plastic items kg^{-1} compost (Gajst, 2016; Bläsing and Amelung, 2018; Weithmann et al., 2018). Bläsing and Amelung (2018) screened compost from bio waste, green cuttings and structure compost and found highest concentrations in compost of biowaste from households, leading to the assumption that the raw material mainly influences the plastic concentration of compost. However, the authors analyzed only one compost per compost type. The authors also stated that the detected concentration must be considered as minimum estimate since only visible plastic items, i.e. mesoplastic (5 25 mm) and macroplastic (>25 mm) have been analyzed. Weithmann et al. (2018), in contrast, quantified also plastic items in the size of large microplastic (1 5 mm) as well as larger items in two composts of one compost plant, contents ranged from 20 to 24 plastic items kg^{-1} compost. Both studies analyzed only a small number of composts from compost plants while compost from hardware stores, which might be a relevant source of plastic in private gardens, had been neglected. Further, until now, an analysis of small microplastic (<1 mm) in compost is still missing. Hence, a comprehensive study about plastic abundance in different types of compost is urgently needed.

Beside uncertainties in the contents, also the composition of plastic in compost, i.e. the size and shape of plastic items, is not known. The shape and size of items significantly determine the environmental fate of plastic in the environment: particularly smaller items in form of fibers, for instance, may interact with soil constituents and can be more easily leached and degraded than larger fragments (e.g. McGechan and Lewis, 2002; Souza Machado et al., 2018). Zubris and Richards (2005) detected plastic fibers in soil under sewage sludge application and found evidence for vertical movement of these fibers. In contrast, Weithmann et al. (2018) found mainly fragments in the two compost types they investigated, suggesting that the shape of plastic in compost differs from that found in sewage sludge. If this also holds true for other compost types remains to be clarified.

The aims of this study were to elucidate the role of compost as plastic source for agricultural and horticultural soils. In detail, we hypothesized that differences in compost production (like raw material, duration of composting process and treatments to remove plastics in compost plants) as well as the origin of compost (compost plants and hardware store) affect the prevalence of plastic in compost. Further we assumed that the composition of compost, i.e. the shape and size of plastic in compost, differs from that of other potential plastic sources, like sewage sludge. Composts were obtained from different compost plants and hardware stores, including compost produced from green cuttings and biowaste as well as fresh and finished compost. The plastic items were determined via density separation and light microscopy.

2. Materials and methods

2.1. Plastic and compost/soil materials for method testing

Prior to quantifying plastics in compost, we conducted recovery experiments to test our extraction method. We spiked plastic items of different type and size with known amount to i) top and subsoil taken from a Luvisol (C_{org} content: 3.1 and 0.29%, respectively; located at Campus Poppelsdorf of the Rheinische Friedrich Wilhelms University of Bonn; 50°43′30.4″N, 7°05′07.0″E), as well as ii) compost, which was purchased at a local hardware store. Soil and compost samples were not checked for plastic residues before recovery tests, instead we added known amounts of plastic particles (known size, shape and color) and considered only these particles for calculation of recovery in our experiments. Primary plastic material was derived from household waste and items purchased at a local hardware store (Photo S1, Table S1), including polyamide (PA), polycarbonate (PC) low density polyethylene (PE LD), high density polyethylene (PE HD), polyethylene terephthalate (PET), polypropylene (PP), polystyrene (PS) and polyvinyl chloride (PVC). Primary plastic material was cut into three different size classes: mesoplastics (5 25 mm) as well as large (1 5 mm) and small microplastic (<1 mm). The size of items was checked via digital video microscope (DVM, Keyence VHX 1000 model, VH Z 20R lens). For these recovery experiments, 500 g of soil and compost were spiked with four particles of every size class of four plastic types in 4 fold replication.

2.2. Compost substrates

After finalizing the method checks (Section 2.1), eight different compost were analyzed in fivefold replication for plastic contamination: three composts were obtained from local hardware stores and five from regional composting plants (Table 1, for organic matter contents of compost see Table S2).

Table 1
Origin, raw material and code of analyzed compost types.

Origin	Description	Raw material	Code
Plant A	Municipal bio-waste derived fresh compost, sieved (0–20 mm)	Biowaste from households	PlantA-Bio-e-20
	Municipal green waste finished compost, sieved (0–20 mm)	Municipal green cuttings	PlantA-GC-i-20
Plant B	Municipal green waste fine finished compost, sieved (0–10 mm)	Municipal green cuttings	PlantB-GC-i-10
	Municipal green waste finished compost, sieved (0–20 mm)	Municipal green cuttings	PlantB-GC-i-20
	Municipal green waste fresh compost, sieved (0–20 mm)	Municipal green cuttings	PlantB-GC-e-20
Hardware store	Commercial garden compost ('Compo Bio Gärtnerkompost')	Municipal green cuttings	HS-GC-I
	Commercial garden compost ('Obi Living Garden Bio Naturkompost')	Municipal green cuttings	HS-GC-II
	Commercial raised bed compost ('Knauber Hochbeet-Kompost torffrei')	Municipal green cuttings	HS-GC-III

The description of the code contains information regarding the origin (plant or hardware store, HS), the raw material (green cuttings, GC, or biowaste, BIO), and the type of compost (fresh, e, or finished, i) as well as the sieving size of compost from compost plants (10 or 20).

