



PFAS, PCBs, PCDD/Fs, PAHs and extractable organic fluorine in bio-based fertilizers, amended soils and plants: Exposure assessment and temporal trends

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HIGHLIGHTS

- Persistent organic contaminants in bio-based fertilizers, amended soils and plants
- Concentrations below thresholds, except in a pyrolyzed sewage sludge
- High EOF concentrations in sewage sludge and chicken manure BBFs
- Decreased concentrations of legacy contaminants in sewage sludge over time
- Low long-term risks for soils and low human exposure through cereal consumption

GRAPHICAL ABSTRACT

PFAS, PCBs, PAHs, dioxins & EOF in bio-based fertilizers				
	Hygienization at low temperature (< 150°C)			Pyrolysis & incineration
	Agricultural & food industry waste	Sewage sludge	Biowaste	
				
Threshold compliance (Target contaminants in BBFs)	✓	✓	✓	⚠ PAHs
Non-target PFAS (EOF in BBFs)	⚠ Manure	⚠ All	✓	✓
Amended soils and plants (Calculated & measured levels)	✓	✓	✓	✓
	Within background levels & minimal contribution to tolerable intake			

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ABSTRACT

Bio-based fertilizers (BBFs) produced from organic waste contribute to closed-loop nutrient cycles and circular agriculture. However, persistent organic contaminants, such as *per*- and poly-fluoroalkyl substances (PFAS), polychlorobiphenyls (PCBs), polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs), as well as poly-aromatic hydrocarbons (PAHs) can be present in organic waste or be formed during valorization processes. Consequently, these hazardous substances may be introduced into agricultural soils and the food chain via BBFs. This study assessed the exposure of 84 target substances and extractable organic fluorine (EOF) in 19 BBFs produced from different types of waste, including agricultural and food industrial waste, sewage sludge, and

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biowaste, and through various types of valorization methods, including hygienization at low temperatures ($<150\text{ }^{\circ}\text{C}$) as well as pyrolysis and incineration at elevated temperatures ($150\text{--}900\text{ }^{\circ}\text{C}$). The concentrations in BBFs (ΣPFOS & PFOA : $<30\text{ }\mu\text{g kg}^{-1}$, ΣPCBs : $<15\text{ }\mu\text{g kg}^{-1}$, Σ11PAHs : $<3\text{ mg kg}^{-1}$, $\Sigma\text{17PCDD/Fs}$: $<4\text{ ng TEQ kg}^{-1}$) were found to be below the strictest thresholds used in individual EU countries, with only one exception (pyrolyzed sewage sludge, Σ11PAHs : 5.9 mg kg^{-1}). Five BBFs produced from sewage sludge or chicken manure contained high concentrations of EOF ($>140\text{ }\mu\text{g kg}^{-1}$), so monitoring of more PFAS is recommended. The calculated expected concentrations in soils after one BBF application (e.g. PFOS: $<0.05\text{ }\mu\text{g kg}^{-1}$) fell below background contamination levels (PFOS: $2.7\text{ }\mu\text{g kg}^{-1}$) elsewhere in the literature. This was confirmed by the analysis of BBF-amended soils from field experiments (Finland and Austria). Studies on target legacy contaminants in sewage sludge were reviewed, indicating a general decreasing trend in concentration with an apparent half-life ranging from 4 (PFOS) to 9 (PCDD/Fs) years. Modelled cumulative concentrations of the target contaminants in agricultural soils indicated low long-term risks. Concentrations estimated and analyzed in cereal grains were low, indicating that exposure by cereal consumption is well below tolerable daily intakes.

1. Introduction

The global population is expected to reach 9.7 billion by 2050 (UNDESA, 2022). Meeting food demands requires increasing agriculture production, and fertilizers play an essential role in this effort (Alexandratos and Bruinsma, 2012; Byrnes and Bumb, 1998). Conventional limited (phosphorus) and energy-intensive (nitrogen) inorganic fertilizers are not sustainable and ill-suited to meet this challenge (Cordell et al., 2009; Erisman et al., 2008). Bio-based fertilizers (BBFs) are products obtained by recycling nutrient-rich side streams, and constitute a valid alternative to conventional fertilizers (Babcock-Jackson et al., 2023; Chojnacka et al., 2020; Svanbäck et al., 2019). BBFs can be derived from biomaterials of various origins (industry, agriculture, society) and many of them have a similar agronomical effectiveness to that of conventional fertilizers (Kurniawati et al., 2023; Sigurnjak et al., 2019, 2016; Vaneckhaute et al., 2013; Wester-Larsen et al., 2024, 2022). Promoting the use of BBFs can thus significantly reduce dependence on conventional inorganic fertilizers, supporting the transition to a circular economy (Chojnacka et al., 2020; Svanbäck et al., 2019). However, to allow for a safe circular economy, it is essential to avoid recirculating persistent organic contaminants that can be present in the organic waste and that may accumulate in the environment and food chain. Historical applications of sewage sludges have been reported to contaminate agricultural land, groundwater, and adjacent bodies of water with *per*- and poly-fluoroalkyl substances (PFAS), polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs) and polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) (Johnson, 2022; Lindstrom et al., 2011; Mackiewicz-Walec and Krzbiec, 2020; Pepper et al., 2021a; Röhrer et al., 2021; Washington et al., 2010a; Weber et al., 2018a). Concerns about the presence of these contaminants in BBFs have been raised because exposure to them is associated with a variety of health issues including various cancers and effects on the immune, nervous, and endocrine systems (Faroon et al., 2003; Fenton et al., 2021; Steenland et al., 2020; Van den Berg et al., 2006).

PFAS are a group of thousands of compounds that are composed of a fluorinated alkyl chain and a polar head group that give them surfactant-like properties and extreme chemical and thermal stability (Buck et al., 2011; Dickman and Aga, 2022). They have been used in a wide range of products including fire-fighting foams, non-stick cookware, and fast-food containers (Glüge et al., 2020; Prevedouros et al., 2006). The growing health concerns associated with PFAS have been reflected in the drastic decrease of the tolerable intake set by the European Food Safety Authority (EFSA), i.e., from 150 and $1500\text{ ng kg}^{-1}\text{ body weight (kg}^{-1}\text{ bw}^{-1})$ per day for perfluorooctanoic acid (PFOA) and perfluorooctanesulfonic acid (PFOS), respectively, in 2013, to $4.4\text{ ng kg}^{-1}\text{ bw}^{-1}$ per week for the sum of PFOS, PFOA, perfluorononanoic acid (PFNA) and perfluorohexanesulfonic acid (PFHxS) in 2020 (Schrenk et al., 2020). Some legacy PFAS have been regulated and their use restricted (e.g., PFOA, PFOS, and PFHxS are listed in the Stockholm Convention on persistent organic pollutants (POPs)), but these have often been substituted by other, probably equally problematic, PFAS

such as 6:2 chlorinated polyfluoroalkyl ether sulfonic acid (F-53B), hexafluoropropylene oxide dimer acid (GenX) or dodecafluoro-3H-4,8-dioxananoic acid (ADONA) (Munoz et al., 2019). Due to the very large number of different PFAS, it has been shown that the usual 30 to 60 target compounds account for only a small fraction of all fluorinated compounds present in various samples including human serum (Aro et al., 2021b) and sewage sludges (Aro et al., 2021a; Spaan et al., 2023). For these reasons, a proposal for a general restriction of all PFAS has been submitted to the European Chemical Agency (ECHA) in 2023. PCBs are also a group of synthetic compounds. They have been used extensively as insulating fluid in capacitors and transformers, as well as plasticizers in building materials such as paints and sealants (Erickson and Kaley, 2011; Reddy et al., 2019). Their ability to biomagnify and the extreme toxicity of the dioxin-like congeners led to a general ban in the 1980s. However, because of their persistence, they are still ubiquitous in the environment. PAHs and PCDD/Fs - contrary to PFAS and PCBs - are not manufactured intentionally but are mostly formed as unintended by-products from incomplete combustion and, for PAHs, can be naturally present in coal derivatives and petroleum. Humans are mostly exposed to PAHs and PCDD/Fs via inhalation and diet (ATSDR, 1995; Marquès and Domingo, 2019).

PFAS, PCBs, PAHs and PCDD/Fs end up in organic waste (e.g., wastewater, manure, municipal organic waste) through various processes including excretion, laundry washing, food or green waste, improper sorting, and release from industrial point sources producing, using or treating these compounds (Andersen et al., 2008; Bolan et al., 2021; Gottschall et al., 2017; O'Connor et al., 2022; Thakali et al., 2022; Thompson et al., 2022). Due to their persistence, conditions encountered in many valorization methods (e.g., drying, composting and anaerobic digestion processes) result in little ($<50\%$) (Patureau and Trably, 2006; Siebielska and Sidelko, 2015) to no reduction in the amounts of persistent substances (Brändli et al., 2007, 2005; Lakhdar et al., 2009; Lazzari et al., 1999). In contrast, thermal conversion of organic waste by pyrolysis or incineration has been shown to provide good removal efficiency. Only small percentages of PFAS, PCBs and PCDD/Fs initially present in the waste have been measured in chars produced by different pyrolysis systems ($500\text{--}800\text{ }^{\circ}\text{C}$ in anoxic conditions) (Kundu et al., 2021; McNamara et al., 2023; Sørmo et al., 2024, 2023), and a significant reduction was reported after combustion in a full-scale incinerator ($850\text{--}1100\text{ }^{\circ}\text{C}$ in oxic conditions) (Björklund et al., 2023; Loganathan et al., 2007). Depending on the operating conditions (and the types of feedstocks), by-products such as PAHs and PCDD/Fs can form during thermal conversion, with pyrolytic conditions favouring the formation of PAHs and PCDD/Fs (Altarawneh et al., 2009; Chagger et al., 2000; Sørmo et al., 2024; Wang et al., 2017). Concentrations in BBFs are thus expected to depend on the waste origin and the valorization process (especially the temperature), but not in the same way for all persistent organic substances. Studies investigating PFAS in diverse BBFs are rare (Kim Lazzano et al., 2020), and none have focused simultaneously on PFAS, PCBs, PAHs and PCDD/Fs in the same BBFs. Moreover, limit values of these substances in BBFs have only been

defined at country levels but not in EU regulations. Directive 86/278/EEC (biosolids) and Regulation 2019/1009 (fertilizing products) regulate heavy metals but not organic pollutants. Concentrations of regulated/legacy contaminants such as PFOS, PCBs, PCDD/Fs, and PAHs are expected to decrease with time (e.g., an apparent half-life of 10 for PCBs and 12 years for PCDD/Fs was reported for the 1993–2012 period in sewage sludges (Zennegg et al., 2013)) but reported temporal trends in organic waste are scarce (Gewurtz et al., 2024; Ulrich et al., 2016; Zennegg et al., 2013).

The use of contaminated BBFs could constitute a risk for human and ecosystem health because plants grown in amended soils have been shown to accumulate POPs and other substances, thus representing a pathway for the trophic transfer into higher organisms, including humans (Blaine et al., 2013; Olowoyo and Mugivhisa, 2019; Pullagurala et al., 2018; Rorat et al., 2019; Wang et al., 2020). The total amount of persistent substances transferred to agricultural soils depends on BBF application rates and the pollutant concentrations in the BBF. The fraction of persistent substances available for plant uptake depends on contaminant physicochemical properties (e.g., hydrophobicity, water solubility, potential biodegradation), soil characteristics (e.g., amount and type of organic matter) and environmental conditions (e.g., precipitation and temperature) (Reid et al., 2000). For example, the amounts of bioavailable PCBs, PAHs or PCDD/Fs are lower in soils rich in organic matter and black carbon because of the strong sorption of the contaminants to these matrices (Cornelissen et al., 2005; Ortega-Calvo et al., 2015) while the amounts of short-chain PFAS rapidly decrease in rainy conditions because of their high solubility (McLachlan et al., 2019; Stahl et al., 2013a; Weidemann et al., 2022). The transfer of bioavailable substances from soils to crops, and their translocation within plants vary significantly among contaminants, plant species and plant parts (Collins et al., 2006; Ghisi et al., 2019; Lesmeister et al., 2021). More hydrophobic substances are expected to accumulate significantly in plant roots (Collins et al., 2006; Duarte-Davidson and Jones, 1996) although some of them have also been shown to translocate to other (edible) parts of the plants (Sun et al., 2019). Smaller and more hydrophilic persistent substances – such as short-chain PFAS – are more likely to be translocated to shoots (Adu et al., 2023; Krippner et al., 2014; Lesmeister et al., 2021). For PFAS, this is of particular concern because short-chain PFAS tend to be used as substitutes for long-chain PFAS, and an increasing volume of recent studies has detected PFAS in edible parts of plants (Bao et al., 2020; Brendel et al., 2018). Very few studies have assessed human exposure to POPs and other persistent substances through BBF-amended plants, representing a knowledge gap for the large-scale application of such fertilizers in the EU. Furthermore, estimations of long-term build-up of persistent substances in soils as a result of repetitive BBF amendments are lacking.

To address these collective concerns related to persistent organic substance contamination in BBF-amended soils and plants, this study assessed, for the first time, the exposure of 84 persistent organic substances from 4 classes – i.e. 7 PCBs, 16 PAHs, 17 PCDD/Fs, 44 PFAS – in 19 BBFs produced by different methods (including incineration, pyrolysis, and hygienization at lower temperatures) and from various waste materials (including sewage sludge, biowaste, and agricultural and food industry waste). Another novel element was the quantification of extractable organic fluorine (EOF) in BBFs to account for non-targeted fluorinated compounds. Estimated contents of the studied contaminants in soils and plants amended with BBFs were verified by analyzing BBF-amended soils. Literature providing concentrations of legacy contaminants in sewage sludges was reviewed to provide temporal trends and model the build-up of these compounds in BBF-amended soils. The specific aims of the study were to (i) evaluate the impact of valorization methods and waste origins on the target pollutants in BBFs, (ii) assess the compliance of concentrations in BBFs with existing national threshold values, (iii) predict the concentrations in agricultural soils after one BBF application and over time, (iv) assess the risk for consumers by comparing expected concentrations in plants with tolerable

intakes, (v) validate the estimates by measuring real samples from field trials, and (vi) review the literature on legacy PFOS, PCBs, PCDD/Fs and PAHs in sewage sludge to determine temporal change rates and assess the long-term risk for amended soils. Addressing these aims will aid the assessment of whether the use of BBFs can be considered a safe circular economic alternative, and whether specific waste types and valorization techniques require further development.

