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Exposure assessment of plastics, phthalate plasticizers and their transformation products in diverse bio-based fertilizers

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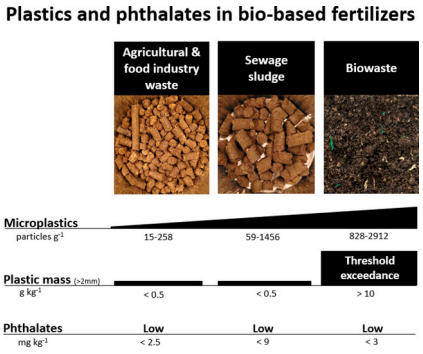
HIGHLIGHTS

- Fertilizers from biowaste, sewage sludge and agro/food industry waste were assessed.
- Packaging plastic concentrations in biowaste fertilizers exceeded the EU limit.
- Small amounts of non-packaging plastics were found in other fertilizers.
- Low concentrations of DEHP and phthalate metabolites were measured in all fertilizers.
- Phthalate metabolites bioaccumulated in earthworms.

GRAPHICAL ABSTRACT



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ABSTRACT

Bio-based fertilizers (BBFs) produced from organic waste have the potential to reduce societal dependence on limited and energy-intensive mineral fertilizers. BBFs, thereby, contribute to a circular economy for fertilizers. However, BBFs can contain plastic fragments and hazardous additives such as phthalate plasticizers, which could constitute a risk for agricultural soils and the environment. This study assessed the exposure associated with plastic and phthalates in BBFs from three types of organic wastes: agricultural and food industry waste (AgriFoodInduWaste), sewage sludge (SewSludge), and biowaste (i.e., garden, park, food and kitchen waste). The wastes were associated with various treatments like drying, anaerobic digestion, and vermicomposting. The number of microplastics (0.045–5 mm) increased from AgriFoodInduWaste-BBFs (15–258 particles g^{-1}), to SewSludge-BBFs (59–1456 particles g^{-1}) and then to Biowaste-BBFs (828–2912 particles g^{-1}). Biowaste-BBFs

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mostly contained packaging plastics (e.g., polyethylene terephthalate), with the mass of plastic (>10 g kg^{-1}) exceeding the EU threshold (3 g kg^{-1}, plastics >2 mm). Other BBFs mostly contained small (< 1 mm) non-packaging plastics in amounts below the EU limit. The calculated numbers of microplastics entering agricultural soils via BBF application was high (10^7–10^{10} microplastics $ha^{-1}y^{-1}$), but the mass of plastic released from AgriFoodInduWaste-BBFs and SewSludge-BBFs was limited (< 1 and <7 kg $ha^{-1}y^{-1}$) compared to Biowaste-BBFs (95–156 kg $ha^{-1}y^{-1}$). The concentrations of di(2-ethylhexyl)phthalate (DEHP; < 2.5 mg kg^{-1}) and phthalate transformation products (< 8 mg kg^{-1}) were low (< benchmark of 50 mg kg^{-1} for DEHP), attributable to both the current phase-out of DEHP as well as phthalate degradation during waste treatment. The Biowaste-BBF exposed to vermicomposting indicated that worms accumulated phthalate transformation products (4 mg kg^{-1}). These results are overall positive for the implementation of the studied AgriFoodInduWaste-BBFs and SewSludge-BBFs. However, the safe use of the studied Biowaste-BBFs requires reducing plastic use and improving sorting methods to minimize plastic contamination, in order to protect agricultural soils and reduce the environmental impact of Biowaste-BBFs.

1. Introduction

Agriculture in the EU strongly relies on imported mineral fertilizers, especially nitrogen (N) and phosphorus (P) which are the dominant rate-limiting nutrients (Guignard et al., 2017; Stark and Richards, 2008). Mineral fertilizers are, however, associated with various environmental concerns. The production of N fertilizer through the Haber-Bosch process is very energy-intensive and global P resources (present as phosphate rocks) are limited (Cordell et al., 2009; Erisman et al., 2008; Kuokkanen et al., 2017; Villalba et al., 2008). Paradoxically, large amounts of N and P nutrients are lost because organic waste is underused. In addition, N and P dispersion into the environment contributes to soil, water and air pollution (Cameron et al., 2013; Carpenter, 2008; Kominko et al., 2021; Svanbäck et al., 2019). The use of bio-based fertilizers (BBFs) can significantly reduce dependence on mineral fertilizers and close N and P cycles (Chojnacka et al., 2020; Svanbäck et al., 2019). BBFs are derived from biomaterials of various origins - often wastes or side-streams from agriculture, industry or society - with a content of bioavailable plant nutrients suitable to serve as crop fertilizers (Wester-Larsen et al., 2022). The agronomical efficiency of BBFs has been reported to be similar to that of mineral fertilizers (Kurniawati et al., 2023; Sigurnjak et al., 2019, 2016; Vaneeckhaute et al., 2013). Promoting the use of BBFs can therefore support the transition from a linear to circular economy for fertilizers (Chojnacka et al., 2020). Concerns have been raised, however, about potential environmental risks associated with the presence of plastics in some organic fertilizers, including microplastics (plastics with size <5 mm).

The application of sewage sludges (biosolids) and biowaste-based composts and digestates to farmland were reported to be a significant input of microplastics to the terrestrial environment (Bläsing and Amelung, 2018; Weithmann et al., 2018; Zhang et al., 2022; Zhu et al., 2019a). Biowaste, i.e., biodegradable garden and park waste, as well as food and kitchen waste, has been reported previously to be contaminated by plastics sourced from bags and foils from packaging (Porterfield et al., 2023; Steiner et al., 2023; Weithmann et al., 2018). Wastewater, for its part, is commonly contaminated by plastics coming from textiles (detachment during washing) and personal care products that end up in sewage sludges produced by wastewater treatment plants (Carr et al., 2016; Golwala et al., 2021; Mahon et al., 2017; Murphy et al., 2016; Reddy and Nair, 2022; Sun et al., 2019). Nizzetto et al. (2016) estimated yearly microplastic inputs to European and American farmlands (up to 430′000 and 300′000 tons, respectively) exceeded the total amount of microplastics estimated in the marine aquatic environment (93′000–236′000 tons). Quantitative and definitely qualitative studies on the presence of plastic residues in BBFs are still limited (Bläsing and Amelung, 2018; Braun et al., 2021; O'Connor et al., 2022; Scopetani et al., 2022). To our knowledge, prior to this study data for BBFs produced from agricultural and food industry waste are almost non-existent.

Microplastics not only impact soil properties and their microbiology and water holding capacity (de Souza Machado et al., 2018; Rillig et al., 2023; Wan et al., 2019; Yao et al., 2023), but also their residues can be ingested by soil-dwelling organisms (Huerta Lwanga et al., 2016; Zhang et al., 2020), transferred to higher trophic levels (Huerta Lwanga et al., 2017), and alter organism growth and reproduction (Agathokleous et al., 2021; Besseling et al., 2014; Huerta Lwanga et al., 2016; O'Connor et al., 2022). In addition to mechanical effects from the physical plastics themselves (Wright et al., 2013), concern has been raised regarding the role of microplastics as vector of contaminants in soils (Bhagat et al., 2021), both added during the manufacturing (intentional and unintentional additives) and sorbed from the surrounding environment (e.g. persistent organic pollutants) (Bridson et al., 2021; Fauser et al., 2022; Ge et al., 2021; Hartmann et al., 2017; Mammo et al., 2020).

Plastic additives, such as phthalate plasticizers, have drawn particular global attention because they are present in plastics at much higher levels than sorbed substances (Campanale et al., 2020; Gallo et al., 2018). For example, di (2-ethylhexyl) phthalate (DEHP) was until recently one of the most commonly used plasticizer (50 % of the 8 million tons of phthalate produced globally in 2015) and can reach over 50 % of the mass of polyvinyl chloride (PVC) plastics (Bernard et al., 2014; Ito et al., 2019; Latini et al., 2010; Tumu et al., 2023). Many of these phthalate plasticizers are toxic. DEHP has been shown to be an endocrine disruptor with genetic toxicity and is classified as a substance of very high concern (SVHC) under the EU REACH regulation (Sun et al., 2022; Tumu et al., 2023). As they are not chemically bound to polymers, phthalates can relatively easily leach out into the environment, constituting a risk even if the plastics are not directly ingested (Groh et al., 2019; Hahladakis et al., 2018; Rowdhwal and Chen, 2018). Upon uptake in organisms, phthalates are quickly metabolized to monoesters. For example, DEHP is hydrolysed to mono(2-ethylhexyl)phthalate (mEHP) and further metabolized and/or conjugated with glucuronic acid to be excreted (Carli et al., 2022; Meeker et al., 2009). mEHP and other metabolites have been found to be more toxic than the parent phthalates (Zhu et al., 2019b; Zolfaghari et al., 2014). A recent study has reported that DEHP degradation in wastewater could generate monoesters (He et al., 2021), potentially contributing to the total level of these toxic compounds in organic waste used to produce BBFs. Exposure to phthalates around the globe is now widespread (Koch and Calafat, 2009) and thus monitoring these contaminants in BBFs is essential to ensure their safety. To address these concerns, new regulations are being enacted in the European Union which state that fertilizers should not contain >3 g kg^{-1} dry weight (d.w.) of plastics above 2 mm (regulation 2019/1009 (EU, 2019)).

Quantifying (micro)plastics in solid organic matrices such as fertilizers (or soils) involves isolation and identification steps. For now, there is no standardized analytical method for detection of microplastic in such matrices. Methods used for the isolation step of microplastics includes techniques such as digestion (e.g. H_2O_2 or Fenton's reagent), density separation (e.g. Zn_2Cl_2), or centrifugation (Junhao et al., 2021; Ruggero et al., 2020), and identification relies on techniques such as spectroscopy (e.g., Fourier transform infrared spectroscopy (FTIR), thermal degradation (e.g. pyrolysis gas chromatography), or visual

Table 1
Waste origins and valorisation methods of the twelve selected bio-based fertilizers (BBFs).