2.3. Extraction

Extraction of plastic was conducted via density separation using $ZnCl_2$ solution as previously described by, e.g., Liebezeit and Dubaish (2012) and Imhof et al. (2013). In detail, 500 g of soil and compost (dry weight) was used for the recovery experiments, and for monitoring, 200 g dry weight compost was carefully homogenized with a ceramic pestle and mortar and thereafter sieved into three size fractions of >5 mm (mesoplastic), 1–5 mm (large microplastic), and < 1 mm (small microplastic). In the fraction >5 mm, we isolated the plastic items manually with a pincer, cleaned them in an ultrasound bath (Bandelin Sonorex RK 102), dried them at 40 °C and weighed them. The two smaller microplastic fractions were then subject to density separation. For this purpose, we prepared the density solution using distilled water and technical grade zinc chloride powder ($ZnCl_2$, UN 2331 zinc chloride, unhydrous, VWR Life Science, Radnor, USA) to obtain a density of 1.8 kg L^{-1}. Sieved compost material was then filled into a 500 mL amber glass reaction bottle, and 400 mL of the $ZnCl_2$ density solution was added. Samples were shaken for 30 min using a horizontal shaker (GFL analogues back and forth shaker, type 3018) at a speed of 200 rpm. Afterwards the sample was poured into a 1000 mL glass beaker to allow heavier particles to settle for a period of 2 h. After this sedimentation process, the supernatant at top of the solution was vacuum filtrated (vacuumbrand vacuum pump ME 2C NT) using moistened quartz filter (Macherey Nagel, MN QF 10, diameter 47 mm and 125 mm, particle retention: 0.3 μm). Extraction residues on the filter were rinsed with distilled water to remove remaining $ZnCl_2$ density solution and dried; to avoid contaminations the filters were then placed in a glass container. We then subjected filter including sample to a digital video microscope (DVM, Keyence VHX 1000 model, VH-Z 20R lens) for digital counting of plastic particles. The plastic particles were photographed, then separated from the sample with a fine tweezer and placed in a container for further analysis. We identified the particles as plastic by their color, shape and elasticity. We chose a light microscope for plastics analysis because FT-IR does not work for plastic particles <1 mm (Weithmann et al., 2018) and the autofluorescence of the organic matter prevents the detection of plastic signals by Raman spectroscopy (own unpublished data).

To avoid systematic errors all analyses were conducted by one and the same person, carrying nitrile gloves and a lab coat during the whole sample treatment (clothing made from synthetic fiber was avoided). In addition, most work was conducted under a fume hood to minimize contamination risks. During extraction the samples were covered with aluminum foil whenever possible. After extraction the samples were stored in glass containers. Laboratory blanks were processed with each batch of samples. Blanks showed a maximum contamination of 2 plastic items, mainly as fibers of 0.3 to 2 mm length. Accordingly, plastic contamination during lab work could be neglected.

2.4. Attempts to separate and remove organic matter

For the recovery experiments, we tested ultrasound treatment before sedimentation process to ensure better separation of plastic items and adhesive organic and mineral material. For this purpose we used 60 J ml^{-1} energy input (Branson Digital Sonifier W 250) as suggestion for careful destruction of macroaggregates (Amelung and Zech, 1999). To avoid formation of secondary microplastic, we did not test higher sonification levels. To test whether the separation process could be improved by removal of organic matter, we treated selected samples with 30% H_2O_2 solution and left them to stand for up to 48 h, with progress being checked in regular intervals. To maximize efficiency of this organic matter destruction, the H_2O_2 treatment was conducted after density separation and filtration.

2.5. Calculation of plastic masses

To translate the number of plastic items into masses, i.e. a concentration unit, we calculated concentrations (g plastics kg^{-1} compost) for mesoplastic after cleaning and weighing of the isolated plastic items. For smaller items, i.e. small and large microplastic, this procedure could not be used without destroying plastic items. For such small items we thus calculated the mean volume of plastic particles for each compost type (Table S4). Considering the lowest and highest plastic density (0.85 g cm^{-3} for PP and 1.45 g cm^{-3} for PET and PVC) and the number of plastic particles found, we then estimated a minimum and maximum plastic concentration of small and large microplastic in each compost type. We are aware that this is only a rough estimation, and consider it like this in the further discussion. However, as available analytical methods fail to determine exact plastic concentrations for such small particles in compost (Weithmann et al., 2018) we wanted at least give a hint about the contribution of microplastic to plastic contamination of compost. Such estimations can also help to interlink number of particles and plastic masses detected by other studies.

3. Results

3.1. Method performance

The method test using sieving and afterwards density separation with $ZnCl_2$, yet without ultrasonic and H_2O_2 treatment, recovered, on average, 80.2 ± 27.9% of the added plastic items. The variation reflects that the overall recovery decreased with decreasing size of items (recovery of mesoplastic: 96 ± 12.2%, of large microplastic: 82.4 ± 22.8%, and of and small microplastic: 62.3 ± 32.9%). No difference was observed between soil and compost. However, recovery differed between plastic types. It ranged from 90.1 ± 23.8% for PE-LD to 69.3 ± 32% for PVC (Table S3). Especially small microplastic particles made of dark colored PS, PET and PVC were only halfway recovered, likely because they were overseen by the light microscope. Sieving, however, was effective in sorting plastic particles in the respective sizes, i.e. most plastic items found in the respective fractions fitted to the defined sieve size. Only in very few cases, especially for small microplastic items, also particles larger than 1 mm were found, in this case the length was exceeded in one dimension only (Photo 1).