2. Materials and methods

2.1. Selection of bio-based fertilizers

Nineteen BBFs were selected to cover the main categories of waste valorization methods, i.e., hygienization techniques at temperature < 150 °C such as composting, biogasification and drying (12 BBFs), pyrolysis (3 BBFs), incineration (3 BBFs), and crystallisation (1 BBF) (see Table 1 and Supporting Information SI.1). BBFs produced by hygienization at low temperature (<150 °C) – hereafter referred to as “hygienization” – dominated the sample set because of their generally higher fertilizing values (see SI.1), especially in terms of N supply, which is the foremost limiting nutrient for crop yield (Fageria and Baligar, 2005). Crystallisation is expected to provide BBFs with low concentrations of contaminants (de Boer et al., 2018; Dong et al., 2023; Ronteltap et al., 2007), and for this reason only one struvite material was tested. The 19 selected BBFs also covered the three main categories of waste origin, i.e., green waste and livestock residues from the agricultural and food industries (11 AgriFoodInduWaste-BBFs), sewage sludge (6 SewSludge-BBFs) and biowaste (2 Biowaste-BBFs) (Table 1). A stronger focus was placed on the two first categories because the resulting BBFs – often in form of pellets – are more suitable for large-scale commercialization, compared to the digestates or composts that are usually generated from the treatment of biowaste. Information on the nutrient content, dry matter and application rates of the tested BBFs can be found in the Supporting Information (SI.1). Analyses of 7 PCBs, 16 PAHs, 17 PCDD/Fs, and 44 PFAS were first conducted on one replicate of each BBF to assess the contamination levels. Then, triplicate analyses of PCBs and PAHs as well as PFAS and EOF were conducted for the nine and thirteen most contaminated BBFs, respectively.

2.2. Preparation of BBFs

Samples were ground and homogenized using an agate mortar and pestle, which was cleaned 3 times with methanol (VWR) and pentane (Merck) between each sample. Procedural blanks were done by grinding a certified reference material made of a clean loamy soil (CLNLOAM6, Merck, Norway), at least every 10 samples. Samples were stored in the freezer in glass containers (PCBs, PAHs and PCDD/Fs) or polypropylene (PP) tubes (PFAS and EOF) until analysis.

2.3. Target PCBs, PAHs and PCDD/Fs

Samples were freeze-dried using method DIN 38414-22 (2000-09). The quantification of seven PCBs (PCBs 28, 52, 101, 118, 138, 153 and 180, additional information in SI.2) and 17 PCDD/Fs (full list in SI.2) was conducted based on the method DIN EN 16190 (2019–10). Briefly, after the addition of 13C-labeled standards (1 for each congener) to 5 g of sample, extraction was completed by accelerated solvent extraction (ASE, 180 °C, 140 bar, 10 min) with toluene. Then, the extracts were purified by column chromatography using mixed silica columns (acid, neutral, basic) and aluminium oxide column, before being measured by gas chromatography coupled to high-resolution mass spectrometry (GC/HRMS). Results for PCDD/Fs were expressed as WHO toxic equivalent (WHO-TEQ) by multiplying the concentrations of each compound by its corresponding WHO-05 toxic equivalency factor (TEF) (SI.2). The quantification of 16 PAHs (full list in SI.2) – was achieved following method DIN EN 16181 (2019–08). After the addition of seven

Table 1

Valorization methods and waste origins of the 19 selected BBFs of European origin.

Acronym ^a	Valorization method		Waste origin	
	Category	Short description	Category ^b	Short description
CGO	Crystallisation	Struvite precipitation	Sewage Sludge	Wastewater supernatant
EPH	Incineration	>850 °C, granulating	Agriculture & food industry	Sunflower husk ash
PLA	Incineration	>850 °C	Agriculture & food industry	Poultry litter ash
ADC	Incineration	900–950 °C	Sewage Sludge	Calcined phosphate from sewage sludge ash ^c
CRA	Pyrolysis	HTC, 190 °C	Agriculture & food industry	Sludge from juice-making industry
MBC	Pyrolysis	300–450 °C	Agriculture & food industry	Chicken manure
BAG	Pyrolysis	650 °C	Sewage sludge	Sewage sludge
BA1	Hygienization	Fermentation & distillation	Agriculture & food industry	Wheat and maize
MO14	Hygienization	Pelletising	Agriculture & food industry	Vegetable by-products from food industry & animal proteins
BIO	Hygienization	Pelletising	Agriculture & food industry	Meat and bones, apatite, vinasse, chicken manure, K ₂ SO ₄
OPU	Hygienization	Pelletising	Agriculture & food industry	Chicken manure
FEK	Hygienization	Drying and pressing	Agriculture & food industry	Chicken manure
OG2	Hygienization	Hydrolysis	Agriculture & food industry	Horn meal (pig bristles)
ECO	Hygienization	Pelletising	Agriculture & food industry	Blood and feather meal
RAN	Hygienization	Drying & granulating	Sewage sludge	Sewage sludge and biowaste
PRV	Hygienization	Biogasification & hygienisation	Sewage sludge	Sewage sludge and biowaste
NNP	Hygienization	Infrared drying	Sewage sludge	Sewage sludge and industrial sludge
VERMI	Hygienization	Biogasification & vermicomposting	Biowaste	Biowaste and manure
PLP	Hygienization	Composting	Biowaste	Biowaste, peat and wood chips

^a For confidentiality reasons, acronyms are used instead of full names.

^b The waste origin category refers to the characterizing raw material; some BBFs contained mixtures of wastes as they were real-world, commercially available (or in development) products.

^c Calcination is a thermal treatment whereby the substrate is exposed to very high temperatures (usually >800 °C) without melting under restricted supply of ambient oxygen, generally for the purpose of removing impurities or volatile substances.

deuterated standards (SI.2), an extraction was conducted by ASE with toluene. The extracts were purified using deactivated silica columns, dimethylformamide clean-up, and aluminium oxide columns. The 16 PAHs were quantified by GC coupled to tandem mass spectrometry (MS/MS). Limits of quantification for PCBs, PAHs, and PCDD/Fs are given in SI.2.

2.4. Target PFAS

Extraction of target PFAS was performed according to a method adapted from Knight et al. (2021) and Bräunig et al. (2019). Freeze-dried samples (1.5–2 g) were placed into 15 mL PP tubes and an internal standard mixture was added (0.03 mL of 0.1 ppm). After a minimum of 30 min, 5 mL of methanol/NH₃ solution (99:1) was added to the samples before samples were shaken on a side-to-side shaker for 30 min, then sonicated for 20 min and then centrifuged for 10 min at 2700 rpm. A second extraction was performed with 3 mL of methanol/NH₃ solution, after which the supernatants were combined and concentrated to 1 mL under a gentle nitrogen (N₂) flow at 40 °C and acidified with 0.01 mL of acetic acid. The extracts were passed through pre-conditioned (1 mL of methanol) Bond Elut Carbon cartridges (100 mg, Agilent) and collected in 1.5 mL PP vials. The extract tubes were rinsed with 0.5 mL and passed through the same cartridge and collected into the same 1.5 mL vial. Samples were concentrated to 1 mL under N₂ at 40 °C, and a 0.2 mL aliquot was transferred into a SpinX centrifuge tube (Merck), buffered (0.1 mL of 5.2 mM ammonium acetate in ultra-pure water), centrifuged (1 min, 10'000 x g), transferred to 1.5 mL PP vials and stored at –20 °C.

A total of 44 PFAS compounds were quantified using liquid chromatography (UPLC, Acquity Ultra Performance HPLC system) coupled with quadrupole time-of-flight mass spectrometry (QToF-HRMS, Xevo G2-S instrument) from Waters (Milford, MA, U.S.). Analysis included 13 perfluoroalkyl carboxylic acids (PFCAs), 10 perfluorinated alkyl sulfonic acids (PFSA), six perfluorooctane sulfonamido substances (pre-FOS), four fluorotelomer sulfonic acids (FTSA), three polyfluoroalkyl phosphate diesters (diPAP) and eight other PFAS of interest, e.g., substitutes of PFOS or PFOA (the whole list of target compounds is provided in Supplementary Information SI.3). Separation was carried out on a Acquity BEH C8 reversed phase column (100 × 2.1 mm, 1.8 µm, Waters). Acetonitrile and water with 5.2 mM NH₄OAc were used as mobile phases for chromatographic separation, using a flow rate of 0.5 mL min^{–1}. Negative ion electrospray was used as ionization source. The analytical method parameters are detailed in Supplementary Information SI.3. Three procedural blanks were subjected to the whole analysis procedure, using a certified reference material (Clean loam soil CLNLOAM6, Supelco) as matrix. In all of the procedural blanks, concentrations of target PFAS were below the analytical limit of quantification (LOQ) of the method; perfluorobutanesulfonic acid (PFBS) was found at a very low concentration (0.12 µg kg^{–1}) in one of the three blanks (SI.3). When replicate(s) presented a value <LOQ, a value of ½ LOQ was assigned to calculate the average value reported for individual and total PFAS. Recoveries were determined by spiking one sample of each matrix type (i.e., AgriFoodInduWaste-BBFs, Biowaste-BBFs, SewSludge-BBFs, soils and grains) with 2 ng of native PFAS (0.1 mL of 20 ng/mL). The recoveries of PFAS with > three fluorinated carbon atoms were acceptable in all matrices (mean recoveries in each group >77 %, see Supplementary Information SI.3). For PFAS with three fluorinated carbon atoms (i.e., PFBA and PFPrS), chromatographic quality criteria were not met. Thus an alternative extraction method based on acetonitrile - was used for these compounds (Langberg et al., 2020). Briefly, the samples (1.5–2 g) were extracted twice with acetonitrile (8 + 6 mL) both times using an ultrasonic bath (30 min) and shaking (30 min) and then concentrated under N₂. The quantification by LC-QToF-HRMS was performed using the same parameters described above; the recoveries were 104 % for PFBA and 115 % for PFPrS.

2.5. Extractable organic fluorine (EOF)

Non-target extractable organic fluorine (EOF) analysis was conducted on separate portions of all samples (1–5 g) by using the same extraction and clean-up procedures described above for the targeted PFAS analysis without the addition of internal standards. The EOF in the extracts was measured as inorganic fluoride (ions) on a Combustion Ion Chromatography (CIC) system. The CIC system consisted of an Analytik Jena combustion unit (Jena, Germany) coupled with a 920 Absorber Module and a 930 Compact Ion Chromatography (IC) Flex from Metrohm (Herisau, Switzerland). Aliquots of 0.1 mL were combusted in quartz boats at 1050 °C (several boats were needed as co-extracted alkali metals devitrified the quartz) and the combusted fluorine was absorbed in deionized water and transferred to the IC. Measured fluoride peak areas were corrected by subtracting the peak area of empty boat blanks that were injected immediately before or after the extract. Concentrations of EOF were determined from the area of the fluorine peak using PFOA (perfluorooctanoic acid) standards in methanol ($R^2 > 0.99$). Calibration quality control samples were run throughout the worklist and were within $\pm 15\%$ of the nominal concentration. The detection limit (calculated as blank mean of five procedural blanks plus three times their standard deviation) was $20.8 \mu\text{g kg}^{-1}$. Quality control samples included three procedural extraction blanks (certified reference material, CLNLOAM6, Supelco) and multiple methanol blanks analyzed together with the BBF extracts. It was verified that inorganic fluorine was not co-extracted with EOF using a sodium fluoride spike in the sample replicates. Organofluorine recovery was performed using PFOA spiked in samples replicates (BIO, ECO, RAN). The recoveries determined from the spiked samples with PFOA ranged from 83 % for SewSludge-BBFs to 7–12 % for AgriFoodInduWaste-BBFs. The low recoveries observed for the latter were slightly improved to 21–28 % when working with acetonitrile extraction.

To obtain a fluorine mass balance, the concentrations of the measured target PFAS (C_{PFAS}) were converted into fluoride concentrations and summed up (C_{F^-}) using Eqs. (1) and (2) (Aro et al., 2021a):

$$\%_{\text{F}^-} = \frac{n_{\text{F}} M_{\text{F}}}{M_{\text{PFAS}}} \quad (1)$$

$$C_{\text{F}^-} = \sum C_{\text{PFAS}} \%_{\text{F}^-} \quad (2)$$

where $\%_{\text{F}^-}$ is the mass fraction of fluorine in PFAS, n_{F} is the number of fluorine atoms in a PFAS molecule, M_{F} is the atomic mass of fluorine and M_{PFAS} is the molecular mass of PFAS.

2.6. Impact of waste origins and valorization methods, and compliance assessment

The comparisons of PCB, PAH, PCDD/F, PFAS, EOF concentrations between groups (or sub-groups) of BBFs were conducted using one-way ANOVA ($p = 0.05$) followed by a post-hoc Tukey's test ($\alpha = 0.05$) using R 4.3.2 software. Limit values of the target pollutants in fertilizers have not been defined in EU regulation. The strictest values used in individual EU countries were used for compliance assessment, i.e., $200 \mu\text{g kg}^{-1}$ for $\sum 6\text{PCBs}$ (Luxemburg, PCB 118 not included), 3 mg kg^{-1} for $\sum 11\text{PAH}$ (Denmark, naphthalene (NAP), acenaphthylene (ACY), anthracene (ANT), benzo[a]anthracene (BaA), chrysene (CHR), and dibenz[a,h]anthracene (DBaA) not included), $20 \text{ ng TEQ kg}^{-1}$ for $\sum 17\text{PCDD/Fs}$ (Luxemburg, same compounds as those studied in the present study), and $100 \mu\text{g kg}^{-1}$ for $\sum \text{PFOS}$ and PFOA (Germany) (Collivignarelli et al., 2019a; Hall et al., 2020). Note that for PCBs and PAHs, some countries have set thresholds for individual compounds that, in certain situations, may be the limiting values ($100 \mu\text{g kg}^{-1}$ for individual PCB in Germany and Croatia, 1 mg kg^{-1} for benzo[a]pyrene (BaP) in Germany, see SI.4).