BBF	Waste origin category (Characterizing raw material)	Description of raw material	Technology
BA1	Agriculture & food industry waste	Wheat and maize	Fermentation & distillation
MO14	Agriculture & food industry waste	Vegetable by-products from food industry, animal proteins	Pelletising
BIO	Agriculture & food industry waste	Meat and bones, apatite, vinasse, chicken manure, K2SO4	Pelletising
OPU	Agriculture & food industry waste	Chicken manure	Pelletising
FEK	Agriculture & food industry waste	Chicken manure	Drying and pressing (extrusion)
OG2	Agriculture & food industry waste	Crushed and dried dead animals	Hydrolysis
ECO	Agriculture & food industry waste	Blood and feather meal	Pelletising
NNP	Sewage sludge	Sewage sludge and industrial sludge	Infrared drying
RAN	Sewage sludge	Sewage sludge and biowaste	Drying and granulating
PRV	Sewage sludge	Sewage sludge and biowaste	Biogasification and hygienisation
VERMI[a]	Biowaste[b]	Biowaste (household and commercial origins) and manure	Biogasification and vermicomposting
PLP	Biowaste[b]	Biowaste (household and commercial origins), peat and wood chips	Composting

[a] Additional information about VERMI is available in Section 2.2 and in Supplementary Information SI.6.
[b] Biowaste is defined as *biodegradable garden and park waste, as well as food and kitchen waste.*

analysis (e.g. light microscopy). The pore sizes of meshes used to retain or filtrate samples during the isolation step can significantly vary, impacting the minimal size of microplastics that are analysed. The absence of standard methods complicates data comparison between studies and hinders progress in understanding the microplastic transfer to soils, its distribution and risks (Gui et al., 2021; Porterfield et al., 2023). In addition, few studies have reported plastic abundance on a weight/weight basis (Porterfield et al., 2023; Zhang et al., 2019), thereby preventing assessment of compliance with ecotoxicity or regulatory thresholds which are delineated on this basis. Studies on plastic additives in fertilizers are also very scarce, although these chemicals can affect nutrient transmission, growth of crops and soil living organisms (e.g. earthworms) (Le et al., 2023). Addressing these knowledge gasps is critical. Indeed, to enable a safe circular economy by reintroducing nutrients back to farm ecosystems through the use of BBFs, it must be ensured that this is not also introducing contaminants into the environment and food chain (Nizzetto et al., 2016).

Due to the concern of risk associated with plastics and phthalate plasticizers in BBFs from various waste materials, this study approached several aspects that have not yet been investigated. The specific aims of the study were to determine (i) microplastics in BBFs obtained from agricultural and food industry waste (AgriFoodInduWaste), sewage sludge (SewSludge) and biowaste (Biowaste) using the same method (digestion of organic matter followed by FTIR), (ii) the mass of plastic in the most contaminated BBFs, and (iii) concentrations of DEHP and phthalate transformation products. Addressing these aims allowed for a consistent comparison between BBFs, compliance assessment with limit values, and an estimation of the amount of contaminants entering agricultural soils via BBF applications. In addition a fourth aim was to (iv) quantify DEHP and phthalate transformation products in plastics and earthworms of a vermicompost to assess the degradation of DEHP and the resulting generation of monoesters and phthalic acid. Addressing this fourth aim allows for a better understanding of the behaviour and risks of these contaminants. Novel elements of the study are the determination of plastics in AgriFoodInduWaste-BBFs, the comparison of plastic levels in various types of BBF using the same method, the simultaneous analysis of plastics and phthalate additives, the generation of phthalate monoesters in BBFs, and the estimation of amounts of these entering soil systems via BBFs.

2. Materials and methods

2.1. Selection of BBFs and analysis strategy

Twelve BBFs were selected from three main categories of waste origin, i.e., agricultural and food industry waste (7 AgriFoodInduWaste-BBFs), sewage sludge (3 SewSludge-BBFs) and biowaste (2 Biowaste-BBFs). Table 1 presents the BBF code letter (full name is not given for confidentiality reasons), waste origin category, description of the raw material constituting the waste, and technology used to valorise the waste. Complementary information (e.g., NPK content, dry matter content, and recommended application rates) is given in the Supporting Information (SI.1). Note that the BBFs were real-world, commercially available (or in development) products that sometimes may contain mixtures of wastes. Although none of the AgriFoodInduWaste-BBFs was reported to be mixed with other types of waste, some of the SewSludge-BBFs and Biowaste-BBFs were combined with other wastes (e.g., biowaste in RAN and NNP, or manure in VERMI). Analyses of microplastics and phthalate transformation products were first done on one replicate of each BBF to assess the contamination levels. Then, triplicate analyses of microplastics, bigger plastics (> 1 mm) and phthalate transformation products were conducted for the five most contaminated BBFs.

2.2. Sampling analysis of earthworms in one BBF

The Biowaste-BBF that was a vermicompost (VERMI, Table 1) utilized earthworms (mostly *Eisenia Fetida*) to assist with composting. The earthworms were sampled to better understand how they mix plastics as well as DEHP and transformation products. This vermicompost contained a substantial amount of plastic from green low density polyethylene (LDPE) bags used to collect organic household waste (OHW). Fagerheim (2020) reported that the OHW plastic bags represent about 25 % of the plastics entering the treatment process for Biowaste-BBF VERMI, while the rest of plastics derive from incorrectly sorted waste in OHW (19 %) and from plastic packaging in commercial organic waste (56 %). Almost half of the OHW plastic bags were made of LDPE that was coloured green; whose fragments are easily identifiable in the vermicompost, allowing a comparison of contaminant concentrations between the pristine bags and the fragments. Therefore "pristine green PE bags" were sampled separately. The vermicompost was produced in a reactor (80 cm depth) where the solid fraction of anaerobically digestated organic waste was added on the top and processed vermicompost was removed from the bottom (through a grid), with a residence time of 2–3 months. About 10 kg of vermicompost were collected from the bottom of the reactor using small metallic shovels and transported to the lab using a stainless-steel container. Earthworms were collected from 3 kg of vermicompost by using a 4-mm sieve. Worms were rinsed with Milli-Q water on a 1-mm mesh size sieve, quickly dried on a tissue paper and weighed. Worms heavier than 0.1 g were individually depurated for 48 h in 380 mL glass containers containing a paper filter (70 mm diameter, grade 1, Whatman, UK) moistened with 0.5 mL of Milli-Q water which was changed every 12 to 16 h (to prevent coprophagy). The containers were closed with perforated aluminium foil (to allow sufficient oxygenation) and were kept at 18 °C (see Supplementary information SI.5). At the end of the depuration, worms were transferred to 20 mL glass containers, weighed, and directly stored in freezer (−18 °C).

2.3. Chemical digestion of organic matter

A two-step chemical digestion of organic material – that involves a dissolution of cellulosic material followed by an oxidation –- was slightly adapted from Olsen et al. (2020) (see Supplementary Information SI.2 for details). Briefly, stainless steel mesh foils (45 μm, The Mesh Company, UK) were cut into 10 × 6 cm rectangles that were ultrasonicated for 30 min in a sodium dodecyl sulphate solution (Merck, Norway) and methanol (VWR, Norway), successively. After drying (60 °C), the rectangular meshes were sandwiched between two glass plates and macroscopically inspected (Olympus SZX16) to ensure the absence of particles. The meshes were then folded into envelopes and made tight by double folding each edge of the envelopes. Seven of the twelve BBFs were in pellet or granulate form and needed to be powdered to allow an efficient digestion of the organic matter. For this purpose, BBFs were softened in Milli-Q water for 4 h in 10-mL vials (to avoid breaking plastic fragments by grinding). After quantitative transfer to the envelopes, they were secured by winding a pre-weighed nickel-copper wire (Alloy Wire, China) and then oven dried overnight (60 °C). The steel-mesh envelopes containing the dried samples were weighed and placed in 380 mL glass jars along with magnetic stirring bars. To dissolve the cellulosic material (1st step of the chemical digestion), an aqueous solution of 8 % NaOH, 8 % urea and 6.5 % thiourea (by weight) was added in a ratio of 40 mL per 0.1 g d.w. sample, and the jars were stored at −18 °C for 45 min (to allow mini-crystal formation). After magnetic stirring (1 h) at room temperature, the envelopes were rinsed with Milli-Q water (15 × 30 mL). The oxidation (2nd step of the chemical digestion) was done by adding 30 % H_2O_2 and 10 M NaOH to the jars in ratios of 30 mL and 0.75 mL per 0.1 g d.w. sample, respectively. After magnetic stirring (3–4 h), the envelopes were rinsed with Milli-Q water (10 × 30 mL), oven dried at 60 °C overnight and weighed the next day. The two-step digestion was repeated until the reduction of mass from one day to the next was below 4 %. At the end of the chemical digestion, >75 % of the samples had a mass < 0.005 g (i.e., < 2.5 % of the initial mass), meaning that the mass of potential minerals was very low. The use of density separation was thus considered non-relevant as it would present more risks (potential loss of plastic particles and contamination of the sample) than benefits (only a slightly reduction of particles to identify by FTIR); tests conducted on samples with the highest amount of mineral confirmed that density separation was not necessary (Supplementary Information SI.2).

2.4. Microplastic quantification

The quantification method was also slightly adapted from Olsen et al. (2020) (see Supplementary Information SI.2 for details). The envelopes were opened in 500 mL beakers using tweezers, then rinsed and soaked with Milli-Q (300 mL), and sonicated for 30 min. Samples were thus transferred to 20 mm diameter circular 45 μm stainless steel meshes using a vacuum filtration system. The sonication and filtration steps were reconducted and the particles collected on a second circular mesh. The two meshes were put on a stainless-steel plate that could hold the two 20 mm meshes, and oven dried at 60 °C overnight in a glass Petri dish. Identification of microplastics was performed on both meshes using a micro FTIR imaging system (Perkin Elmer Spotlight 200i FT-IR microscope) operating in transmittance mode (wave number range: 4000–650 cm^{-1}, accumulations: 4 scans per spectra). The spectra were identified using FTIR libraries "Polymer", "ATR-Spectra", "Transmission-Spectra" and "Fluka" provided by PerkinElmer, considering spectra matches above 0.7 (Olsen et al., 2020), and were classified into 18 plastic categories (see Supplementary Material SI.3). For copolymers, results indicating the predominance of one of the polymers (i.e., > 50 %), were classified in the category of the predominant plastic; when no indication was provided, they were classified as "copolymers". The FTIR spectra for chlorinated polyethylene and polyvinyl chloride (PVC) cannot be distinguished from each other, and both were thus classified as "PVC".

