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Substituting fossil-based mineral fertilizers with bio-based products – impacts on potentially toxic elements in soil and crops

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Similar to conventional mineral fertilizers, circular bio-based fertilizer (BBF) products may contain potentially toxic elements (PTEs). In this study, conducted as part of the LEX4BIO project, the contents of arsenic (As), cadmium (Cd), nickel (Ni) and lead (Pb) were surveyed in 37 phosphorus (P) BBFs and 7 nitrogen (N) BBFs representing different product function and component material categories. In addition, mass balances of these elements were calculated over 100-year scenarios of P fertilization with a product that was low or high in the assessed elements, a common mineral phosphate fertilizer (MPF), and a hypothetical fertilizer containing the maximum amount of these PTEs allowed by the EU regulations. The examination was performed using case-specific conditions for Finland, Denmark, France, Spain, Hungary, and Germany, in which countries the products had been tested. Although in some individual products the current maximum EU limits were exceeded for Cd and Ni, the contents of the targeted PTEs were overall low in the studied products. The dominant trends in the contents and fluxes of As, Ni, and Pb were decreasing in all fertilizer-use scenarios with observed BBF concentrations, but the maximum allowed rates would instead commonly lead to prominent increases. The highest sensitivity to variation in fertilizer input rates, along with some substantial increases in the environmental contents and fluxes was observed for Cd. However, with low-Cd BBFs, favorable development is also achievable for Cd fluxes. It is recommended that PTE levels remain well below the maximum EU limits.

Key words: mass balance modelling, arsenic, cadmium, lead, nickel

Introduction

Adverse leakage of nutrients from agricultural systems into the environment, together with the excessive use of natural resources pushes the agricultural sector toward closing nutrient cycles (Chojnacka et al. 2020). The desired circularity calls for the recovery of nutrients from various waste streams, such as agricultural, forest, and industrial residues, as well as wastewater sludges (Rosemarin et al. 2020). Biomasses and products derived from the waste streams may contain harmful elemental impurities (Möller and Schultheiβ 2015, Gong et al. 2019). Variability in the contents of potentially toxic elements (PTEs) in different waste materials is generally high. For instance, sewage sludge and wood ash may contain elevated levels of arsenic (As), cadmium (Cd), and lead (Pb) (Reimann et al. 2008, Dahl et al. 2010, Lopes et al. 2011). On the other hand, unwanted elements are also found in conventional fertilizers. In particular, phosphate rock-derived products often contain Cd, As, chromium (Cr), and Pb (Gupta et al. 2014). Thus, new circular products and processing technologies may either increase or decrease PTE loads associated with fertilization.

Fertilization-induced PTE pollution is regulated in many countries through maximum allowed limits set by national legislation to ensure the quality and safety of fertilizer products. At the EU level, Regulation 2019/1009, which lays down rules on the availability of EU fertilizing products on the market, establishes common rules for safety, quality, and labeling of inorganic, organic, and organo-mineral fertilizers, soil improvers, plant biostimulants, and growing media. The EU regulation specifies product function category (PFC)-specific maximum limits for Cd, Cr(VI), mercury (Hg), nickel (Ni), Pb, inorganic As, copper (Cu), and zinc (Zn). Among these PTEs, As, Cd, Ni, and Pb were considered in this study. The selection of elements prioritized their current relevance to food safety, as this study served as the basis for a separate dietary exposure assessment (Domínguez Carrasco et al. 2025). For Hg, dietary exposure risks are largely confined to methyl-Hg in seafood (El-Kady and Abdel-Wahhab 2018, Suomi et al. 2021), whereas Cr occurs mainly as Cr(III), which is of low toxicity in food, and no maximum levels for Cr in food have been set (EFSA 2014). Due to the low risk of exposure via food and analytical challenges of Hg and Cr speciation, these two elements were excluded from the study, along with the plant nutrients Cu and Zn.

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A mass balance modelling approach was applied to assess the effects of substituting mineral fertilizers with bio-based fertilizers (BBFs) on the occurrence and movement of the target PTEs in selected cropping systems. Mass balance modelling is a well-established tool for assessing the partitioning and transport of target elements or compounds across environmental compartments, such as air, water, soil, and plants (Wania and Mackay 1999). Models based on mass balance principles are characterized by low spatial and temporal resolution and simplifying assumptions that constrain the environmental system. Despite inevitable uncertainties, they are recognized as valuable tools for both scientific research and decision-support applications (MacLeod et al. 2010). This type of model typically uses constant empirical distribution coefficients (K_d), which describe the ratio of the total content in the solid phase to the total concentration in solution ($l\ kg^{-1}$) to assess the migration of PTEs. This simplistic approach does not account for the complexity of sorption processes or for variations across space and time (Degryse et al. 2009, Stockmann et al. 2017).

In reality, the fate of PTEs introduced into soils depends on both their inherent characteristics and environmental conditions, such as mineralogy, pH, redox potential, organic matter content, microbial activity, temperature, and moisture (Fageria et al. 2002, Kabata-Pendias 2004). Furthermore, measured K_d values tend to exhibit considerable variability (Sheppard 2011). When considering PTE uptake by plants, simple soil-to-plant transfer factors (TF), which express the ratio of total PTE content in plants to the total or available PTE content in soil, are often applied (e.g., Tome et al. 2003, Adamo et al. 2014). Comprehensive examples of risk assessments of PTEs based on mass balance modelling have been presented, for example, by VKM (2022) in Norway and Salo et al. (2018) in Finland. These reports also provide a comprehensive description of the behavior of the target PTEs (As, Cd, Ni, and Pb) in the environment.