2.7. Concentration in BBF-amended soils and plants

Concentrations expected in BBF-amended soils were calculated for a worst-case scenario using maximum allowed application rates of fertilizers. The EU Nitrate Directive 91/676/EEC allows a maximum of $170 \text{ kg N ha}^{-1} \text{ y}^{-1}$ as manure-based N, with N generally representing the limiting factor for application rates (Amery and Schoumans, 2014; Collivignarelli et al., 2019b). Also applying the $170 \text{ kg N ha}^{-1} \text{ y}^{-1}$ threshold to non-manure-based BBFs (see N content in BBFs in Supplementary Information SI.1), maximum allowed application rates were calculated to range from 1.1 (OG2, N content of 15 %) to $13.1 \text{ t ha}^{-1} \text{ y}^{-1}$ (VERMI, N content of 1.3 %). As BBFs with a very low N content ($\leq 1\%$) are primarily intended to supply P, applying the limit for N was deemed irrelevant. Instead in those cases, an application rate of $50 \text{ kg P ha}^{-1} \text{ y}^{-1}$ was used as a basis for the calculations (upper end of the range of allowed rates in several EU countries; Amery and Schoumans (2014)), resulting in application rates between 0.6 (ADC, P content of 8.1 %) and $1.0 \text{ t ha}^{-1} \text{ y}^{-1}$ (PLA and BAG, P content of 5.2 %). The mass of persistent organic substances entering agricultural soils was determined by multiplying these rates with the concentrations measured in BBFs. Then, expected concentrations in (initially not contaminated) soils after one BBF application were calculated by dividing the mass of contaminants entering the soils by the amount of soil ($3'900'000 \text{ kg ha}^{-1}$ for soil with a density of 1.3 g cm^{-3}), assuming that most of the studied pollutants accumulated in the top 30-cm surface layer (see discussion in Section 3.4 regarding more mobile PFAS) (Di Guardo et al., 2020; Wellmitz et al., 2023). The concentrations expected in plants were calculated based on the calculated concentrations in soils and literature-based factors (BAF). Literature-based soil porewater partition coefficient (K_d) values were used to discuss the sorption of these contaminants to soil. K_d and BAF can be found in SI.5.

To verify the calculated concentration in soils and plants, these estimates were compared to values measured in samples from two field trials conducted with a selection of the studied BBFs in 2021. Soils and maize amended with the BBFs EPH and OPU were obtained from a field trial in Langenlebern, Austria (48.32093, 16.10166), and soils and barley amended with PLP and OPU from a field trial in Jokioinen, Finland (60.863839; 23.521162); see Supplementary Information SI.6 for additional information about the sites. At both sites, each of the studied BBFs or control (no BBF application) treatments were replicated four times (complete randomized block design) and application rates were based on total P rates of 30 kg ha^{-1} . The size of each plot was $5 \text{ m} \times 10 \text{ m}$ (Jokioinen) or $6 \text{ m} \times 9 \text{ m}$ (Langenlebern). Both maize and barley were grown up to maturity and harvested. Composite soil samples after harvest from the topsoil layer were collected from each plot. The soil and grain samples were dried at 40 and 60 °C, respectively. Composite soil and grain samples from the four replicates of the specific BBF and control treatments were analyzed for PCBs and PFAS in triplicate using the same method as for BBFs (Sections 2.2 to 2.4). The comparisons of concentrations of these contaminants between amended and non-amended soils were done using a Student's *t*-test ($p = 0.05$).

2.8. Temporal trends

Studies reporting 'PFOS', 'PCB', 'PCDD/F', and 'PAH' concentrations in 'sewage sludge' and 'biosolid(s)' were investigated using these keywords in Google Scholar. This selection (37 studies for PFOS, 13 for PCBs, 11 for PCDD/Fs, 17 for PAHs, see SI.9 and SI.10) is not claimed to be fully exhaustive. However, it is expected to be representative of the last two decades of PFOS data (most recently regulated compound), and three decades of PCB, PCDD/F and PAH data (compounds banned or regulated for a longer time). Individual sample years were used when contaminant concentrations were reported annually; an average value was used for studies mentioning sampling campaigns conducted over a range of years without providing concentrations for individual years (see details in SI). In those cases where mean and median concentrations

of contaminants were not directly reported in the studies, they were calculated from individual concentrations.

3. Results and discussion

3.1. Impact of waste origins and valorization methods

The average concentrations measured in the 19 BBFs ranged from <LOQ to 14.8 $\mu\text{g kg}^{-1}$ for $\Sigma 7\text{PCBs}$, 0.01 to 23.2 mg kg^{-1} for $\Sigma 16\text{PAHs}$, <LOQ to 3.7 ng TEQ kg^{-1} for $\Sigma 17\text{PCDD/Fs}$ (Fig. 1), and < LOQ to 29.0 $\mu\text{g kg}^{-1}$ for $\Sigma 44\text{PFAS}$ (Fig. 2, all individual substance concentrations in SI.7). The results of the EOF analysis (Fig. 2) indicated that substantial amounts of fluorinated compounds are present in some BBFs (up to 492 $\mu\text{g F kg}^{-1}$), as a large fraction of F (> 85 %) was not explained by the 44 PFAS targeted in the present study (Fig. 3).

3.1.1. Hygienization

Significant differences were observed between the BBFs, depending on the valorization methods and waste origins. Among the BBFs that underwent a hygienization at low temperature (<150 °C), the average concentration was significantly lower in AgriFoodInduWaste-BBFs than in SewSludge-BBFs, by a factor of 59 for $\Sigma 7\text{PCBs}$, 5.4 for $\Sigma 16\text{PAHs}$, 10 for $\Sigma 17\text{PCDD/Fs}$, 5.4 for $\Sigma 44\text{PFAS}$ and 2.2 for EOF. Biowaste-BBF presented intermediate concentrations for all substances, neither of which were significantly different from those in either AgriFoodInduWaste-BBFs or SewSludge-BBFs, except PCB concentrations, which were significantly higher in Biowaste-BBFs (SI.8). The overall elevated concentrations observed in SewSludge-BBFs can probably be explained by the fact that the majority of PCBs, PAHs, PCDD/Fs and longer-chain PFAS present in wastewater are distributed in sludge because of their high sorption potential (Sinclair and Kannan, 2006;

Tian et al., 2012; Urbaniak et al., 2017; Zhang et al., 2019). For PCBs, the higher average contamination level in biowaste-BBFs was caused by the elevated concentrations measured in one specific BBF (PLP), which could be explained by random organic contaminant impurities that typically occur in some biowaste streams (Amlinger et al., 2004).

3.1.2. Pyrolysis and calcination

The contamination levels in BBFs obtained by pyrolysis (CRA, MBC and BAG) were quite variable due to the variety of pyrolysis methods utilized. The relatively low temperature used to produce CRA (190 °C, hydrothermal carbonization) and MBC (300–450 °C) did not effectively remove all persistent organic substances (> 6 $\mu\text{g kg}^{-1}$ of $\Sigma 7\text{PCBs}$ and $\Sigma 44\text{PFAS}$ in CRA, > 5 $\mu\text{g kg}^{-1}$ of $\Sigma 44\text{PFAS}$ in MBC). The deployment of higher pyrolysis temperatures >600 °C usually achieves this goal (Sørmo et al., 2024, 2023), as exemplified by BAG (650 °C) for PCBs, PCDD/Fs and PFAS. However, this does not guarantee the absence of unintentionally formed compounds such as PAHs (23.2 mg kg^{-1} $\Sigma 16\text{PAHs}$ in BAG) (Dai et al., 2014; Sørmo et al., 2024). The importance of using high temperatures to remove the target persistent organic substances was further confirmed by the results obtained for BBFs produced by incineration at temperatures >850 °C (PLA, ADC, EPH). These BBFs contained concentrations close to or below LOQ. The same three currently studied BBFs (PLA, ADC, EPH) were also shown to be free of pesticides and pharmaceuticals in a previous study (Dong et al., 2023). The removal efficiency of organic contaminants by high-temperature incineration is known to be very high (Björklund et al., 2023; Loganthan et al., 2007).

3.1.3. Crystallisation

The BBF obtained by crystallisation (CGO) was almost free of targeted PCBs, PAHs, PCDD/Fs and PFAS. Previous studies have shown that

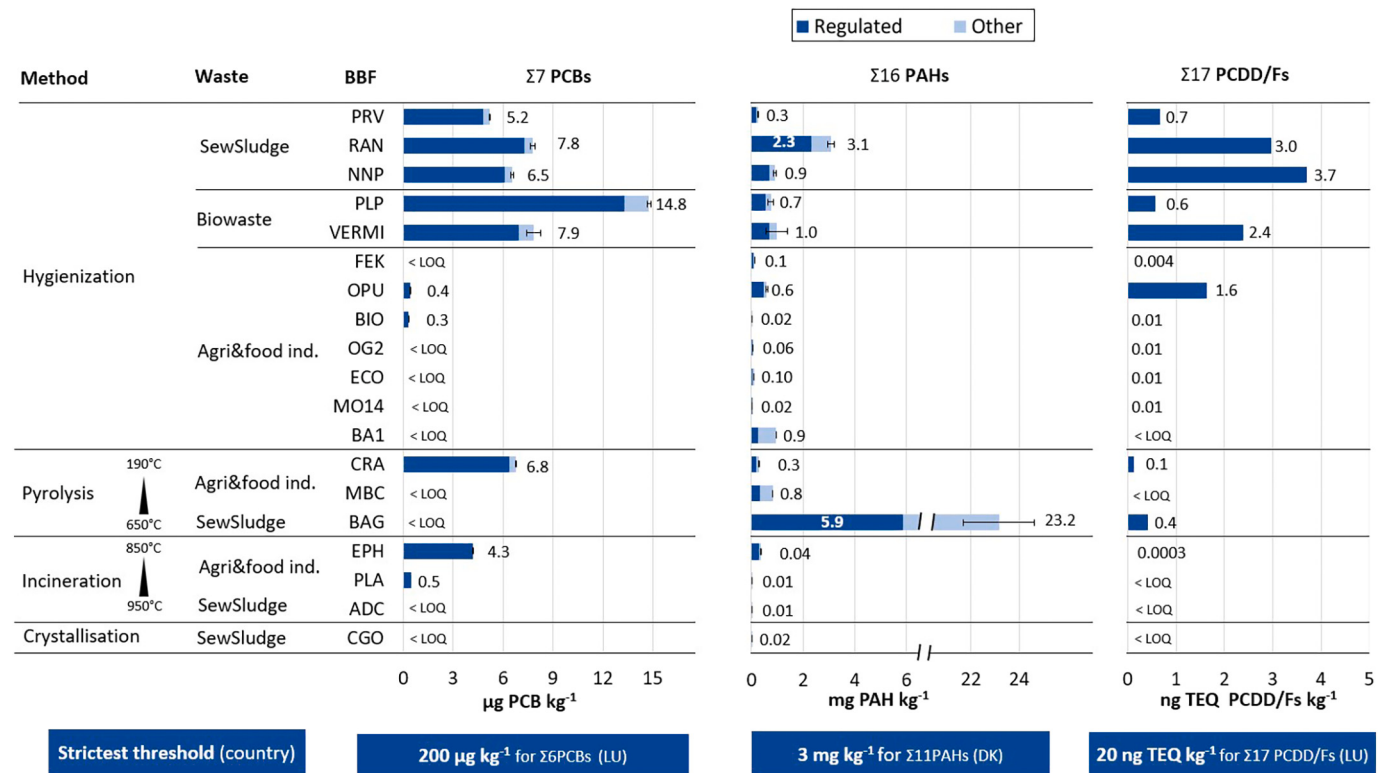


Fig. 1. Concentrations of $\Sigma 7\text{PCBs}$ (PCBs 28, 52, 101, 118, 138, 153 and 180), $\Sigma 16\text{PAHs}$ (NAP, ACE, ACY, FLE, PHE, ANT, FLU, PYE, BaA, CHR, BbF, BkF, BaP, IDP, BghiP, DBaH) and $\Sigma 17\text{PCDD/Fs}$ (see entire list in SI.2) in 19 BBFs produced by different methods and from various waste materials. The concentrations of compounds regulated by the strictest thresholds (in Luxembourg for PCBs and PCDD/Fs, in Denmark for PAHs) are represented as dark blue bars, the concentrations of substances not included in the thresholds (i.e., PCB 118, and PAHs NAP, ACY, ANT, BaA, CHR, DBaH) are represented as light blue bars. See Table 1 and SI.1 for details about the valorization methods and waste origins of BBFs.

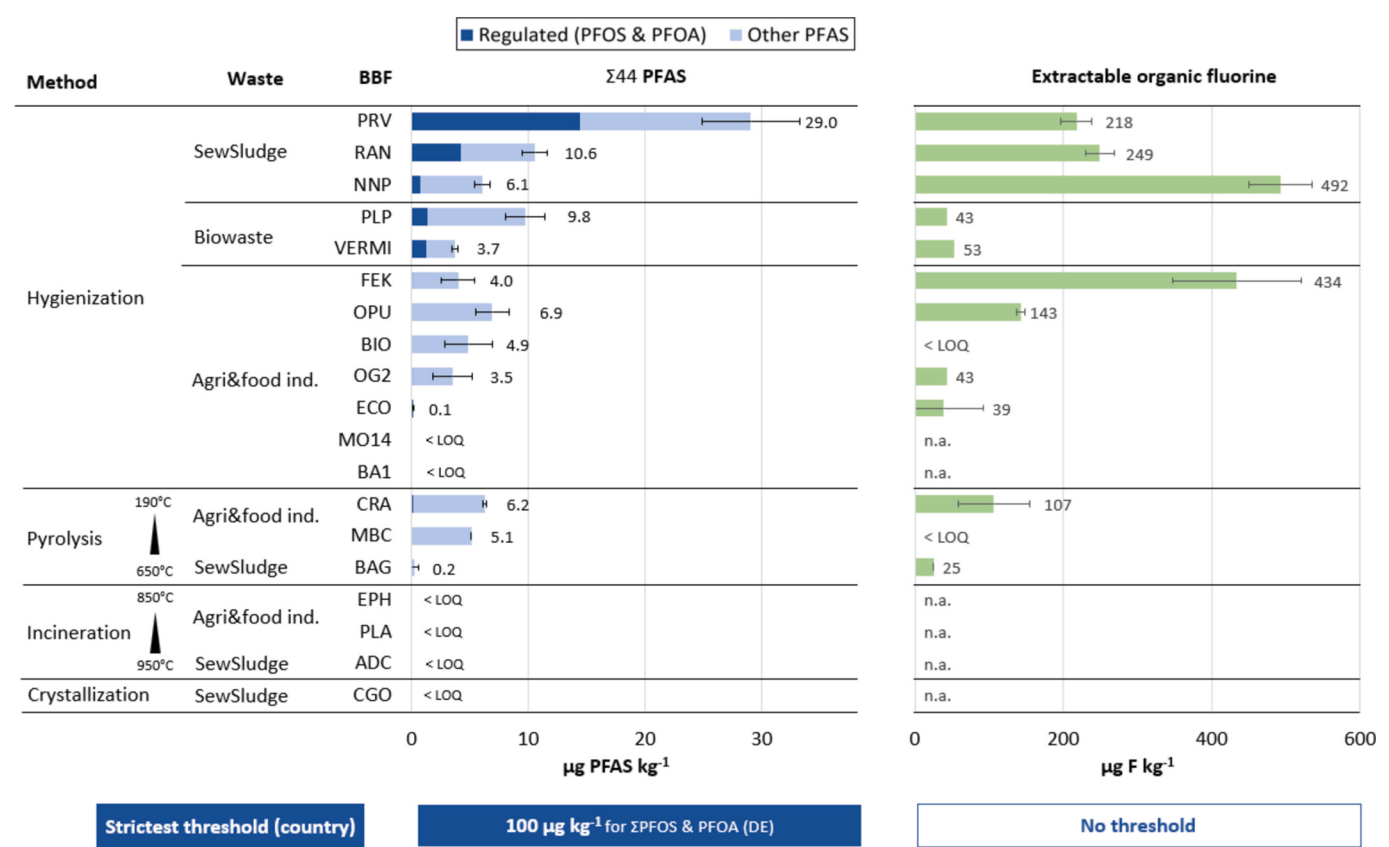


Fig. 2. Concentrations of $\Sigma 44$ PFAS, including 13 PFCAs, 10 PFSAAs, 13 PFAA precursors (6 preFOS, 4 FTSAAs, 3 diPAPs), and 8 other PFAS (see entire list in SI.3), and extractable organic fluorine in 19 BBFs produced by different methods from various waste materials. The concentrations of the regulated PFOS and PFOA (threshold of $100 \mu\text{g kg}^{-1}$ in some German regions) are represented as dark blue bars, the concentrations of the other 42 PFAS are represented as light blue bars. Note the different x-axis scale for target PFAS and EOF. See Table 1 and SI.1 for details about the valorization methods and waste origins of BBFs.