Because especially organic matter hinders plastic analysis in organic rich materials like compost and soil (He et al., 2018) we tried in addition to density separation different attempts, i.e. ultrasonic treatment before the sedimentation process as well as H_2O_2 treatment after the sedimentation process to remove this fraction from the sample. However, both attempts were ultimately dismissed due to ineffectiveness. Sonification did not lead to enhanced dispersion of soil or compost macroaggregates, most likely because $ZnCl_2$ is a potent dispersion agent on its own. Instead formation of foam was observed, which is presumably caused by destruction of cells and accompanied releases of intercellular proteins (Imhof et al., 2012). The treatment with H_2O_2 lead to a visible alteration in color of the organic material, which may have simplified the separation process later on. However, it did not seem to reduce the volume of organic material and cellulose structures remained unaffected; in addition, also the color of plastic items was altered by simultaneous formation of foam. Accordingly, sonification as well as oxidation was not used for further analysis.

Surprisingly, the content of organic matter did not influence the recovery of added plastics in our method tests as the recovery of plastic did not differ significantly between compost (highest content of organic matter) and topsoil and subsoil (lowest content of organic matter). Instead, recovery depended on the size and especially the color of plastic. Nevertheless, with an overall recovery of 80.2 ± 27.9% our method performed acceptably well for the different plastic types tested. Also, laboratory blanks showed only marginal contamination with fibers. Consequently, we suggest that simple density fractionation and digital imaging is sufficient for reliable quantification of microplastic and larger plastic items in soil and compost, even without destroying organic matter.

3.2. Results of plastic assessment in compost

3.2.1. Number and weight of plastic items in compost

On average, composts contained 28 ± 14 plastic items kg^{-1} dry weight. The highest mean number of plastic items was found in commercial compost obtained from hardware store HS GC I (46 ± 8 items kg^{-1}); the fresh compost derived from municipal green waste (PlantB GC e 20) showed the lowest mean plastic content (12 ± 8 items kg^{-1}; Fig. S1; Supplementary data). This was also the only compost where only in 80% of samples plastic was found, in all other samples the detection frequency was 100%. Consequently, with exception of one subsample, all analyzed samples contained plastic materials (Fig. S1; Supplementary data).

The plastic contents of compost were highly variable within single compost types, pointing to large heterogeneity of plastic contamination (Fig. 1A C). Hence, neither substantial differences between plastic contents from different composting plants nor between plastic contents of composting plants and compost derived from hardware stores were found.

To translate the number of items into a concentration unit (g plastics kg^{-1} compost), we calculated concentrations for mesoplastic after cleaning and weighing of the isolated plastic items. The compost produced from biowaste (PlantA BIO e 20), which contained also the second highest number of plastic items (Fig. 1D), was most enriched in mesoplastic, i.e. it contained mesoplastic with concentrations of 1.35 ± 0.59 g mesoplastic items kg^{-1} at a detection frequency of 100% (Fig. 1D). This concentration was more than twice as high as the second highest plastic concentration (PlantB GC e 20: 0.62 ± 0.65 g