The main aim of this study was to evaluate the impacts of adopting BBFs as a substitute for virgin mineral fertilizers on the loads of As, Cd, Ni, and Pb in a European context. Mass balance calculations accounting for sources, transport, and sinks were conducted over a 100-year timeframe for a case-specific field soil in Finland, Denmark, France, Spain, Hungary, and Germany, where BBF products were tested under field conditions as part of the LEX4BIO project (Frick et al. 2025). Predictive simulations were conducted for high- and low-PTE products within the tested set of BBFs, as well as for the mineral phosphate fertilizer (MPF) representative of each country. For comparison, maximum PTE-contents were applied in the calculations using the maximum allowed limits under EU legislation (EU 2019/1009) and the P content of the high-PTE products. The crops considered included wheat, barley, oats, rye, maize, and sunflower grown in a monoculture, and potato or carrot in rotation with barley. Representative values for soil PTE status, annual aerial deposition, runoff and erosion, rates of fertilizer and lime use, and harvestable yield were collected for each target country with support from local project partners. Where possible, soil PTE contents, transfer factors from soil to crops, and soil adsorption coefficients (K_d) were derived from field and laboratory experiments conducted as part of the LEX4BIO project; otherwise, literature values were used. The study produced estimates of the annual balances of the target PTEs ($g\ ha^{-1}$), their concentrations in soil water ($\mu g\ l^{-1}$), grain yield, and soil content ($mg\ kg^{-1}$), and the amounts ($g\ ha^{-1}$) leached, lost via erosion, and removed with the yield. The results provide insight into PTE-related risks associated with BBF use and serve as a basis for exposure assessments.

Material and methods

Studied fertilizer products and element analyses

Samples of 37 bio-based phosphorus (P) fertilizers and 7 nitrogen (N) fertilizers were collected from the target countries for total element analysis. Selected properties of the products are presented in the Supplementary material (Appendix 1 Table S1). The selection of both N- and P-fertilizers was based on representing different types of BBFs, classified according to their PFC and component material category (CMC) as defined in the Fertilising Products Regulation (EU 2019/1009). Some of the selected BBFs were commercially available, while others were still in the development stage but were considered to have potential for enhancing nutrient recycling in the future. Agronomic efficiency of most of the P-BBFs and mineralization potential of the N-BBFs have been assessed earlier by Hernandez-Mora et al. (2024) and Agostini et al. (2024), respectively. The majority of the products (n = 18) were solid organic fertilizers (PFC 1 A-I), followed by inorganic macronutrient fertilizers (PFC 1 C-I; n = 17) and solid organo-mineral fertilizers (PFC 1 B-I; n = 9). The feedstock materials included plant- and animal-derived side-streams, such as, biowaste, crop residues, meat and bone meal, manures, and sewage sludge.

Before analysis, all samples were ground to pass through a 1-mm sieve. Each sample was then digested in duplicate using microwave-assisted aqua regia digestion. Finally, the total element contents were measured by ICP-MS. The limits of detection were 0.02 mg As kg^{-1}, 0.001 mg Cd kg^{-1}, 0.09 mg Ni kg^{-1}, and 0.03 mg Pb kg^{-1}. The analyses were performed in the laboratory of the Natural Resources Institute Finland, located in Jokioinen.

Mass balance model structure

Mass balance calculations were based on estimated input rates of Cd, Pb, As, and Ni to cropland and losses from it, under monocultures of wheat, barley, oats, rye, maize, and sunflower, as well as rotations of 2-year potato or 2-year carrot with 3-year barley, in Finland, Denmark, France, Spain, Hungary, and Germany. The effects of the mineral phosphate fertilizer (MPF) typical for each country and two recycled fertilizer products with low (BBF_L) or high (BBF_H) PTE content (Fig. 1) on PTE fluxes were modelled over a 100-year period. Cd, Pb, As and Ni concentrations in MBF in each country were taken from the survey by Verbeeck et al. (2020), in which P concentrations of MPFs averaged 9%. The detailed PTE concentrations are presented in Appendix 2. BBFs were selected from those used in the P field experiments of the LEX4BIO project, resulting in the lowest or highest doses of the studied PTEs. PTE dosage to soil from fertilizers was assumed to origin only from P fertilizers, that would be applied by crop and country-wise recommendations based on soils with a satisfactory soil P availability (details in Appendix 2.) The P fertilizer rate ranged from 8 to 55 P kg ha^{-1}.