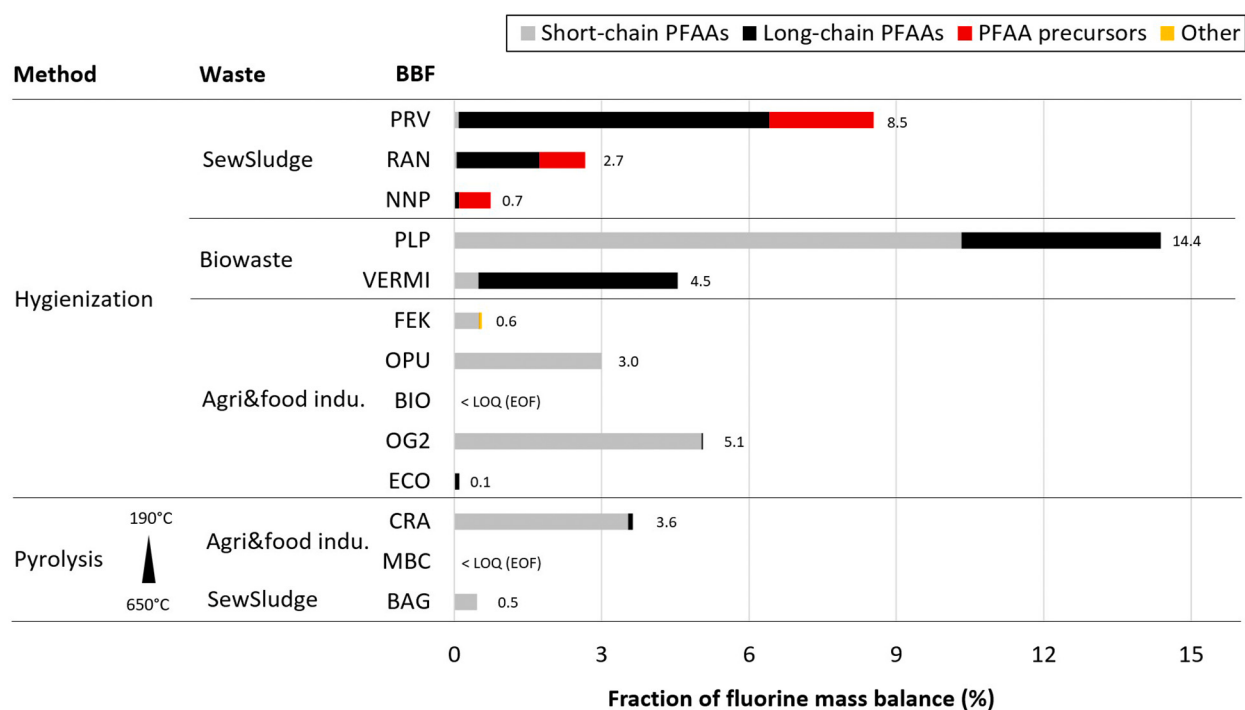


Fig. 3. Fraction of organic fluorine explained by the 44 PFAS targeted in the study, i.e., 5 short-chain perfluoroalkyl acids (short-chain PFAAs), 18 long-chain perfluoroalkyl acids (long-chain PFAAs), 13 PFAA precursors (6 preFOS, 4 FTSA, 3 diPAP), and 8 other PFAS (see entire list in SI.3) in 13 BBFs produced with different methods from various waste materials (see details about BBFs in Table 1 and SI.1).

struvite precipitation produces fertilizers and amended plants with low concentrations of organic micropollutants (de Boer et al., 2018; Ronteltap et al., 2007). In the CGO struvite, pesticides and pharmaceuticals were also found to be < LOQ in a previous study (Dong et al., 2023).

3.1.4. Valorization comparison

Considering all BBFs, no statistically significant differences in pollutant concentrations were found between the three main groups of valorization methods, i.e., hygienization, pyrolysis, and incineration (crystallisation was not considered for statistical analysis as only one such BBF was investigated). This can be explained by the high variability caused by the waste origin in the hygienization group and by the higher PCB or PAH concentrations in some samples of the pyrolysis and incineration groups. When considering the SewSludge-BBFs only (overall highest contaminated BBFs and triplicates available for most of BBFs), concentrations of PCBs (ANOVA, $F(2,12) = 54.1$, $p < 0.01$) and PFAS (ANOVA, $F(1,10) = 5.16$, $p = 0.042$) in BBFs that went through hygienization were significantly higher than those in BBFs obtained through pyrolysis (Tukey, PCBs: $p < 0.01$, PFAS: $p = 0.041$) and incineration (PCBs: Tukey, $p = 0.011$). This underscores the potential of high temperature thermal processes (i.e., pyrolysis and incineration) for persistent organic substance removal. In contrast, concentrations of PAHs in pyrolyzed products were statistically significantly higher (ANOVA, $F(2,12) = 52.9$, $p < 0.01$) than in BBFs obtained through hygienization and incineration (Tukey, $p < 0.01$ for both comparisons), confirming the earlier trade-off described between organohalogen pollutant removal and PAH generation (Sørmo et al., 2024). However, it should be noted that the amount and bioavailability of PAHs produced is very dependent on the pyrolysis technology (Hale et al., 2012).

3.2. Compliance assessment

All measured concentrations of PCBs, PAHs, PCDD/Fs and PFAS in BBFs were well below the limit values set in individual EU countries except in one case (PAHs in the pyrolyzed product BAG) (Figs. 1 and 2). High concentrations of PAHs in pyrolyzed products (such as in BAG) are often explained by uneven heat distribution and vapor trapping during pyrolysis or cool zones in the post-pyrolysis area, and can often be drastically reduced by modifying the pyrolysis unit design (Buss et al., 2022). Post-treatment of pyrolyzed products at moderated temperature (100–300 °C) can be used to thermally desorb PAHs (Kołtowski and Oleszczuk, 2015). Moreover, the bioavailability of PAHs in pyrolyzed products has been shown to be generally low (Hale et al., 2012). The present results are thus encouraging for the application of the studied BBFs as an alternative to conventional inorganic fertilizers. However, it is important to target – and regulate – more PFAS in the future, since a large fraction of total organic fluorine (>85 %) was not explained by the target PFAS in this study. This total organic fluorine can originate from non-target precursors that are known to be predominant in some organic waste such as sewage sludge (e.g., fluorotelomer alcohols, see section 3.1.1). This can also be explained by unconventional PFAS and non-PFAS organofluoride substances widely used in products such as pharmaceuticals (Spaan et al., 2023), batteries (Guelfo et al., 2024), or pesticides (Lasee et al., 2022) that could contaminate various types of organic waste.

3.3. Comparisons to current trends in contamination of organic waste

3.3.1. PFAS and EOF

Overall, PFAS patterns in SewSludge-BBFs were dominated by PFOS (0.8–14.7 $\mu\text{g kg}^{-1}$), perfluorohexanoic acid (PFHxA, < LOQ – 5.4 $\mu\text{g kg}^{-1}$), ethylperfluorooctane sulfonamidoacetic acid (EtFOSAA, 1.3–2.9 $\mu\text{g kg}^{-1}$), methylperfluorooctane sulfonamidoacetic acid (MeFOSAA, 0.6–2.1 $\mu\text{g kg}^{-1}$), 12:2 fluorotelomer sulfonate (12:2 FTS, 0.7–1.6 $\mu\text{g kg}^{-1}$), 10:2 FTS (0.4–1.5 $\mu\text{g kg}^{-1}$) and PFOA (< LOQ – 2.4 $\mu\text{g kg}^{-1}$). In raw sludges, the predominance of PFOS and its precursors MeFOSAA

and EtFOSAA (as well as some other long-chain PFAS such as PFOA) has been reported by many studies (Higgins et al., 2005; Schultz et al., 2006; Sepulvado et al., 2011). PFAA precursors (entire list in SI.3) represented a significant fraction of target PFAS in SewSludge-BBFs (25–86 %) (Fig. 3). In contrast to recent studies that reported high concentrations of fluorotelomer phosphate diesters (diPAPs) in raw sludges (> 50 % of the targeted PFAS) (Aro et al., 2021a; Thompson et al., 2023), the concentrations of these precursors were < LOQ in the three SewSludge-BBFs. An explanation could be that most of the diPAPs had already been transformed to fluorotelomer alcohols (FTOHs, not measured in this study) and perfluorocarboxylic acids (PFCAs) (Butt et al., 2014; D'eon and Mabury, 2007; Lee et al., 2010a; Yoo et al., 2010). This assumption is supported when considering that mostly even chain-length PFCAs (PFHxA, PFOA, PFDA) were quantified in PRV and RAN, which is consistent with the biological production of PFCAs from fluorotelomer-based compounds (Lee et al., 2010a). Moreover, PFHxA presented the second-highest concentrations of all the targeted PFAS (up to 5.4 $\mu\text{g kg}^{-1}$). Unlike diPAPs, FTSs – which are also precursors of PFCAs – were detected in SewSludge-BBFs. The presence of FTSs could be explained by the slower degradation of these compounds (Wang et al., 2011; Zhang et al., 2016) and/or by their potentially initially higher concentrations (compared to diPAPs). The relatively high EOF concentrations in SewSludge-BBFs ($320 \pm 133 \mu\text{g kg}^{-1}$) – which were consistent with levels reported in sludges (Aro et al., 2021a) – support that PFAA precursors and other non-targeted PFAS, such as the above-mentioned FTOHs, can be present in high concentrations in these matrices.

3.3.2. PFAS temporal trends

Although the degradation of PFAA precursors is expected to extend in time the occurrence of PFAA in organic waste, a decrease of regulated PFAS has been reported in sludges (Fredriksson et al., 2022; Gewurtz et al., 2024; Ulrich et al., 2016) and sludge based-products (Kim Lazzano et al., 2020). Between 2000 and 2010, average concentrations of PFOS in sludges were often reported to reach 100 $\mu\text{g kg}^{-1}$ (Becker et al., 2008; Higgins et al., 2005; Schultz et al., 2006; Sepulvado et al., 2011) whereas average concentrations are currently getting closer to 10 $\mu\text{g kg}^{-1}$ (Aro et al., 2021a; Eriksson et al., 2017; Fredriksson et al., 2022; Sørmo et al., 2023; Ulrich et al., 2016). Considering the concentrations of PFOS reported in the literature (Table 2), it appears that in sewage sludge in general, concentrations of PFOS have decreased by about 50 % every four years in Europe (or every six years in America), whether using the mean (Fig. 4) or median (SI.9) values. These data represent diffusely polluted municipal sewage sludges that – to the best of our knowledge – were not affected by major PFAS hotspots (see information about wastewater treatment plants and potential local PFAS sources in Table 2). A similar decrease has been reported in Europe by time-trend monitoring campaigns conducted by Fredriksson et al. (2022) and Ulrich et al. (2016); the same trend seems to apply to other long-chain PFAS (e.g., PFOA) and precursors because these PFAS have been replaced by industry with short-chain PFASs (Fredriksson et al., 2022). Recently, in Canada, Gewurtz et al. (2024) reported a decrease of regulated PFAS concentrations in sewage sludges, except for PFOS, for which a slower response to regulations/phase-outs seems to occur in this country.

In Biowaste-BBFs, PFOA was present at higher concentrations than PFOS, and short-chain PFAS were predominant (Fig. 3), in line with previous studies and in contrast to profiles measured in SewSludge-BBFs (Choi et al., 2019a; Stahl et al., 2018; Thakali et al., 2022). Another main difference with SewSludge-BBFs was that PFAA precursors were not detected in Biowaste-BBFs. These two differences in profile could potentially be explained by the higher uptake of short-chain PFAS by plants which constitute a large fraction of the biowaste (Ghisi et al., 2019; Stahl et al., 2018); in addition short-chain PFAS likely accumulate less in sludge than long-chain ones in a wastewater treatment plant. However, there are also potentially many different sources of contamination from other inputs to biowaste streams, for example the high presence of packaging plastics in the two Biowaste-BBFs (Estoppey et al.,

Table 2

PFOS concentrations in sewage sludges reported by European and American studies. WWTP: wastewater treatment plant.

