

# Long-term soil accumulation of potentially toxic elements and selected organic pollutants through application of recycled phosphorus fertilizers for organic farming conditions

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**Abstract** The pollutant input and the risk of contamination by long-term application of recycled P fertilizers (RPFs) in European agricultural soils were estimated by a mass balance approach. Calculations based on literature data were carried out for the potentially toxic elements (PTEs) Cd, Cr, Cu, Ni, Pb, Zn and for the persistent organic pollutants (POPs) PCBs, PAHs and polychlorinated dibenzo-dioxins and -furans. The PTE accumulation estimation during 200 years of fertilizer application, equivalent to  $11 \text{ kg P ha}^{-1} \text{ year}^{-1}$ , the mean P export via harvested products on European stockless organic farms,

regarded soil background concentrations and proposed threshold concentrations. For PTEs, inputs were fertilizer application, atmospheric deposition and liming, output processes were leaching and crop harvest. The effect of varying site conditions was assessed by considering two precipitation excess (F) values and two soil pH values. For POPs, fertilizer application and the half-life time were considered. The PTE accumulation risk was low for most RPFs. For the analysed POPs no accumulation was found. The highest accumulation was found for all PTEs at pH 7 and  $F = 0.1 \text{ m year}^{-1}$ , the lowest at pH 5 and  $F = 0.3 \text{ m year}^{-1}$ . A high P concentration in fertilizers resulted in a low PTE flow per unit of P. Composts had the highest PTE accumulation risk due to lowest P contents. Struvite, meat and bone meal, digestate of catering waste, ash and biosolids would be better suited as P fertilizers. The use of fertilizers should be regulated based on their pollutant-to-nutrient ratio.

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## Introduction

Phosphorus (P) is an essential element for all living organisms and food production, but it is not treated sustainably in Europe. Through fertilization it ends up

in surface water bodies or it is irretrievably lost by co-incineration in municipal solid waste incinerators, power plants or cement kilns (Egle et al. 2015, 2016). At the current rate of extraction, phosphate rock reserves could peak in 2033 (Cordell and White 2011) and will be exhausted in the next few centuries (Elser 2012; Desmidt et al. 2014). This demonstrates that the reliance on non-renewable P sources for production of P fertilizers directly contradicts both the principles of sustainable development and the principle of care in organic farming (IFOAM 2017). Furthermore, phosphate rock sources are increasingly polluted with cadmium (Cd), thallium and the radioactive elements uranium and thorium (Egle et al. 2016). Therefore, we need a more efficient and sustainable P recycling within our societies.

The P status of long-term organically managed farms is currently becoming an issue of increasing concern (Nesme et al. 2012; Kolbe 2015). Phosphorus balances for organically managed arable cropping systems indicate that often more P is removed with the products than recycled via external inputs (Watson et al. 2002; Berry et al. 2003; Kolbe 2015; Ohm et al. 2017), leading to decreasing soil status in plant available soil P (Løes and Ogaard 1997; Kolbe 2015) which threatens the long-term sustainability of organic farming systems. Moreover, P shortage may also limit other processes such as symbiotic N<sub>2</sub> fixation (Römer and Lehne 2004; Oberson et al. 2013) that have a direct impact on the farm nitrogen cycle and on yields. Therefore, outbalancing P removals via exported products and appropriate P fertilization is a major challenge in the organic farming sector for preservation of the long-term soil fertility. According to the principles of organic farming, non-renewable inputs should be reduced by reuse, recycling and efficient management of materials and energy in order to maintain and improve environmental quality, therefore, organic farming should decrease dependency on phosphate rock.

The most important potential recycling sources of P in our societies are (Ott and Rechtberger 2012; Zoboli et al. 2016):

- Wastewater based residues.
- Slaughterhouse wastes (e.g. meat and bone meal).
- Urban organic waste.

These P sources, provided an efficient recycling, might replace approximately 50–70% of the current P import from phosphate rock in European societies (Klinglmair et al. 2015; Zoboli et al. 2016). Koppelaar and Weikard (2013) analyzed the global P flow and found a substantial scope for P use reduction by 32% of phosphate rock supply in 2009, and 28% technologically recyclable P from waterways and wastewaters, but at high costs. However, any recycling may be hazardous due to the risk of the transfer of potentially toxic elements (PTEs), pathogens and contaminants, which may harm the product quality, the long-term soil fertility and the environment. These concerns for example have led to a ban of sewage sludge based recycled P fertilizers (RPFs) in organic farming in the seventies of last century. There is a huge body of publications addressing the potential consequences of nutrient recycling on soil contamination. Most of the publications address biosolids obtained after stabilization and hygienization of wastewaters (Sekhar et al. 2002; Herter et al. 2003; Udom et al. 2004; Dai et al. 2006; Eljarrat et al. 2008; Nogueira et al. 2009; Wu et al. 2012; Tavazzi et al. 2012), the most relevant potential P source. The loads of PTEs and organic contaminants of composts and digestates obtained from organic household wastes were also an issue of discussion (e.g. Amlinger et al. 2004; Berset and Holzer 1995; Brändli et al. 2007a, b; LfU 2007; Smith 2009a; Govasmark et al. 2011; Mairitsch et al. 2011). The two main groups of concern are PTEs and persistent organic pollutants (POPs). Potentially toxic elements may accumulate in the soil and lead to phytotoxic effects reducing the productivity of the soil, or they are taken up by the crops compromising the food quality and animal as well human health (Puschenreiter and Horak 2000; Kabata-Pendias 2011; de Vries et al. 2013). Persistent organic pollutants are substances that persist in the environment and pose a risk of causing adverse effects to human health and the environment. This group of priority pollutants consists of pesticides, industrial chemicals (such as polychlorinated biphenyls, PCBs) and unintentional by-products of industrial processes (such as dioxins and furans) (EU 2016). Polychlorinated biphenyls are a group of 209 congeners that were used in transformers, sealing compounds, isolators, coolants, flame retardants, plasticisers and other applications (Leschber 2006). Polychlorinated dibenzo-*p*-dioxins (PCDDs) include 75 congeners, polychlorinated dibenzo-furans

(PCDFs) 135 congeners, together they are addressed as polychlorinated dibenzo-dioxins and -furans (PCDD/Fs) (Leschber 2006). They are emitted from metal industry, chemical industry, waste combustion plants and illegal waste combustion, in which the purification of waste gases was a big contributor to the reduction of pollution (Herter et al. 2003; Leschber 2006). Polycyclic aromatic hydrocarbons (PAHs) include several hundred congeners and have a wide diversity of physical, chemical and toxicological characteristics. They are found as by-products of incomplete combustion (Leschber 2006; Lerda 2011).

Currently, 80–90% of societal recoverable P sources are not permitted in organic farming due to public or private legislation standards (Løes et al. 2017). The average net P export of the organic sector accounted for approximately 6–7 kg P ha<sup>-1</sup> year<sup>-1</sup> (Kolbe 2015; Cooper et al. 2017). The remaining 10–20% of the societal recoverable P sources, currently permitted in organic farming, will not meet the P needs of the entire organic sector, even assuming that they are exclusively used on organically managed fields. This situation asks for a re-design of the P recycling and P fertilization approach within the organic community. The challenge is to increase the P recycling efficiency without compromising the long-term sustainability by accumulation of harmful substances. A holistic approach should consider the options and consequences of recycling of the major potential societal P sources. Furthermore, it should compare the effects of different potential P recycling sources on long-term soil fertility and soil contamination. A re-consideration of the appropriate P recycling for organic farming should also consider the products obtained from more advanced treatment technologies of societal organic wastes. For example, in the last two decades several new approaches were developed to treat urban organic household wastes (e.g. by anaerobic digestion instead of composting) or urban wastewaters. The two current main approaches of urban wastewaters treatment are (Herzel et al. 2015b; Egle et al. 2016; Kraus et al. 2017):

- Chemical approaches resulting in the production of struvite (magnesium ammonium phosphate) or different forms of water- and non water-soluble calcium phosphates.
- Thermal approaches aiming to recover P as ashes (with or without PTE depollution), chars, P-rich

converter slags (CaSiPO<sub>4</sub>) or calcined phosphates like ashes (CaNaPO<sub>4</sub>) among others.

Some methods represent a combination of thermal and chemical approaches (e.g. Leachphos<sup>®</sup>, where ashes are solubilized with acids). The aim of these treatments is to reduce the load of contaminants, either by selective P removal (chemical P recovery approaches), or by decontamination of organic contaminants due to the incineration process itself (thermal P recovery approaches). Several methods also reduce the specific PTE loads, e.g. by selective P precipitation, or by volatilization as chlorines at higher incineration temperatures (Kraus and Seis 2015).

The objective of this study was to assess the input and to estimate the long-term accumulation risk in European agricultural soils of PTEs and selected POPs related to the annual application of different societal P recycling sources and to assess how decontamination by thermal, chemical or chemico-physical approaches influences the soil accumulation risk of the addressed pollutants, in order to test following hypotheses:

1. The PTE and POP concentrations in fertilizers are the main factor for their accumulation in soil.
2. The source material and the production process of RPFs determine their contamination and the risk of PTE and POP accumulation in soil.
3. The regular use of recycled phosphorus fertilizers causes higher soil pollution risks (PTEs and POPs) than that of commonly used fertilizers.
4. The soil and climatic conditions have a minor relevance on the soil accumulation risk of PTEs.