The BBF with the lowest content of all PTEs was struvite (CGO). With its high P content of 22.8% and an exemplary P application rate of 60 kg ha^{-1}, it led to PTE doses of 0, 0.004, 0.224, and 0.055 g ha^{-1}, for As, Cd, Ni, and Pb, respectively. The BBF with the highest content of Cd, Ni and Pb was ash derived from husks (P content 2.0%), which, at an exemplary P application rate of 60 kg ha^{-1}, resulted in PTE doses of 4.65, 79.7, and 9.51 g ha^{-1} for Cd, Ni, and Pb, respectively. The highest As dose was associated with sewage sludge-based ash (P content 8.1%), which, at an exemplary P application rate of 60 kg ha^{-1}, resulted in an As dose of 4.2 g ha^{-1}. In order to evaluate the effect of the maximum limits set by EU legislation (EU 2019/1009), the impact of the BBF fertilizers was calculated using the maximum PTE limits for inorganic fertilizers (MAX), including ashes with a sufficiently high P content: 40 mg kg^{-1} for As, 3 mg kg^{-1} for Cd, 100 mg kg^{-1} for Ni, and 120 mg kg^{-1} for Pb on a dry matter basis.

A list of parameters is provided in Table 1, with example values for Cd in a wheat monoculture in Finland. Parameter tables for all considered countries, elements, and crops are presented in the Supplementary material (Appendix 2). The parameter values were derived from data generated in laboratory and field experiments of the LEX4BIO project (e.g. Ylivainio et al. 2024), as well as from scientific literature and national recommendations provided by partners in the target countries. Inputs were allocated to the 0–20 cm topsoil with a soil bulk density of 1.0 kg dm^{-3}, and all outputs from the soil were also deducted from this topsoil layer (Fig. 1). The model calculations were conducted using a SAS program for a 100-year forward projection. An example of the SAS code is included in the Supplementary material (Appendix 3).

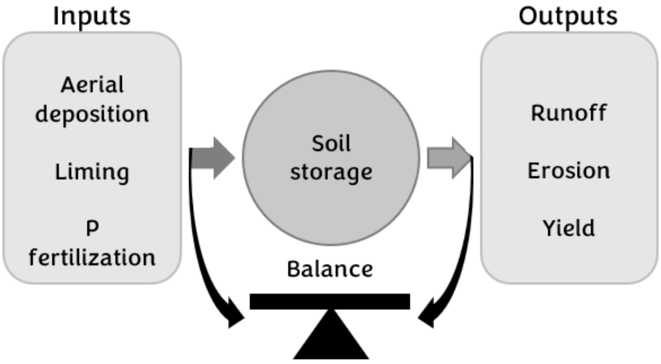

Fig. 1. Schematic presentation of the mass balance model

From the model calculations, values after the first-year simulation were documented to assess the initial stage following one year of applying the selected fertilizers. For these initial values, soil storage was the most decisive factor for most contents and fluxes, except for crop content and uptake. Fertilizer had negligible effects on the values after the first year. In order to assess the effect of continuous use of the selected fertilizers, the relative change was calculated as the percentage of contents or fluxes in the hundredth year relative to those in the first year. Since all the parameters used to create fluxes are coefficients that extract a proportion from soil storage, PTE contents and fluxes for each country, crop and fertilizer combination change by the same magnitude.

Table 1. Parameters used in annual mass balance (g ha^{-1}) calculations[1] for As, Cd, Ni, and Pb in croplands across Finland, Denmark, France, Germany, Hungary, and Spain. Calculations were performed under monocultures of wheat, barley, oats, rye, maize, and sunflower, as well as for potato or carrot in rotation with barley. Example values are provided for Cd in a wheat monoculture in Finland. Corresponding values for the other crops and countries are available in the Supplementary material (Appendix 2).

Model parameter	Unit	Value (FIN)	Reference
Total content in soil	mg kg^{-1} dry mass	0.173	Mäkelä-Kurtto et al. 2003, Reimann et al. 2014, Salo et al. 2018
Aerial deposition	g ha^{-1} y^{-1}	0.35	Mäkelä-Kurtto et al. 2007
Annual runoff	mm	200	Finnish Environment Institute 2000
Annual erosion	kg dry mass ha^{-1} y^{-1}	500	Finnish Environment Institute 2000
Content in liming materials	mg kg^{-1} dry mass	0.15	Mäkelä-Kurtto et al. 2007
Average liming rate[1]	kg ha^{-1} y^{-1}	400	annual recommendation
Content in mineral-P fertilizer[2]	mg kg^{-1} P	2.5	Mäkelä-Kurtto et al. 2003
Average P fertilization rate	kg ha^{-1} y^{-1}	10	annual recommendation
Average yield level	kg dry mass ha^{-1} y^{-1}	3225	Eurostat 2023, yield average 2013–2022
Soil-to-plant transfer factor (TF)	-	0.1296	Smolders 2013
Soil adsorption coefficient (K_d)	l kg^{-1}	1213	LEX4BIO-data

[1] The average liming rate was expressed on an annual basis, although in practice, liming would be applied at five- or ten-year intervals.

[2] The calculation was conducted for a biobased P-fertilizer low in PTEs (BBF_L), a biobased P-fertilizer high in PTEs (BBF_H) or a P-fertilizer meeting the maximum limits set by EU fertilizer legislation, and a conventional mineral-P fertilizer (MPF). For each of the PTEs considered, the computation produced annual estimates of mass balance, uptake in yield, concentrations in soil water, soil, and yield, as well as annual losses through leaching and erosion.