Study	Sample year	Median ($\mu\text{g kg}^{-1}$)	Mean ($\mu\text{g kg}^{-1}$)	Number of WWTPs (and country) Potential local PFAS sources
Bossi et al. (2008)	2004	–	18.4	Six WWTPs (Denmark).
Becker et al. (2008)	2006	100	100	One WWTP (Germany). 2/3 of wastewater came from commercial and industrial sources including breweries, food, plastics and tobacco industries.
Zhang et al. (2010)	2008	213	333	Three WWTPs (Switzerland).
Navarro et al. (2011)	2006	28.3	63.9	Twenty WWTPs (Spain).
Llorca et al. (2011)	2010	73.5	84.2	One WWTP (Spain).
Esparza et al. (2011)	2009	39.5	40.5	Four WWTPs (The Netherlands).
Sun et al. (2011)	2008	78.0	139	Twenty WWTPs (Switzerland). The sewage sludges from three WWTPs presented PFOS levels ca. $8\times$ higher than sewage sludges from the 17 other WWTPs. These three WWTPs were probably impacted by local wastewater sources (incl. Chromium electroplating and surface finishing industries).
Gómez-Canela et al. (2012)	2012	1.91	1.94	Fifteen WWTPs (Spain (12) and Germany (3)). The three (Spanish) sewage sludges presenting the highest PFOS concentrations were produced by WWTPs treating wastewater from industrial sectors (with vehicle, textile, and chemical industries).
Arvaniti et al. (2012)	2009	4.3	4.3	Two WWTPs (Greece). One received 80 % domestic wastewater and 20 % industrial wastewater, the other only domestic wastewater.
Stasinakis et al. (2013)	2011	6.5	7.3	One WWTP (Greece).
Martínez-Moral and Tena (2013)	2013	1.38	1.32	Different WWTPs (Spain).
Perkola and Sainio (2013)	2010	63.0	63.0	One WWTP (Finland).
Campo et al. (2014)	2010	51.7	229.1	Sixteen WWTPs 16 (Spain).
	2011	0.01	38.0	
Filipovic and Berger (2015)	2013	2.9	4.2	Three WWTPs (Sweden).
Alder and van der Voet (2015)	2011	75	177.1	PFAS-related industrial and commercial activities in the catchment of 35 of the 45 WWTPs: metal plating industries (22), fire brigade training sites and foam suppliers (8), textile/textile finishing industries (9), landfill leachates (5), paper manufactures (5), packaging supplier (1), airports (2). Several WWTPs (Germany).
Ulrich et al. (2016)	2008	–	48.0	
	2009	–	21.5	
	2010	–	18.0	
	2011	–	23.0	
	2012	–	24.0	
	2013	–	15.5	
Navarro et al. (2016)	2011	8.2	14.8	Sixteen WWTPs (Spain).
Zacs and Bartkevics (2016)	2015	0.16	0.27	WWTPs from the Baltic area.
Eriksson et al. (2017)	2012	4.7	5.2	Three WWTPs (Sweden)
	2014	2.7	3.2	The three WWTPs received domestic wastewater and water from hospitals. One WWTP received wastewater from textile and chemical industries.
	2015	3.0	2.4	
Stahl et al. (2018)	2013	7.3	23.1	Different sewage treatment plants (Gemrnay).
Abril et al. (2020)	2018	20.1	20.4	Ten sewage treatment plants (Spain).
Aro et al. (2021a)	2017	3.9	4.9	Ten WWTPs (Finland, Sweden, Denmark, Norway, Faroe Islands).
Fredriksson et al. (2022)	2004	27.5	27.5	Two WWTPs (Sweden).
	2005	30.0	30.0	
	2007	26.0	26.0	
	2008	27.5	27.5	
	2009	27.5	27.5	
	2010	20.0	20.0	
	2011	15.0	15.0	
	2012	19.8	19.8	
	2013	10.1	10.1	
	2014	13.0	13.0	
	2015	12.0	12.0	
	2016	6.7	6.7	
Sørmo et al. (2023)	2021	24.5	25.0	Three WWTPs (Norway).
Higgins et al. (2005)	2001	77.3	400	Eight WWTPs (USA). All the WWTPs received at least 50 % domestic waste. One WWTP received papermill effluent; it however presented some of the lowest PFOS concentrations.
Schultz et al. (2006)	2004	–	100	One WWTP (USA).
Sinclair and Kannan (2006)	2004	30	31	Two WWTPs (USA). One WWTP influenced by domestic and commercial discharge, the other WWTP had an additional industrial discharge. Only small difference in PFOS concentrations between sludges (mean: 37 vs. 25 $\mu\text{g kg}^{-1}$, median: 28 vs. 32 $\mu\text{g kg}^{-1}$)
Loganathan et al. (2007)	2005	61.0	61.7	Two WWTPs (USA).
D'eon et al. (2009)	2002	41.0	95.2	Six WWTPs (Canada).
Sepulvado et al. (2011)	2005	145	143	Biosolids (USA). No information about WWTPs (biosolids obtained from Metropolitan Water Reclamation District of Greater Chicago).
Venkatesan and Halden (2013)	2001	–	403	Ninety-four WWTPs (USA).

(continued on next page)

Table 2 (continued)

Study	Sample year	Median ($\mu\text{g kg}^{-1}$)	Mean ($\mu\text{g kg}^{-1}$)	Number of WWTPs (and country) Potential local PFAS sources
Guerra et al. (2014)	2010	13	534 ^a or 29.6	Fifteen WWTPs (Canada). Sludges from one WWTP receiving industrial wastewater contained very high PFOS concentrations (13'100 and 2099 $\mu\text{g kg}^{-1}$). These concentrations impact very much the mean value: 534.2 $\mu\text{g kg}^{-1}$ (considered) vs. 29.6 $\mu\text{g kg}^{-1}$ (not considered)
Armstrong et al. (2016)	2005	1.0	1.1	One WWTP (USA).
	2006	50.4	51.7	
	2007	28.8	27.3	
	2008	21.1	21.3	
	2009	19.0	19.3	
	2010	18.8	15.6	
	2011	13.5	15.6	
	2012	12.6	12.1	
	2013	17.0	21.2	
Gottschall et al. (2017)	2008	7.2	7.2	One WWTP (Canada). The WWTP processed domestic, commercial and industrial wastewater
Kim Lazcano et al. (2020)	2018	9.9	10.1	Four WWTPs (USA). PFOS concentrations in sludges prior studied treatment process have been used to determine the mean and median values.
Letcher et al. (2020)	2017	5.7	10.9	Twenty WWTPs (Canada).
Kim Lazcano et al. (2020)	2014	10.3	18.2	Eleven commercially available bio-based products (USA).
Gewurtz et al. (2024)	2009	22.8	1072 ^a or 45.8	Twenty-seven WWTPs (Canada). Sludges from one WWTP presented high PFOS concentrations in 2009 (7617.5 $\mu\text{g kg}^{-1}$); no significant industrial source was known. These concentrations impact very much the mean value: 1072 $\mu\text{g kg}^{-1}$ (considered) vs. 45.8 $\mu\text{g kg}^{-1}$ (not considered).
	2010	8.3	10.5	
	2011	14.6	12.8	
	2013	14.7	37.7	
	2014	3.54	62.3	
	2015	6.0	8.4	
	2016	9.4	11.0	
	2018	12.1	15.6	
	2019	9.9	17.7	
	2021	7.9	10.3	

^a Value not considered in the determination of the temporal trend (Fig. 4) because the contribution of a local hotspot was suspected.

2024). These plastics were shown to be correlated with high contents of short-chain PFAS in composts (Choi et al., 2019b). Total concentrations of target $\Sigma 44\text{PFAS}$ ($6.7 \pm 3.5 \mu\text{g kg}^{-1}$) were similar with recent data reported in literature (Bolan et al., 2021; Choi et al., 2019a; Kim Lazcano et al., 2020; O'Connor et al., 2022; Sivaram et al., 2022; Stahl et al., 2018; Thakali et al., 2022). The relatively low EOF concentrations ($48 \pm 7 \mu\text{g kg}^{-1}$, $\mu\text{g F kg}^{-1}$) in Biowaste-BBFs indicate that large amounts of precursors and long-chain PFAS are not expected in Biowaste-BBFs. In AgriFoodInduWaste-BBFs, the low concentrations of $\Sigma 44\text{PFAS}$ in ($2.8 \pm 2.8 \mu\text{g kg}^{-1}$) were in accordance with results from Munoz et al. (2022) who reported median $\Sigma 44\text{PFAS}$ concentrations of $0.66 \mu\text{g kg}^{-1}$ for pig slurry, poultry manure and dairy cattle. Interestingly, EOF concentrations were relatively high in chicken manure-based FEK and OPU (434 and $143 \mu\text{g kg}^{-1}$, respectively). Soils on which (free-ranging) chicken are grown are often enriched in organic carbon through the build-up of feed waste and manure; these soils can sorb PFAS strongly and be an important sink, exposing the chickens through digestion of contaminated soil particles and intake of soil organisms (Lasters et al., 2022). The closeness to point pollution sources cannot be excluded, which would amplify the contamination of chickens.

Concentrations of PFAS in BBFs produced by pyrolysis and incineration were $> 5 \mu\text{g kg}^{-1}$ at an operating temperature $< 500^\circ\text{C}$ (CRA, MBC), and $< 0.3 \mu\text{g kg}^{-1}$ for combustion conducted at $> 600^\circ\text{C}$ (BAG, PLA, ADC, EPH). This is consistent with recent studies reporting PFAS removals $> 90\%$ when pyrolysis temperatures were $> 500^\circ\text{C}$ (Kundu et al., 2021; McNamara et al., 2023; Sørmo et al., 2023) and very low concentrations in the bottom ashes after incineration (Björklund et al., 2023; Loganathan et al., 2007). EOF concentrations were higher in CRA produced at 190°C ($107 \mu\text{g F kg}^{-1}$) than in BAG produced at 650°C ($25 \mu\text{g F kg}^{-1}$), indicating a beneficial effect of high temperature to reduce PFAS concentrations. PFAS removal occurring during thermal processes does not mean that PFASs are fully destroyed. At pyrolysis temperatures

between 500 and 800°C , Sørmo et al. (2023) showed that moderate amounts of (shorter chain) PFAS were present in the flue gas ($< 3\%$ of the total PFAS-mass in the waste) and high amounts of (longer chain) PFAS were expected to be concentrated in condensation oils, requiring treatment at higher temperature to be destroyed (McNamara et al., 2023). In contrast, incineration temperatures ($> 800^\circ\text{C}$) were shown to allow for mineralization of PFAS (Gehrmann et al., 2024; RIVM, 2021) although small amounts of PFAS were measured in flue gas (4.0 – 5.6 ng m^{-3}) (Björklund et al., 2023).

3.3.3. PCBs and PCDD/Fs

In SewSludge-BBFs, PCB profiles were dominated by the hexachlorinated PCBs 138 and 153 (> 35 – 46% of $\Sigma 7\text{PCBs}$), as reported by previous studies on raw sewage sludges (Antolín-Rodríguez et al., 2016; Urbaniak et al., 2017; Zennegg et al., 2013). PCDD/F profiles were dominated by OCDD (73%) and HpCDD (9%), similar to what was found in sewage sludges (contribution of around 80% for these two congeners, (Elskens et al., 2013; Zennegg et al., 2013) and in background air in Europe (contribution of $> 60\%$) (Degrendele et al., 2020). Atmospheric deposition and wash-off by rain into combined sewer systems is one of the main sources of these compounds (Zennegg et al., 2013). Concentrations of $\Sigma 7\text{PCBs}$ ($6.5 \pm 1.1 \mu\text{g kg}^{-1}$) and $\Sigma 7\text{PCDD/Fs}$ ($2.4 \pm 1.6 \text{ ng TEQ kg}^{-1}$) in SewSludge-BBFs were at the low end of the range of concentrations reported in sewage sludges since the 1980s (Fig. 5). This underscores the drastic decrease of PCB and PCDD/F contamination in sewage sludge over the last decades due to the phasing-out of PCBs and the introduction of PCDD/F restrictions (UNEP, 2001). Since the wash-off of atmospheric deposition into combined sewer systems is a primary source of PCBs and PCDD/Fs in sewage sludge (Zennegg et al., 2013), the observed decreasing trend in this organic waste is most likely due to the reduction of emissions of these compounds into the air (EEA, 2024). Fig. 5 shows the projected

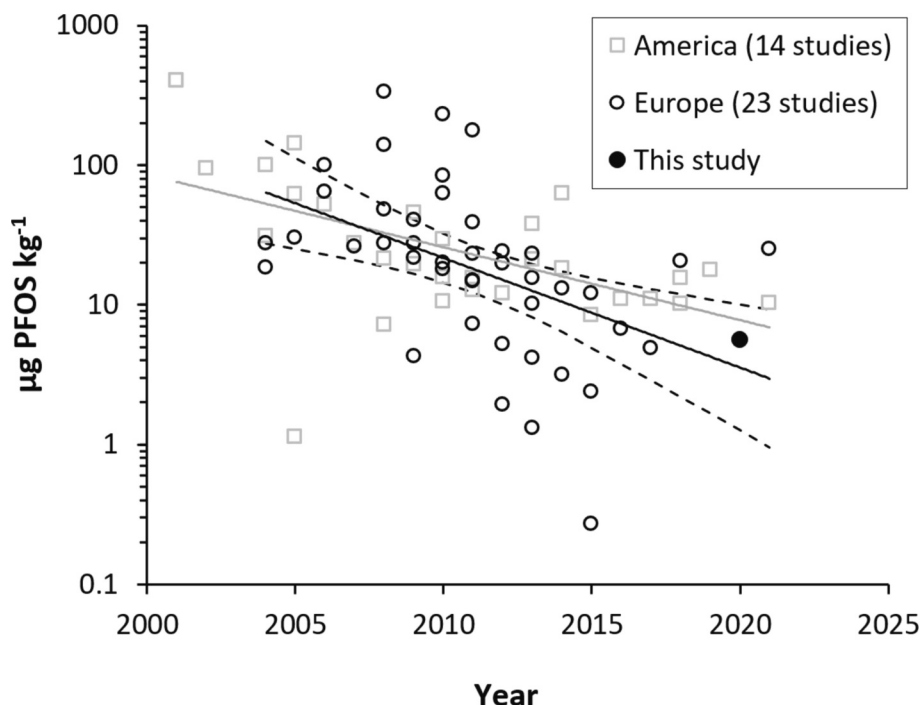


Fig. 4. Mean PFOS concentrations in sewage sludge reported in the literature from Europe (open black circles, 23 studies) and America (open grey squares, 14 studies) and measured in SewSludge-BBFs (filled black circle, this study). The solid black and grey lines are the fits of European and American data and represent decreases of PFOS concentration with an apparent half-life of 4 and 6 years, respectively; dashed black lines show a 95 % confidence interval for European values. All concentrations are given in Table 2 and graphs with median values are presented in SI.9. The samples for the current study were taken in 2020. Note the logarithmic y-axis.

evolution of PCB and PCDD/F concentration in sewage sludge-based fertilizers, using (i) the apparent half-life reported by Zennegg et al. (2013) (10 years for PCBs, 12 years for PCDD/Fs) and (ii) the apparent half-life found when fitting literature data to sludges sampled between 1990 and 2023 (6 years for PCBs, 9 years for PCDD/Fs) (Abad et al., 2005; Alcock and Jones, 1993; Blanchard et al., 2004; Eljarrat et al., 2003, 1999; Elskens et al., 2013; Fijalkowski et al., 2017; Fuentes et al., 2007; Katsoyiannis and Samara, 2004; Kaya et al., 2015; Martínez et al., 2007; McGrath et al., 2000; Roskosch and Heidecke, 2018; Sava et al., 2024; Sørmo et al., 2024; Stevens et al., 2001, 2003; Zennegg et al., 2013).