2.5. Quality control for microplastic analysis

To minimize plastic contamination, only metal tools and glass vessels were used. They were rinsed thoroughly with MeOH and Milli-Q water, and immediately wrapped with aluminium foil. Nitrile gloves and cotton lab coats were worn during the laboratory work. To detect potential contamination, procedural blanks were done by applying the same procedure (i.e., sample preparation, digestion of organic matter, transfer to small meshes, FTIR analysis) to empty envelopes. Three procedural blanks were analysed during the first screening of the 12 BBFs and another three during the triplicate analyses of the five most contaminated BBFs. In addition, 12 blanks spiked with PE powder (0.02 g), PE fibres (0.02 g), PE granulates (0.2 g), and PET fibres (0.02 g) were processed (in triplicates for each type of plastic) to determine the method weight recoveries (Olsen et al., 2020). The same procedure as for samples was used except that the plastics recovered after filtration were only weighed (not analysed by FTIR).

The six procedural blanks presented very low amounts of plastic particles, i.e., 0, 3 and 0 microplastics in the blanks of the first screening, and 3, 8 and 4 in the blanks processed alongside the most contaminated samples (see Supplementary Information SI.3). Limits of quantification (LOQ) for individual plastic polymers were set at average plus three standard deviations in blanks and ranged between 2.1 and 4.1 microplastic particles $sample^{-1}$; a LOQ of 1 microplastic $sample^{-1}$ was used for plastic types absent from blanks (SI.3). When a plastic type was found in a sample with a microplastic count ≤ LOQ, a value of ½ LOQ was assigned. Note that variable and sometimes significant number of polytetrafluoroethylene (PTFE) fragments (2 to 106 fragments) were found in procedural blanks. This contamination was attributed to the PTFE-coating of stirring bars and potentially PTFE-tubing of the Milli-Q water device. Therefore, PTFE was not quantified in samples. Recoveries were shown to be slightly dependent on the shape of microplastics - PE powder (84 ± 7 %) < PE fibres (94 ± 5 %) < PE granulates (101 ± 1 %) – and on the types of plastic - PET fibres (70 ± 2 %). PET is known to be (one of) the most sensitive polymer to alkaline treatment, especially for fibres, and a recovery of 70 % was thus considered acceptable (Hurley et al., 2018; Olsen et al., 2020; Pfeiffer and Fischer, 2020). Results presented hereafter have not been corrected for potential losses.

2.6. Mass of plastics above 1 mm in most contaminated BBFs

Assessing the compliance of BBFs with regulations in terms of plastic contamination requires expressing results in 'mass of plastic per mass of fertilizer' (and not only in the more frequently used 'number of fragments per mass of fertilizers'). For this purpose, 110 to 350 g (equivalent to 100 g d.w.) of the 5 most contaminated BBFs were weighed, pellets (when applicable) were crushed using metallic bucket and pestle, and samples were oven dried at 60 °C overnight. Then, samples were sieved using 4, 2 and 1 mm mesh sizes and all fragments suspected to be plastics were collected. The fragments were analysed by Frontier ATR (Attenuated total reflection) assembly FTIR (PerkinElmer), sorted by type of plastic, and their surface was measured. Fragments of the same plastic type were weighed together. In addition, for the Biowaste-BBF VERMI, 800 g of the previously treated vermicompost (without worms) were sorted in "green PE" and "other plastic" fragments (see photographs in Supplementary Information SI.5), weighed and stored in glass containers at −18 °C).

2.7. DEHP and phthalate transformation products analyses

Phthalate transformation products were analysed in addition to DEHP for two reasons. First, DEHP and other phthalates are expected to degrade to monoesters in organic waste, and then to phthalic acid, the common breakdown product of phthalates (Carli et al., 2022; Meeker

et al., 2009). Second, the quantification of parent phthalates is known to suffer from severe background contamination because these compounds are ubiquitous (AMAP, 2017; Guo and Kannan, 2012). Targeting phthalate transformation products thus allows for the measurement of actual phthalate content - including monoesters which can be more toxic than the parent molecules - and for the drastic reduction of background contamination in samples. Extraction of DEHP and phthalate transformation products were performed according to a method adapted from Rian et al. (2020); details are given in Supplementary Information SI.4. The BBF samples in pellet form were grinded using a porcelain mortar and a pestle. All samples were freeze-dried (Labconco FreeZone 2.5 L) and further homogenised (mortar and pestle), after which 0.1 g was placed in a 15 mL glass tube. Internal standard (20 μL of 1 μg mL^{-1}) was added, and enzymatic digestion (deconjugation of glucuronides) was done with 1 mL of 1 M ammonium acetate solution containing 44 units of β-glucuronidase (prepared by spiking 50 μL of β-glucuronidase into 50 mL of 1.0 M ammonium acetate solution), in an incubator shaker (150 RPM) at 37 °C for 12 h. Extraction was done with 5 mL of MeOH/Milli-Q water solution (1:1 *v*/v) acidified to pH < 3 (HCl) under ultrasonication (45 min, 35 °C). After centrifugation (4000 rpm; 5 min) and dilution to 50 mL with HCl-acidified Milli-Q (pH < 3), a solid-phase extraction was performed using 200 mg STRATA X-RP cartridges (Phenomenex). Elution of compounds was done with 10 mL of MeOH: acetonitrile (1:1 v/v) and collected into a 15 mL glass tube. The eluent was then concentrated to 0.1 mL at 45 °C under a gentle nitrogen flow (TurboVap LV), diluted to 1 mL with Milli-Q water and transferred to LC vials.

DEHP and 17 phthalate transformation products (see Supplementary Information SI.4) were analysed by ultra-high performance liquid chromatography (Acquity UPLC® I-Class system) coupled to tandem mass spectrometry (Xevo TQ-S triple quadrupole mass spectrometer) from Waters (Milford, MA, U.S.). Separation was carried out on a Kinetex C18 column (30 × 2.1 mm, 1.3 μm, 100 Å Phenomenex) connected to a C18 Phenomenex guard column (2 × 2.1 mm). Milli-Q water and acetonitrile, both acidified with 0.1 % acetic acid, were used as mobile phases for chromatographic separation; an isolator column (Acquity UPLC, 2.1 × 50 mm, Waters) was placed in the flow path between the mobile phase mixer and the sample manager injector to eliminate the potential phthalate contamination commonly found in the solvents of the mobile phases and tubing of the instrument. Electrospray was used as ionization source. The analytical method parameters are detailed in SI.4. Quantification was accomplished based on the internal standard method and matrix-matched calibration standards (Asimakopoulos et al., 2016), using one matrix pool for each of the three BBF types (i.e., AgriFoodInduWaste-BBFs, SewSludge-BBFs and Biowaste-BBFs) and 4 to 5 fortification concentrations per pool (5, 10, 25, 50 and 300 ng mL^{-1}). Absolute recoveries, determined by spiking matrix pools before and after extraction, are provided in SI.4. Whereas recoveries for phthalate transformation products were acceptable (average > 60 %), recovery for DEHP was low (< 10 %), therefore DEHP was semi-quantified. DEHP concentration was assessed as the sum of DEHP and its major hydrolysed transformation product mEHP. For each pool, limits of quantification were determined from the procedural blanks conducted along the samples (10*standard deviation in procedural blanks/slope of calibration curve) and were set as LOQ if they exceeded the analytical LOQ (SI.4). When one (or two) replicate(s) presented a value <LOQ, a value of ½ LOQ was assigned to calculate the average.

For the study of vermicompost (VERMI), phthalate concentrations in the pristine green PE bags (cut in tiny pieces, using clean scissors and tweezers) were compared with those measured in microplastics that formed from this material ("green PE fragments") along with "other plastic fragments" and depurated earthworms. Depurated worms and plastics were analysed in triplicate for DEHP and phthalate transformation products using the method described above. Worms underwent the same sample preparation as BBFs (freeze-drying, homogenization, enzymatic digestion).

2.8. Statistical tests

The comparisons of plastic counts, plastic masses, DEHP concentrations and transformation product concentrations between groups (or sub-groups) of BBFs were conducted using one-way ANOVA ($p = 0.05$) followed by a post-hoc Tukey test ($\alpha = 0.05$).

3. Results and discussion

3.1. Number of microplastics in BBFs

Overall, the total number of microplastics was shown to significantly increase (ANOVA, $F(2,19) = 15.95$, $p < 0.01$, Tukey test, $p < 0.04$) from AgriFoodInduWaste-BBFs (15 - 258 microplastic particles g^{-1} d.w.) to SewSludge-BBFs (59 - 1456 microplastic particles g^{-1} d.w.) to Biowaste-BBFs (828 - 2912 microplastic particles g^{-1} d.w.) (Fig. 1, SI.3 for all fragment counts). The 5 BBFs within the two last categories (i.e., 3 SewSludge-BBFs and 2 Biowaste-BBFs) were analysed in triplicates. The variability in total fragment counts among replicates was lower for the two BBFs that went through biogasification (relative standard deviation, RSD, VERMI: 2 % and PRV: 16 %) than for those composted (PLP: 54 %) or dried (NNP: 54 % and RAN: 94 %). A similar trend was observed when considering the variability in fragment counts for individual plastic types (SI.3); mean RSDs of the dominant plastic types (i.e., PE, PVC, PS, PET, PACR, copolymers) in case of digested material (VERMI: 35 %, PRV: 51 %) were lower than RSDs for non-digested material (NNP: 69 %, PLP: 108 %, RAN: 109 %). This is most probably because anaerobic digester content is mixed to provide uniformity of the process (Kumar and Ramanathan, 2021).