Results

PTE contents in bio-based fertilizers

The minimum, median, and maximum values of total Cd, Pb, As, and Ni in the assessed N and P fertilizer products are shown by PFC in Table 2. In order to present the composition of the products comprehensively, Table 2 also contains the corresponding values for silver (Ag), beryllium (Be), cobalt (Co), Cr, Cu, manganese (Mn), molybdenum (Mo), rubidium (Rb), selenium (Se), strontium (Sr), uranium (U), vanadium (V), and Zn, though these elements are not discussed further in this paper. Overall, the data indicate a wide range of contents, from below the limit of detection to >100 mg kg^{-1} on a dry weight basis. No clear trend in element levels between the PFCs was observed. A preliminary analysis of the raw materials and processing technologies used in the BBFs revealed that products containing sewage and/or industrial sludges generally had the highest concentrations of As, Cd, Ni, and Pb. Additionally, pig- and poultry-manure-based products were frequently among those with elevated Cd levels (Supplementary material, Appendix 1 Table S1). Treatments that concentrate biomass, such as incineration, charring, composting, and digestion, were found to produce PTE-rich products.

Table 2. Minimum, median, and maximum total contents (mg kg^{-1} dry weight) of selected elements in bio-based fertilizers by product function category (PFC): solid organic fertilizers (n = 18), solid organo-mineral fertilizers (n = 9), and inorganic macronutrient fertilizers (n = 17)

Element (mg kg^{-1} dw)	Solid organic fertilizers (PFC 1 A-I)			Solid organo-mineral fertilizers (PFC 1 B-1)			Inorganic macronutrient fertilizers (PFC 1 C-I)		
	Min	Med	Max	Min	Med	Max	Min	Med	Max
Silver (Ag)	<0.04	0.1	3.9	<0.04	<0.04	3.3	<0.04	0.1	9
Arsenic (As)	<0.02	0.7	10	<0.02	0.7	19	<0.02	0.7	10
Beryllium (Be)	<0.002	0.1	1.1	<0.002	0.1	1.0	<0.002	0.1	1.8
Cadmium (Cd)[1]	<0.001	0.1	15	<0.001	0.6	4.2	<0.001	0.1	2.8
Cobalt (Co)	0.1	1.2	14	0.1	1.6	18	<0.08	1.5	13
Chromium (Cr)	0.4	10	128	0.7	15	79	0.2	22	628
Copper (Cu)	1.9	54	928	3.5	20	353	0.5	84	555
Manganese (Mn)	2.1	160	1305	4.1	82	943	<12	406	3425
Molybdenum (Mo)	<0.01	2.0	16	0.2	3.1	8.7	<0.01	3.1	22
Nickel (Ni)	0.6	11	72	0.2	7.6	49	0.1	17	159
Lead (Pb)	<0.03	1.5	75	0.2	1.4	23	<0.03	3.9	17
Rubidium (Rb)	1.2	14	61	3.7	12	36	<0.4	9	310
Selenium (Se)	<0.01	1.1	72	0.2	1.2	7.6	<0.01	0.9	159
Strontium (Sr)	6.5	65	1325	20	91	493	<0.6	102	851
Uranium (U)	<0.001	0.3	27	<0.001	2.4	62	<0.001	0.3	34
Vanadium (V)	<0.8	4.2	61	<0.8	7.2	45	<0.8	4.6	53
Zinc (Zn)	101	285	1135	89	118	736	<10	133	1947

[1] The total P content exceeded 5% phosphorus pentoxide (P_2O_5) equivalent by mass in seven organo-mineral fertilizers and 15 inorganic fertilizers. The minimum, median and maximum values for these products calculated as mg Cd kg^{-1} P_2O_5 were 0.1, 9.6, and 36 for organo-mineral fertilizers and < 0.001, 0.2, and 11 for mineral fertilizers.

Effects of fertilizer use on PTE fluxes

The initial values generated by the mass balance model calculation (at the 1-year time point) for As, Cd, Ni, and Pb in soil (mg kg^{-1}), soil water (µg l^{-1}), and crop yield (µg kg^{-1}), as well as the fluxes of these PTEs (g ha^{-1}) via leaching, erosion, and crop harvest in the target countries are given in Appendix 1, Tables S2 and S3. In Finland, maize is rarely grown; therefore, it was not included. At the 1-year time point, the rotations and fertilization scenarios had negligible effects on soil content, soil water, leaching and erosion; thus, common values are provided for each element and country (Appendix 1, Table S2). PTE content in yield and uptake of PTEs are shown for wheat, maize and the carrot–barley rotation in Appendix 1, Table S3, and for other crops in Appendix 4. Prominent differences between countries were evident, reflecting geographical variation in PTE occurrence across Europe as well as variation in distribution coefficients from soil to soil water and to plants.

The effects of a 100-year application of MPF, BBF$_H$, BBF$_{L,}$ and the hypothetical BBF MAX on the annual BBF balance (g ha^{-1}) and the relative change (%) induced in the amounts and fluxes of the studied PTEs due to different fertilizer applications are shown by PTE in Tables 3–6 for wheat, maize, and carrot–barley rotation. Because of the structure of the model, the relative change is equal for all fluxes and contents. The corresponding values of other modelled crops (barley, oats, rye, sunflower, and potato–barley rotation) are given in the Supplementary material (Appendix 4).