## Materials and methods

### Recycled P fertilizers

The assessed RPFs derived from municipal organic waste, urban wastewater and meat production were compared to composts, cattle manure from conventional agriculture, phosphate rock and triple superphosphate as references. Information regarding the raw material of fertilizers and the manufacturing process is provided in Table 1. Central European average values for P and PTEs in all fertilizers are summarized in Table 2. The available fertilizers were

**Table 1** Raw material and manufacturing process of fertilizers

Fertilizers	Main raw material	Treatment
Organic household waste digestate OF	Organic household waste	Anaerobic digestion
Catering waste digestate	Catering waste	Anaerobic digestion
Biosolid dewatered	Sewage sludge	Hygienization, stabilization, dewatering
Meat and bone meal OF	Slaughterhouse waste	Sterilization, drying, grinding
Struvite Stuttgart	Urban wastewater	Sludge leaching
Struvite Airprex <sup>®</sup>	Urban wastewater	Sludge precipitation
Biosolid ash untreated	Sewage sludge	Incineration
Biosolid ash Leachphos <sup>®</sup>	Sewage sludge ash	Ash solubilization with acids
Calcined ash AshDec <sup>®</sup>	Sewage sludge ash	Calcination with Na or K
Mephrec converter slag	Sewage sludge/ash	Metallurgical treatment
<i>Reference fertilizers</i>		
Green waste compost OF <sup>a</sup>	Plant residues of gardens	Composting
Organic household waste compost OF	Organic household waste	Composting
Catering waste compost	Food waste	Composting
Cattle solid manure OF	Manure	–
Phosphate rock OF	Phosphate rock	–
Triple superphosphate	Phosphate rock	Chemical, Ca(H <sub>2</sub> PO <sub>4</sub> ) <sub>2</sub>

<sup>a</sup>OF allowed in organic farming according to EU regulation EC 889/2008

**Table 2** Mean concentration of potentially toxic elements and phosphorus in fertilizers on a dry weight base (mg kg<sup>-1</sup> DM)

	Cd	Cr	Ni	Pb	Cu	Zn	P	References
Organic household waste digestate OF	0.4	19	13	20	49	200	6800	1
Catering waste digestate	0.3	8	9	4	40	177	22,100	1
Biosolid dewatered	1.0	37	29	37	327	937	35,700	3
Meat and bone meal OF	0.2	14	3	3	11	109	53,100	1
Struvite Stuttgart	0.4	14	5	6	19	260	169,630	1, 4, 5
Struvite Airprex <sup>®</sup>	0.3	16	16	60	42	86	91,686	1, 6, 7
Biosolid ash untreated	2.8	96	37	129	875	2479	70,900	7, 5
Biosolid ash Leachphos <sup>®</sup>	2.6	34	14	25	851	1390	131,000	5, 8
Calcined ash AshDec <sup>®</sup>	0.3	127	56	57	441	1710	77,000	5, 8
Mephrec converter slags	0.3	89	14	13	107	69	51,976	8, 9, 10
Green waste compost OF	0.4	23	14	32	34	154	2200	1
Organic household waste compost OF	0.4	22	12	28	41	150	3100	1
Catering waste compost	0.5	28	18	45	54	213	3600	1
Cattle solid manure OF	0.8	28	19	9	32	107	6313	2
Phosphate rock OF	19	152	28	12	17	221	124,431	11
Triple superphosphate	62	229	21	15	15	354	198,653	12

1 Möller and Schultheiß (2014), 2 AROMIS (2016), 3 Wollmann and Möller (2015), 4 Pinnekamp et al. (2011), 5 Steinmetz et al. (2014), 6 Kraus and Seis (2015), 7 Langeveld (2014), 8 Herzel et al. (2015a), 9 Scheidig (2009), 10 Scheidig et al. (2014), 11 Kratz and Schnug (2005), 12 Dittrich and Klose (2008). For abbreviations see Table 1

considered on a high and a low POP contamination level (Table 3).

The following fertilizers are currently allowed for use in organic farming according to EU regulation EC (2008): Green waste composts, organic household waste compost, digestate, meat and bone meal, conventional animal manures and phosphate rock. These fertilizers are indicated in the following text with the additional specification “OF”.

The calculations of pollutant inputs and accumulation risks were performed for a time span of 200 years of fertilizer application equivalent to 11 kg P ha<sup>-1</sup> year<sup>-1</sup> (25.2 kg P<sub>2</sub>O<sub>5</sub> ha<sup>-1</sup> year<sup>-1</sup>), which is an average P export via crops on European organic arable stockless farms (Lindenthal 2000; Berry et al. 2003; Möller and Stinner 2010; Cooper et al. 2017).

Model description for the risk assessment of potentially toxic element accumulation in soils

The included PTE input routes to soil were atmospheric deposition, liming and fertilization. Input of PTEs and their accumulation potential were compared to the current soil pollution (soil background concentration) and to proposed soil threshold values.

This study included the elements regulated in the sludge directive (EU Directive 86/278/EEC 1986) except mercury, because it is volatile (Steinnes 2013a) and is not covered completely by the model. The assessment of the PTE accumulation risks in European agricultural top soils was based on a mass balance approach applied by Smolders (2013), Six and Smolders (2014) and the European Scientific

Committee on Toxicology, Ecotoxicology and the Environment, CSTE (2002) for assessing the cadmium accumulation in European agricultural soils. The model considered the input of pollutants by fertilizer application, atmospheric deposition and liming and the output via leaching, plant uptake and crop removal.

$$\Delta C_{soil} = INPUT - OUTPUT \tag{1}$$

$\Delta C_{soil}$  was the change in PTE concentrations in the plough layer (25 cm) in g ha<sup>-1</sup> year<sup>-1</sup>. The concentration of PTEs in soil at year *i* ( $C_{soil\ i}$  in mg kg<sup>-1</sup>) was calculated from the annual net PTE balance (input – output in g PTEs ha<sup>-1</sup>) in year<sub>*i*-1</sub> and soil PTE concentration in year<sub>*i*-1</sub> as:

$$C_{soil\ i} = C_{soil\ i-1} + \frac{INPUT - OUTPUT}{M_{soil}} \tag{2}$$

For  $C_{soil\ i-1}$  = PTE concentration in soil in the previous year (mg kg<sup>-1</sup>), Input = the annual sum of PTEs in fertilizer, atmospheric deposition and lime (g ha<sup>-1</sup>), Output = the annual sum of PTEs in leaching losses and harvested crop parts (g ha<sup>-1</sup>),  $M_{soil}$  = soil mass in the plough layer (Mg ha<sup>-1</sup>), The calculation refers to the soil mass  $M_{soil}$  in the plough layer with a bulk density of 1.3 kg dm<sup>3</sup> and a depth of 25 cm, which are 3250 Mg ha<sup>-1</sup>. The PTE content in harvested crop parts in year *i* (Crop export<sub>*i*</sub>) was calculated as a function of PTE concentration in soil:

$$Crop\ export_i = TF * C_{soil\ i-1} * crop\ yield \tag{3}$$

**Table 3** Organic pollutant and phosphorus concentration in fertilizer dry weight for two contamination levels

Contamination level	PCB (mg kg <sup>-1</sup> )		PAH (mg kg <sup>-1</sup> )		PCDD/F (ng TEQ kg <sup>-1</sup> )		References
	Low	High	Low	High	Low	High	
Green waste compost OF	0.01	0.04	0.38	6.40	0.003	0.005	4, 1, 8
Organic household waste compost OF	0.01	0.09	0.38	22.0	0.004	0.005	4, 1, 8
Biosolid	0.02	0.18	18.0	46.0	0.018	0.180	6, 3, 11
Organic household waste digestate OF	0.02	0.03	0.12	1.60	0.003	0.015	7, 1, 8
Meat and bone meal	n.a.	n.a.	n.a.	n.a.	0.0001	0.001	1, 12
Solid farmyard manure	0.01	0.04	0.01	0.14	0.004	0.020	5, 2, 9, 10, 11

For abbreviations: see Tables 1 and 4. n.a. not available; 1 Möller and Schultheiß (2014); 2 AROMIS (2016); 3 Wollmann and Möller (2015); 4 Brändli et al. 2007a; 5 Herter et al. (2003); 6 Gawlik and Bidoglio (2006); 7 Lindenblatt et al. (2007); 8 Beníšek et al. (2015); 9 Rey-Salgueiro et al. (2008); 10 Hough et al. (2012); 11 Stevens and Jones (2003); 12 FAO (2004)

**Table 4** Transfer factors (TF) of wheat grain and the partitioning coefficients (KD) used in the model

	TF	References TF	
Cd	0.15	Puschenreiter and Horak (2000)	
Cr	0.04	Lavado et al. (2007)	
Cu	0.30	Puschenreiter and Horak (2000)	
Ni	0.07	Lavado et al. (2007)	
Pb	0.08	Lavado et al. (2007)	
Zn	0.36	Puschenreiter and Horak (2000)	
	KD in soil pH 5	KD in soil pH 7	References KD
Cd	59	818	EPA (1999, Appendix C)
Cr(III)	1048	28,649	Hassan and Garrison (1996)
Cu	900	2000	Sauvé et al. (2000)
Ni	90	2400	Sauvé et al. (2000)
Pb	5532	21,750	EPA (1999, Appendix F)
Zn	150	3000	Sauvé et al. (2000)

Units: TF is total PTE concentration in plant divided by the total concentration in soil ( $\text{mg kg}^{-1}$ ); KD is in ( $\text{L kg}^{-1}$ )

Crop export<sub>i</sub> = annual PTE uptake into wheat grain in year *i* ( $\text{g ha}^{-1}$ ), TF = transfer factor for wheat grain, Crop yield = average annual yield of wheat grain  $3 \text{ Mg ha}^{-1}$ .

