

249. LCA and Risk Assessment of Recycled Phosphorous Fertilisers

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ABSTRACT

An efficient phosphorus (P) recycling from urban areas is becoming an increasing issue due to the scarcity of natural P deposits. In order to assess the environmental performance of different approaches of P recycling, a LCA assessment and risk assessment studies were carried out. Generally, we found the supply of recycled P-fertilisers (RPFs) to be competitive as compared to conventional sources in terms of LCA results analysed in this contribution: per kg P and with exclusion of infrastructure processes, the highest abiotic depletion potential is shown for conventional mineral fertilisers based on phosphate rocks due to the finite mineral P resources. For fossil primary energy demand, the recycled fertilisers (struvites and an ash-based fertiliser analysed) had highest impacts per kg P. Relatively high acidification and eutrophication potentials from the supply of P-fertilisers are related to composts, triple-superphosphate and struvites. For the global warming potential per kg P, compost presents the worst results again. However, if co-products of organic fertilisers are considered (i.e. N- and K-contents and the humus sequestration potential), most organic fertilisers are in advantage for a number of indicators – with the exception of conventional composting. The low emission compost and the stabilised sewage sludge present moderate to good overall results. The best relative results for all indicators were found for biogas digestate.

To assess the risk of soil contamination related to the long-term application of RPFs, accumulation scenarios in soil were calculated with a mass balance approach for the potentially toxic elements (PTEs) Cd, Cr, Cu, Ni, Pb, and Zn and for the persistent organic pollutants (POPs) polychlorinated biphenyls (PCB), polycyclic aromatic hydrocarbons (PAH) and polychlorinated dibenzodioxins and dibenzofurans (PCDD/F) in composts, digestates and other RPFs derived from sewage sludge. For all calculations, a fertiliser application over 200 years equivalent to 11 kg P ha⁻¹ yr⁻¹ was assumed. Dependent on PTE mobility in soil due to pH and precipitation excess F, an accumulation or depletion compared to the soil background values was found. Highest accumulation was found in scenario pH 7 F 0,1 m yr⁻¹, lowest in scenario pH 5 F 0,3 m yr⁻¹. Fertilisers like composts, with low P content compared to PTE load, had a higher accumulation potential than fertilisers like struvite, meat and bone meal, sewage sludge ash, sewage sludge and digestates, rock phosphate and triple super-phosphate. Only Cd accumulation with TSP was higher than that with compost. For POPs no accumulation risk in soil was found.

Keywords: life cycle assessment, . potential toxic elements (PTEs), contaminants, recycled P fertilisers

1. Introduction

Phosphorous (P), an essential element in plant nutrition and agricultural management is a globally limited resource. Due to a permanent export of nutrients with biomass harvests and because of the fact that phosphorous (P) is not fixed from the air such as nitrogen and must be imported as a bought-in fertiliser, soils of organic and conventional farms are in danger to loose P soil contents. Negative P balances were found for eight of the 27 EU countries for the average agricultural area for the period 2005 to 2008 (on average -4.8 kg P per hectare; Eurostat 2015). The average for all agriculturally utilised land in the EU-27 shows a positive balance of 1.8 kg P per hectare. Thus and due to the fact that P from fossil deposits is more and more depleted, it is important to close farm's P-cycles and the societal cycle by recycling residential organic wastes, e.g. organic household wastes or (stabilised) sewage sludge and its products. The focus of this contribution is the analysis of important

environmental impacts of the production, supply and fertilisation of selected recycled P fertilisers (RPFs) by a life cycle assessment (LCA) and the accumulation potential of potentially toxic elements (PTEs, often referred as heavy metals) and three persistent organic pollutants in soil after long-term fertiliser application.

2. Methods

We analysed and compared a number of recycled P-fertilisers: Firstly organic fertilisers produced from urban organic wastes, which are composts (with different composting techniques and substrates) and biogas digestates¹, as well as stabilised sewage sludge. Secondly a recycled fertiliser based on sewage sludge (SS) ash, two different struvites (with different technologies; from SS) and meat and bone meal. Thirdly, the conventional and commonly used references rock phosphate (assumed to be imported from Morocco for the LCA) and triple-superphosphate (TSP; based on phosphate rocks mined in Morocco and partly produced in Morocco as well as the European Union for the LCA).