With regard to As, differences in the relative change in contents and fluxes between the MPF, BBF_L, and BBF_H were overall small in all target countries (Table 3). The use of these fertilizers led to a decreasing trend in As occurrence with only one marginally positive value recorded for maize in Denmark. The highest decrease, approximately, 30%, was recorded in Spain while in Germany and France, the decrease was also prominent, at nearly 20%. In Finland and Denmark, these fertilizer-use scenarios induced changes of only a few percentage points in As contents and fluxes. However, in these countries the hypothetical scenario with a fertilizer containing the maximum allowed amount of As lead to a clearly positive As balance, whereas in the other countries, the As balance remained negative even in this scenario.

Table 3. Annual As balance (g ha^{-1}) after a 100-year simulation of various P-fertilizer use scenarios, including a mineral phosphate fertilizer typical for the country in question (MPF), a low-As BBF_L, a high-As BBF_H, and a hypothetical fertilizer containing the maximum allowed As content (MAX) in monocultures of spring wheat and maize and in carrot–barley rotation. The relative change represents the difference from the initial status in year 1 and is equal for all modelled parameters.

		Wheat		Maize		Carrot	
Country	Scenario	Annual balance (g ha^{-1})	Relative change (%)	Annual balance (g ha^{-1})	Relative change (%)	Annual balance (g ha^{-1})	Relative change (%)
Denmark	MPF	-1.5	-1.7	-0.2	-0.2	-0.4	-0.9
	BBF_L	-2.6	-3.0	-1.9	-2.2	-2.0	-2.6
	BBF_H	-1.3	-1.4	0.1	0.2	-0.2	-0.6
	MAX	6.5	7.5	12	14.3	11.1	11.2
Finland	MPF	-2.2	-3.2			-2.2	-3.4
	BBF_L	-2.3	-3.3			-2.5	-3.6
	BBF_H	-1.6	-2.3			0.3	-1.3
	MAX	2.5	3.7			16.7	12.2
France	MPF	-126	-17	-122	-17	-124	-17
	BBF_L	-128	-18	-124	-17	-125	-18
	BBF_H	-127	-17	-122	-17	-124	-17
	MAX	-118	-16	-115	-16	-117	-17
Germany	MPF	-52	-18	-50	-17	-51	-18
	BBF_L	-53	-18	-51	-18	-52	-18
	BBF_H	-51	-18	-50	-17	-50	-18
	MAX	-40	-14	-47	-16	-42	-14
Hungary	MPF	-13	-6.8	-13	-7.0	-13	-7.1
	BBF_L	-14	-7.1	-14	-7.1	-14	-7.4
	BBF_H	-12	-6.2	-13	-6.8	-13	-6.6
	MAX	-0.85	-0.45	-9.9	-5.2	-4.3	-1.6
Spain	MPF	-79	-34	-74	-33	-75	-34
	BBF_L	-80	-35	-76	-34	-76	-34
	BBF_H	-79	-34	-74	-33	-75	-34
	MAX	-70	-31	-67	-30	-67	-31

The relative changes in contents and fluxes of Cd in the 100-year fertilizer-use scenarios exhibited different patterns in different countries (Table 4). In Spain, Hungary, Denmark, and to a lesser extent in France, fertilization with conventional MPF led to a notable increase in Cd contents and fluxes, whereas in Finland and Germany the corresponding values showed a decrease. In all countries except Hungary, the use of $\mathrm{BBF_L}$ led to a clear decrease in the Cd contents and fluxes. The use of $\mathrm{BBF_H}$, increased the Cd contents and fluxes in all countries instead. In Spain, the increase in Cd induced by $\mathrm{BBF_H}$ was smaller than the increase caused by MPF use, whereas in Hungary, Denmark, and France the opposite was observed. The scenario with the maximum permitted Cd content (MAX) led to substantial increases in Cd occurrence and environmental fluxes in all the target countries.

Table 4. Annual Cd balance (g ha^{-1}) after 100-year simulation of various P-fertilizer use scenarios, including a mineral phosphate fertilizer typical for the country in question (MPF), a low-Cd $\mathrm{BBF_L}$, a high-Cd $\mathrm{BBF_H}$, and a hypothetical fertilizer containing the maximum allowed Cd content (MAX) in monocultures of spring wheat and maize, and in carrot–barley rotation. The relative change represents the difference from the initial status in year 1 and is equal for all modelled parameters.