In order to assess the PTE removal via exported products, the transfer factor for wheat grain (Puschenreiter and Horak 2000; Lavado et al. 2007) and a continuous cropping of wheat with an average annual yield of  $3 \text{ Mg ha}^{-1}$  was assumed. This is a low average wheat yield in organic production (Beckmann et al. 2001) taking into account that ley cropping without nutrient removal are typically part of the crop rotation on organic farms. The average annual yield of  $3 \text{ Mg ha}^{-1}$  equals a P export from the field of  $11 \text{ kg P ha}^{-1} \text{ year}^{-1}$  (Stein-Bachinger et al. 2004) and also corresponds to the above-mentioned mean P export in harvested crop parts in stockless organic farms. The transfer factor (TF) is the PTE concentration of an element in the crop divided by the PTE concentration in the soil. The TF values are shown in Table 4. Transfer factors for wheat grain were used because it is an important commodity and carbohydrate source in Europe.

Leaching is the outflow of PTEs dissolved in soil pore water from the topsoil. It is a function of water percolation through the soil—expressed as precipitation excess *F* in  $\text{m year}^{-1}$ —and the PTE concentration in pore water. To predict the PTE concentration in pore water, a distribution coefficient (KD in  $\text{L kg}^{-1}$ ) was used. KD is a measure for the relative partitioning of an element between the solid phase ( $C_{\text{soil}}$ ) and the solution phase ( $C_{\text{solution}}$ ):

$$\text{Leaching (g ha}^{-1} \text{ year}^{-1}) = C_{\text{soil } i-1} / \text{KD} \\ * \text{ precipitation excess } F \\ * 10,000 \quad (4)$$

$C_{\text{soil } i-1}$  = Compound concentration in soil ( $\text{mg kg}^{-1}$ ), KD = distribution coefficient ( $(\text{mg kg}^{-1}) / (\text{mg L}^{-1}) = \text{L kg}^{-1} = \text{dm}^3 \text{ kg}^{-1}$ ), *F* = precipitation excess ( $\text{m year}^{-1}$ ), Leaching ( $\text{mg kg}^{-1} / (\text{dm}^3 \text{ kg}^{-1}) * \text{m year}^{-1} * 10,000 = \text{mg m}^{-2} \text{ year}^{-1} * 10 = \text{g ha}^{-1} \text{ year}^{-1}$ ).

The KD is the PTE concentration in the solid phase divided by the PTE concentration in the solution phase (Table 4). It varies widely according to pH, clay content, organic carbon, iron content, PTE concentration in soil and—if predicted and not measured—on the empirical regression model. Among these factors, the pH value had the largest effect (Young 2013; Kabata-Pendias 2011). Therefore, the mean KD values from literature for pH 5 and for pH 7 were used in this calculation (EPA 1999; Sauvé et al. 2000; Hassan and Garrison 1996). To calculate the water percolation in the soil, a high rainfall scenario ( $F = 0.3 \text{ m year}^{-1}$ ) and a low rainfall scenario ( $F = 0.1 \text{ m year}^{-1}$ ) were calculated. The European average precipitation excess is  $0.2 \text{ m year}^{-1}$ , in Mediterranean region it can be reduced to  $0.05 \text{ m year}^{-1}$  and for regions with high rainfall a precipitation excess of  $0.3 \text{ m year}^{-1}$  is possible (Smolders 2013; Six and Smolders 2014).

The calculations were based on the soil background concentrations of PTEs already present in the soil. These values for European soils were taken from an European survey (Salminen et al. 2005). The PTE threshold values were taken from a recommendation paper by the Joint Research Centre, the European Commission'' in-house science service (Gawlik and Bidoglio 2006) because no legal European thresholds exist (Table 5).

**Table 5** Input of potential toxic elements to soil via atmospheric deposition, lime and fertilizers (referring to 11 kg P ha<sup>-1</sup> year<sup>-1</sup>) after 200 years compared to soil background concentration and threshold values

Soil background concentrations and proposed threshold values (aqua regia basis) were converted from mg kg<sup>-1</sup> to kg ha<sup>-1</sup> assuming a soil bulk density in the plough horizon (25 cm) of 1.3 g cm<sup>-3</sup>. 1 Salminen et al. (2005); 2 Gawlik and Bidoglio (2006), 3 Smolders (2013), 4 Eriksen et al. (2009). For abbreviations and fertilizer references see Tables 1 and 2

PTE input to soil (kg ha <sup>-1</sup> )	Cd	Cr	Cu	Ni	Pb	Zn	References
Soil background concentration	0.92	106	53.3	100	78	198	1
Proposed threshold values	3.25	244	163	163	228	488	2
Input via atmospheric deposition	0.07	1.85	6.77	1.99	2.38	45.2	3, 4
Input via lime	0.03	0.18	0.27	0.15	0.19	0.40	2
<i>Input via fertilizers</i>							
Organic household waste digestate OF	0.13	6.08	15.8	4.06	6.47	64.4	
Catering waste digestate	0.03	0.75	3.96	0.94	0.38	17.5	
Biosolid dewatered	0.06	2.25	20.1	1.80	2.25	57.5	
Meat and bone meal OF	0.01	0.56	0.47	0.14	0.12	4.49	
Struvite Stuttgart	0.01	0.18	0.25	0.06	0.08	3.36	
Struvite Airprex <sup>®</sup>	0.01	0.39	1.01	0.38	1.44	2.05	
Biosolid ash untreated	0.09	2.95	27.0	1.16	3.98	76.5	
Biosolid ash Leachphos <sup>®</sup>	0.04	0.57	14.2	0.23	0.42	23.2	
Calcined ash AshDec <sup>®</sup>	0.01	3.61	12.5	1.60	1.61	48.6	
Mephrec converter slag	0.01	3.75	4.50	0.57	0.53	2.91	
Green waste compost OF	0.40	22.7	34.2	13.7	32.2	153	
Organic household compost OF	0.25	15.2	28.6	8.61	19.7	106	
Catering waste compost	0.28	17.1	33.0	10.6	27.2	130	
Cattle solid manure OF	0.28	9.57	11.1	6.65	3.15	37.1	
Phosphate rock OF	0.34	2.68	0.30	0.50	0.21	3.89	
Triple superphosphate	0.68	2.52	0.17	0.23	0.16	3.90	

Model description for accumulation of persistent organic pollutants in soil

The three groups PAH, PCB and PCDD/F were considered as an example for POPs, because they are still important pollutants in soil and the database for calculations is available. A mass balance model according to Amlinger et al. (2004) was used to assess the accumulation of persistent organic pollutants in soil. Accumulation problems of organics arise, if the persistence of compounds in soil is high (EU 2016). The accumulation was computed with the following iterative formula:

$$C_{soil} \text{ (mg kg}^{-1}\text{)} = C_0 * X^n + ((If + Id)/M_{soil}) * ((1 - X^n)/(1 - X))$$

(Amlinger et al. 2004)

(5)

C<sub>0</sub> = Soil background concentration mg kg<sup>-1</sup>, X = e<sup>-ln2/(T1/2)</sup>, T<sub>1/2</sub> = Half-life time of compound in soil in years, n = year of which accumulation is considered, If = Compound input with fertilizer (g ha<sup>-1</sup> year<sup>-1</sup>), Id = Compound input with

atmospheric deposition (g ha<sup>-1</sup> year<sup>-1</sup>), M<sub>soil</sub> = Soil mass plough horizon (Mg ha<sup>-1</sup>).

The most critical parameter was the assumed half-life time which is usually determined in lab experiments (OECD 2005), and the bibliographical references vary widely (Table 6). In the calculation, mean values of literature data were used. Also the range of PCB, PAH and PCDD/F content in fertilizers was broad, therefore scenarios with fertilizers of low and high contamination level were calculated as shown in Table 3. Soil guidance values (according to the Austrian threshold values ÖNORM S2088-2 2004) and soil background concentrations for organic pollutants are shown in Table 6. Leaching and plant uptake of POPs were not considered. The assessed POPs—as apolar molecules—are highly bound to the organic matter, reducing their availability to plant roots (Smith 2009b) and leaching into groundwater is unlikely (Leschber 2006; Gawlik and Bidoglio 2006; Clarke and Cummins 2015; Tavazzi et al. 2012).