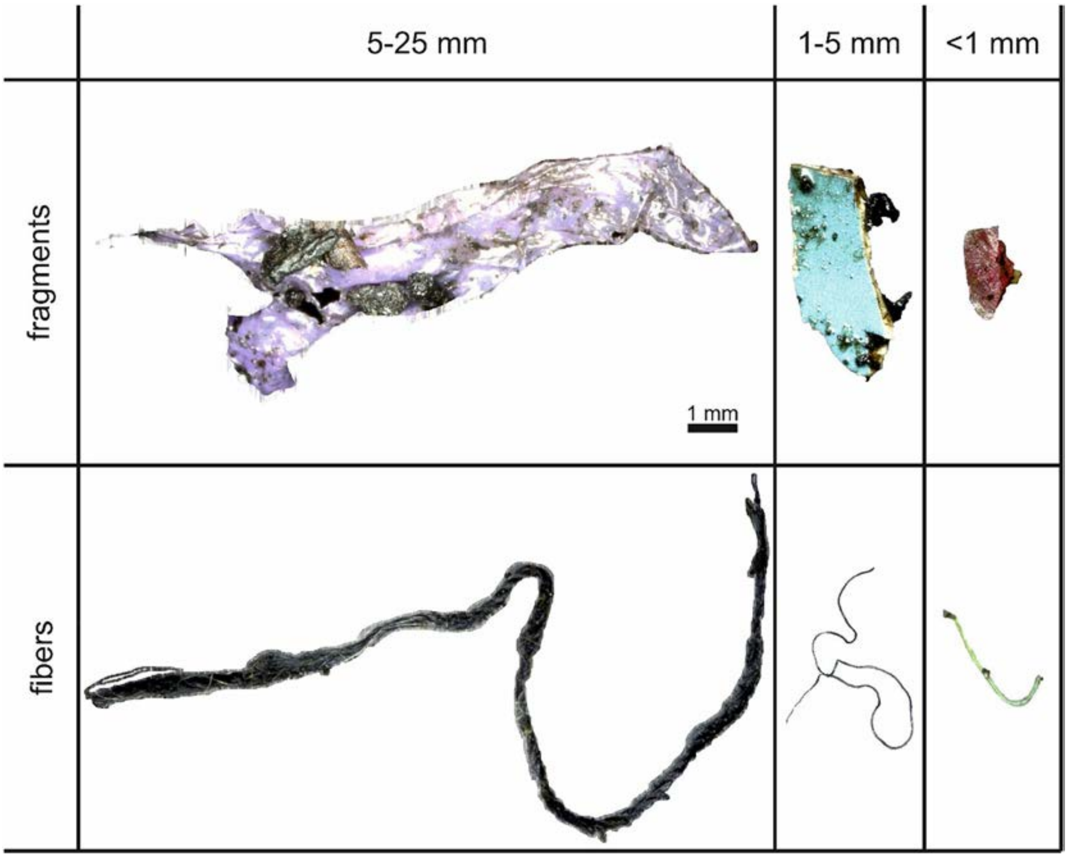

Photo 1. Examples for mesoplastic (5–25 mm) as well as large and small microplastic (1–5 mm and < 1 mm, respectively) in form of fragments and fibers found in compost.

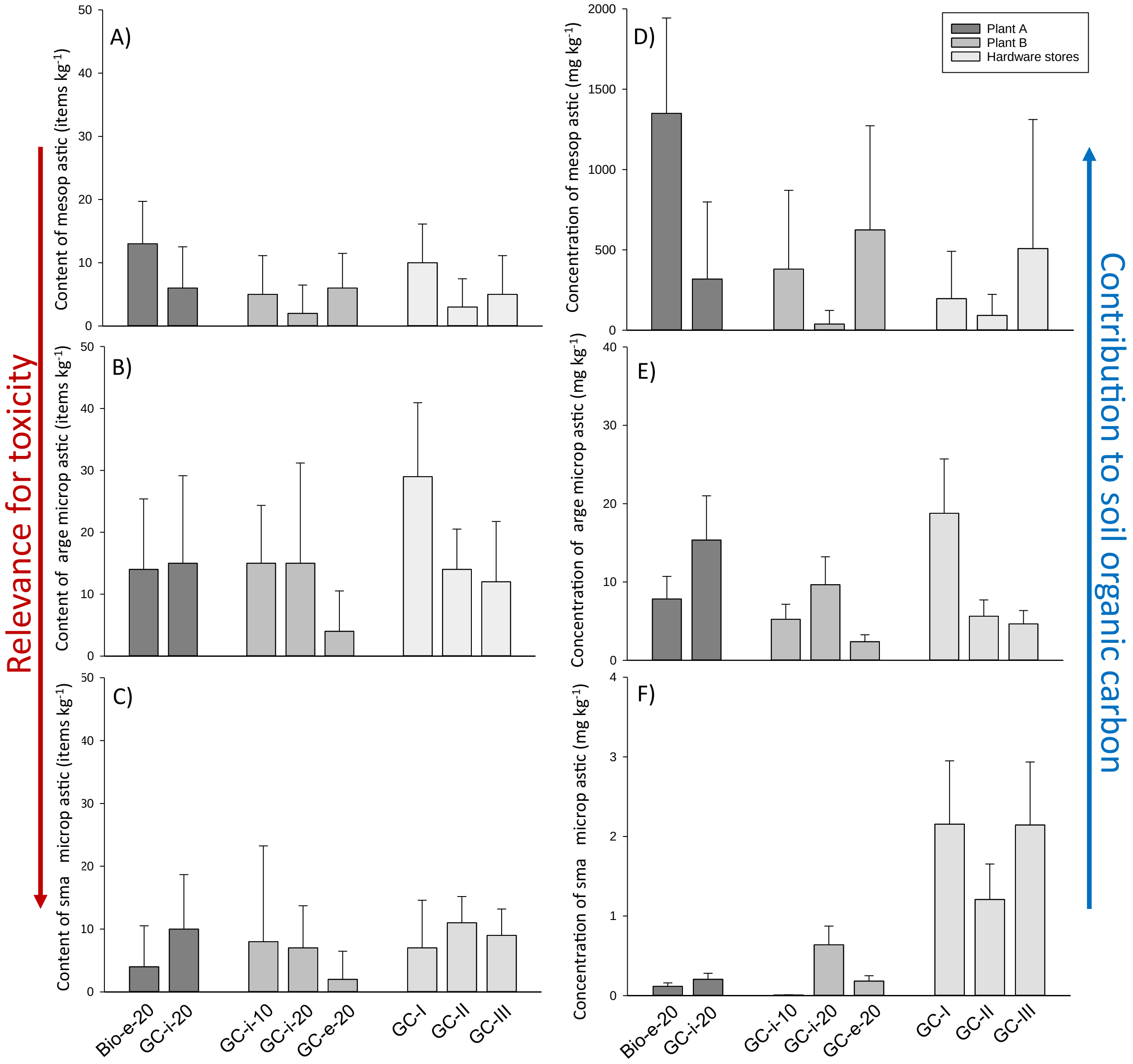

Fig. 1. Plastic content and concentration of mesoplastic (5–25 mm; A, D), large microplastic (1–5 mm; B; E) and small microplastic (<1 mm; C, F) of analyzed compost produced from green cuttings (GC) and biowaste (Bio) shown on the x-axis. For compost from composting plants the letters i and e refers to type of compost (i: finished and e: fresh); numbers behind the compost name on the x-axis denote the sieving size after compost treatment in mm. Roman numerals after compost from hardware stores refer to the description. Note different y-axes scales for concentrations (D–F). The side arrows are intended to highlight the significance for toxicity and the contribution to soil organic carbon. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