		Wheat		Maize		Carrot	
Country	Scenario	Annual balance (g ha^{-1})	Relative change (%)	Annual balance (g ha^{-1})	Relative change (%)	Annual balance (g ha^{-1})	Relative change (%)
Denmark	MPF	0.94	24	1.7	43	1.4	29
	$\mathrm{BBF_L}$	-0.22	-5.6	-0.18	-4.6	-0.4	-9.1
	$\mathrm{BBF_H}$	1.3	32	2.1	55	1.9	40
	MAX	2.7	69	4.4	110	4.2	87
Finland	MPF	-0.01	-0.24			-0.41	-3.8
	$\mathrm{BBF_L}$	-0.03	-0.90			-0.51	-5.2
	$\mathrm{BBF_H}$	0.74	20			2.6	40
	MAX	1.5	40			5.6	84
France	MPF	0.76	12	0.54	8.5	0.55	4.81
	$\mathrm{BBF_L}$	-0.79	-12	-0.80	-13	-0.77	-14
	$\mathrm{BBF_H}$	0.84	13	0.61	9.6	0.62	5.8
	MAX	2.45	38	2.0	31	1.99	25
Germany	MPF	-0.48	-7.7	-0.77	-12	-0.57	-9.2
	$\mathrm{BBF_L}$	-0.80	-13	-0.86	-14	-0.81	-14
	$\mathrm{BBF_H}$	1.3	20	-0.27	-4.4	0.74	15
	MAX	3.3	53	0.30	4.9	2.3	44
Hungary	MPF	1.7	40	0.39	9.2	1.3	34
	$\mathrm{BBF_L}$	0.06	1.4	-0.07	-1.7	0.03	0.35
	$\mathrm{BBF_H}$	2.1	50	0.51	12	1.6	43
	MAX	4.1	98	1.1	26	3.1	85
Spain	MPF	2.0	44	1.4	31	1.5	28
	$\mathrm{BBF_L}$	-0.61	-14	-0.81	-18	-0.7	-16
	$\mathrm{BBF_H}$	1.0	23	0.60	13	0.69	12
	MAX	2.6	59	2.0	43	2.0	39

Ni contents and fluxes generally showed a decreasing trend in all countries and fertilizer-use scenarios, except under the MAX Ni application. The decrease was smaller with BBF$_{H}$ than with MPF or BBF$_{L}$ (Table 5). In Finland, an increase in Ni was recorded with the BBF$_{H}$, and in all target countries except Spain, the MAX scenario prominently increased Ni amounts and fluxes.

Table 5. Annual Ni balance (g ha^{-1}) after a 100-year simulation of various P-fertilizer use scenarios, including a mineral phosphate fertilizer typical for the country in question (MPF), a low-Ni BBF$_{L}$, a high-Ni BBF$_{H}$, and a hypothetical fertilizer containing the maximum allowed Ni content (MAX), in monocultures of spring wheat, maize, and in carrot–barley rotation. The relative change represents the difference from the initial status in year 1 and is equal for all modelled parameters.

		Wheat		Maize		Carrot	
Country	Scenario	Annual balance (g ha^{-1})	Relative change (%)	Annual balance (g ha^{-1})	Relative change (%)	Annual balance (g ha^{-1})	Relative change (%)
Denmark	MPF	-35	-25	-31	-23	-23	-26
	BBF$_{L}$	-41	-30	-41	-30	-30	-31
	BBF$_{H}$	-16	-12	-1.3	-1.0	-2.5	-7.7
	MAX	56	40	112	80	77	59
Finland	MPF	-6.2	-2.1			-8.0	-2.4
	BBF$_{L}$	-6.3	-2.2			-8.3	-2.4
	BBF$_{H}$	7.0	2.4			45	7.9
	MAX	45	15			197	37
France	MPF	-34	-7.8	-34	-8.0	-46	-9.3
	BBF$_{L}$	-38	-8.8	-38	-8.8	-50	-10
	BBF$_{H}$	-9.8	-2.3	-14	-3.2	-26	-5.1
	MAX	70	16	56	13	42	9.3
Germany	MPF	-76	-13	-75	-13	-98	-15
	BBF$_{L}$	-78	-14	-76	-13	-100	-15
	BBF$_{H}$	-43	-7.4	-66	-11	-73	-9.9
	MAX	58	9.9	-38	-6.5	3.3	5.2
Hungary	MPF	-34	-6.0	-35	-6.3	-51	-7.5
	BBF$_{L}$	-37	-6.6	-36	-6.5	-54	-8.0
	BBF$_{H}$	-1.9	-0.3	-26	-4.7	-27	-2.5
	MAX	99	18	2.6	0.5	50	13
Spain	MPF	-120	-19	-124	-19	-140	-20
	BBF$_{L}$	-130	-20	-129	-20	-140	-21
	BBF$_{H}$	-100	-16	-105	-16	-120	-18
	MAX	-21	-3.3	-35	-5.5	-49	-8.0

Pb contents and fluxes decreased in all fertilizer-use scenarios, except for MAX, in Germany, Spain, France, and to a lesser extent, in Denmark (Table 6). In contrast, in Finland and Hungary, the corresponding values increased, and the increase was slightly higher with BBF$_{H}$ than with MPF or BBF$_{L}$. Applying a fertilizer containing the maximum permitted amount of Pb would prominently increase Pb occurrence and fluxes in all target countries.

Considering the assessed crops, the model calculation showed no clear systematic differences in either the annual balances or the relative changes of PTE contents and fluxes between rotations of wheat, maize, and carrot–barley (Tables 3–6). There were some isolated deviations in the responses of Cd and Ni, but they differed between crops and countries. For example, in Finland, larger changes in Cd were observed with carrot than with wheat, whereas in Spain, the trend was reversed.

Table 6. Annual Pb balance (g ha^{-1}) after a 100-year simulation of various P-fertilizer use scenarios, including a mineral phosphate fertilizer typical for the country in question (MPF), a low-Pb BBF$_L$, a high-Pb BBF$_H$, and a hypothetical fertilizer containing the maximum allowed Pb content (MAX) in monocultures of spring wheat and maize, and in carrot–barley rotation. The relative change represents the difference from the initial status in year 1 and is equal for all modelled parameters.