**Table 6** Persistent organic pollutant input to soil in 200 years via fertilizers (referring to 11 kg P ha<sup>-1</sup> year<sup>-1</sup>) and atmospheric deposition, compared to half-life time, soil background concentration and guidance values

	Unit		References
<i>PCB</i>			
Digestate	kg ha <sup>-1</sup>	0.003	For fertilizer references see Table 3
Sewage sludge	kg ha <sup>-1</sup>	0.011	
Farmyard manure	kg ha <sup>-1</sup>	0.015	
Green waste compost	kg ha <sup>-1</sup>	0.043	
Organic waste compost	kg ha <sup>-1</sup>	0.054	
Atmospheric deposition	kg ha <sup>-1</sup>	0.070	Suominen et al. (2014), Amlinger et al. (2004), Brändli et al. (2007b) and Arellano et al. 2015
Half-life time in soil	years (mean)	14	Leschber (2006), Amlinger et al. (2004), Creaser et al. 2007 and Bruckmann et al. (2013)
	years (max.)	31	
	years (min.)	1	
Initial soil background concentration	kg ha <sup>-1</sup>	0.045	Amlinger et al. (2004) and Creaser et al. (2007)
Soil guidance value	kg ha <sup>-1</sup>	0.975	ÖNORM S 2088-2 (2004)
Digestate	kg ha <sup>-1</sup>	0.16	For fertilizer references see Table 3
Sewage sludge	kg ha <sup>-1</sup>	2.83	
Farmyard manure	kg ha <sup>-1</sup>	0.05	
Green waste compost	kg ha <sup>-1</sup>	6.40	
Organic waste compost	kg ha <sup>-1</sup>	13.4	
Atmospheric deposition	kg ha <sup>-1</sup>	0.53	Amlinger et al. (2004), Suominen et al. (2014) and Brändli et al. (2007b)
Half-life time in soil	years (mean)	12	Leschber (2006), Amlinger et al. (2004), Creaser et al. (2007) and Bruckmann et al. (2013)
	years (max.)	32	
	years (min.)	5	
Initial soil background concentration	kg ha <sup>-1</sup>	1.38	Amlinger et al. (2004) and Brändli et al. (2007b)
Soil guidance value	kg ha <sup>-1</sup>	3.25	ÖNORM S 2088-2 (2004)
<i>PCDD/F</i>			
Digestate	mg TEQ ha <sup>-1</sup>	1.49	For fertilizer references see Table 3
Sewage sludge	mg TEQ ha <sup>-1</sup>	11.1	
Farmyard manure	mg TEQ ha <sup>-1</sup>	6.97	
Meat and bone meal	mg TEQ ha <sup>-1</sup>	0.02	
Green waste compost	mg TEQ ha <sup>-1</sup>	5.20	
Organic waste compost	mg TEQ ha <sup>-1</sup>	3.30	
Atmospheric deposition	mg TEQ ha <sup>-1</sup>	0.38	Amlinger et al. (2004), Suominen et al. (2014) and Brändli et al. (2007b)
Half-life time in soil	years (mean)	30	Leschber (2006), Amlinger et al. (2004), Umlauf et al. (2010) and Bruckmann et al. (2013)
	years (max.)	100	
	years (min.)	5	
Initial soil background concentration	mg TEQ ha <sup>-1</sup>	18.6	Amlinger et al. (2004) and Brändli et al. (2007b)
Soil guidance value	mg TEQ ha <sup>-1</sup>	32,500	ÖNORM S 2088-2 (2004)

Polychlorinated biphenyls (PCB), polycyclic aromatic hydrocarbons (PAH) and polychlorinated dibenzo-dioxins and -furans (PCDD/F)

## System boundaries

Volatilization of PTEs was not considered by the model as well as other interactions which could influence the mobility of PTEs and the degradation of POPs, like fixation of PTEs through P added by fertilizers or site-specific differences. Also other risks like metal accumulation into soil organisms, leaching to groundwater, transport with surface water or wind, direct uptake of soil by humans and animals have not been considered here.

## Results

### Input routes of pollutants to soil

The assessment of the 200-years PTE inputs by fertilizers, liming and atmospheric deposition indicated that these inputs are expected to be small compared to the already existing PTE background concentrations in soil (Table 5). Between 28% (Cd) and 61% (Ni) of the proposed thresholds were already filled with the existing soil background concentration. Also the assessed sum of all inputs over 200 years was little compared to soil background concentration and would not reach the proposed threshold values. Liming contributed less to the pollution than most fertilizers. The expected atmospheric deposition of pollutants was considerable, however, most fertilizers would have a higher input at least for some elements. With input rates below the atmospheric deposition, meat and bone meal, struvite, digestate of catering waste and treated ashes turned out to be the cleanest fertilizers in relation to their P content. The variability among the fertilizers can be compared on the basis of the ratio of PTE input by fertilizers and atmospheric input (Fig. 1). PTEs in composts and cattle manure, and some elements in phosphate rock, triple superphosphate and untreated biosolid ash exceeded the atmospheric input by far. Fertilizers with a high PTE-to-phosphorus ratio, like composts, showed the highest PTE input to soil.

POPs entered the agricultural soil via fertilization and atmospheric deposition more or less to the same extent. The fertilizer input and atmospheric deposition of POPs in 200 years was compared to the initial soil background concentration, guidance values and the half-life time (Table 6). Soil background

concentration was low compared to the guidance values, it reached 5% for PCB, 43% for PAH and 0.06% for PDCC/F. For PCB, atmospheric deposition was the main route compared to fertilizer input. Main PHA input was caused by organic waste compost, green waste compost and sewage sludge, digestate and farmyard manure were below the atmospheric deposition. For PCDD/F the soil guidance value was far higher than any input. Expected PCDD/F input via atmospheric deposition was considerably lower than via fertilizers, except for meat and bone meal. PCDD/F contamination level by fertilizer application declined as following in descending order: sewage sludge, farmyard manure, green waste compost, organic waste compost and digestate (Table 6).

The PTE and POP inputs provided information about the pollution loads in fertilizers and input routes. Additional information was gained from calculating the soil accumulation risk with the mass balance models which included the PTE output by leaching and harvested crop parts and the POP decomposition in soil.

### Accumulation risk of pollutants in soil estimated by the mass balance models

The model estimates for PTE accumulation risks after 200 years of fertilizer application (equivalent to  $11 \text{ kg P ha}^{-1}$ ) indicated that with none of the assessed fertilizers the proposed soil threshold values would be reached (Fig. 2). However, the results revealed strong differences in the accumulation risk for the individual PTEs. An overall low Cd accumulation risk in soil, in relation to the threshold values, was predicted for all RPFs except for triple superphosphate, a moderate accumulation risk for Cu, Cr and Pb, and the highest accumulation risk for Ni and Zn (Fig. 2; Table 8). Soil pH and precipitation excess, affecting the element output from the soil via leaching and harvested crop parts, is expected to have a stronger effect on Cd, Ni and Zn accumulation than the type of fertilizer. For Cr, Cu and Pb, it was the opposite: the PTE amount in fertilizers had a stronger effect on their accumulation than the output losses, varying with soil pH and precipitation excess (Fig. 2).

For POPs, a mass balance model considering the soil background concentration, the contamination level of the fertilisers, the atmospheric deposition, and their half-life time was used. The mass balance

**Table 7** Total output (kg ha<sup>-1</sup>) of PTEs after 200 years of fertilization (equivalent to 11 kg P ha<sup>-1</sup>) by leaching and in harvested crop parts

(a)

	Cd pH 5 F 0.1	Cd pH 5 F 0.3	Cd pH 7 F 0.1	Cd pH 7 F 0.3	Cr pH 5 F 0.1	Cr pH 5 F 0.3	Cr pH 7 F 0.1	Cr pH 7 F 0.3	Cu pH 5 F 0.1	Cu pH 5 F 0.3	Cu pH 7 F 0.1	Cu pH 7 F 0.3
No fertilizer	0.64	0.95	0.09	0.22	7.03	18.03	1.03	1.03	6.34	13.34	4.34	7.34
Organic household waste digestate OF	0.69	1.04	0.05	0.23	7.11	18.11	1.11	1.11	7.14	15.14	5.14	9.14
Catering waste digestate	0.65	0.97	0.09	0.22	6.78	17.78	0.78	1.78	7.30	13.30	5.30	8.30
Biosolid dewatered	0.66	0.99	0.10	0.22	7.28	18.28	1.28	1.28	7.44	15.44	5.44	9.44
Meat and bone meal OF	0.65	0.96	0.09	0.22	6.59	17.59	0.59	1.59	6.81	12.81	4.81	7.81
Struvite Stuttgart	0.65	0.96	0.10	0.22	7.21	18.21	1.21	1.21	6.59	12.59	4.59	7.59
Struvite Airprex <sup>®</sup>	0.65	0.96	0.10	0.22	6.42	18.42	1.42	1.42	6.35	13.35	4.35	7.35
Biosolid ash untreated	0.68	1.01	0.11	0.23	6.98	17.98	0.98	1.98	8.34	16.34	5.34	9.34
Biosolid ash Leachphos <sup>®</sup>	0.65	0.98	0.09	0.22	6.60	17.60	0.60	1.60	7.54	14.54	5.54	8.54
Calcined ash AshDec <sup>®</sup>	0.65	0.96	0.09	0.22	6.64	18.64	0.64	1.64	6.84	14.84	4.84	8.84
Mephrec converter slags	0.64	0.96	0.09	0.22	6.78	18.78	0.78	1.78	6.84	13.84	4.84	7.84
Green waste compost OF	0.79	1.23	0.12	0.22	7.73	19.73	0.73	1.73	8.54	16.54	5.54	9.54
Organic household waste compost OF	0.74	1.12	0.07	0.27	7.23	19.23	1.23	1.23	7.94	15.94	5.94	9.94
Catering waste compost	0.75	1.14	0.10	0.20	7.13	19.13	1.13	1.13	8.34	17.34	6.34	10.34
Cattle solid manure OF	0.75	1.14	0.10	0.20	7.60	18.60	0.60	1.60	7.44	14.44	5.44	8.44
Phosphate rock OF	0.77	1.19	0.06	0.26	6.71	17.71	0.71	1.71	6.64	12.64	4.64	7.64
Triple Superphosphate	0.90	1.42	0.10	0.30	6.55	18.55	0.55	1.55	6.51	13.51	4.51	7.51