In Biowaste-BBFs, concentrations of $\sum 7$ PCBs ($11.3 \pm 3.8 \mu\text{g kg}^{-1}$) were at the low end of the range of concentrations in composts or digestates from organic household and green wastes reported between 2000 and 2020 ($7.9\text{--}63 \mu\text{g kg}^{-1}$) (Antolín-Rodríguez et al., 2016; Barcauskaitė, 2019; Beníšek et al., 2015; Brändli et al., 2007, 2005; Govasmark et al., 2011; Hellström et al., 2011; O'Connor et al., 2022; Višniauskė et al., 2018). For $\sum 17$ PCDD/Fs, concentrations in biowaste-BBFs ($1.5 \pm 1.3 \text{ ng TEQ kg}^{-1}$) were not significantly different from those measured in SewSludge-BBFs, supporting findings from Elskens et al. (2013). The obtained concentrations are slightly lower than those measured in composts in the 2000s ($8.5\text{--}9.5 \text{ ng TEQ kg}^{-1}$) (Brändli et al., 2005) and in the 2010s ($4.1 \pm 1.5 \text{ ng TEQ kg}^{-1}$), most probably because of the general decrease of PCDD/Fs emissions.

AgriFoodInduWaste-BBFs have been poorly investigated in terms of PCB and PCDD/F contamination. Low PCB concentrations have been reported in manure ($0.99 \mu\text{g kg}^{-1}$, or $0.15 \text{ ng TEQ kg}^{-1}$) or in manure compost ($2.7 \mu\text{g kg}^{-1}$) (Barcauskaitė, 2019; Elskens et al., 2013; Ng et al., 2008) which is in accordance with manure-based BBFs included in this study ($< 0.5 \mu\text{g kg}^{-1}$ in FEK, OPU, BIO). Concentrations of $\sum 17$ PCDD/Fs in AgriFoodInduWaste ($0.19 \text{ ng TEQ kg}^{-1}$) were in accordance with the most recent concentrations reported in manure ($0.15 \text{ ng TEQ kg}^{-1}$) (Elskens et al., 2013) and slightly lower than those reported for manure in the 2000s ($0.1\text{--}4 \text{ ng TEQ kg}^{-1}$) (Ng et al., 2008;

Stevens and Jones, 2003; Welsch-Pausch and McLachlan, 1998). The presence of PCBs and PCDD/Fs in manure can be explained by the fact that livestock farming accumulates these substances from soils (contaminated from past releases or emissions from buildings and construction works) and that significant fractions of these contaminants are expected to leave animals in manure (Weber et al., 2018a, 2018b; Welsch-Pausch and McLachlan, 1998). This is especially true for free-range chickens that take up more soil than other farm animals per body weight (Weber et al., 2018a, 2018b). The PCB concentrations $< \text{LOQ}$ in animal (OG2, ECO) and plant (MO14, BA1) based BBFs are in accordance with low PCB levels being observed in meat, cereals, vegetables and fruits ($0.011\text{--}2.26 \mu\text{g kg}^{-1}$) (Esposito et al., 2017; Schwind et al., 2009; Zhang et al., 2008). Elskens et al. (2013) showed that dioxin-like PCB concentrations in plant and animal-based fertilizers ($0.05\text{--}0.06 \text{ ng TEQ kg}^{-1}$) were significantly below those in sewage sludges and composts ($1.6\text{--}2.2 \text{ ng TEQ kg}^{-1}$), confirming that PCB levels in AgriFoodInduWaste-BBFs are at least 10 times lower than in SewSludge-BBFs and Biowaste-BBFs.

In BBFs produced by pyrolysis, concentrations of $\sum 7$ PCBs ranged from $< \text{LOQ}$, when obtained by dry pyrolysis ($> 300^\circ\text{C}$), to concentrations similar to common feedstock ($6.8 \mu\text{g kg}^{-1}$), when obtained by wet pyrolysis ($< 200^\circ\text{C}$). This is in line with previous studies that report a 1–2 order magnitude reduction of $\sum 7$ PCB concentrations with dry pyrolysis of sewage sludges (from 7.6 to $20.7 \mu\text{g kg}^{-1}$ (Sørmo et al., 2024) or $274.2 \mu\text{g kg}^{-1}$ (Moško et al., 2021) in sludges to $< 0.25\text{--}1.7 \mu\text{g kg}^{-1}$ or $< \text{LOQ} - 26.6 \mu\text{g kg}^{-1}$ in pyrolyzed products, respectively) and no significant reduction (or generation) of PCBs by wet pyrolysis (HTC) (Brookman et al., 2018; Tasca et al., 2022). Concentrations of $\sum 7$ PCDD/Fs were very low in most of the BBFs ($< 0.005 \text{ ng TEQ kg}^{-1}$) and slightly higher in BAG obtained by dry pyrolysis at 600°C ($0.3 \text{ ng TEQ kg}^{-1}$). Conversion of PCBs into PCDD/Fs in BBFs is unlikely due to the low levels of PCBs. These results show that processes involving thermal treatment do not generally generate by-products, but require close monitoring of operating conditions to avoid uneven heat distribution or

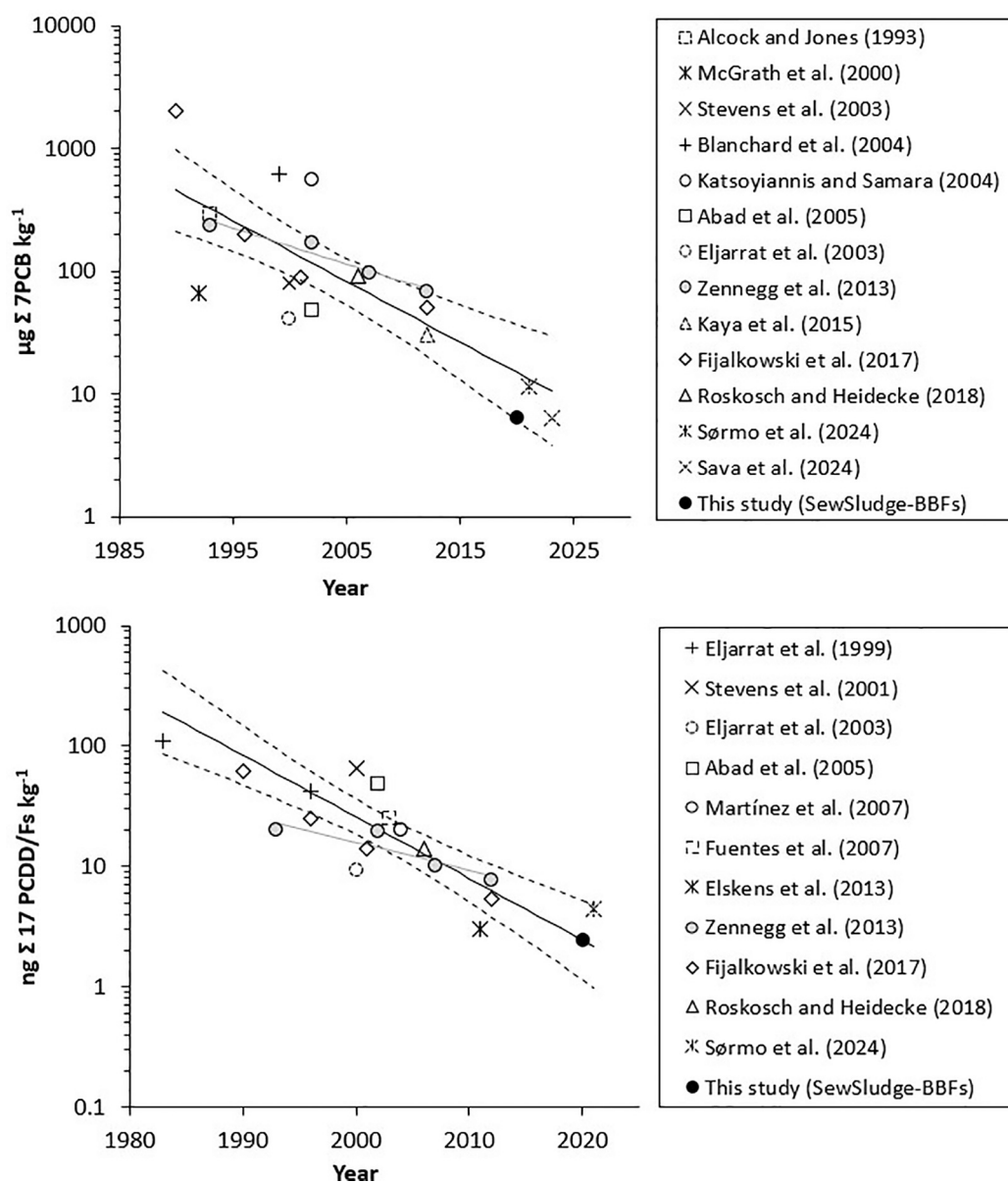


Fig. 5. Concentrations of Σ PCBs 28, 52, 101, 138, 153, 180 and 118 when measured - (upper panel) and of Σ 17 PCDD/Fs (lower panel) in sewage sludge-based fertilizers reported in the literature and measured in SewSludge-BBFs (this study). The solid grey line is the decrease of concentrations in sewage sludge reported by Zennegg et al. (2013) with an apparent half-life of 10 years for PCBs and 13 years for PCDD/Fs. The solid black line is the fit of all data presented and shows a decrease of concentrations with an apparent half-life of 6 years for PCBs and 9 years for PCDD/Fs; dashed black lines show a 95 % confidence interval. All numerical data can be found in Supplementary Information SI.10. Note the logarithmic y-axis.

cool zones in the post-pyrolysis area, especially when substantial chlorine sources are present in the feedstocks (Altarawneh et al., 2009; Buss et al., 2022; Chagger et al., 2000; Wang et al., 2017).

3.3.4. PAHs

In SewSludge-BBFs, PAH profiles were dominated by phenanthrene (16 %), fluoranthene (15 %), and pyrene (18 %), consistent with earlier results (Chen et al., 2019). Concentrations of Σ 16PAHs in SewSludge-BBFs ($1.41 \pm 1.28 \text{ mg kg}^{-1}$) were in line with concentrations reported in sewage sludges throughout Europe from 2010 (close to 1 mg kg^{-1}) and about an order of magnitude lower than those measured in the beginning of the century (see SI.10) (Alhafez et al., 2013; Baran and Oleszczuk, 2003; Berset and Holzer, 1999; Boruszko, 2017; Busetti et al., 2006; Pérez et al., 2001; Roskosch and Heidecke, 2018; Sørmo et al., 2024; Stevens et al., 2003; Suciu et al., 2015; Villar et al., 2006). Thus, similarly to the other substances, a general decrease in PAH

concentrations seems to occur with a half-life of <10 years (SI-10). Furthermore, the hygienization process used to produce the studied BBFs (i.e., drying and/or anaerobic digestions) is expected to decrease the amount of PAHs; a reduction of >60 % was indeed reported when applying aerobic stabilization (Trably et al., 2005; Włodarczyk-Makula et al., 2021). Concentrations of Σ 16PAHs in Biowaste-BBFs ($0.86 \pm 0.3 \text{ mg kg}^{-1}$) were similar to those measured in composts from organic household waste and green waste ($1.7\text{--}1.9 \text{ mg kg}^{-1}$) (Brändli et al., 2005; Farrell and Jones, 2009). Low concentrations in Biowaste-BBFs can be explained by the degradation of some PAHs in composts, especially low molecular weight PAHs (up to 90 % reduction) (Brändli et al., 2007; Houot et al., 2012). The low concentrations of Σ 16PAHs in AgriFoodInduWaste-BBFs ($0.26 \pm 0.36 \text{ mg kg}^{-1}$) are in good agreement with the very low concentrations reported in cereals ($< 0.001 \text{ mg kg}^{-1}$) (Einolghozati et al., 2022) or manure ($< 0.5 \text{ mg kg}^{-1}$) (Mackiewicz-Walec and Krzbiec, 2020).

Table 3

Expected concentrations in soils amended with SewSludge-BBFs, Biowaste-BBFs, AgriFoodInduWaste-BBFs, as well as BBFs obtained through pyrolysis and incineration, when maximum allowed application rates are used ($0.6\text{--}13.1\text{ t ha}^{-1}\text{ y}^{-1}$, see SI.1 for individual application rates). The amount of soil in the top layer (30 cm) was determined using a density of 1.3 g cm^{-3} (i.e., $3'900'000\text{ kg ha}^{-1}$).

	Hygienization			Pyrolysis	Incineration	Background concentration in soils ^a
	SewSludge-BBFs	Biowaste-BBFs	AgriFoodInduWaste-BBFs			
$\sum 15\text{PAHs}$ (mg kg^{-1})	< 0.004	< 0.004	< 0.001	< 0.002	< 0.0003	0.004–7.3 (mean: 0.37)
$\sum 7\text{PCBs}$ ($\mu\text{g kg}^{-1}$)	< 0.011	< 0.033	< 0.001	< 0.005	< 0.003	0.01–58 (mean: 2.9)
$\sum 44\text{PFAS}$ ($\mu\text{g kg}^{-1}$)	< 0.036	< 0.022	< 0.013	< 0.008	< LOQ	
PFOS ($\mu\text{g kg}^{-1}$)	< 0.016	< 0.002	< 0.00008	< 0.0002	< LOQ	0.02–162 (median: 2.7)
PFOA ($\mu\text{g kg}^{-1}$)	< 0.003	< 0.003	< LOQ	< LOQ	< LOQ	0.02–124 (median: 2.7)
$\sum 17\text{PCDD/Fs}$ (ng TEQ kg^{-1})	< 0.004	< 0.009	< 0.003	< 0.0002	< 0.000003	0.5–28.9 (mean: 3.18)

^a Background concentrations are from Meijer et al. (2003) for PCBs, Vives et al. (2008) for PCDD/Fs, Nam et al. (2008) for PAHs, Brusseau et al. (2020) for PFOS and PFOA. For PCFF/Fs values reported are for Italy only, but studies conducted in other countries fell in this range (Environment Agency, 2009).