		Wheat		Maize		Carrot	
Country	Scenario	Annual balance (g ha^{-1})	Relative change (%)	Annual balance (g ha^{-1})	Relative change (%)	Annual balance (g ha^{-1})	Relative change (%)
Denmark	MPF	-4.2	-1.7	-3.7	-1.5	-5.3	-2.0
	BBF$_L$	-5.1	-2.1	-5.1	-2.1	-6.7	-2.5
	BBF$_H$	-2.1	-0.9	-0.4	-0.2	-2.1	-0.8
	MAX	112	46	180	74	170	61
Finland	MPF	3.2	1.5			3.1	1.7
	BBF$_L$	3.1	1.5			3.1	1.6
	BBF$_H$	4.7	2.3			9.4	3.4
	MAX	65	32			250	69
France	MPF	-43	-6.3	-43	-6.3	-47	-6.7
	BBF$_L$	-44	-6.4	-43	-6.4	-48	-6.8
	BBF$_H$	-40	-5.9	-41	-6.0	-45	-6.4
	MAX	86	13	69	10	63	8.0
Germany	MPF	-61	-11	-61	-12	-65	-12
	BBF$_L$	-61	-12	-61	-12	-65	-12
	BBF$_H$	-57	-11	-60	-11	-62	-11
	MAX	102	19	-15	-2.8	59	15
Hungary	MPF	12	3.5	8.4	2.5	9.6	3.3
	BBF$_L$	11	3.3	8.2	2.4	9.1	3.1
	BBF$_H$	15	4.5	9.4	2.8	12	4.2
	MAX	170	51	55	16	133	45
Spain	MPF	-58	-12	-58	-12	-60	-13
	BBF$_L$	-60	-13	-60	-13	-62	-13
	BBF$_H$	-57	-12	-57	-12	-60	-13
	MAX	69	15	52	11	48	8.0

Discussion

PTE contents in bio-based fertilizers

In all the studied fertilizers, Pb content was clearly below the common maximum limit of 120 mg kg^{-1} (dry matter) set by the EU Regulation (EU 2019/1009). For As, the maximum EU limit of 40 mg kg^{-1} (dm) applies to inorganic As rather than total content, but in all current products, total As content was far below this limit. Cd content fell within the acceptable range (<1.5 mg kg^{-1} (dm) in organic fertilizers and <3 mg kg^{-1} (dm) or <60 mg kg^{-1} P_2O_5 (dm) in organo-mineral and inorganic fertilizers containing less than or more than 5 mass% of P_2O_5-equivalent P, respectively). Only one plant-based organic BBF had an exceptionally high Cd content, with a P_2O_5 content of 15.5% (dm) and a Cd content of 15 mg kg^{-1}, due to the addition of apatite to increase P content (personal communication). Discarding the extreme outlier would have led to a maximum value of 0.9 mg Cd kg^{-1} (dm) in PFC 1 A-I. For Ni, the maximum regulatory limits are 50 mg kg^{-1} (dm) in organic and organo-mineral fertilizers and 100 mg kg^{-1} (dm) in inorganic macronutrient fertilizers. These limits were exceeded in one charred organic fertilizer and two inorganic struvite products.

Despite occasional breaches of the maximum EU limits (EU 2019/1009), the contents of As, Cd, Ni, and Pb were overall low in the studied products. The samples were collected before the EU fertilizer regulation entered into force, which may explain the few unacceptable PTE levels recorded. A previous survey on compost and digestate samples across Europe (Tavazzi et al. 2013) reported higher maximum total contents for As (40 mg kg^{-1}), Ni (250 mg kg^{-1}), and Pb (270 mg kg^{-1}), but a somewhat lower maximum value for Cd (2.8 mg kg^{-1}) compared to the current data of N- and P-BBFs. In comparison to an extensive study (n = 117) on PTE contents of organic fertilizers produced in northern China, the current EU median and maximum values were comparable in magnitude to Ni, and lower for As, Cd, and Pb except for the exceptionally high maximum value for Cd, which clearly exceeded the 5 mg kg^{-1} recorded in the Chinese data (Gong et al. 2019). Individual PTE contents exceeding or approaching the maximum allowed limits in the current data emphasize the need for regular quality control in BBF production and surveillance.

Conventional mineral fertilizers, especially those containing phosphates, also contain PTEs derived from the rock source. When comparing EU mineral P fertilizers (Verbeeck et al. 2020) and the current circular macronutrient P fertilizers, the median and maximum contents were clearly higher in the mineral fertilizers than in the BBFs for As and Cd. For Ni and Pb, the inorganic BBFs exhibited higher median values but lower or similar maximum values compared to the mineral P fertilizers. This highlights that the inclusion of mineral P sources to fortify the P content of a BBF, which is rare but does occur, may introduce PTE contamination into the final product.

In the current data, products derived from or containing sewage sludge tended to be relatively rich in PTEs. Furthermore, poultry manure was identified as a raw material potentially high in PTEs. Sewage sludge is known to contain various amounts of PTEs, mostly derived from industrial wastewater and surface runoff (Fijalkowski et al. 2017), whereas mineral supplements in animal feeds are the main sources of PTEs in animal manure (Nicholson et al. 1999). Among processing technologies, the present data showed that thermal treatments (incineration and charring) produced products with relatively high PTE contents. As thermal processing efficiently reduces the volume of the treated biomass via decomposition of the organic fraction, thermally stable PTEs, which do not readily volatilize, concentrate in the residue (Udayanga et al. 2018).