mesoplastic items kg^{-1}). The lowest concentration (0.04 ± 0.08 g mesoplastic items kg^{-1}) was found in compost PlantB GC i 20 (Fig. 1D). Here, only 20% of the samples contained mesoplastics, the rest of the samples rather contained smaller particles. Compost HS GC I from a local hardware store, which contained the highest number of total plastic items (Fig. S1), showed with 0.217 ± 0.303 g kg^{-1} the third lowest plastic concentration (Fig. S2), i.e., this compost was preferably contaminated with large microplastic particles that contributed only minor and in the mg range to total plastic concentration. Both fresh composts (Bio e 20 and GC e 20) had the highest total plastic concentration, mainly caused by high concentrations of mesoplastic (Fig. 1D and Fig. S2). However, due to high variability no substantial differences were found.

All composts showed high variability in plastic concentrations inasmuch as they also showed high variations in the number of particles, as indicted by large standard deviations. Overall, this interspecific variability among particular composts was not significantly lower than the intraspecific variability between different compost types (Fig. 1, Fig. S3). In addition, no substantial differences between plastic concentrations from different composting plants and between plastic contents of composting plants and compost derived from hardware stores were found. The calculated concentrations of microplastic ranged from 2.4 ± 0.9 mg kg^{-1} (PlantB GC e 20) to 18.8 ± 6.9 mg kg^{-1} (HS GC I) for large microplastics (Fig. 1E) and from 0.01 ± 0.0 mg kg^{-1} (PlantB GC i 10) to 2.16 ± 0.80 mg kg^{-1} (HS GC I) for small microplastic (Fig. 1F). Overall, microplastic contributed only minimal to the total plastic concentrations for most composts. Small microplastic accounted for 0.01 wt% (PlantA Bio e 10 & PlantB GC e 20) to 1.3 wt% (PlantB GC i 20) of total plastic concentrations (Fig. 1F). Large microplastic contributed 0.4 wt% (PlantB GC e 20) to 8.7 wt% (HS GC I) to total plastic concentrations, except for PlantB GC i 20, here large microplastic

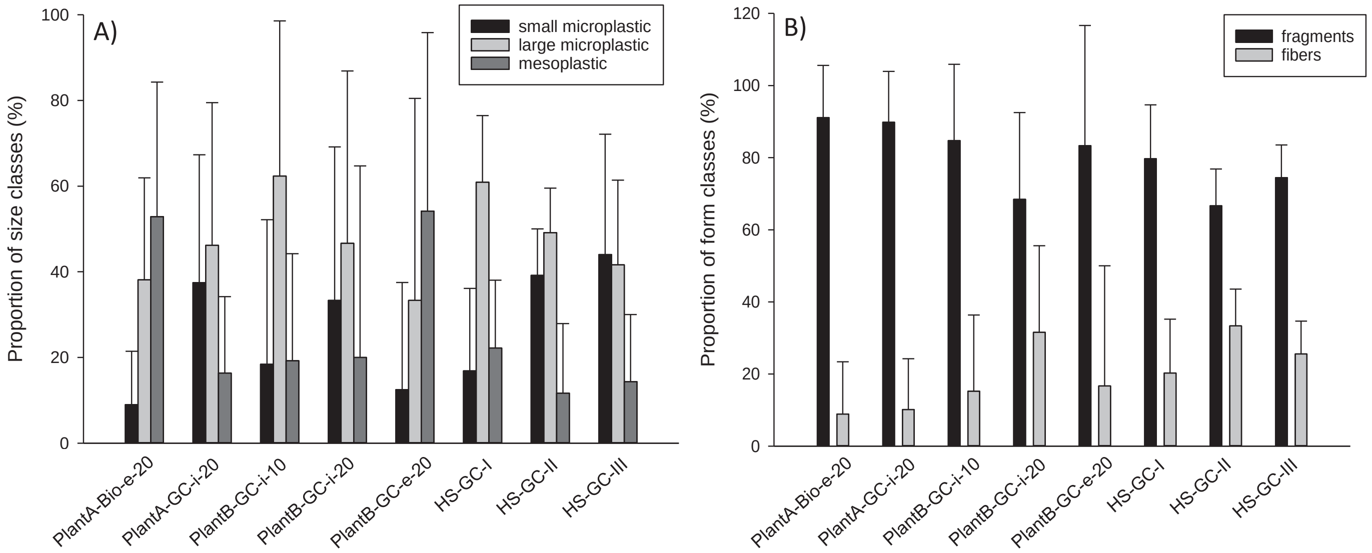

Fig. 2. Proportions of size classes (**A**) and form classes (**B**) of plastic items in analyzed compost produced from green cuttings (GC) and biowaste (Bio). For compost from composting plants numbers behind the compost name on the x-axis denote the sieving size after compost treatment in mm, roman numerals after compost from hardware stores (HS) refer to the description. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

made up 20 wt% of total concentration. Including calculated plastic concentrations of microplastic, total plastic load of compost ranged from 48.3 ± 88.8 mg kg^{-1} (PlantB GC i 20) to 1357.9 ± 596.0 mg kg^{-1} (PlantA Bio e 20, Fig. S2).