LCA – data, impact categories, system boundaries and sensitivity analysis: LCA-relevant data on P concentration in the different substrates (e.g. household wastes, sewage) and their dry matter content were taken from Möller & Schultheiss (2014) and Möller (2015). For the demand on inputs for the production process, i.e. energy (carriers) and chemicals, data from studies and databases were used including measured data from pilot plants, e.g. Remy & Jossa (2015) and Ecoinvent (2014). Emissions from the production of fertilisers, for example during composting were taken from Ecoinvent (2014) or calculated from Pardo et al. (2014). For emissions from biogas plants, we assumed a leakage of 2% of CH₄ but no losses of N₂O and used IPCC (2006) guidelines for calculation. Credits for substituted energy from biogas plants were derived with the German energy mix according to Ecoinvent (2014) data.

Five LCA impact categories were calculated for each of the P-fertilisers and referred to the functional unit of 1 kg P: abiotic resources depletion potential (ADP; Sb-eq), fossil primary energy demand (FED; MJ), global warming potential (GWP, CO₂-eq), acidification potential (AP; SO₂-eq) and the eutrophication potential (EP; PO₄³⁻).

ADP considers the use of resources from the lithosphere and also includes fossil energy carriers. The latter are also included in the FED. The EP covers potential losses of phosphate (PO₄) and nitrogen (as NO₃, NH₃, NO_x) during the production processes and distribution of fertilisers with their potential negative impacts on aquatic and terrestrial ecosystems. The AP analyses acidifying substances (SO₂, NH₃ and NO_x) during the production and transportation of fertilisers and their inputs. The GWP reflects emissions relevant for the climate change: methane (CH₄), nitrous monoxide (N₂O) und carbon dioxide (CO₂).

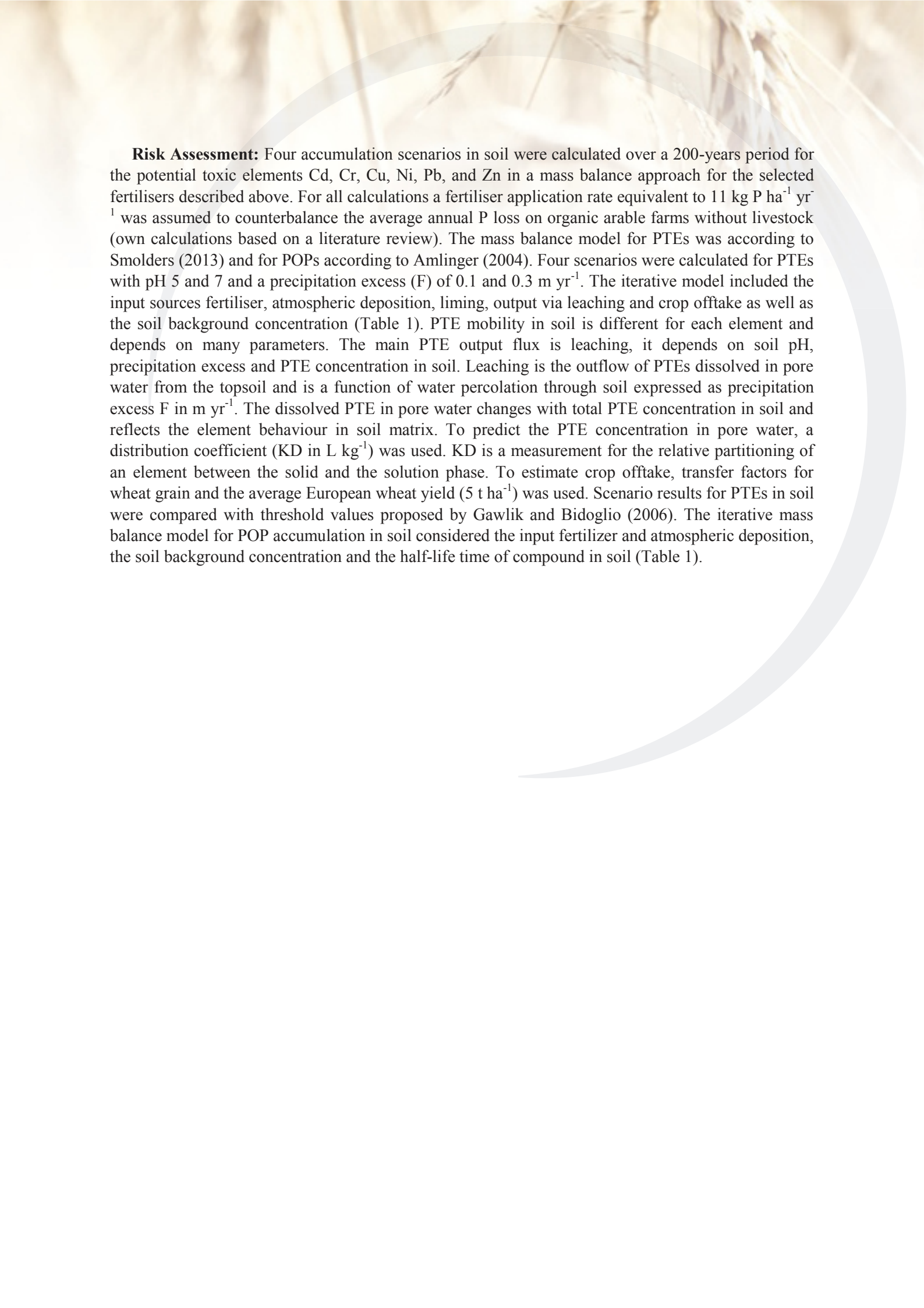
All recycled P-sources are evaluated in the software SimaPro (v 7.3; PRé Consultants) along their whole life cycle until application and they are compared with each other as well as with the conventional references of mineral P fertilisers. For characterisation, the method "CML" (Guinée et al., 2002; with updated characterisation factors) was selected.