The lower level of microplastics found in AgriFoodInduWaste-BBFs is consistent with earlier observations made by Steiner et al. (2023) and Weithmann et al. (2018) who found that agricultural energy crop digesters contained significantly less plastics than fertilizers from biowaste treatment plants. The high level of microplastics in Biowaste-BBFs can be explained by the frequent contamination of biowaste by plastics sourced from bags and foils from packaging, resulting from improper disposal (Porterfield et al., 2023; Steiner et al., 2023; Weithmann et al., 2018). Improper sorting at the household level is not the only cause of plastics in AgriFoodInduWaste-BBFs (Alvarez et al., 2008; Friege and Eger, 2022); the unpacking process of wasted food from industrial and commercial settings (e.g., date-expired food) results in imperfect separation efficiency (do Carmo Precci Lopes et al., 2019; Öling-Wärnå et al., 2023; Porterfield et al., 2023). With regard to SewSludge-BBFs, the relatively high levels of microplastics can be explained by the fact that wastewater treated by WWTPs is contaminated by various types of microplastics coming from synthetic textiles (detachment during washing) and personal care products such as soaps, toothpaste, and facial scrubs. Most of them are retained in the sewage sludge because of the high removal efficiency of treatment processes (35–58 % in preliminary treatment techniques in the form of screening and skimming, 97.8 % after primary/secondary treatment such as decantation and biological treatment, and >99 % when a tertiary treatment, e.g., ultrafiltration, is added) (Carr et al., 2016; Golwala et al., 2021; Mahon et al., 2017; Murphy et al., 2016; Reddy and Nair, 2022; Sun et al., 2019).

Comparing the numbers of microplastics present in the studied BBFs with those provided in literature on fertilizers is difficult because standard microplastic detection methods (e.g., minimal size of fragments) have not yet been established (Gui et al., 2021; Porterfield et al., 2023), and because most of these types of fertilizers have not yet been analysed for microplastics (previous studies have mostly focused on raw sludges and biowaste). A recent review showed that particle count in composts and digestates from green and food waste (i.e., biowaste) spanned as much as seven orders of magnitude, ranging from 0.012 to 43′000 particles g^{-1} d.w. for composts, and from 0.07 to 39′000 particles g^{-1} d.w.

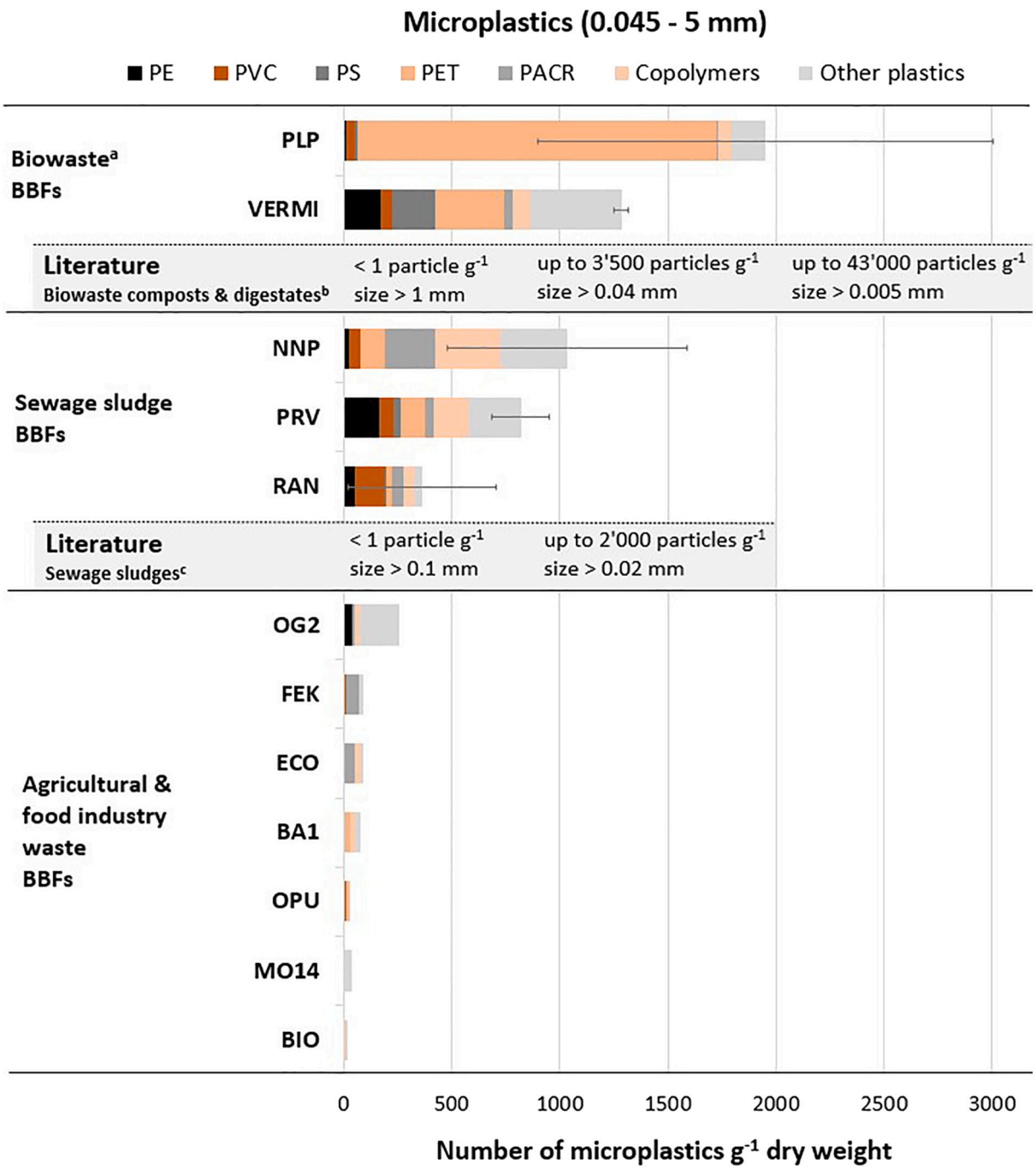

Fig. 1. Microplastic (0.045–5 mm) count (microplastic particles g^{-1}) in 12 bio-based fertilizers issued from biowaste (Biowaste-BBFs), sewage sludge (SewSludge-BBFs), and agricultural & food industry waste (AgriFoodInduWaste-BBFs). PE: polyethylene, PVC: polyvinylchloride, PS: polystyrene, PET: polyethylene terephthalate, PACR: polyacrylate. [a] Biowaste was defined as biodegradable garden and park waste, as well as food and kitchen waste. [b] Meixner et al., 2020; Öling-Wärnå et al., 2023; Porterfield et al., 2023; Schwinghammer et al., 2020; Weithmann et al., 2018. [c] Cunsolo et al., 2021; Liu et al., 2019; Öling-Wärnå et al., 2023; Sujathan et al., 2017; Zhang et al., 2021, 2020. The total number of microplastic particles is significantly different between the three groups of BBFs (ANOVA, $F(2,19) = 15.95$, $p < 0.01$, Tukey test, $p < 0.04$).

for digestates (Porterfield et al., 2023). Unsurprisingly, the particle count depended on the studied range of particle size: only a maximum of 0.024 and 0.19 particles per kg were reported by Schwinghammer et al. (2020) and Weithmann et al. (2018) for plastics >1 mm, whereas 30 and 39′000 particles g^{-1} were reported by Edo et al. (2022) and Meixner et al. (2020) for plastics >0.025 and 0.005 mm, respectively. A study on biowaste-based digestates that focused on a similar range of particle size as the present study (0.045 to 5 mm) reported up to 3′500 and 240 particles g^{-1} before and after a sieve step over 8 mm, respectively (Öling-Wärnå et al., 2023). With about 1′300 (VERMI) and 2′000 (PLP) counts g^{-1}, the results obtained for the biowaste-BBFs here are thus in accordance with this data from literature. In sewage sludges, literature values of microplastics concentration varied from 0.024 to 2′000 particle g^{-1} d.w. (Collivignarelli et al., 2021; Hatinoğlu and Sanin, 2021; Rolsky et al., 2020; Sun et al., 2019). The highest number of microplastics (> 200 microplastic particles g^{-1} d.w.) were found in studies that included microplastic sizes <0.1 mm (Cunsolo et al., 2021; Liu et al., 2019; Sujathan et al., 2017; Sun et al., 2021) whereas the lowest number of

microplastics (< 1 microplastic particle g^{-1} d.w.) were most of the times found in studies that focused on higher size ranges (e.g. > 0.1 mm) (Zhang et al., 2020, 2021). In a recent study using a range of particle size (0.04 to 5 mm) similar to the present study, sewage sludge-based digestates were shown to contain 75 to 1′500 microplastic particles g^{-1} d.w. (Öling-Wärnå et al., 2023). With about 370 (RAN), 820 (PRV), and 1′000 (NNP) microplastic particles g^{-1} d.w., the SewSludge-BBFs were thus in the range reported for sludges. The technologies used to produce BBFs from sludges do not seem to reduce the number of microplastics, confirming speculations in Nizzetto et al. (2016) regarding the inability of common processing steps (e.g., drying, pasteurization, composting, etc.) to reduce the microplastic content of WWTP sludges during the production of SewSludge-BBFs.

A striking contrast between Biowaste-BBFs and SewSludge-BBFs is the 2 to 3 order of magnitude difference (ANOVA, $F(1,12) = 7.82$, $p < 0.01$, Tukey test, $p = 0.016$) regarding the number of plastics >1 mm (Fig. 2A). Previous studies also reported dominance of microplastics <1 mm in sewage sludges (> 85 % of all microplastics) (Bretas Alvim et al., 2020; Liu et al., 2019; Magni et al., 2019) whereas plastics >1 mm were higher in number and mass in biowaste-based fertilizers (Braun et al., 2021). This difference can most probably be attributed to the input paths. Wastewaters receive plastics mainly from street runoff that contain small items from abrasion, or from municipal and industrial effluents that contain small plastics from personal care products and washing machines (Braun et al., 2021; Carr et al., 2016; Ziajahromi et al., 2016), whereas biowaste receives bigger plastics that are originated from improper waste disposal and are fragmented during composting/digestating processes (Gui et al., 2021; O'Connor et al., 2022).