3.2.2. Size and form of plastic items

Variations in numbers and concentrations of plastic particles in the compost were accompanied by variations in the size of the particles (examples for found plastic sizes and forms are given in Photo 1). Mesoplastic was the most dominant plastic size in both fresh composts: produced from biowaste (PlantA Bio e 20: 53 ± 31% of all plastic items found) and green cuttings (PlantB GC e 20: 54 ± 42% of all plastic items found). In most other composts large microplastic was most abundant (Fig. 2A), only in one compost from a hardware store small microplastic dominated (HS GC III, Fig. 2A). Due to these large variabilities, no statistical differences in the size distribution of particles among composts from different composting plants and between composts of composting plants and those derived from hardware stores were found.

All composts contained, however, significantly ($p > 0.01$) more fragments (68 ± 24% to 91 ± 15% of all plastic items) than fibers (5 ± 7% to 13 ± 22% of all plastic items). Noteworthy, no spheres were detected (Fig. 2B).

4. Discussion

4.1. Plastic abundance in compost

Based on our earlier study and other findings (Gajst, 2016; Weithmann et al., 2018; Bläsing and Amelung, 2018) we hypothesized that compost may contain plastic to a severe extent, and that this amount is influenced by the production of compost, including used raw materials or treatment of compost like the size of sieving after composting. Here, we found plastic in all analyzed composts, only in one out of 40 analyzed subsamples no plastic was detected (Fig. 1A). Further we assumed that compost made from biowaste contains more plastic than compost made from green cuttings. Higher plastic loads of biowaste compost is mainly caused by improper waste disposal of plastic materials in biowaste, as stated in interviews with compost plant managers and confirmed by own observations during delivery of biowaste in a compost plant (Bläsing and Amelung, 2018). Indeed, the compost from biowaste contained the highest total plastic concentration, as well as the second highest number of plastic items of all compost types (Fig. 1; Fig. S2). However, due to the high intra specific variability no substantial differences between compost from biowaste and green cuttings were found. Surprisingly, compost treatment to remove foreign matter, i.e. the size of sieving after compositing did not generally influence the abundance of plastic in compost. For example, the compost made of green cuttings and sieved to 10 mm from plant B (PlantB GC i 10) contained a higher number of plastic items than two other composts sieved to 20 mm from the same plant (Fig. 1, Fig. S1) and also a higher plastic concentration than the finished compost from this plant (Fig. S2). However, the duration of compost process seemed to affect plastic abundance of larger plastic items in compost, as both fresh composts (PlantA Bio e 20 and PlantB GC e 20) showed the highest concentration of mesoplastic, while this was not the case for smaller plastic items (Fig. 1). Here, perhaps longer composting duration of finished compost favor fragmentation of larger plastic items into secondary microplastics.

The high variability of plastic contaminations did neither allow to ascertain clear differences in plastic numbers, size distributions and concentrations between the material obtained from different compost plants nor between the compost obtained from plants and local hardware stores. As no larger plastic items than mesoplastic were found, manual sorting and sieving procedures after composting were apparently effective in removing macroplastic (> 2.5 cm) from compost. This, however, was not the case for smaller plastics. To avoid significant plastic contamination of soil by using compost as important tool for soil amelioration, therefore, novel efficient pre cleaning procedures are needed, that are not yet routinely in place.

Comparable data about plastic in compost are scarce. Weithmann et al. (2018) analyzed two composts produced from biowaste and found 20 to 24 plastic items kg^{-1}; considering plastic items >1 mm. Most composts of our study contained similar plastic amounts, however, two composts of our study, the biowaste compost and a compost from a hardware store (HS GC I), showed even larger numbers of 27 ± 17 and 39 ± 14 items (>1 mm) kg^{-1} compost, whereas one compost from a local compost plant (plantB GC e 20) contained less (10 ± 6 items kg^{-1}).

While the number of small items is important for toxicity considerations, plastic concentrations determine the contribution of plastic to soil carbon. Schleiss (2017) analyzed plastic items >1 mm in 91 Swiss composts and found 0.2 and 0.48 g plastic kg^{-1} in composts for horticultural and agricultural use, respectively. These concentrations are in the same range than the concentrations detected in our study, only the compost produced from biowaste had higher plastic concentrations. Noteworthy, most plastic concentrations determined in this study exceeded those summarized in earlier studies (2.38 to180 mg kg^{-1}; Bläsing and Amelung, 2018) by a factor of up to 10. However, with exception of the compost produced from biowaste all compost are still below the mean concentrations determined by Gajst (2016) for Slovenian composts (1.2 g kg^{-1}). On the one hand methodological differences most likely account for different amounts of plastic particles recovered. In this study, for instance, we sieved the compost to a size of <5 mm before searching for larger plastic items, while formerly compost was directly searched manually for >5 mm plastic items (Bläsing and Amelung, 2018). On the other hand, these data clearly confirm that large variations of plastics items seem to be normal for different compost types, likely due to different care of people for plastic disposal into the raw materials that are finally selected for the composting process.

The high heterogeneity of plastic load will lead to heterogenous plastic pollution of compost treated soil. The sampling design of future studies should account for this high variability by analyzing a sufficiently high number of replications as well as a high sample weight to produce reliable data. In this study we analyzed 5 replications per compost with each 200 g sample weight, which was substantially more than used in former studies (e.g. Dumichen et al., 2017; Bläsing and Amelung, 2018; Weithmann et al., 2018; Corradini et al., 2019), but is still not sufficient to delineate clear differences among different compost producers.