¹ Fertilisers that may be used in organic farming are designated with „(OF)“

System boundaries were chosen to include the phases of supply and production of RPFs. This includes acquisition of inputs such as raw materials and (co-) products from previous processes (e.g. materials for chemical processes), the demand on energy carriers, etc. Infrastructural processes i.e. capital goods such as buildings and roads for transports were excluded. The collection of substrates (e.g. organic wastes from households) was considered for the recycled P-fertilisers where it is specifically needed for the production processes. In cases where the collection of substrates is independent from production and has to be done in any case (e.g. the transportation of urban household wastes to a collection site), this first transportation was not considered for the LCA of the APFs. For these cases, the effort for the disposal of wastes was attributed to the end of life stage of the previous main product, i.e. to its main use phase; the disposal of biogenic household waste, for instance, is therefore attributed to food production.

Short as well as long process chains were found for the APPs' calculation: (1) for stabilised SS, for instance, the process chain covered by the LCA contains only the two processes of transports to a farm and field application. Emissions from SS until transports to a farm were allocated to the disposal phase (especially for the case of anaerobic treatment, where the potential net-energy gain from the process is also accounted for the disposal phase of SS and not to the fertiliser). (2) The example of the RPF biogas digestate covers more processes. The system boundary starts with a treatment of the substrate and includes the demand on auxiliary material or energy. On the contrary, the process of digestion leads to biogas which is used as fuel (to produce electricity and heat); consequently energy is produced that replaces other energy and a credit is accounted for this substitution. As above for the example SS, the household wastes' collection (transport) before their digestion is not accounted for, but allocated to the food waste disposal (food's end of life). (Modern) biogas plants are built as airtight systems, hence, losses of CH₄ and N-containing gases (NH₃, NO_x, N₂O) are assumed to be low. However, a small proportion of gases passes off and is accounted for within the process of the biogas plant. System boundaries end with the inclusion of energy demand for the process of fertilising: The application (spreading) of RPFs onto fields is also accounted for within the system boundaries for all P-fertilisers. Further emissions from fertilising or from the fertilised soil are not accounted for, to obtain comparability due to varying emissions, which highly depend on used techniques or processes for P recovery, the content and availability of (reactive) nitrogen and carbon in the fertilisers, etc. However, for the organically-based RPFs which contain other nutrients such as nitrogen (N) and carbon (C), an additional calculation addressed the credits for a humus sequestration potential and for substituted N- and K-fertilisers, the latter according to Ecoinvent (2014). The credit for the humus sequestration potential was calculated based on the assumption that a long-term organic fertilisation (50 years) increases the content of humus at most by 10% of the initial value (for compost) until humus saturation. Credits were specifically determined for organic fertilisers considering their respective P- and C_{org}-contents. Due to different types (liquid or solid) and varying application machinery as well as different concentrations of P in a certain fertilisers (for dry matter) and due to the RPFs specific water contents, impacts of transports and fertilising vary among the RPFs. For a useful comparison of the different RPFs, comparable transport distances were used. For an industrial production of a P-fertiliser (this is the case for recycled fertilisers, for meat and bone meal, rock phosphate and TSP), in total a 500 km transport by a lorry (16-32 t; fleet average concerning loading) was assumed from the production to a regional storehouse. Furthermore, a 20 km transport from the storehouse to the farm by tractor and trailer was assumed for these industrial P-fertilisers. For regionally produced RPFs (composts, biogas slurry or untreated SS), in total 50 km with tractor and a trailer (or a tanker for liquids) were assumed. For transports of auxiliary inputs (which are also considered within the category "transport"), e.g. chemicals for recycled RPFs, each 100 km of lorry and freight train were assumed.