3.2. Types of polymers in BBFs

The types of plastics found in BBFs presented differences between the waste origin categories. Polyethylene terephthalate (PET) was the predominant polymer type in Biowaste-BBFs (85 % in PLP, 25 % in VERMI), and was mostly present in the form of fibres (58 % for PLP, 51 % for VERMI). Polystyrene (PS), polyethylene (PE) and polypropylene (PP) were also notably present in VERMI (15 %, 13 %, 11 %, respectively), with low fibre fractions in PS (6 %) and PE (22 %) and a high fibre fraction in PP (71 %). Previous studies reported PE, PP and PS as the most frequent polymers in biowaste-based fertilizers; PET was also frequently identified, although in lower proportions (4–16 %; Schwinghammer et al. (2020); Sholokhova et al. (2022); Weithmann et al. (2018)) than in the present study (25–85 %). The dominance of these four polymers in Biowaste-BBFs is consistent with them being frequently used as food packaging plastics which are often not efficiently removed before composting/digestion (see Section 3.1). The high fraction of PET fibres in the studied Biowaste-BBFs could indicate a hitherto unreported issue. It is speculated that particularly high PET contamination in the Biowaste-BBF could stem from i) wrongly sorted packaging (e.g., bottles, jars, tubes, trays, blisters, bags and snack food wrappers), and ii) the presence of biodegradable plastics with chemical structure close to PET (e.g., Polybutylene Adipate Terephthalate which also has ester bonds and a benzene ring structure), such that their true polymer composition could not be isolated with the FT-IR method used. Biodegradable plastics were shown to produce microplastics upon suboptimal biodegradation conditions in the environment (Griffin-LaHue et al., 2022; Liao and Chen, 2021); the residence time of all microplastics in a digestion system or soil before complete mineralization would

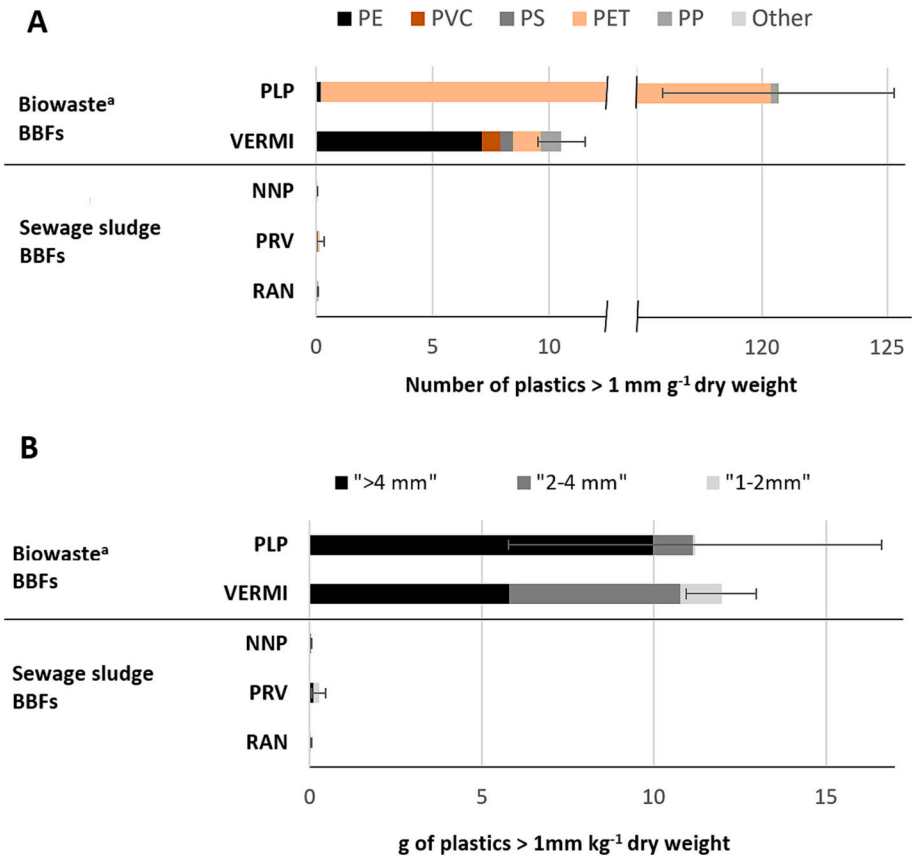

Fig. 2. Count (A) and mass (B) of plastic >1 mm in the five most contaminated bio-based fertilizers (BBFs), i.e., biowaste BBFs (PLP and VERMI) and sewage sludge BBFs (NNP, PRV and RAN). PE: polyethylene, PVC: polyvinylchloride, PS: polystyrene, PET: polyethylene terephthalate, PP: polypropylene. [a] Biowaste was defined as biodegradable garden and park waste, as well as food and kitchen waste. Both the count (ANOVA, $F(1,12) = 7.82$, $p < 0.01$, Tukey test, $p = 0.016$) and mass (ANOVA, $F(1,13) = 134$, $p < 0.01$, Tukey test, $p < 0.01$) of plastics were significantly higher in Biowaste BBFs than in Sewage Sludge BBFs.

depend on local conditions. As a consequence, biodegradable plastics were previously found in Biowaste-BBFs (Quecholac-Piña et al., 2020; Steiner et al., 2023, 2022; Zhang et al., 2018). These microplastic might be (wrongly) identified as PET by FT-IR because of the similar structure as shown in Supplementary Information SI.7 where spectra of PET and Mater-Bi bio bags are compared (Mater-Bi bio bags are used to collect one quarter of the household organic waste of VERMI).

In contrast to the two Biowaste-BBFs, the three SewSludge-BBFs (i.e., RAN, PRV and NNP) were not particularly dominated by food packaging polymers (in average PE < 20 %, PP < 4 %, PS < 5 %, PET <15 %), especially for exclusively sludge-based NNP (PE: 2 %, PP: 1 %, PS: 0 %, PET: 11 %). Microplastics in NNP predominantly consisted of ethylene acrylic copolymer (AEM) and poly(11-bromoundecyl acrylate) (PBA). PRV was also significantly contaminated by AEM (19 %; in addition to PE, 20 %, and PET, 14 %), whereas RAN was characterised by high proportions of polyvinyl chloride (PVC; > 50 % in one replicate) and polyacrylates (PACR; > 30 % in one replicate). AEM is used in (automotive) seals, gaskets and hoses that have to be resistant to transmission fluids; their strong presence has been shown in sludges in a previous study (Magni et al., 2019). PBA has previously been reported as a dominant plastic in non-digested and non-refined sludges, with unclear potential sources (Xu et al., 2020); like other PACR, it could derive from daily products such as shower gels, peelings, waterproof sunscreen, lipsticks, and paints (Bayo et al., 2020). PVC raised concern because it is seen as the most hazardous microplastic, with strong mutagenicity and carcinogenicity (Wei et al., 2019). These present results indicate that heavier and potentially more toxic microplastics can be found in SewSludge-BBFs than in Biowaste-BBFs, contrary to what has been reported by previous studies (Golwala et al., 2021; Hatinoğlu and Sanin, 2021). The types of plastics depend on the initial characteristics of wastewaters. In the present study, the contribution of plastics from grey water was probably low, as indicated by the low percentage of fibres (NNP, 18 %, PRV: 19 %, RAN: 31 %) that usually derived mostly from the washing of synthetic clothing (Collivignarelli et al., 2021; Prata, 2018).

As AgriFoodInduWaste-BBFs contained low number of microplastics, trends in the types of polymers were difficult to detect. However, non-packaging plastics dominated microplastic contents in these BBFs (e.g., > 50 % melamine in OG2, > 45 % of PBA in FEK and ECO), except for OPU which was dominated by PE, PP and PET.

3.3. Mass of plastic in BBFs

3.3.1. Measured mass of larger (> 1 mm) plastics

The mass of plastics >1 mm was low in SewSludge-BBFs (< 0.3 g kg^{-1}), but significantly higher in Biowaste-BBFs (> 10 g kg^{-1}) (ANOVA, $F(1,13) = 134$, $p < 0.01$, Tukey test, $p < 0.01$). In the European Union, fertilizers should not contain >3 g kg^{-1} d.w. of plastics above 2 mm (regulation 2019/1009 EU, 2019). The regulation states that the limit will be reduced to 2.5 g kg^{-1} d.w. from 16 July 2026, and will be re-assessed by 16 July 2029 to take into account the progress made with regards to separate collection of biowaste. In some countries, stricter regulation on the amount of plastic (and foreign matter) is enforced. In Austria, the limit for plastics >2 mm is set at 2 g kg^{-1} d.w. (BGBl. II Nr. 292/2001), while in Germany, a limit of 1 g kg^{-1} d.w. for particles >1 mm is used (Düngemittelverordnung, DüMV). Norway set up a higher limit, i.e., plastic, glass or metal pieces >4 mm shall not amount to >5 g kg^{-1} d.w. (Forskrift om gjødsel varer mv. av. organisk opphav, 2023). The two Biowaste-BBFs (VERMI and PLP) exceeded the EU threshold, with the Norwegian BBF (VERMI) also exceeding the less restrictive national limit. In contrast, the SewSludge-BBFs (NNP, PRV, RAN) did not exceed any of the above-mentioned limits (Fig. 2B, SI.8).

Studies that measured (or calculated) the mass of plastics in biowaste-based fertilizers often reported high concentrations of plastics. While Braun et al. (2021) and O'Brien (2019) reported mass of plastics below the 3 g kg^{-1} EU threshold (1.35 g kg^{-1} and 2.5 g kg^{-1} of plastics >1 mm in biowaste-based composts and manure/biowaste-based digestates, respectively), Scopetani et al. (2022) found mass of plastics of 6.53 g kg^{-1} for plastics >5 mm in biowaste-based composts and Öling-Wärnå et al. (2023) measured 11.2 g kg^{-1} plastic in biowaste digestates (before sieving). In the latter case, all plastic sizes were included (> 0.05 mm) but the contribution of small microplastics (< 1 mm) has been shown to be negligible in Biowaste-BBFs (0.01 % to 1.3 % of the total plastic mass) (Braun et al., 2021). Although biowaste is often mechanically sorted before composting/digestion, the initial mass of plastics can be so high that thresholds are exceeded. For example, do Carmo Precci Lopes et al. (2019) calculated concentrations up to 56 g kg^{-1} in the initial biowaste and about 6 g kg^{-1} in the final Biowaste-BBFs. Source-sorting is thus extremely important (Rodrigues et al., 2020), as without it plastic content in mechanically sorted organic waste can exceed 100 g kg^{-1} (Cesaro et al., 2016).