(b)

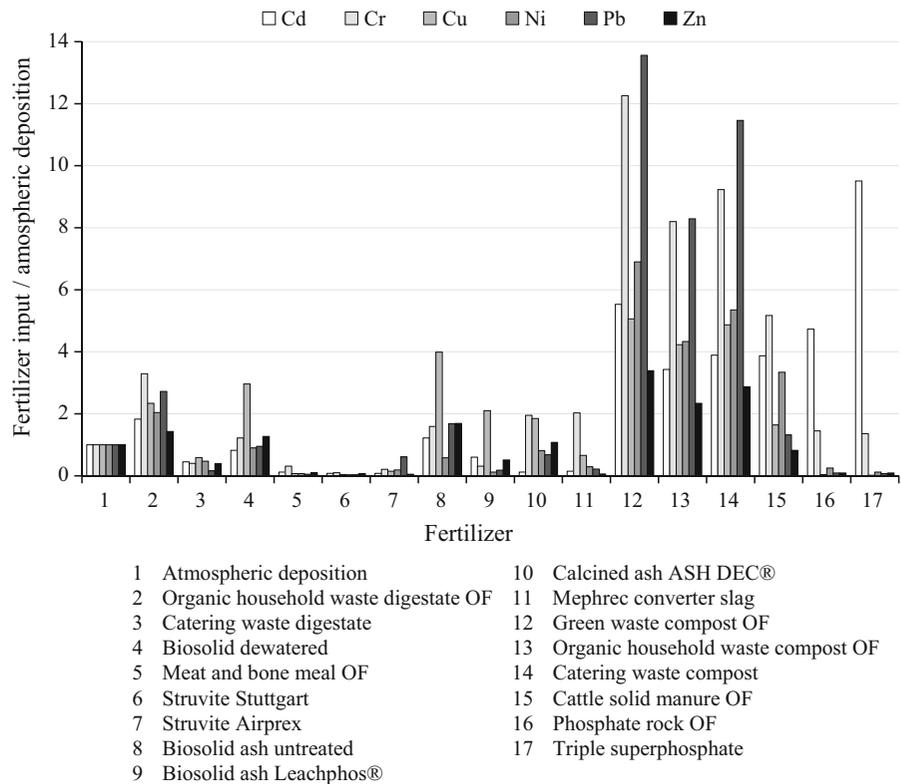
	Ni pH 5 F 0.1	Ni pH 5 F 0.3	Ni pH 7 F 0.1	Ni pH 7 F 0.3	Pb pH 5 F 0.1	Pb pH 5 F 0.3	Pb pH 7 F 0.1	Pb pH 7 F 0.3	Zn pH 5 F 0.1	Zn pH 5 F 0.3	Zn pH 7 F 0.1	Zn pH 7 F 0.3
No fertilizer	51.1	89.1	4.14	9.14	2.57	3.57	1.57	2.57	84.6	164	18.6	26.6
Organic household waste digestate OF	52.2	91.2	4.20	9.20	2.04	4.04	2.04	2.04	97.0	192	21.0	30.0
Catering waste digestate	51.1	89.1	4.08	9.08	1.95	3.95	1.95	1.95	88.1	172	19.1	27.1
Biosolid dewatered	50.9	89.9	3.94	8.94	2.82	3.82	1.82	1.82	96.1	189	20.1	30.1
Meat and bone meal OF	51.3	89.3	4.28	9.28	2.69	3.69	1.69	1.69	85.1	166	18.1	27.1
Struvite Stuttgart	51.2	89.2	4.20	9.20	2.65	3.65	1.65	1.65	85.0	166	18.0	27.0
Struvite Airprex <sup>®</sup>	51.5	88.5	4.52	8.52	2.01	4.01	2.01	2.01	84.7	165	18.7	26.7
Biosolid ash untreated	51.3	89.3	4.30	9.30	2.55	4.55	1.55	2.55	99.1	197	21.1	31.1
Biosolid ash Leachphos <sup>®</sup>	51.4	88.4	4.37	9.37	1.99	3.99	1.99	1.99	88.8	174	18.8	27.8
Calcined ash AshDec <sup>®</sup>	51.7	89.7	3.74	8.74	2.18	4.18	2.18	2.18	94.2	185	20.2	29.2
Mephrec converter slags	50.7	88.7	3.71	8.71	2.10	4.10	2.10	2.10	84.5	166	18.5	26.5
Green waste compost OF	54.8	96.8	3.84	9.84	2.77	4.77	1.77	2.77	115	231	23.6	34.6
Organic household waste compost OF	53.8	93.8	3.75	8.75	2.27	4.27	1.27	2.27	106	211	22.6	32.6
Catering waste compost	53.7	94.7	3.74	9.74	2.77	4.77	1.77	2.77	111	221	23.6	34.6
Cattle solid manure OF	52.8	92.8	3.79	8.79	2.72	3.72	1.72	1.72	91.7	181	19.7	28.7
Phosphate rock OF	50.6	88.6	3.64	8.64	2.78	3.78	1.78	1.78	85.5	165	18.5	26.5
Triple Superphosphate	51.4	88.4	4.37	9.37	2.73	3.73	1.73	1.73	85.5	166	18.5	26.5

**Table 8** Modelled soil accumulation of potentially toxic elements (kg ha<sup>-1</sup>) after 200 years of fertilizer application equivalent to 11 kg phosphorus per ha and year at four levels of pH and precipitation excess F values

	Cd pH 5 F 0.1	Cd pH 5 F 0.3	Cd pH 7 F 0.1	Cd pH 7 F 0.3	Cr pH 5 F 0.1	Cr pH 5 F 0.3	Cr pH 7 F 0.1	Cr pH 7 F 0.3	Cu pH 5 F 0.1	Cu pH 5 F 0.3	Cu pH 7 F 0.1	Cu pH 7 F 0.3
No fertilizer	0.38	0.07	0.93	0.80	101	90	107	107	54	47	56	53
Organic household waste digestate OF	0.46	0.11	1.1	0.92	107	96	113	113	69	61	71	67
Catering waste digestate	0.40	0.08	0.96	0.83	102	91	108	107	57	51	59	56
Biosolid dewatered	0.42	0.09	0.98	0.86	103	92	109	109	73	65	75	71
Meat and bone meal OF	0.38	0.07	0.94	0.81	102	91	108	107	54	48	56	53
Struvite Stuttgart	0.38	0.07	0.93	0.81	101	90	107	107	54	48	56	53
Struvite Airprex®	0.38	0.07	0.93	0.81	102	90	107	107	55	48	57	54
Biosolid ash untreated	0.43	0.10	1.0	0.88	104	93	110	109	79	71	82	78
Biosolid ash Leachphos®	0.41	0.08	0.97	0.84	102	91	108	107	67	60	69	66
Calcined ash AshDec®	0.38	0.07	0.94	0.81	105	93	111	110	66	58	68	64
Mephrec converter slags	0.39	0.07	0.94	0.81	105	93	111	110	58	51	60	57
Green waste compost OF	0.63	0.19	1.3	1.2	<b>123</b>	<b>111</b>	<b>130</b>	<b>129</b>	<b>86</b>	<b>78</b>	<b>89</b>	<b>85</b>
Organic household waste compost OF	0.53	0.15	1.2	1.0	116	104	122	122	81	73	83	79
Catering waste compost	0.55	0.16	1.2	1.1	118	106	124	124	85	76	87	83
Cattle solid manure OF	0.55	0.16	1.2	1.1	110	99	117	116	64	57	66	63
Phosphate rock OF	0.59	0.17	1.3	1.1	104	93	110	109	54	48	56	53
Triple superphosphate	<b>0.80</b>	<b>0.28</b>	<b>1.6</b>	<b>1.4</b>	104	92	110	109	54	47	56	53
Soil background value <sup>a</sup>	0.92	0.92	0.92	0.92	106	106	106	106	53	53	53	53
Threshold <sup>a</sup>	3.25	3.25	3.25	3.25	244	244	244	244	163	163	163	163
	Ni pH 5 F 0.1	Ni pH 5 F 0.3	Ni pH 7 F 0.1	Ni pH 7 F 0.3	Pb pH 5 F 0.1	Pb pH 5 F 0.3	Pb pH 7 F 0.1	Pb pH 7 F 0.3	Zn pH 5 F 0.1	Zn pH 5 F 0.3	Zn pH 7 F 0.1	Zn pH 7 F 0.3
No fertilizer	51	13	98	93	78	77	79	78	159	80	225	217
Organic household waste digestate OF	54	15	102	97	85	83	85	85	211	116	287	278
Catering waste digestate	52	14	99	94	79	77	79	79	173	89	242	234
Biosolid dewatered	53	14	100	95	80	79	81	81	205	112	281	271
Meat and bone meal OF	51	13	98	93	78	77	79	79	163	82	230	221
Struvite Stuttgart	51	13	98	93	78	77	79	79	162	81	229	220
Struvite Airprex®	51	14	98	94	80	78	80	80	161	81	227	219
Biosolid ash untreated	52	14	99	94	82	80	83	82	221	123	299	289
Biosolid ash Leachphos®	51	14	98	93	79	77	79	79	178	93	248	239
Calcined ash AshDec®	52	14	100	95	80	78	80	80	198	107	272	263
Mephrec converter slags	52	14	99	94	79	77	79	79	162	81	228	220
Green waste compost OF	<b>61</b>	<b>19</b>	<b>112</b>	<b>106</b>	<b>110</b>	<b>108</b>	<b>111</b>	<b>110</b>	<b>282</b>	<b>166</b>	<b>373</b>	<b>362</b>
Organic household waste compost OF	57	17	107	102	98	96	99	98	244	139	327	317
Catering waste compost	59	18	109	103	105	103	106	105	263	153	350	339
Cattle solid manure OF	56	16	105	100	81	80	82	82	189	100	261	252
Phosphate rock OF	52	14	99	94	78	77	79	79	162	82	229	221
Triple superphosphate	51	14	98	93	78	77	79	79	162	82	229	221
Soil background value <sup>a</sup>	100	100	100	100	78	78	78	78	198	198	198	198
Threshold <sup>a</sup>	163	163	163	163	228	228	228	228	488	488	488	488