Considering plastic as soil constituent, the contribution of plastic to soil organic carbon (SOC) is negligibly small, above all when compost is added to the huge reservoir of SOC and considering the minor contribution of especially small microplastics to total plastic concentration (<0.01 1.3 wt%, Fig. 1). Yet, if plastic is produced from petroleum it is free of ^{14}C, it will hence dilute ^{14}C signal of SOC and could, therefore, falsify soil radiocarbon ages. The negligible importance of small microplastic might also be assumed for risk assessment, i.e. the calculation of predicted environmental concentrations (PECs). However, as already stated by Backhaus and Wagner (2019), the classical PECs might not work for the heterogeneous group of plastic materials. The PEC concept does also not account that toxicity or any other effects of plastic particles likely increase with decreasing particle size (e.g. Jacob et al., 2020), whereas at a given number of particles the contribution of the small particles to total plastic concentration may be negligibly small. Looking at the size distribution of plastic in compost, small microplastic accounted for 9 to 44% of all counted items (Fig. 2A), which are likely toxically much more relevant than detected majority of larger plastic items.

Plastics in compost might additional affect toxicity when altered during composting in a way that toxic additives might be easily introduced into soil. Also, a potential faster degradation of such altered particles in soil, caused by degradation or at least surface alteration of these particles during composting and enhanced microbial activity in compost seems possible.

4.2. Compost as source of plastic for soils

Application rates of compost vary widely: while recommended application rates are typically in the range of 30 35 t ha^{-1} annually (WRAP, 2015), some countries like Germany restrict application rates to 20 or 30 t ha^{-1} within 3 years (BioAbfV, 2013). Based on these application rates and the plastic contents found in this study (mean: 12 to 46 items kg^{-1}), between 84,000 320,000 and between 420,000 1,610,000 plastic items will reach agricultural fields per ha and year for compost applications of 7 and 35 t ha^{-1}, respectively. Using the data of this study to translate items to plastic loads, this results in an annual export of 0.34 9.51 kg plastic per ha (for 7 t compost application) to 1.69 47.53 kg plastic per ha (for 35 t compost application) to agricultural fields. Consequently, plastic loads may be even 7.5 to 8 times higher than formerly estimated (Bläsing and Amelung, 2018). Nevertheless, compared to the potential plastic input via sewage sludge application and other potential plastic sources for agricultural fields, this amount is still low. Sewage sludge, for instance, may contain 1000 to 24,000 plastic items kg^{-1} (Mahon et al., 2017; Mintenig et al., 2017). These plastic contents can lead to a yearly input of 1.67 to 40.8 Mio. plastic items ha^{-1}, even if considering only restricted application of 5 t dry mass of sewage sludge ha^{-1} within 3 years (Bläsing and Amelung, 2018). Until now, data about plastic pollution caused by plastic mulching are scarce, especially for Europe. For agricultural fields under plastic mulching in China up to 259 to 381 kg of large plastic per ha was found (Changrong et al., 2014; Zhang et al., 2016), which corresponds to the plastic pollution caused by 5 to 225 years of compost application, considering a high application rate of 35 t ha^{-1} and year. Also, irrigation with untreated wastewater (2.2 Mio. 3.32 Mio. items per ha and growing season) and treated wastewater (up to 275 Mio. to 625 Mio. items per per ha and growing season) will lead to considerably higher plastic input than compost application (Bläsing and Amelung, 2018).

Proportions of both, size and shape classes in compost differ substantially from that found in sewage sludge. Mahon et al. (2017) analyzed plastic in seven different sewage sludges with different sludge treatments and found that in all substrates items <1 mm and fibers were dominant, thus contrasting our findings for compost (Figs. 1 and 2). We attribute this difference to the input paths; wastewater and later on sewage sludge receive plastic mainly from street runoff containing small items from tire abrasion or effluents of, e.g., households including fibers from washing machines and small plastic in cosmetic products (e.g. Napper and Thompson, 2016; Hartline et al., 2016; Ziajahromi et al., 2016). Plastic in compost, in contrast, mostly originates from improper waste disposal and fragmentation of this waste during composting, which produces larger plastic particles. This difference presumably leads to different pattern of plastic contamination of fields under compost and sludge application. The size of plastic might be also essential for the fate of plastic in soil. Smaller plastic items, for instance, more likely undergo vertical movement and colloidal leaching than larger ones (Rillig et al., 2017; O'Connor et al., 2019). First hints of such leaching were already found by Zubris and Richards (2005) and Zhang and Liu (2018) who detected fibers from sludge in deeper soil horizons. Further, the shape of plastic items might determine interactions with soil mineral and organic phases and thus related soil physical properties (Souza Machado et al., 2018; Zhang and Liu, 2018).