3.3.2. Calculated mass of small (< 1 mm) microplastics

Only very few studies have so far determined the mass of plastics in sewage sludge (or biosolids). Ng et al. (2018) estimated concentrations between 9 and 63 g of microplastics kg^{-1} d.w. in Australian biosolids, far above the concentrations of plastics >1 mm presently measured in the SewSludge-BBFs (NNP: 0.04 g kg^{-1}, PRV: 0.26 g kg^{-1}, RAN: 0.03 g kg^{-1}). This raises the question of the magnitude of the mass contribution of smaller microplastics (<1 mm) in SewSludge-BBFs. To this end, herein the mass of small plastics (<1 mm) from the plastic counts was conservatively estimated. Liu et al. (2019) showed that 80 % of the small microplastics (<1 mm) in sludges consisted of those in the size range 0.02–0.3 mm. Thus, an average surface area of $0.15 \times 0.15 = 0.0225$ mm^2 was used (see photographs in SI.9). As all plastics >1 mm were encountered in the form of thin layers or fibres, it was assumed that this was also the case for plastics <1 mm and a value of 0.05 mm for the thickness was used. Thus, a volume of $0.0225 \times 0.05 = 0.001125$ mm^3 was used as a proxy for each particle. This volume was multiplied by the density of the respective polymer and the number of plastics >0.045 mm (after subtraction of the number of plastics >1 mm). Estimated masses of small microplastics (< 1 mm) were 1.40, 1.05 and 0.51 g kg^{-1} for NNP, PRV and RAN, respectively. The total mass of plastics >0.045 mm in SewSludge-BBFs (NNP: 1.44, PRV: 1.31, RAN: 0.54 g kg^{-1}) was thus lower than the actual (3 g kg^{-1}) and future (2.5 g kg^{-1}) EU limits for plastics >2 mm. Using the same approach to estimate the mass of microplastics in AgriFoodInduWaste-BBFs resulted in values between 0.02 and 0.3 g kg^{-1}, revealing that even conservative estimates were at least 10 times lower than the EU thresholds for plastics >2 mm.

3.4. Estimated amounts of plastics on agricultural soils

As a worst case, the estimations of the number and mass of plastics entering agricultural soils through BBFs were based on maximum allowed application rates. The EU Nitrate Directive 91/676/EEC allows a maximum of 170 kg N ha^{-1} y^{-1}, generally representing the limiting factor for application rates (Amery and Schoumans, 2014; Collivignarelli et al., 2019). Taking into account N content in BBFs (see SI.1), maximum allowed application rates were calculated to range from 1.1 (OG2, N content of 15 %) to 13.1 t ha^{-1} y^{-1} (VERMI, N content of 1.3 %). Note that recommended BBF application rates are approximately half these values for AgriFoodIndustWaste-BBFs and SewSludge-BBFs (see recommended values provided by producers in SI.1) and twice these values for biowaste-BBFs (Braun et al., 2021). Based on these rates and the number of microplastics counted in BBFs (15'000 to 2'912'000 microplastics kg^{-1}, see Section 3.2), the estimated number of microplastics (0.045–5 mm) entering agricultural soils ranged between 3.3×10^7 and 3.9×10^8 microplastics ha^{-1} y^{-1} for AgriFoodInduWaste-BBFs, were in the 10^9 range for SewSludge-BBF, and in the 10^{10} range for Biowaste-BBFs (Table 2). Multiplying the maximum allowed BBF application rates by the plastic concentrations measured for plastics >1 mm (0.03 to 12.0 g kg^{-1}, see Section 3.3) resulted in releases of 95 to

Table 2
Estimated number of microplastics and mass of plastic released to agricultural soils via agricultural & food industry waste BBFs, sewage sludge BBFs and biowaste BBFs when maximum allowed application rates are used (1.1–13.1 t^{-1} ha^{-1} y^{-1}), and resulting expected number of microplastics and mass of plastic in the top layer (30 cm) of soil.

		Agricultural & food industry waste BBFs	Sewage sludge BBFs	Biowaste BBFs
Number of microplastics released to soil (microplastic particles ha^{-1} y^{-1})		3.3×10^7	1.9×10^9	1.1×10^{10}
		-3.9×10^8	-3.9×10^9	-1.7×10^{10}
Mass of plastic released to soil (kg ha^{-1} y^{-1})[a]	> 1 mm	n.d.	0.1–1.2	95–156
	> 0.045 mm	0.04–0.6	2.8–6.2	n.d.
Expected number of microplastics in soil (microplastic particles kg^{-1})		8–101	478–992	4303–4231
Expected mass of plastic in soil (mg kg^{-1})[a]	> 1 mm	n.d.	0.03–0.31	24.3–40.1
	> 0.045 mm	0.01–0.15	0.7–1.6	n.d.

The amount of soil in the top layer (30 cm) was determined using a density of 1.3 g cm^{-3} (i.e., 3′900’000 kg ha^{-1}).
[a] Mass of plastic >1 mm were measured, and mass of plastic between 0.045 and 1 mm was calculated from the number of particles, the density of the polymers and the estimated average volume of particles (see Section 3.3).

156 kg ha^{-1} y^{-1} for Biowaste-BBFs and between 0.1 and 1.2 kg ha^{-1} y^{-1} for SewSludge-BBFs. When considering plastic concentrations estimated for all plastics (0.54 to 1.44 g kg^{-1} for SewSludge-BBFs 0.02 to 0.3 g kg^{-1} for AgriFoodInduWaste-BBFs, see Section 3.3), releases of plastics from SewSludge-BBFs ranged from 2.8 to 6.2 kg ha^{-1} y^{-1} and those from AgriFoodInduWaste-BBFs from 0.04 to 0.6 kg ha^{-1} y^{-1}.

A few studies previously estimated the number and mass of plastics entering agricultural soils via Biowaste-BBFs or SewSludge-BBFs. Based on relatively high application rates (up to 35 t ha^{-1} y^{-1}), Braun et al. (2021) estimated that composts could release up to 1.6×10^6 plastic items ha^{-1} y^{-1}, amounting to 47.5 kg ha^{-1} y^{-1}. This is respectively 10′000 and 2–3 times lower than our estimates for Biowaste-BBFs (i.e., 10^{10} microplastics ha^{-1} y^{-1} and 95–156 kg ha^{-1} y^{-1} at an application rate of 8.5 to 13.1 t ha^{-1} y^{-1}), probably because smaller microplastics (< 0.3 mm) – that do have a high impact on the total microplastic count but not on the plastic mass – were not considered in Braun et al. (2021). Using a restricted application rate (1.7 t ha^{-1} y^{-1}), Bläsing and Amelung (2018) estimated that plastics entering agricultural soils via sludges could reach 40.8×10^6 microplastics ha^{-1} y^{-1}, which is 25 times lower than our estimate of SewSludge-BBFs application (i.e., 10^9 microplastics ha^{-1} y^{-1}). Their estimates were however based on plastic concentrations (1×10^3 to 2.4×10^4 g kg^{-1}) that did not take fibres into account (Mintenig et al., 2017). These examples underscore the issue of the lack of standardized methods for interstudy comparability.

Expected plastic concentrations in agricultural soils after one BBF application were calculated assuming that plastic items do not infiltrate to deeper soils and are mostly restricted to the 30-cm surface layer (Sa’adu and Farsang, 2023; Schell et al., 2022; Zhu et al., 2019a). The number and mass of plastic entering the soils (when maximum allowed applications rates are used) were divided by the amount of soil (3′900’000 kg ha^{-1} for a soil with a density of 1.3 g cm^{-3}). Calculated concentrations in soils ranged between 8 and 101 microplastics kg^{-1} for AgriFoodInduWaste-BBFs, 478 and 992 microplastics kg^{-1} for SewSludge-BBFs, 4303 and 4231 microplastics kg^{-1} for Biowaste-BBFs. These concentrations were in accordance with results previously reported for agricultural soils, i.e., typically between 10 and 5×10^3 (Corradini et al., 2021, 2019; Porterfield et al., 2023; Wang et al., 2021; Yang et al., 2021) and up to 4.3×10^4 and 6.2×10^5 microplastics kg^{-1} in some studies (Zhang and Liu, 2018; Zhou et al., 2019). On a mass basis, expected concentrations of plastics (> 1 mm) in amended soils ranged between 0.03 and 0.31 mg kg^{-1} for SewSludge-BBFs and between 24.3 and 40.1 mg kg^{-1} for Biowaste-BBFs. When considering estimated releases for all plastics (> 0.045 mm), expected concentrations in amended soils ranged from 0.7 to 1.6 mg kg^{-1} for SewSludge-BBFs and from 0.01 to 0.15 mg kg^{-1} with AgriFoodInduWaste-BBFs. Note that these expected soil concentrations do not take into account the accumulation of plastics over time; concentrations have indeed been shown to increase with the number of sludge applications (Corradini et al., 2019; Zhang et al., 2020).