<sup>a</sup>Soil background values. Thresholds: see Table 3. Italics Minimum values. Bold Maximum values. Abbreviations: see Table 1

**Fig. 1** Ratio between fertilizer input and atmospheric deposition of potentially toxic elements for the fertilizer application rate equivalent to  $11 \text{ kg P ha}^{-1} \text{ year}^{-1}$



model for PCB, PAH and PCDD/F showed no accumulation potentials for these substances. A decrease of the POP amounts in soil was calculated during the assessed period, as indicated by lower values after 200 years than the initial soil background concentration, which means that imported and already accumulated POPs could degrade. For PCB, the amount of fertilizer input was very similar to the atmospheric deposition (Fig. 3). The contamination level (high or low) of PCB in fertilizers did not make any difference in accumulation.

For PAH, the contamination level mattered to some extent and resulted in higher PAH accumulation for composts compared to other fertilizers and atmospheric deposition. PCDD/F accumulation was negligible compared to the guidance value and the initial soil background concentration (Fig. 3).

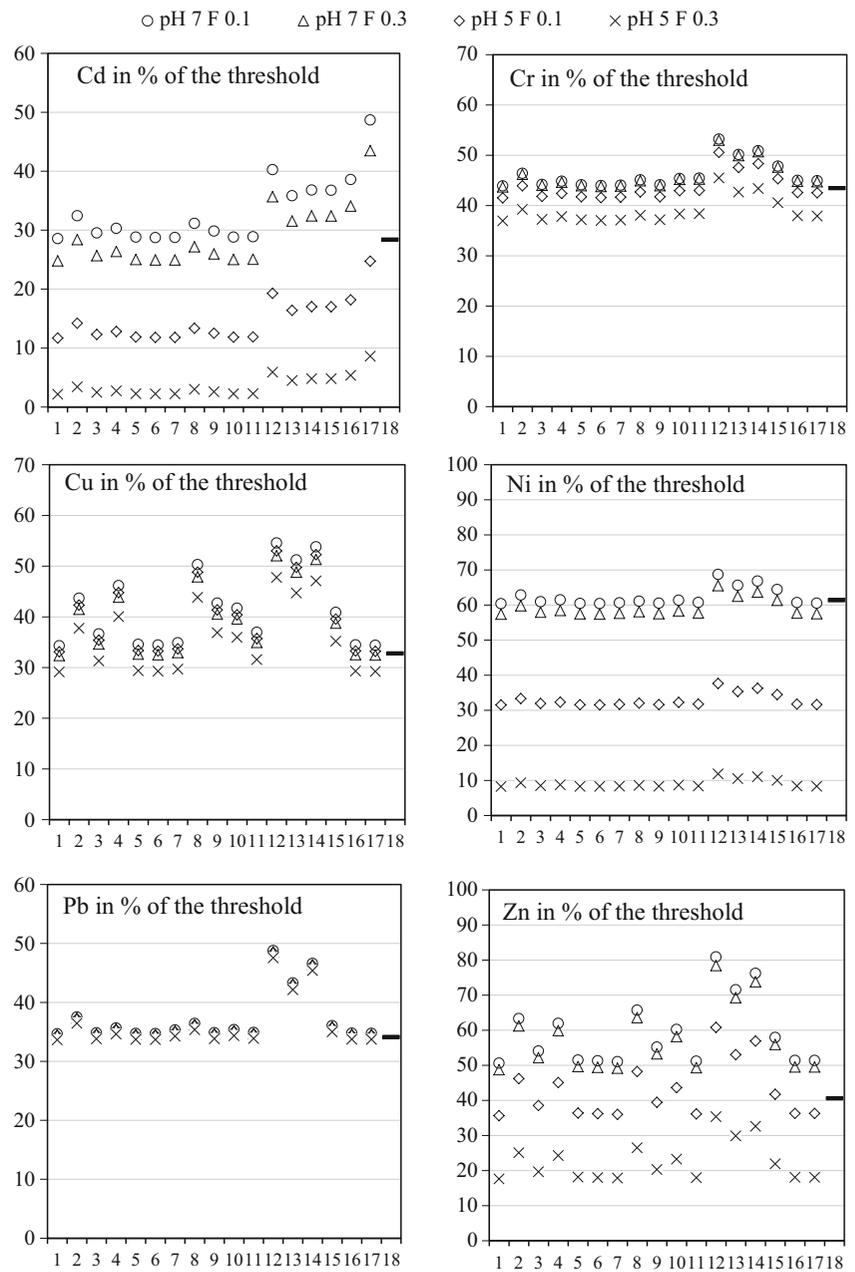
#### Relevance of soil pH and precipitation excess for soil pollution

The partitioning coefficient  $KD$  varied strongly with pH value for all regarded PTEs. Among the PTEs, the

ratio of  $KD$  at pH 7 and pH 5 was narrowest for Cu, indicating the least pH dependence (Table 4).

Over all elements, the greatest accumulation was predicted in the scenario with pH 7 and precipitation excess  $F = 0.1 \text{ m year}^{-1}$ , the lowest in the scenario with pH 5 and precipitation excess  $F = 0.3 \text{ m year}^{-1}$ . The model calculation indicated a major influence of pH value and the precipitation excess  $F$  on soil PTE accumulation risk, mainly on Cd, Ni and Zn accumulation, whereas Cr, Cu and Pb were less influenced (Fig. 2). This differentiation among the PTEs could not be clearly inferred from their  $KD$  values at varying pH value (Table 4). Comparing the scenarios, a high accumulation of PTEs was related to a low output by leaching and in harvested crop parts, and vice versa (Fig. 2; Table 7). However, for Cr, Cu, Ni, Pb and Zn the influence of precipitation excess on the accumulation risk was negligible at pH 7. The PTE accumulation was clearly higher than the output for Cr, Cu, Pb and Zn at both pH 5 and pH 7. For Cd and Ni this was the case only at pH 7, while in soil with pH 5 the reverse was observed (Tables 7, 8).

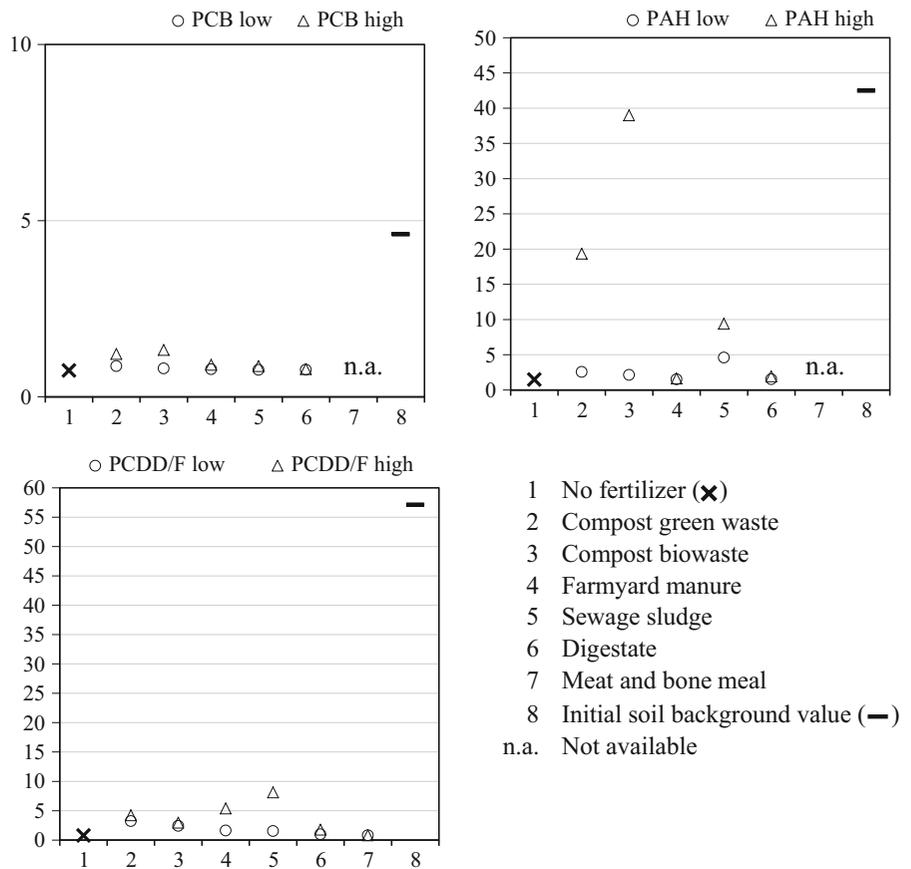
**Fig. 2** Modelled soil accumulation of potentially toxic elements (y-axis) in % of proposed threshold values after 200 years of fertilizer application equivalent to 11 kg P ha<sup>-1</sup> year<sup>-1</sup> at four levels of pH and precipitation excess F