Effects of fertilizer use on PTE fluxes

The estimates of soil PTE contents, atmospheric deposition, and rates of runoff and erosion presented in Table 1 and Supplementary material (Appendix 2) and used in the mass-balance model in assessing the BBF-induced changes in the PTE contents and fluxes, were derived from small and often local datasets and may even contain point values. The assessment thus fails to capture nationwide variations in environmental characteristics such as soil texture and mineralogy, acidity, organic carbon, temperature, and moisture (Shi et al. 2018). Furthermore, the soil-to-plant transfer and solid phase-to-solution distribution of elements were simplified to constant empirical coefficients without accounting for the complexity of the sorption processes and their variations in space and time (Degryse et al. 2009, Adamo et al. 2014). Therefore, the model outcomes should be taken as indicative approximations of the magnitude and direction of change in different European conditions and interpreted as relative differences from the conventional practice of mineral-P fertilization.

For As, Ni, and Pb, the fertilizer-use scenarios with observed BBF PTE contents gave favorable results, with decreasing PTE occurrence and fluxes, except for Pb in Finland and Hungary. The PTE contents in European soils vary widely, primarily due to differences in natural geochemical background (Tóth et al. 2016, Reimann et al. 2018). However, the observed decreases can be interpreted as reflecting general reductions in PTE inputs. Both atmospheric metal deposition (Harmens et al. 2010, Türtscher et al. 2017) and P fertilizer consumption (van Dijk et al. 2016, Panagos et al. 2022) have declined from the peak levels observed in previous decades. It should be noted that, though the BBF$_H$ represented a product from the upper range of current As, Ni, and Pb contents, the observed values were far from the maximum levels allowed by the EU regulation (EU 2019/1009). Using the maximum allowed PTE content led to marked increases in the occurrence and fluxes of Pb in all target countries, Ni in all countries except Spain, and of As in Denmark and Finland. Maintaining PTE contents well below the maximum allowed limits can thus be recommended in BBF production to ensure the desired decreasing trend in PTE fluxes.

Among the studied PTEs, Cd showed the highest sensitivity to variation in the fertilizer inputs. The amount of PTEs introduced to soil through fertilizers constituted a significant share of the total PTE input to the crop rotations, but it was still low compared to the total amount of these elements in soil. While Cd contents in soil were 50–90 times lower than those of As, Ni, and Pb, Cd input in fertilizers was only approximately 10 times lower than that of the other studied PTEs. Cadmium in fertilizers thus had a greater effect on soil fluxes than As, Ni, or Pb. Moreover, fertilizers containing relatively high amounts of Cd (BBF$_H$, the hypothetical MAX, and in some countries MPF) induced marked increases in Cd contents and fluxes. Six and Smolders (2014) calculated the mass balance of Cd for European agricultural soils under cereal or potato cultivation and predicted a decrease of 15% in topsoil Cd over the next 100 years as an EU average. However, they reported regional variation, with a predicted increase in soil Cd by 15% in Spain, which is still lower than the estimates in the current study. In a detailed mass balance calculation from France, Sterckeman et al. (2018) reported that P fertilizer applications accounted for approximately

74% of soil Cd inputs. According to their calculations, current cultivation practices (12 kg P ha^{-1}) are expected to lead to an increase of 3–5% in soil Cd in a century, which is just below the 5–12% range obtained for France in the present study with P input rates between 10 and 21 kg P ha^{-1}. In Sterckeman et al. (2018), a small decrease of 3–5% in Cd was achieved in a scenario restricting MPF use to levels that met crop needs (9 kg P ha^{-1}) and limiting the Cd content in fertilizers. The present calculations suggest a more prominent decrease of approximately 13% if MPF were replaced with a BBF low in Cd. In agreement with the current results, Sterckeman et al. (2018) found crop rotations to have less influence on the Cd balance than fertilizer Cd content. Since it is well established that Cd accumulation poses risks to soil, the aquatic environment, and human health (Clemens et al. 2013, Salo et al. 2018), restricting Cd levels in circular products is essential. Excessive exposure to Cd through diet has been identified as a cause of kidney damage (EFSA 2009) and is also linked to an increased risk of osteoporosis in women (Engström et al. 2011).

Conclusions

The current study showed European BBFs to contain mainly low or moderate amounts of As, Cd, Ni, and Pb, though some individual products high in these PTEs emphasized the need for regulations and surveys. Substituting conventional mineral phosphates, which also contain PTE impurities, with BBFs does not appear to pose a risk of increased As, Ni, and Pb fluxes in the environment. Cd showed the highest sensitivity to variation in fertilizer input rates, with substantial increases in Cd contents and fluxes associated with the application of products with relatively high Cd levels. Application of fertilizer products containing the maximum allowed levels of As, Ni, Cd, and Pb (EU 2019/1009) resulted in significant increases compared to current levels in the occurrence and fluxes of these elements. Therefore, maintaining PTE levels well below the maximum allowed limits can be recommended to support favorable trend in agricultural PTE fluxes.

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