3.5. DEHP and phthalate transformation products in BBFs

DEHP concentrations ranged from <LOQ (0.18) to 0.69 mg kg^{-1} in AgriFoodInduWaste-BBFs, from 0.35 to 0.73 mg kg^{-1} in SewSludge-BBFs and from 1.22 to–2.5 mg kg^{-1} in Biowaste-BBFs (Fig. 3), following a similar relative sequence to the number and mass of plastics (see Sections 3.2 and 3.4, respectively). Note that DEHP concentration in Biowaste-BBFs was significantly higher than in AgriFoodInduWaste-BBFs and SewSludge-BBFs (ANOVA, $F(2,19) = 9.66$, Tukey test, $p < 0.01$ for both comparisons), but the difference of concentration between AgriFoodInduWaste-BBFs and SewSludge-BBFs was not statistically significant (Tukey test, $p = 0.73$). The EU regulation 2019/1009 (EU, 2019) does not include limit values for DEHP (and other phthalates) in fertilizers. A working document on sludge (ENV.E.3/LM) proposed a value of 100 mg kg^{-1} but was subsequently withdrawn (Hudcova et al., 2019). However some countries (e.g. Denmark and Norway) have set or recommended a limit of 50 mg kg^{-1} (Eggen et al., 2019). In all BBFs, the DEHP concentration determined in the present study was well below this limit. It was also the case when the sum of DEHP and its major transformation product, mEHP, was considered (∑DEHP & mEHP <6 mg kg^{-1}) and when all transformation products were taken into account (∑DEHP & transformation products <9 mg kg^{-1}). This is in accordance with DEHP concentration previously reported in biowaste composts (0.28–9.6 mg kg^{-1} d.w. (2007)) (Brändli et al., 2007; Marb et al., 2001; Pollak et al., 2004), but in contrast to raw sewage sludges in which the limit value of 50 mg kg^{-1} was often exceeded (6–345 mg kg^{-1} d.w.) (Anne and Paulauskiene, 2021; Aparicio et al., 2009; Eggen et al., 2019; Fromme et al., 2002; Marttinen et al., 2003). This discrepancy can be explained by two main reasons. First, aerobic biodegradation of DEHP is much faster than anaerobic biodegradation (Nas et al., 2022; Reeh and Møller, 2002). Reported half-life ($t_{1/2}$) of DEHP in food waste composting is as low as 5 days (Tran et al., 2023). Also Moeller and Reeh (2003) showed that 96 to 99 % of DEHP was degraded within standard compost retention times of 25 days, independent of process temperature. Thus, low concentration of DEHP in BBFs, even those containing significant amounts of plastics, can be explained by degradation during composting (e.g., VERMI and PLP). In contrast, aerobic processes in wastewater treatment processes are short (i.e., several hours) and sewage sludges are often anaerobically digested (Di Costanzo et al., 2021), thus explaining high DEHP concentrations reported in literature for raw sludges or sludges only undergoing anaerobic digestion. The low DEHP concentrations observed in SewSludge-BBFs might be attributed to drying and hygienisation processes. Drying and aeration of sewage sludge have been shown to significant reduce DEHP content (Gibson et al., 2007; Marttinen et al., 2004). The second reason for low DEHP concentration in all BBFs may be that the 2020 EU REACH regulation has restricted DEHP and other phthalates (DBP, BBP and DIBP) to <0.1 % by weight (EC 1907/2006, Annex XVII) in most articles containing plasticized materials. A decrease over time of DEHP concentrations in sewage sludges and other organic waste is thus expected, although to

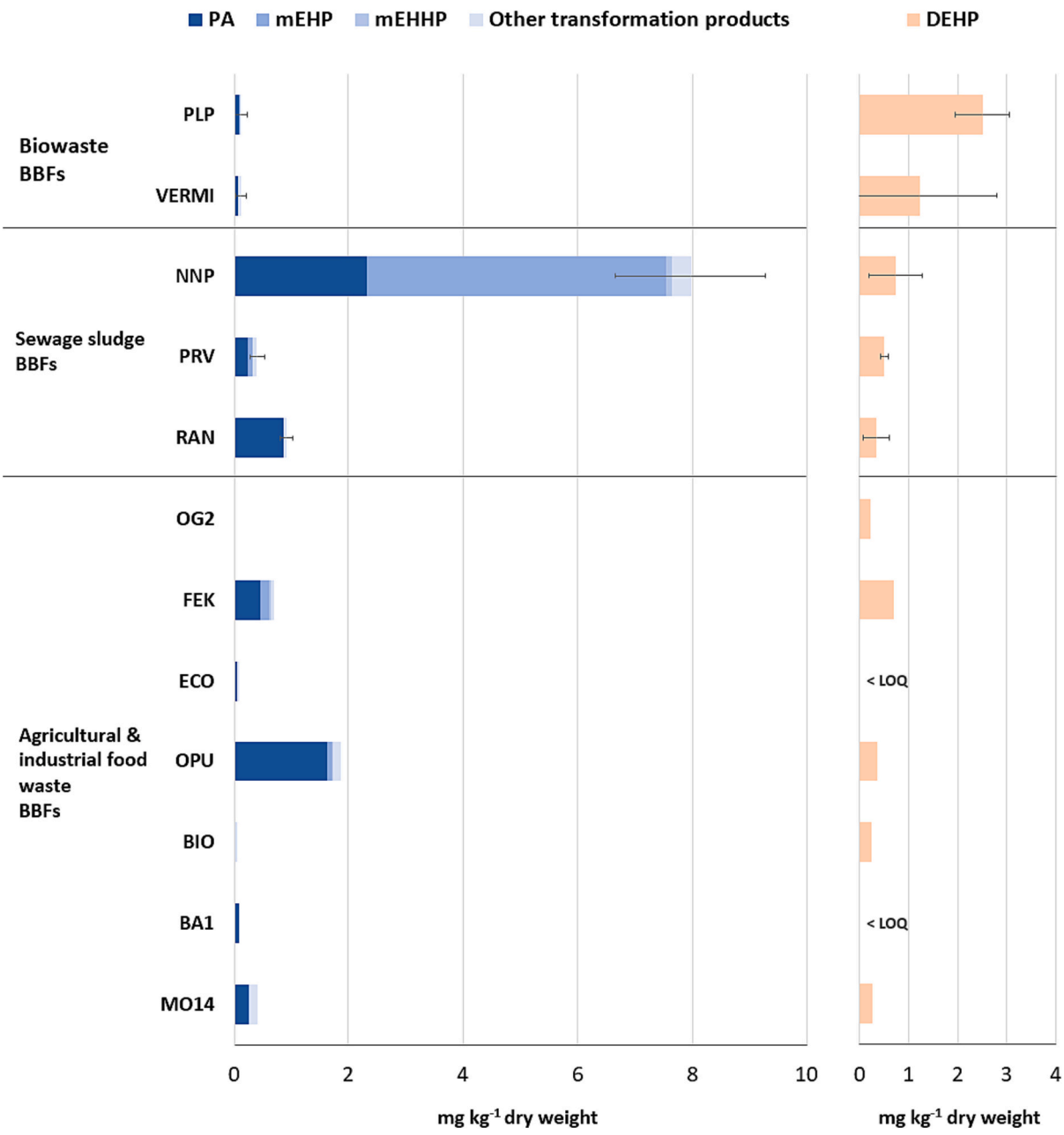

Fig. 3. Concentrations of phthalate transformation products and di(2-ethylhexyl)phthalate (DEHP) in 12 bio-based fertilizers (BBFs) from biowaste (Biowaste-BBFs), sewage sludge (SewSludge-BBFs), and agricultural & industrial food waste (AgriFoodInduWaste-BBFs). PA: phthalic acid, mEHP: mono(2-ethylhexyl)phthalate, mEHHP: mono(2-ethyl-5-hydroxyhexyl)phthalate. DEHP concentration in Biowaste-BBFs was significantly higher than in AgriFoodInduWaste-BBFs and SewSludge-BBFs (ANOVA, $F(2,19) = 9.66$, $p < 0.01$, Tukey test, $p < 0.01$), whereas the increase of concentration between AgriFoodInduWaste-BBFs and SewSludge-BBFs was not statistically significant (Tukey test, $p = 0.73$). The sum of transformation products was not significantly different between the three groups of BBFs (ANOVA, $F(2,19) = 3.59$, $p = 0.047$, Tukey test, $p > 0.05$).

our knowledge no trends have been reported since these restrictions. The amount of DEHP entering agricultural soils through maximum allowed BBF application rates has been calculated to be between 10 and 21 g ha^{-1} for Biowaste-BBFs and <3 g ha^{-1} for the other BBFs, leading to expected concentrations in soils (30 cm top layer) that are <0.01 mg kg^{-1}, and <0.001 mg kg^{-1}, respectively. This falls in the low end of ranges reported for soils (Li et al., 2023; Xu et al., 2018), and well below the (few) existing benchmarks (e.g., 2.8 mg kg^{-1} for Norway or 4.35 mg kg^{-1} in the USA) (Eggen et al., 2019; Tao et al., 2022).

The sum of the studied phthalate transformation products was very low in Biowaste-BBFs (0.1 mg kg^{-1}), reached 8 mg kg^{-1} in one SewSludge-BBFs (0.4–8 mg kg^{-1}), and varied between low and intermediate levels in AgriFoodInduWaste-BBFs (0.02–1.9 mg kg^{-1}) (Fig. 3). No significant differences between the three groups of BBFs were observed (ANOVA, $F(2,19) = 3.59$, $p = 0.047$, Tukey test, $p > 0.05$), because of the high variability withing the SewSludge and AgriFoodInduWaste groups. Except for NNP, the predominant metabolite was phthalic acid (PA), the common final metabolite of phthalates, followed by mEHP (Bang et al., 2011). This observation is in accordance with the proposed biodegradation pathway for DEHP which consists of the de-esterification of DEHP into mEHP, followed by a second de-esterification into PA (Fu et al., 2013). PA is then further oxidized (Wang et al., 2022). In Biowaste-BBFs, the very low concentrations of phthalate transformation products were probably explained by the high

residence time in composts, allowing for the biodegradation of most of mEHP and PA. Composting – in addition to significant degradation of DEHP – has thus the additional advantage of providing fertilizers with low concentrations of potentially toxic phthalate transformation products. PA concentrations in sewage sludge-based NNP and RAN (ANOVA, $F(4, 13) = 300.7$, $p < 0.01$, Tukey test, $p < 0.01$) and mEHP concentration in NNP (ANOVA, $F(4, 13) = 74.62$, $p < 0.01$, Tukey test, $p < 0.01$) were significantly higher than concentrations of PA and mEHP in Biowaste-BBFs. The first reason can be that these compounds were initially present in wastewater; phthalate transformation products are indeed excreted into the urine as metabolites (either freely or conjugated as glucuronides; Silva et al. (2003)) but might also be formed from the hydrolysis of parent phthalates in the sewer (González-Mariño et al., 2017; Tang et al., 2020). The second reason can be that biodegradation is less efficient during wastewater treatment and sludge valorisation. This is especially true in NNP where mEHP - the primary metabolite of DEHP - was more dominant than PA. Contrary to the two other SewSludge-BBFs (i.e., PRV and RAN), this BBF is only constituted of sludges. In addition, the NNP drying process, that destroys microorganisms via infrared drying, probably reduces the potential of biodegradation. An additional (aerobic) treatment should ideally be conducted to reduce mEHP as it is the most toxic metabolite of DEHP (Inada et al., 2012). Regarding AgriFoodInduWaste-BBFs, the two BBFs that are entirely made of chicken manure (OPU and FEK) contained more phthalate transformation products (1.9 and 0.7 mg kg^{-1}) than the other BBFs mostly constituted of dead animals, blood meal, meat, bone meal, or vegetables (< 0.5 mg kg^{-1}). Although phthalate transformation products can be found at low concentrations in animal tissues (Hu et al., 2016; Valton et al., 2014), they are expected to be mostly excreted via urine and thus present at higher concentration in this fluid. As avian species do not have a urinary bladder, urine is mixed with faeces, and both wastes are voided together (Pudelkiewicz et al., 1968), thus explaining why BBFs produced from chicken manure have the highest phthalates transformation products among AgriFoodInduWaste-BBFs.