Up to now, there are no overall strict threshold values for maximum plastic concentrations in agricultural fields, and a such also not in Germany. However, in some countries, fertilizer ordinances include maximum permissible values for foreign matter, including plastic. In Germany, a maximum of 0.5% of foreign matter, considering particles >2 mm, in compost are allowed, which corresponds to a quantity of 5 g plastic kg^{-1}compost. This is even larger than the maximum amount of plastic concentrations detected here (1.36 ± 0.6 g kg^{-1}), i.e., even maximum plastic loads do at present not restrict the use of compost in agriculture. While this is reasonable in terms of soil fertility, it seems questionable in terms of potential long term accrual of stable foreign materials. In contrast, in Switzerland the content of foreign matter in compost is more restricted, i.e. a maximum of 0.1% foreign matter is allowed. Interestingly, here since 2015 the maximum content of 0.1% is not restricted to items >2 mm but includes all sizes (Der Schweizerische Bundesrat, 2015). According to the Swiss legislation the compost made of biowaste analyzed in this study exceeds the threshold, while all other compost showed plastic concentrations below 0.1%.

Table 2
Potential plastic input via compost application in horticultural soils.

Field of application		Recommended fertilizer rate	Calculated fertilizer rate	Potential plastic input	
				Content	Concentration
		(L m^{-2})	(t ha^{-1})	(items ha^{-1})	(kg ha^{-1})
Vegetable bed	Plants with low nutrient demand	1	6.48	77,760–298,080	0.31–8.80
	Plants with medium nutrient demand	2	12.96	155,520–569,160	0.63–17.6
	Plants with high nutrient demand	3	19.44	233,280–894,240	0.94–26.4
Grove		1	6.48	77,760–298,080	0.31–8.80
Shrubs	Slow-growing	1	6.48	77,760–298,080	0.31–8.80
	Fast-growing	2	12.96	155,520–569,160	0.63–17.6
Roses		2	12.96	155,520–569,160	0.63–17.6
Lawn		2	12.96	155,520–569,160	0.63–17.6
Meadow flower		1	6.48	77,760–298,080	0.31–8.80
New installation of gardens		50	324	3,888,000–14,904,000	15.65–439.97

Values indicate yearly compost application and resulting yearly plastic input, only for vegetable values are given per culture. New installation of gardens is a single event. Recommended application rates after Fischer and Jauch (1999).

Composts are not only applied in agricultural fields, but commonly also in horticultural soils. Nevertheless, a detailed estimation for horticultural soils was still missing. Yet, such data can be estimated using the reported range of plastic items in compost. Because most recommendations for compost applications in horticulture are given in L m^{-2} (e.g. Fischer and Jauch, 1999), we used a mean compost density of 0.648 kg L^{-1} (BGK, 2018) to transfer values in kg ha^{-1} (Table 2).

For cultivation of vegetables, the amount of compost and according its potential of plastic inputs increase with nutrient demand of plant, i.e., the largest potential plastic input (233,280–894,240 plastic items meaning 0.94–26.4 kg ha^{-1} per culture) is estimated for vegetables with high nutrient demand, like pumpkin, tomatoes, savoy, broccoli or leek. The cultivation of vegetables with medium (e.g. cucumber, onion, potatoes) and low (many types of beans and salads, spinach and peas) nutrient demand will presumably be accompanied by lower plastic input (Table 2). Additional inputs likely occur upon the establishment of such horticultural gardens: for example, during new installations of gardens, sometimes single doses of compost of up to 50 L m^{-2}, equivalent to 324 t ha^{-1} are given, which lead to potential plastic input of 3,888,000 to 14,904,000 items and 15.65 to 439.97 kg plastic ha^{-1} (Table 2). Additionally, also some potting soil contain proportions of compost and accordingly plastic. Noteworthy, highest plastic concentration was measured in compost produced from biowaste, which is usually not used for domestic gardens; presumably plastic input into domestic gardens might be rather lower than the maximum value calculated here. However, such compost might be used for e.g. commercial cultivation of cut flowers. In summary, horticultural soils receive similar amounts of plastic as agricultural soil with low to medium compost application rates, and hence, compost has to be considered as a major entry path for plastic not only into agricultural but also into horticultural soil.

5. Conclusion

In the present study, we analyzed eight different compost, from both, compost plants and hardware stores. We could confirm that compost application is a major input path for plastic in soil. Based on our results we calculated potential plastic inputs into agricultural and for the first time also horticultural soil. As manual sorting and sieving in compost plants is effective for larger plastic items like macroplastic, plastic composition is dominated by especially microplastic items or even smaller particles not considered here. However, we have to refute the part of our hypothesis that procedures to remove foreign matter after composting like size of sieving affected the plastic load. Instead, compost sieved to 10 mm contained more plastic than compost sieved to 20 mm. However, we can support the other part of our main hypothesis that the duration of composting seemed to influence the abundance of larger plastic items: both fresh composts contained more mesoplastic than the finished ones. Biowaste had the highest plastic concentration and exceeded as only compost the threshold for allowed plastic in compost of Switzerland, however, not the German one, thus highlighting also the need to harmonize regulations across countries. Due to the high inter- and intraspecific variability of plastic items in compost subsamples, using the “right” compost subsample when fertilizing a given field remains thus a challenge for the farmers or owners of allotment gardens.

Credit author statement

Melanie Braun: Conceptualization, Validation, Investigation, Writing. **Matthias Mail:** Conceptualization, Validation, Investigation, Writing. **Rene Heyse:** Conceptualization, Validation, Investigation, Writing. **Wulf Amelung:** Conceptualization, Validation, Investigation, Writing.

Declaration of competing interest

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

Acknowledgements

This study was supported by the Argelander Grant from the University of Bonn, Germany. We also thank the Nees Institute, especially Prof. Barthlott for the opportunity to use his equipment, e.g. the microscope.

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