Legend for the x-axis:

- |                                        |                                       |
|----------------------------------------|---------------------------------------|
| 1 No fertilizer                        | 10 Calcined ash ASH DEC®              |
| 2 Organic household waste digestate OF | 11 Mephrec converter slags            |
| 3 Catering waste digestate             | 12 Green waste compost OF             |
| 4 Biosolid dewatered                   | 13 Organic household waste compost OF |
| 5 Meat and bone meal OF                | 14 Catering waste compost             |
| 6 Struvite Stuttgart                   | 15 Cattle solid manure OF             |
| 7 Struvite Airprex®                    | 16 Phosphate rock OF                  |
| 8 Biosolid ash untreated               | 17 Triple Superphosphate              |
| 9 Biosolid ash Leachphos®              | 18 Initial soil background value      |

**Fig. 3** Modelled soil accumulation of PCB, PAH and PCDD/F (y-axis) in % of soil guidance values after 200 years of fertilizer application equivalent to  $11 \text{ kg P ha}^{-1} \text{ year}^{-1}$  at two contamination levels



### Relevance of PTE load of fertilizers for soil pollution

The soil accumulation risk of PTEs also varied depending on the PTE load applied with the fertilizers. More total PTE input led to a higher accumulation in soil but as well to a higher output via leaching and crop uptake. The estimated overall output of PTEs over 200 years was very similar among the fertilizers, but strongly related to soil reaction and precipitation (Table 7). For all fertilizers, a soil pH of 7 in combination with a low precipitation excess ( $F 1.0 \text{ m year}^{-1}$ ) led to the highest accumulation potential, and this resulted in the lowest PTE output (Fig. 2; Table 7).

Triple superphosphate, green waste compost and phosphate rock had the highest Cd accumulation risk and the highest Cd output (Tables 7, 8). Lowest Cd accumulation and output risk can be expected from the use of struvites, meat and bone meal and calcined ash (AshDec<sup>®</sup>). For Cr, the highest accumulation and

output risk was predicted by the model with composts, and the least accumulation and output risk with struvites, meat and bone meal, catering waste digestate and biosolid ash Leachphos<sup>®</sup>. The elements Cu, Ni, Pb and Zn likewise showed the highest accumulation risk with composts. The maximum accumulation can be expected for the use of green waste composts. Calculations showed that the lowest potential Cu accumulation and lowest output is achieved with the use of triple superphosphate, struvites and phosphate rock. Lowest Zn accumulation and output was found for struvites meat and bone meal, Mephrec converter slags phosphate rock and triple superphosphate (Fig. 2; Table 7). Lowest Pb and Ni accumulation and output was achieved with the fertilization of struvites, meat and bone meal, catering waste digestate and triple superphosphate.

Fertilizers with a high PTE-to-phosphorus ratio, like composts, showed the highest potential for PTE accumulation in soil. Except for Cd, the use of mined P sources (phosphate rock, triple superphosphate) was

related to very low PTE inputs and a negligible soil accumulation risk (Fig. 2; Table 7).

## Discussion

### Pollutant input into soil

Comparing the inputs of PTEs and POPs over 200 years gave a good overview over the routes to soil and over conventional and recycled phosphorus fertilizers (RPFs). The inputs for PTEs (Table 5) point out that the current concentration of pollutants in soil has the largest share of all PTE sources in the 200-years simulation and that atmospheric deposition in this period was partly higher than inputs via fertilizers. Also for POPs atmospheric deposition was an important route to soil, comparable to fertilizer input (Table 6). The calculated total PTE inputs over 200 years were below the proposed thresholds in all cases (Table 5). From the variation in KD values with soil pH (Table 4), a lower pH effect on Cu leaching can be assumed compared to the other PTEs. The total inputs of the POPs PCB and PCDD/F also were lower than the guidance values. Fertilization with organic waste compost, however, resulted in a PAH input surpassing the guidance values (Table 6). The long-term accumulation risk of PTEs and POPs in European agricultural soils cannot be estimated by looking at their inputs only; also their output and decomposition have to be considered.

### Implications of pollutant concentrations in P fertilizers

The estimates of the mass balance model showed that with none of the RPFs and POPs included in the model calculations the soil threshold values would be reached, even in a 200-years perspective. Therefore, PTEs are no longer excluding nutrient recycling for the assessed sources. Furthermore, the PTE inputs with fertilizers applied at a constant P fertilisation level were not correlated with the PTE concentrations in the fertilizers (Tables 2, 5). The fertilizer application rate is governed by the P concentration in the fertilizer. Therefore, materials with low P concentrations, such as composts, can result in a higher PTE and POP inputs to soil than fertilizers with higher pollutant and high P concentrations. Consequently, the

pollutant-to-nutrient ratio is the main driving factor for pollutant flows applied to the soil. This implies that hypothesis (i), stating that PTE and POP concentrations in fertilizers are the main factor for their accumulation in soil, can be rejected. Therefore, any regulation aiming at a quality control of fertilizers should not be defined according to pollutant concentrations on a dry matter base but should consider the relation between contaminants and nutrients and their input per area and time.

The results also indicate that RPFs like sewage sludge and products obtained from sewage like struvite or from thermal approaches like the production of calcined phosphates via AshDec<sup>®</sup>, Mephrec converter slags or Leachphos<sup>®</sup> can reduce the PTE accumulation risk. The advantage of thermal approaches is the decomposition of organic pollutants, but carbon and nitrogen are lost as well.

### Accumulation and safety risks related to specific fertilizer sources

Urban organic waste, substrates of waste water treatment and by-products of meat production are the source materials for phosphorus recycling. The quality of the resulting fertilizers varies to a large extent.

#### *Urban organic waste*

As an example, the similar raw material of organic household waste digestate OF and organic household waste compost is compared. The material used for biogas production had twice as much P and consequently a lower PTE input than the composted material, possibly, because in the composting process carbonaceous bulking material is necessary. For anaerobic digestion, organic substances with a high degradability and high nutrient concentrations are suitable. In Vienna, separated organic household waste from the inner part of the city contains more easily degradable, nutrient rich material and is processed by anaerobic digestion due to its water content, whereas material collected in the suburbs is composted based on its content of grass and hedge trimmings.

### Substrate of urban wastewater treatment

The model predicted an unexpected low PTE and POP accumulation risk for biosolids, lower than that of composts. In agreement, some more recent studies on PTEs (Herter et al. 2003; Dittrich and Klose 2008; Eriksen et al. 2009; Magid 2013) found that soil contamination via biosolid application was within the legal framework. Accordingly, also the risk to human health via dietary intake of POPs from crops grown on biosolid treated soils was found to be minimal (Eriksen et al. 2009; Haynes et al. 2009; Smith 2009b). If the pollutant-to-nutrient ratio allows the use of sewage sludge as fertilizer, it is also economically very efficient.

RPFs based on biosolid ash, like untreated ash, Leachphos<sup>®</sup> and Mephrec converter slag have the advantage that organic contaminants are destroyed during the incineration procedure. Products made of biosolid ash are still in a pilot-scale, so uncertainties in data quality are presumed. Still, the rate of P recovery from the raw sludge was high, ranging from 70 to 98% for Biosolid ash Leachphos<sup>®</sup>, Mephrec calcined slags and Mg-treated ash via AshDec<sup>®</sup> (Herzel et al. 2015b). Cleaned ashes had a low PTE and no POP accumulation risk. The AshDec<sup>®</sup> depollution process for production of Mg-ash can reduce Cd, Cu, Hg, Pb, Mo, Sn and Zn, but not Cr and Ni (Herzel et al. 2015a). The PTE removal is selective and depends on the process, it might differ in future full-scale plants (Kraus and Seis 2015).

Magnesium–ammonium–phosphate (struvite) showed the lowest soil PTE accumulation risk among the inorganic fertilizers in this ranking due to a very high P content. Data on POPs were not available. Crystallisation of (struvite) takes place in sewage sludge before dewatering (Stuttgart process) or in processed water after primary clarification (Airprex<sup>®</sup> process) and is a stoichiometric process in which a very little amount of pollutants is co-precipitated in the final product. This low PTE amount in struvite was also found by Kraus and Seis (2015). Struvite production has a P recovery rate of 7% (Airprex<sup>®</sup>) to 45% (Stuttgart Sludge Leaching), also depending on wastewater P content (Herzel et al. 2015b). Struvite would be very suitable for organic farming because of the favourable P-to-contaminant ratio combined with low water solubility and a high P fertilizer effectiveness (Möller et al. 2017).

### Slaughterhouse waste

Meat and bone meal, which is allowed in organic farming, is the fertilizer source with the lowest soil PTE accumulation risk in this estimation because of very low contamination (food grade raw material) and a high P content. In terms of pathogens, meat and bone meal can be considered as much safer than animal manure and several other organic fertilizers (Möller 2015).

### Inorganic reference fertilizers

Triple superphosphate had a higher accumulation estimate for Cd than green waste compost OF, for phosphate rock it was a little lower than for green waste compost, all other PTEs were on a very low level. Nziguheba and Smolders (2008) found a significant correlation between Cd, Ni and P content in phosphorus ore leading to elevated Cd and Ni concentration in P fertilizers. Mineral P fertilizers are a major source of PTEs in the food chain (Kratz and Schnug 2005).