Overall, phthalate plasticizers and their transformation products do not appear to be a major issue for BBFs. For BBFs that present the highest risks regarding the presence of transformation products (i.e., SewSludge-BBFs or manure-based BBFs), an additional aerobic treatment could be easily implemented. In the future, the strict DEHP regulation (EU REACH) will most probably make BBFs even safer, in that there would be lower concentrations of DEHP and phthalate transformation products in organic waste.

3.6. DEHP and phthalate transformation products in plastics and worms of vermicompost

DEHP concentrations were below LOQ in pristine green PE bags and in green PE fragments collected in the vermicompost (Fig. 4). This is not surprising since DEHP has been mostly used in PVC (Bernard et al., 2014), and its use has been substantially restricted in recent years in Europe (EU regulation 2021/2045). DEHP concentration was also below LOQ in fragments of "other plastics". On the one hand, this may be due to strict regulation. On the other hand, as DEHP was measured in VERMI, it implies that DEHP and other phthalates may have been released from plastics during the composting process. This is supported by the relatively high concentration of phthalate transformation products (3.3 mg kg^{-1}) measured in worms which are known to bioaccumulate contaminants and therefore are often used as indicators (Hu et al., 2005). The main transformation products measured in worms were PA (common metabolite of phthalates) and mEHP (major metabolite of DEHP), and - to a lesser extent - mBP (major metabolite of DBP) and mMP (major metabolite of DMP), thus reflecting that DEHP was the predominant phthalate. As DEHP is not chemically bound to polymers (Hahladakis et al., 2018; Rowdhwal and Chen, 2018), it may have been rapidly released from plastics (other than green PE bags) that could not be analysed in their pristine state (e.g., PVC). It can also be hypothesized that worm action facilitates DEHP release from plastics, especially for DEHP embedded in polymer that is otherwise less prone to degradation (Viljoen et al., 2023). The very low concentration of phthalate transformation products in plastic fragments can be explained by the low hydrophobicity of these contaminants (logKow of mBP = 2.84, mEPH = 4.73; Otton et al., 2008). As they did not significantly accumulate in plastics, the risk of plastic as a carrier of phthalate transformation products is thus very limited.

The present findings suggest that long-term impact of BBF use on soils ecosystems should ideally be assessed by determining contaminants and their transformation products in in-situ organisms from the amended soils (e.g., earthworms). Alternatively, this assessment could be conducted with ex-situ ecotoxicological tests, under acute and chronic conditions, to capture the effects of all possible contaminants and metabolites available in the BBFs (Albert and Bloem, 2023). Using calibrated passive samplers to determine the bioavailable

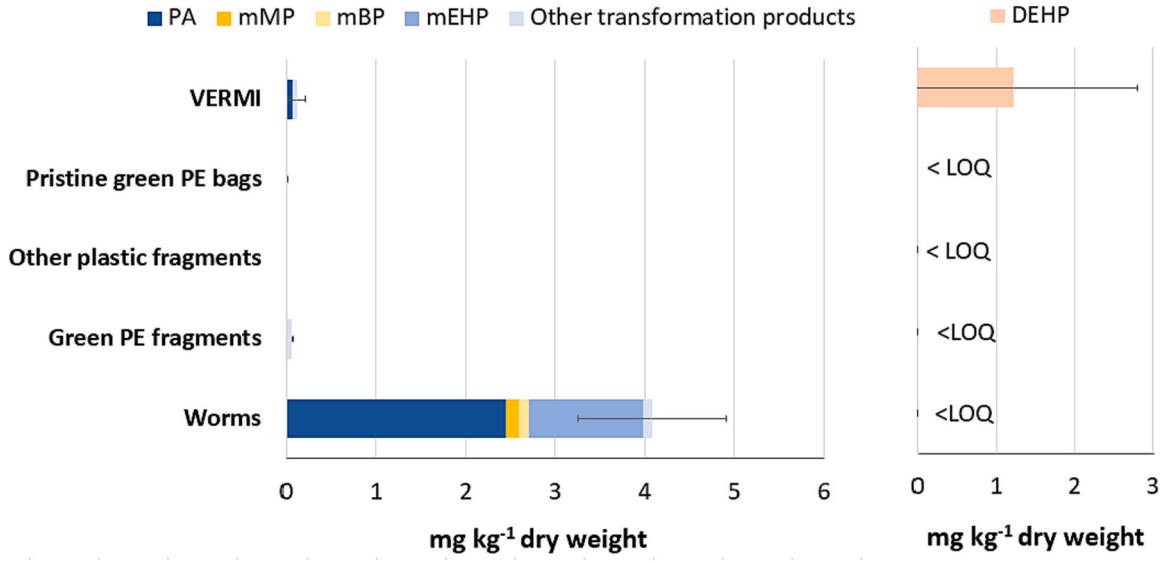

Fig. 4. Concentrations of phthalate transformation products and di(2-ethylhexyl)phthalate (DEHP) in plastics (> 1 mm) and worms from the vermicompost VERMI, as well as in pristine bags used to collect the organic household waste. PA: phthalic acid, mMP: monomethyl phthalate, mBP: mono-n-butyl phthalate, mEHP: mono (2-ethylhexyl)phthalate.

concentrations of plastic-related contaminants would also improve the risk assessment (Runde et al., 2022).

4. Conclusions

Plastics were found in all bio-based fertilizers (BBFs), regardless of waste origin. However, the mass of plastic in BBFs produced from agricultural and food industry waste (AgriFoodInduWaste-BBFs) and from sewage sludge (SewSludge-BBFs) was shown to be much lower than the EU limit value of 3 g kg^{-1} for plastics >2 mm. This is good news for the implementation of these two types of studied BBFs as an alternative to synthetic fertilizers. The recent ban of intentionally added microplastics to cosmetics and household products under EU REACH regulation will make SewSludge-BBFs even safer in the future. In contrast, BBFs derived from biowaste (Biowaste-BBFs) did not comply with the EU limit value. In the EU, biowaste accounts for >34 % of the municipal solid waste generated, representing 86 million tonnes (in 2017) (Alves et al., 2023). Using this organic waste as a fertilizer is essential to address nutrient security and organic matter supply (Cesaro et al., 2015). To prevent the contamination of Biowaste-BBFs, plastic removal efforts should occur across the life cycle of the plastics that end up in biowaste, from using less of these plastics upstream to sorting them out of the biowaste downstream. For all BBFs studied, phthalate plasticizers do not appear to be a major issue, most probably because of the strict DEHP regulations in Europe and the high phthalate degradation during most of the waste treatment processes. This is a positive indicator for the acceptability for large-scale application of BBFs within the European Union, and the safe transition to a circular economy; however, the presence of other contaminants not considered in this study should also be assessed alongside these results.

5. Perspectives

To valorise biowaste into fertilizer, up-stream strategies for preventing plastic use to down-stream strategies for source-separation of this resource need to come in place for the reduction of the amounts of plastic (Cesaro et al., 2019). Results from the present study indicate that plastic contamination in biowaste can mostly be attributed to bags used to collect food waste and packaging containing spoiled food (Cesaro et al., 2019). There could be a reduction of plastics used in food packaging and consumers should have more incentives to sort out plastic from food waste. Waste management companies handling biowaste should also avoid using plastic bags to collect food waste, or at least prioritize sieving of the collected biowaste (rather than shredding), as this has been shown to be efficient for the removal of bigger fragments instead of shredding them into smaller fragments (Braun et al., 2021; Steiner et al., 2023). However, the sustainability of often-promoted biodegradable bags can be questioned because the conditions for degradation are rarely met during composting or anaerobic digestion (do Carmo Precci Lopes et al., 2019; Ōling-Wärnå et al., 2023; Porterfield et al., 2023). Reductions in plastic use – by collecting biowaste without plastic bags and by eliminating unnecessary packaging – should be encouraged (awareness-raising), stimulated (financial incentives) or even imposed (regulations). While phthalates do not appear to be a major issue for BBFs, further work is needed to assess the risk caused by the potential presence of other persistent organic contaminants, such as *per*- and polyfluoroalkyl substances.

CRediT authorship contribution statement

Nicolas Estoppey: Writing – original draft, Supervision, Project administration, Methodology, Investigation, Conceptualization. **Gabriela Castro:** Writing – review & editing, Methodology, Investigation. **Gøril Aasen Slinde:** Writing – review & editing, Project administration, Methodology, Conceptualization. **Caroline Berge Hansen:** Writing – review & editing, Methodology, Investigation. **Mari Engvig Løseth:** Writing – review & editing, Methodology. **Katinka Muri Krahn:** Writing – review & editing, Methodology, Investigation. **Viona Demmer:** Writing – review & editing, Investigation. **Jørgen Svenni:** Investigation. **Teresa-Van-Anh Thi Tran:** Investigation. **Alexandros G. Asimakopoulos:** Writing – review & editing, Supervision, Methodology, Conceptualization. **Hans Peter H. Arp:** Writing – review & editing, Supervision, Methodology, Funding acquisition. **Gerard Cornelissen:** Writing – review & editing, Supervision, Project administration, Methodology, Funding acquisition.

Declaration of competing interest

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Data availability

Data not already provided in the Supplementary data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2024.170501.

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