In BBFs produced by pyrolysis, low concentrations of $\sum 16\text{PAHs}$ in two of the pyrolyzed products ($< 1\text{ mg kg}^{-1}$ for CRA and MBC) and one high concentration exceeding existing thresholds (23.2 mg kg^{-1} for BAG), were consistent with the values reported by Wang et al. (2017) for 102 pyrolyzed products, i.e., $< 1.5\text{ mg kg}^{-1}$ for about 90 % of the pyrolyzed products, and up to 100 mg kg^{-1} in 10 % of the cases. In ashes, the concentrations of $\sum 16\text{PAHs}$ measured in BBFs ($< 0.5\text{ mg kg}^{-1}$) were comparable to those reported in bottom/bed ashes from municipal waste/biomass incineration ($< 1\text{ mg kg}^{-1}$ in most of the cases) (Enell et al., 2008; Masto et al., 2015).

3.4. Expected and measured concentrations of PCBs and PFASs in soil

3.4.1. Contamination after one BBF application

Expected concentrations of the persistent organic substances in amended soils (initially not contaminated) - after one application - were $< 0.033\text{ }\mu\text{g kg}^{-1}$ for $\sum 7\text{PCBs}$, $< 0.009\text{ ng TEQ kg}^{-1}$ for $\sum 17\text{PCDD/Fs}$, $< 0.004\text{ mg kg}^{-1}$ for $\sum 15\text{PAHs}$, $< 0.04\text{ }\mu\text{g kg}^{-1}$ for $\sum 44\text{PFAS}$ (Table 3). All these concentrations were at the low end of the ranges of background concentrations in soils; PFOS and PFOA were used for the comparison with literature since data for these two congeners are the most widely reported (Table 3). These results indicate that one BBF application does not constitute a significant contribution to the contamination of agricultural soils by target substances.

Measurements conducted on soil samples from field trials revealed modest but statistically significant (t -test, $p < 0.05$) increases in PCB concentrations between the control soils and the BBF-amended soils. Indeed, increases of 0.12 and $0.50\text{ }\mu\text{g kg}^{-1}$ ($\sum 7\text{PCBs}$) were measured in PLP-amended soils (Finland) and EPH amended-soil (Austria), respectively (Supplementary Information SI.11). The increase in PCB concentrations in PLP-amended soils was slightly higher than the expected increase (0.12 vs. $0.032\text{ }\mu\text{g kg}^{-1}$) and can probably be explained by the variability of PCB concentrations in the BBFs and soils. In contrast, the measured PCB concentration increase in EPH-amended soil was much higher than expected (0.40 vs. $0.0018\text{ }\mu\text{g kg}^{-1}$). It appeared that some PCBs (e.g. PCB 138 and PCB153, responsible for half of the increase in EPH-amended soil) were $< \text{LOQ}$ in EPH, indicating that the increase of PCB concentrations in the amended soil had most probably another origin than the BBF itself. An explanation could be that 9 months separated the sampling of the pre-trial and amended soils; PCB contamination through the atmosphere or from other plots might be the cause of this increase in Finland. Another explanation could be the heterogeneity and uncertainty for measurements at such low levels. In any case, this measured difference ($0.40\text{ }\mu\text{g kg}^{-1}$) was at the low end of background concentrations in soils ($0.01\text{--}58\text{ }\mu\text{g kg}^{-1}$). Regarding PFAS, PFOS was the only quantifiable congener in soils from the field trials. The concentrations in OPU-amended soils were not higher than in controls or pre-trial soils (Supplementary Information SI.11). In Finland, the concentration of PFOS was higher in the control ($3.1\text{ }\mu\text{g kg}^{-1}$) than in the OPU-amended soil ($0.6\text{ }\mu\text{g kg}^{-1}$), most probably due to soil heterogeneity.

3.4.2. Prediction of pollutant accumulation in amended soils

Persistent organic substances have been shown to accumulate in soils amended with recycled fertilizers (Sepulvado et al., 2011; Umlauf et al., 2011; Washington et al., 2010a, b; Weber et al., 2018a, b). A linear increase with increasing biosolid loading rate was for example reported by Sepulvado et al. (2011). Therefore, the prediction of contamination in amended soils should take into account the expected persistent organic substance concentrations in BBFs in the future (Eq. (3)) as well as the cumulative concentrations over time (Eq. (4)). As discussed in Section 3.3, the concentrations of legacy contaminants in SewSludge-BBFs are expected to decrease with an apparent half-life of < 10 years. Scenarios with half-life values of 10, 15 and 20 years were used for conservative assessment (worst-case scenarios). For contaminant concentrations in BBFs, Eq. (3) was used:

$$C_{\text{cont,BBF},t=n} = C_{\text{cont,BBF},t=0} * 2^{-t/x} \quad (3)$$

where $C_{\text{cont,BBF},t=n}$ is the contaminant concentration in SewSludge-BBFs in n years, $C_{\text{cont,BBF},t=0}$ is the average concentration of contaminant in SewSludge-BBFs at $t = 0$ (concentrations given in Figs. 1 and 2), and x is the half-life of contaminants in BBFs (10, 15 and 20 years for the presented scenarios). For contaminant concentrations in soils, Eq. (4) was used:

$$C_{\text{cont,soil},t=n} = C_{\text{cont,soil},t=0} + \sum_{t=0}^n \frac{m_{\text{BBF}}}{m_{\text{soil}}} C_{\text{cont,BBF},t=n} \quad (4)$$

where $C_{\text{cont,soil},t=n}$ is the accumulated contaminants in amended soil after n year, $C_{\text{cont,soil},t=0}$ is the initial concentration in soils at $t = 0$ (e.g., median values given in Table 3), m_{BBF} is the average mass of BBF applied per hectare annually (4.4 tons; average application rate for SewSludge-BBFs), and m_{soil} is the average mass of soil amended per hectare (3900 tons; 30 cm of soil with density of 1.3 g cm^{-3}). Additional information about the model can be found in SI.12.

For PFAS, the predictions are complicated by the potential leaching of some PFAS and the transformation of precursors (Holly et al., 2024; Pepper et al., 2021b; Ye et al., 2024). While short-chain PFAS ($\text{C}_n\text{F}_{2n+1}\text{COOH}$, $n \leq 6$, and $\text{C}_n\text{F}_{2n+1}\text{SO}_3\text{H}$, $n \leq 5$) are readily mobilized from soils, long-chain product PFAS such as PFOS and its precursors remain mostly in the upper soil horizon because the desorption can be relatively slow (Gellrich et al., 2012; Gnesda et al., 2022; Lee et al., 2010b; Maizel et al., 2021; Sepulvado et al., 2011; Stahl et al., 2013b; Washington et al., 2010b). Therefore, predictions should consider an accumulation of the long-chain PFAS in the top layer of the soils whereas the short-chain will reach the groundwater more rapidly. Regarding the transformation of precursors, previous studies reported that concentrations of MeFOSAA and EtFOSAA measured in amended soils were shown to be much lower than those calculated from the concentration in sludges, suggesting transformation of these precursors (Pepper et al., 2021b; Sepulvado et al., 2011). Thus the predictions for PFOS were based on all precursors (FOSA, EtFOSAA and MeFOSSA, (Kolanczyk

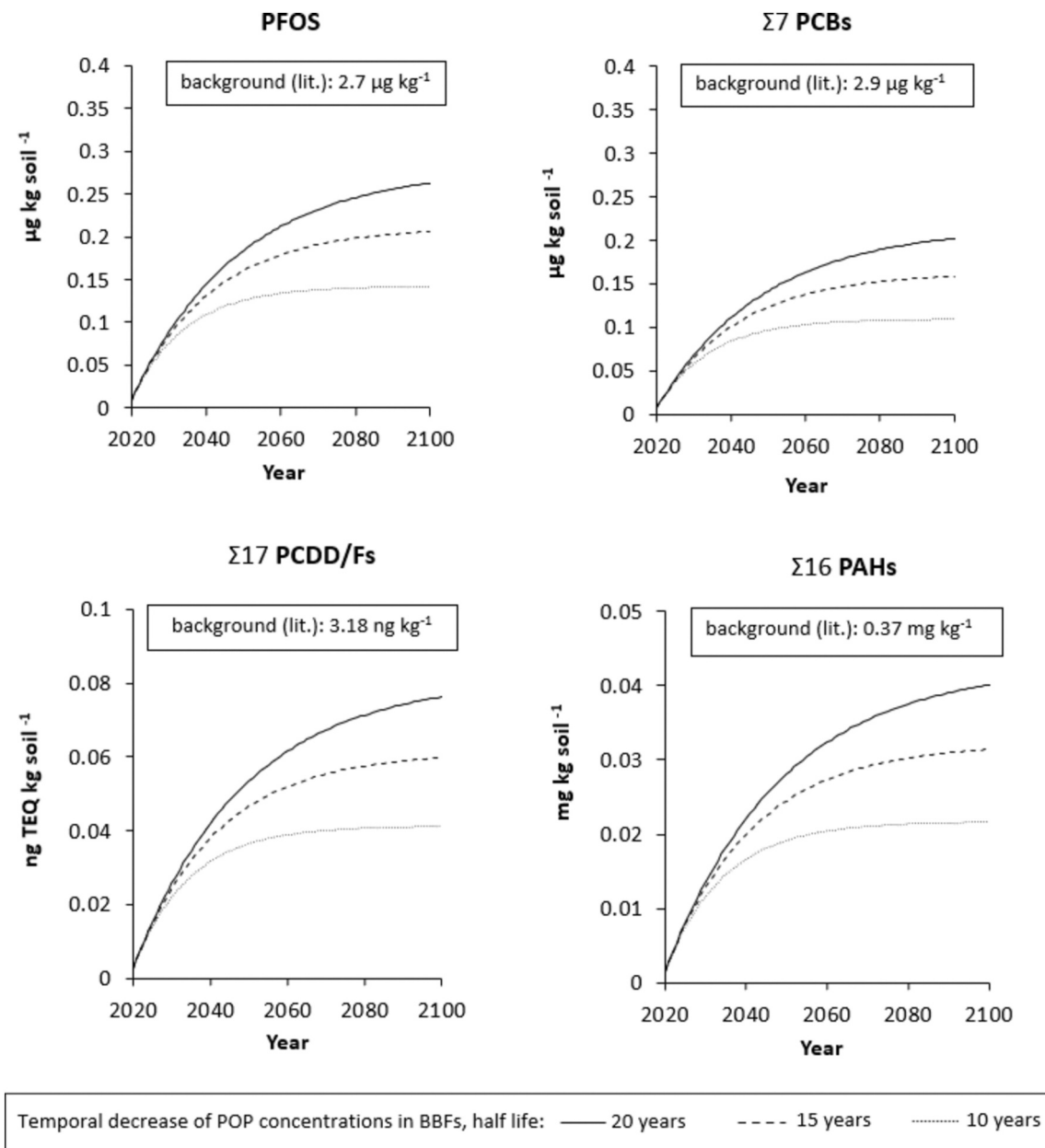


Fig. 6. Expected concentrations of legacy substances in the top layer (30 cm) of initially non-contaminated soils amended with SewSludge-BBFs at an average maximum allowed application rate ($4.4 \text{ t y}^{-1} \text{ ha}^{-1}$) using apparent decreases target substances in BBFs with a half-life of 10, 15 and 20 years (see Figs. 4 and 5, and discussion in Section 3.3). For conservative assessment, it was assumed that all quantified PFOS precursors transform to PFOS, and these contaminants do not significantly transfer in soils, nor degrade. The amount of soil in the top layer (30 cm) was determined using a density of 1.3 g cm^{-3} (i.e., $3'900'000 \text{ kg ha}^{-1}$). Equations and results with initially contaminated soils are given in SI.12.

et al., 2023)) fully transforming to PFOS, in combination with an apparent half-life of 10, 15 or 20 years for both PFOS-precursors and PFOS in BBFs. For all scenarios, the concentration of PFOS would reach a plateau at a value below $0.3 \mu\text{g kg}^{-1}$ in (initially non-contaminated) soils amended with SewSludge-BBFs (Fig. 6). Although these numbers are subject to large uncertainty due to the factors described above, it is apparent that those concentrations are well below the median background concentrations of PFOS in soils ($2.7 \mu\text{g kg}^{-1}$, Table 3) and the limit values set by some countries (the strictest being $0.8 \mu\text{g kg}^{-1}$ in The Netherlands (Hall et al., 2020)). When applying SewSludge-BBFs on soils already contaminated by PFOS (e.g., median background concentration given in the literature), the increase due to BBF application is expected to be $<11\%$ (SI.12). Thus, the application of BBFs is not expected to constitute a risk for contaminating the topsoil layer with PFAS.

However, short-chain PFAS in BBFs could potentially constitute a risk for groundwater contamination.

For PCBs, PCDD/Fs and PAHs, conservatively assuming there is no degradation and transfer of these persistent organic substances in soils (Umlauf et al., 2011), the concentration of $\Sigma 7\text{PCBs}$, $\Sigma 17\text{PCDD/Fs}$ and $\Sigma 16\text{PAHs}$ would reach plateau values of $0.25 \mu\text{g kg}^{-1}$, $0.1 \text{ ng TEQ kg}^{-1}$ and 0.3 mg kg^{-1} , respectively, in (initially non-contaminated) soils amended with SewSludge-BBFs (Fig. 6). Those concentrations are well below the mean background concentrations of PCB, PCDD/Fs and PAHs in soils ($2.9 \mu\text{g kg}^{-1}$, $3.18 \text{ ng TEQ kg}^{-1}$ and 0.37 mg kg^{-1} , respectively). In cases of existing soil contamination by PCBs, PCDD/Fs and PAHs, (e.g., median background concentration given in the literature), the increase due to BBF application is $<8\%$ for PCBs, $<3\%$ for PCDD/Fs and $<12\%$ for PAHs (SI.12).

Table 4

Expected concentrations in grains of cereals grown on BBF-amended soils, when considering maximum allowed application rates (0.6 to 13.1 t⁻¹ ha⁻¹ y⁻¹, see SI.1 for individual application rates) and BAF values of 0.48 for PFBA, 0.11 for PFOA and 0.06 for PFOS (Bizkarguenaga et al., 2016; Blaine et al., 2013; Ghisi et al., 2019; Krippner et al., 2014; Lesmeister et al., 2021; Stahl et al., 2009; Wen et al., 2014) as well as conservative BAF values of 1 for PAHs, 0.005 for PCBs and 0.0005 for PCDD/Fs (Kacálková and Tlustoš, 2011; Paraíba et al., 2010; Strek et al., 1981).