Source material and production process of RPFs determined their pollutant and P concentration, and thereby the pollutant-to-P ratio of the fertilizers. In this way, source material and production process of RPFs, but not their pollutant concentration on a dry matter base, affect the input and soil accumulation risk of PTE and POP.

Hypothesis (ii), stating that the source material and the production process of RPFs determine their contamination and the risk of PTE and POP accumulation in soil, was therefore largely confirmed.

### Comparison of recycled phosphorus fertilizers to commonly used fertilizers

The present estimation indicates a lower PTE input and a lower risk of PTE accumulation in soil by applying RPFs then for most commonly used fertilizers, even in the worst-case scenario with a high pH in soil and low precipitation excess. Therefore, the contamination of RPFs with PTEs is not the main constraint limiting recycling, as also indicated by other recent studies (e.g. Eriksen et al. 2009). Furthermore, this assessment estimated a lower Cd input to soil if commonly used fertilizers like green waste compost, triple superphosphate and phosphate

rock were replaced by RPFs, except composts. Cd poses a health risk at concentrations below those causing phytotoxicity (McLaughlin et al. 1999). Therefore, Cd belongs to the most important elements to consider in terms of food-chain contamination (McLaughlin et al. 1999).

The model estimates contradict several older studies that have shown accumulation of PTEs in soils after long-term application of high amounts of biosolids (McGrath 1984; McGrath et al. 1995; Sekhar et al. 2002; Udom et al. 2004; Dai et al. 2006). In the last three decades there has been a significant reduction of PTEs and POPs in organic residues and the sewage systems due to implementation of environmental regulations in Europe. These include the source separated collection of organic household waste compared to mechanically sorted feedstock (Epstein et al. 1992; Sharma et al. 1997; Amlinger et al. 2004), as well as separation of wastewaters from industry and households and changing waste management technologies (Herter et al. 2003; Amlinger et al. 2004; Magid et al. 2006; Schroder et al. 2008; Eriksen et al. 2009). The quality of recycled P fertilizers has improved, in the case of biosolids with a 3–5-fold decline in the content of PTEs (Cd, Hg, Pb, Zn) since the late seventies (Herter et al. 2003; Amlinger et al. 2004; Kupper 2008; Eriksen et al. 2009; Milieu Ltd et al. 2010). The implementation of source segregated collection of urban organic household wastes has reduced the PTE concentration in compost typically by a factor of 2–10 compared to mechanically separated material (Amlinger et al. 2004). The release of POPs into the environment was reduced by the implementation of the Aarhus Protocol and the Stockholm Convention on Persistent Organic Pollutants and by technical improvements in industry like filters and high temperature combustion (Leschber 2006). These achievements should be considered.

Many fertilizers are not allowed in organic farming, like digestates containing non-certified row material (e.g. catering waste digestate), sewage sludge based fertilizers (struvite, ashes, sewage sludge), or not used because of private association rules, like meat and bone meal (Løes et al. 2017). The present calculation for PTE and POP accumulation in soil showed that the regular use of the P fertilizer mentioned above, implies a lower risk of contaminant input and accumulation than using composts, farmyard manure and phosphate rock, which are mostly used in organic farming. The

RPFs struvite, meat and bone meal, catering waste digestate, the ash products Mephrec, AshDec® and Leachphos® had a very low input and accumulation risk of contaminants compared to commonly used fertilizers. Consequently, hypothesis (iii), stating that the regular use of recycled phosphorus fertilizers causes higher soil pollution than established fertilizers, was disproved with respect to the PTE and POP accumulation risk.

#### The relevance of soil and climatic conditions

The balance model estimated a strong influence of soil reaction (pH value) and precipitation excess on Cd, Ni and Zn mobility in soil and a lesser effect on Cr, Cu and Pb. This diverging effect on pH and precipitation excess among the PTEs was not clearly related to the pH effect on their KD values (Table 4). Cu tends to bind to organic ligands and Pb is strongly absorbed by humic matter at pH 4 and above (Oorts 2013; Steinnes 2013b). Under the assumed conditions, the risk of Cd, Ni and Zn accumulation in the topsoil decreased considerably with decreasing soil pH and increasing precipitation excess (Fig. 2) due to increasing leaching losses (Table 7). Leaching was most relevant for Cd, Cr, Ni and Zn at pH 5 and precipitation excess  $F = 0.3 \text{ m year}^{-1}$ , relocating the problem of environmental pollution to the water bodies. Therefore, hypothesis (iv), stating that the soil and climatic conditions have a minor relevance on the soil accumulation risk of PTEs, was disproved for PTEs.

#### Strengths and limitations of the model approach

The used mass balance models provide an adequate insight into the potential risk of PTE and POP accumulation in an average central and northern European agricultural top soil. In addition to the main PTE input routes, i.e. fertilizers, liming and atmospheric deposition, the PTE model regarded soil background concentrations and the main outputs by leaching and in harvested crop parts. Leaching is mainly affected by the precipitation excess and the partitioning coefficient KD of PTEs between solid phase and the solution phase. Among the factors influencing KD, the pH value had the largest effect (Young 2013; Kabata-Pendias 2011). Therefore the model differentiated between two pH values in soil and two precipitation regimes and combined them to

four scenarios. These scenarios gave estimates of PTE behaviour in European agricultural soils and PTE distribution among the environmental compartments soil, water and crop. In fact, soil is more complex and beside the considered determining factors, the soil type, soil organic matter, redox potential, temperature, Fe/Mn oxides, Ca content, erosion and other variables play a role in PTE behaviour and distribution in soil (Young 2013). Therefore, specific sites can deviate from the simulations. Any comprehensive, site-specific assessment of the accumulation risk of contaminants needs to take the local atmospheric deposition and soil conditions into consideration. In addition, a regular monitoring of the total and soluble contents of potentially harmful substances should complement an improved P recycling approach.

The mass balance calculations were based on an average P withdrawal of  $11 \text{ kg P ha}^{-1} \text{ year}^{-1}$  on European stockless organic farms. The average P fertilizer consumption of arable land in Europe calculated by Smolders (2013) was  $9.6 \text{ kg P ha}^{-1} \text{ year}^{-1}$ . So the estimates of this study can be extended beyond organic farming to the average European situation on arable land. For more intense systems, high fertilisation scenarios should be calculated.

Most POPs are apolar organic compounds adsorbed to the organic matter in the soil and therefore not directly affected by pH and precipitation excess and little prone to leaching (Jones and De Voogt 1999). Although precipitation excess can affect runoff generated erosion and POP transfer into surface waters, already by regarding their half-life time in relation to their input by fertilizer application, their estimated accumulation risk in soil was low to very low. Without the model it would have been impossible, e.g. to realistically estimate PAH accumulation and to predict if it would reach the guidance values in 200 years, because fertilization with organic waste compost would result in PAH input five times above the guidance limit.

#### Environmental cycling of pollutants

There is evidence that PTEs are no longer a major constraint limiting the use of RPFs in agriculture. The systematic use of recycled P fertilizers in Switzerland before the ban of biosolids, for example, contributed to about 17% of the PTEs and 20% of the POPs in the food chain while atmospheric deposition, manure,

mineral fertilizer and pesticides were other important input sources (Herter et al. 2003). For several POPs (e.g. PCBs), environmental cycling, mainly atmospheric deposition, is the main source to biosolids and other waste streams (Smith 2009b; Kupper 2008). Human daily exposure to chemicals (emerging pollutants) contained e.g. in packaging materials (Phthalate, Bisphenol A), cosmetics (UV-filter, fragrances), preservatives, flame retardants or drugs are a much higher risk than a potential contamination through the detour recycled fertilizer (Herter et al. 2003). Only reducing the chemical release into the environment, e.g. by excluding potential point sources from the sewage or other treatment systems, will tackle this problem at its source (Herter et al. 2003; Kupper 2008).

#### Conclusions

The calculated PTE inputs by P fertilizers for substituting phosphorus withdrawal via cash crops on organic farms, by liming and by atmospheric deposition over a 200-years period were small compared to the already existing PTE background concentrations in soil. Fertilizers with a high PTE-to-phosphorus ratio, like composts, showed the highest PTE input to soil. The lowest PTE input was found for meat and bone meal, struvite, digestate of catering waste and treated ashes.

The Cd, Cr, Cu, Ni, Pb, and Zn accumulation in soil estimated by the mass balance model did not exceed proposed soil threshold values. Present results therefore indicate that the PTE content in most of the currently available recycled P sources does not exclude them from being used as P fertilizers. The PTE loads of recycled P fertilizers are no longer a major factor limiting the recycling of nutrients like P from our urban areas back to agriculture. Nevertheless, sustainable RPF application requires a regular PTE monitoring, as a high contamination risk for single batches still persists.

A high P concentration in fertilizers resulted in a considerably lower PTE flow per unit of P. Accordingly, composts had the highest accumulation estimates of PTEs among the fertilizers, except for Cd. Struvite, meat and bone meal and digestate of catering waste were the recycled P fertilizers with the lowest potential of harmful effects in soil. Therefore the

pollutant-to-nutrient ratio is better suited to determine the quality of recycled P fertilizers than the actual standards to measure and assess pollutants on a dry matter base.

The estimated accumulation risk of the addressed persistent organic pollutants was also minor. However, there is a need to continue examining the impact of new contaminants entering the food chain.

To reduce the contamination of waste streams it is necessary to reduce and replace harmful substances at their source.

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