	Hygienization			Pyrolysis	Incineration	Tolerable intake ^a
	SewSludge-BBFs	Biowaste-BBFs	AgriFoodInduWaste-BBFs			
∑15PAHs (μg kg ⁻¹)	0.3–3.7	1.4–3.0	0.01–1	0.1–1.8	< LOQ – 0.3	0.3 μg kg ⁻¹ b.w.d ⁻¹ for BaP (US EPA)
∑7PCBs (ng kg ⁻¹)	< 0.02	< 0.02	< 0.005	< 0.009	< 0.001	20 ng kg ⁻¹ b.w. d ⁻¹ (US EPA)
∑17PCDD/Fs (pg TEQ kg ⁻¹)	< 0.002	< 0.005	< 0.002	< 0.00006	< 0.0000001	2 pg TEQ kg ⁻¹ b.w. week ⁻¹ for PCDD/Fs & dioxin like-PCBs (EFSA)
PFBA (ng kg ⁻¹)	< LOQ	< LOQ – 6.7	< LOQ – 5.4	< LOQ – 3.5	< LOQ	–
PFOS (ng kg ⁻¹)	0.04–0.90	0.086–0.093	< LOQ – 0.004	< LOQ – 0.0073	< LOQ	4.4 ng kg ⁻¹ b.w. week ⁻¹ for ∑PFOS, PFOA, PFNA, PFHxS
PFOA (ng kg ⁻¹)	< LOQ – 0.333	0.219–0.374	< LOQ	< LOQ	< LOQ	

^a Updated from Popli et al. (2022).

This indicates that the application of BBFs is not expected to constitute a soil or crop contamination risk for PCBs, PCDD/Fs and PAHs in the long term. Previous studies also stated that the application of organic fertilizers containing similar levels of PCDD/Fs and PCBs to the studied BBFs does not significantly affect the soil since the contribution from fertilizers is usually very low compared to that of atmospheric depositions (Amlinger et al., 2004; Elskens et al., 2013; Stevens et al., 2003; Timmermann et al., 2003; Umlauf et al., 2011).

3.5. Uptake by plants and risk for humans

Organic pollutant bioavailability in soils and uptake by edible plant parts are usually low for all but the most mobile substances (Clarke et al., 2010). PFAS are the studied compounds that present the lowest sorption to soil (and thus the highest bioavailability), with soil-water partitioning coefficient (K_d) values around 0.3 L kg⁻¹ for PFBA, 1 L kg⁻¹ for PFOA, 8 L kg⁻¹ for PFOS in common soils and under ambient pH conditions (SI.5) (Nguyen et al., 2020). For cereal grains, some of the (maximum) bioaccumulation factor (BAF) values reported are 0.48 for PFBA, 0.16 for PFOA and 0.06 for PFOS (SI.5) (Doucette et al., 2018; Krippner et al., 2014; Stahl et al., 2009; Wen et al., 2014). PFBA, PFOA and PFOS concentrations expected in BBF-amended grains were calculated using these BAF values (Table 4). The data show that consumption of food from BBF-amended plots would only contribute minimally to tolerable daily intake (TDI), confirming results from previous studies on sludges (Gottschall et al., 2017).

The other substances studied herein present much stronger sorption to soil particles. K_d values - calculated from organic carbon to water partitioning coefficient, K_{oc} (log K_{oc} = 0.00028 + (0.983 × log K_{ow}) (Di Toro, 1985)), at an organic carbon content of 3 % - range between 200 and 120'000 L kg⁻¹ for PAHs, 11'000 and 520'000 L kg⁻¹ for PCBs, and 30'000 and 2'200'000 L kg⁻¹ for PCDD/Fs (SI.5). Therefore, reported BAF for these compounds are very low, i.e., < 1 for PAHs, < 0.005 for PCBs and < 0.0005 for PCDD/Fs (see SI.5), the main uptake for these compounds originating from the atmosphere rather than the soil (Nizzetto et al., 2008; Su et al., 2007). Concentrations of PAHs, PCBs and PCDD/Fs in plants were based on conservative BAF values (1, 0.005 and 0.0005, respectively). The consumption of cereals from BBF-amended plots only minimally contributed to tolerable intake levels (Table 4).

Low uptake in crops due to BBF amendment was confirmed by all measured PCB and PFAS concentrations in barley and maize grains being < LOQ, except PCB 28 which was quantified at a very low concentration (0.11 μg kg⁻¹) in barley grains from Finland. The presence of this PCB can be explained by an uptake from the atmosphere which has been shown to be a more important pathway for such compounds (Collins et al., 2006; Nizzetto et al., 2008).

This study assessed the risk of BBFs for cereals. However, BAFs for PFOS for other edible plants could be higher. Values ranging between

0.1 and 1.67 were reported for lettuce, and between 0.07 and 0.7 for radish roots (Blaine et al., 2013). For these vegetables, in the worst case, the concentrations of PFOS could be up to 30 times higher than the PFOS concentrations presented in Table 1 for cereals. Therefore, the use of tested SewSludge-BBFs for vegetables should not be recommended without additional treatment (e.g., thermal treatment). More experimental data on plant uptake would strengthen the conclusion, and are recommended for future studies.

3.6. Further discussion of processes impacting the availability and uptake of contaminants

In this study, the risk of BBFs was assessed using a conservative approach (i.e., assessment of the worst-case scenario) to ensure that contaminants do not enter the food chain at levels of potential health concern. Therefore, for the long-term assessment, we assumed no degradation and migration of the studied legacy contaminants (PCBs, PCDD/Fs, PAHs, PFOS) as well as a full transformation of all measured PFOS precursors (FOSA, MeFOSAA and EtFOSAA) to PFOS. In addition, the highest literature-based BAF values were used to assess the uptake of contaminants in cereals. Hereafter, we discussed to which extent contaminant concentrations would change if these conservative assumptions were not met.

3.6.1. Degradation

Degradation rates of PCBs, PCDD/Fs and PFOS in soils are very low, with half-lives ranging from a few years to hundreds of years (Campanella et al., 2002; Dickman and Aga, 2022; Sinkkonen and Paasivirta, 2000; Terzaghi et al., 2021; Umlauf et al., 2011). Heavier PAHs are also very persistent (e.g., half-life of many years), whereas lower molecular weight PAHs are more prone to biodegradation (e.g., half-life of the three-ring molecule phenanthrene is a few months) (Shuttleworth and Cerniglia, 1995). If degradation occurs, contaminant concentrations in soils reach maximum values after times that depend on the half-life and the scenario studied in section 3.4.2. (see graphs in S.12). These maximum concentrations are about 46 % of the plateau values reported in Section 3.4.2 if the degradation of compounds occurs with a half-life of 20 years, 32 % of the plateau value with a degradation half-life of 10 years, and 23 % of the plateau value with degradation half-life of 5 years.

3.6.2. Migration

The migration of contaminants in (saturated) soils mostly depends on contaminant sorption (K_d values), soil physical properties (bulk density ρ_b and porosity θ), and environmental conditions (precipitations). The retardation factor R_f (i.e., the estimate of how much slower a contaminant moves compared to water) is given by $R_f = 1 + (K_d \rho_b / \theta)$. For PAHs, PCBs, and PCDD/Fs, K_d values range between 200 and 2'000'000 L kg⁻¹

for an organic carbon content of 3 % (field trials: 2–3.1 %). Assuming a bulk density of 1.3 g cm^{-3} and a soil porosity of 50 % (field trials: clay-rich soils), the retardation factor ranges between 500 and $>5 \times 10^9$. If the annual precipitation (field trials: about 750 mm year^{-1}) fully infiltrates into the soil and water moves downward, the migration of contaminants is expected to range between $0.15 \text{ } \mu\text{m year}^{-1}$ (for $R_f = 5 \times 10^9$) and 1.5 mm year^{-1} (for $R_f = 500$). Therefore, assuming no colloidal transport, 200 to 2×10^9 years are needed for PCBs, PCDD/Fs, and PAHs to migrate from the top layer (30 cm), confirming that the effect of migration is negligible for these compounds. For PFOS, the average K_d value is about 8 L kg^{-1} at pH 7.2 (field trials: pH 6.2–6.9) (Nguyen et al., 2020). Using the above-mentioned approach, it can be estimated that 9 years would be required for PFOS to migrate from the top layer (30 cm) with precipitation of 750 mm year^{-1} , confirmed by Sørmo et al. (2024) who showed that 90 % leaching of PFOS from a 1 m soil layer would occur after 15 years (pH 6.2, precipitation 750 mm). The effect of such leaching on PFOS concentrations in soil is similar to a decrease with a half-life of about 10 years as shown in SI.13. In these conditions, concentrations that reach 23 % of the plateau values reported in Section 3.4.2 can be expected. If the top layer is not saturated, the proportion of PFOS retained is expected to be higher because the retention in the vadose zone is stronger due to high adsorption at the air-water and solid-water interfaces (Gnesda et al., 2022).

3.6.3. Precursor transformation

The degradation of PFOS precursors in soils was shown to occur with a half-life in the range of weeks (Lange, 2001; Mejia Avendaño and Liu, 2015; Rhoads et al., 2008; Zabaleta et al., 2018). PFOS yields reported in literature widely vary, from 3 % (Mejia Avendaño and Liu, 2015) to 30 % and more (Zabaleta et al., 2018; Zhao et al., 2016). A PFOS yield of 30 % would lead to PFOS concentrations that equal 72 % of the concentrations presented in Section 3.4.2.

3.6.4. Plant uptake

Available BAF values for cereal grains are very scarce (see SI.5) (Doucette et al., 2018; Krippner et al., 2014; Stahl et al., 2009; Wen et al., 2014). BAF values for wheat grains (0.054–0.062) are the highest BAFs reported in the literature; they were used in Section 3.5 as conservative values. Using the lower BAF values of oat (0.004–0.017) lead to concentrations expected in grains that are 6 % to 28 % of the values provided in Table 4.

In summary, the degradation rates of PCBs, PCDD/Fs, and PFOS in soils are very low, with half-lives ranging from a few years to hundreds of years, while lower molecular weight PAHs degrade faster. Migration of these contaminants in saturated soils is minimal due to high sorption, with PFOS taking some dozens of years to migrate 30 cm. Plant uptake of these contaminants is generally low.

4. Conclusion

To allow a safe circular economy through the use of bio-based fertilizers (BBFs), it is essential to assess the exposure of persistent organic substances in BBFs and the risk they constitute to the environment and food chain. Regarding the 19 studied BBFs - produced with different methods and from various waste materials - PCDD/F, PCB, PAH and PFAS concentrations were below the strictest limit values used in individual EU countries, except in one case. The present results thus represent a positive incentive for the implementation of the studied BBFs as alternatives to conventional inorganic fertilizers. BBFs produced from agricultural and food industry waste through hygienization processes were shown to be particularly promising. They contained extremely low concentrations of target substances, and the absence of thermochemical treatments retains nutrient solubility and fertilizer value of the product. However, attention must be paid to the relatively high concentrations of EOF in chicken-manure based BBFs, and our results suggest that future monitoring studies could target more PFAS, especially PFAA precursors

to ensure the safe use of these BBFs. In addition, the EOF concentrations in BBFs produced from sewage sludge were relatively high; therefore, additional PFAS should be included in the quantification methods as new knowledge increases about PFAA precursors, and fluorinated compounds in general. The use of pyrolysis and incineration to produce BBFs should probably be regarded as the preferred option for the valorization of organic waste containing elevated persistent organic substance concentrations. Expected concentrations of the target substances in soils, even upon maximal allowed BBF application rates, were at the low end of the background soil concentration ranges, indicating that the application of the selected BBFs does not constitute a substantial contribution to the contamination of agricultural soils by these legacy pollutants. This was confirmed by PCB and PFAS analyses conducted on soil samples from BBF field trials. In addition, the consumption of food from BBF-amended plots would only contribute minimally to reaching tolerable intake thresholds, as evidenced by modelling and measurements. Though these results are promising for BBFs considered in this study, it should be kept in mind that contaminated BBFs can reach the market, e.g., in case they are produced near a contaminant hotspot or made from a contaminated waste stream such as paper or sewage sludge. Therefore, close monitoring of contaminant concentrations in BBFs is critical.

The BBFs included in this article were in part selected because of their advanced commercialization stage. This study indicates that long-term use of commercial BBFs does not represent a risk for agricultural soils in terms of contamination with PCBs, PCDD/Fs, PAHs and target PFAS. However, it is still recommended to monitor for these and other hazardous substances in BBFs to ensure that the concentrations measured are in line with those of the BBFs reported in this study. To capture the effects of as many bioavailable contaminants as possible - including their potential interactions and pollutants that may not be detected by chemical analysis alone - it is also recommended to use ecotoxicological assays (Albert and Bloem, 2023).

To reduce the risk that non-monitored contaminants potentially present in BBFs enter the food chain via BBF-amended plants, an avenue for further investigation could be the addition of sorbents, such as bio-char, to the organic waste presenting the highest risk (e.g., manure or sewage sludge) (Kończak and Oleszczuk, 2018; Stefaniuk and Oleszczuk, 2016). The high sorption of contaminants to the sorbents would significantly reduce the availability of contaminants to plants (and other soil organisms). In addition, the plants would benefit from the reduced nutrient leaching in the amended soil (Knowles et al., 2011; Sarkhot et al., 2012).

CRediT authorship contribution statement

Nicolas Estoppey: Writing – original draft, Project administration, Methodology, Investigation, Conceptualization. **Emma R. Knight:** Writing – review & editing, Methodology, Investigation. **Ian J. Allan:** Writing – review & editing, Methodology, Investigation, Conceptualization. **Kuria Ndungu:** Writing – review & editing, Methodology, Investigation. **Gøril Aasen Slinde:** Project administration, Methodology, Investigation, Conceptualization. **Jan Thomas Rundberget:** Methodology, Investigation, Conceptualization. **Kari Ylivainio:** Writing – review & editing, Project administration, Methodology, Investigation. **Alicia Hernandez-Mora:** Writing – review & editing, Methodology, Investigation. **Erlend Sørmo:** Writing – review & editing, Methodology, Conceptualization. **Hans Peter H. Arp:** Writing – review & editing, Methodology, Funding acquisition, Conceptualization. **Gerard Cornelissen:** Writing – review & editing, Methodology, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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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.2024.177347>.

Data availability

Data will be made available on request.

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