Sensitivity analyses were done for the highly effective impacts of C- and N-containing biogenic emissions from the composting processes. We compared data from Ecoinvent (2014) to data calculated for composted food wastes based on Pardo et al. (2014).



Risk Assessment: Four accumulation scenarios in soil were calculated over a 200-years period for the potential toxic elements Cd, Cr, Cu, Ni, Pb, and Zn in a mass balance approach for the selected fertilisers described above. For all calculations a fertiliser application rate equivalent to $11 \text{ kg P ha}^{-1} \text{ yr}^{-1}$ was assumed to counterbalance the average annual P loss on organic arable farms without livestock (own calculations based on a literature review). The mass balance model for PTEs was according to Smolders (2013) and for POPs according to Amlinger (2004). Four scenarios were calculated for PTEs with pH 5 and 7 and a precipitation excess (F) of 0.1 and 0.3 m yr^{-1} . The iterative model included the input sources fertiliser, atmospheric deposition, liming, output via leaching and crop offtake as well as the soil background concentration (Table 1). PTE mobility in soil is different for each element and depends on many parameters. The main PTE output flux is leaching, it depends on soil pH, precipitation excess and PTE concentration in soil. Leaching is the outflow of PTEs dissolved in pore water from the topsoil and is a function of water percolation through soil expressed as precipitation excess F in m yr^{-1} . The dissolved PTE in pore water changes with total PTE concentration in soil and reflects the element behaviour in soil matrix. To predict the PTE concentration in pore water, a distribution coefficient (KD in L kg^{-1}) was used. KD is a measurement for the relative partitioning of an element between the solid and the solution phase. To estimate crop offtake, transfer factors for wheat grain and the average European wheat yield (5 t ha^{-1}) was used. Scenario results for PTEs in soil were compared with threshold values proposed by Gawlik and Bidoglio (2006). The iterative mass balance model for POP accumulation in soil considered the input fertilizer and atmospheric deposition, the soil background concentration and the half-life time of compound in soil (Table 1).

Table 1: Average soil background concentrations in European agricultural topsoil, atmospheric deposition, lime input and proposed threshold values for PTEs and half-life times for POPs.

		Cd	Cr	Cu	Ni	Pb	Zn
Soil background concentration	mg kg ⁻¹	0.28	94.8	17.3	37.3	32.6	68.1
Atmospheric deposition	g ha ⁻¹ yr ⁻¹	0.36	9.30	34.0	10.0	11.9	227
Input with lime	g ha ⁻¹ yr ⁻¹	0.14	0.92	1.38	0.78	0.94	2.00
Proposed threshold values in soil	mg kg ⁻¹	1.00	75.0	50.0	50.0	70.0	150
		PCB	PAH	PCDD/F			
Soil background concentration	mg kg ⁻¹	0.01	0.43	0.000006			
Atmospheric deposition	g ha ⁻¹ yr ⁻¹	0.35	2.67	0.000002			
Half-life time in soil	years (mean)	14.2	12.2	29.7			
Proposed threshold values in soil	mg kg ⁻¹	0.30	1.00	0.01			

PCB: polychlorinated biphenyls, PAH: polycyclic aromatic hydrocarbons, PCDD/F: polychlorinated dibenzodioxins and dibenzofurans

3. Results and Discussions

Detailed results on fossil energy demand and global warming potential: Compost-P results in medium results concerning FED, whereas the two struvites, biogas slurry (due to its transport) and meat and bone meal show much higher demand (Figure 3). Although the same transport distance (50 km by tractor) was assumed for all regionally produced RPFs, digestates shows a substantially higher impact of transportation than other RPFs. This is due to the higher water content of digestates, i.e. that a higher amount of fertiliser (water) has to be transported. However, a part of biogas' FED is substituted by the net-energy produced within the biogas process. Stabilised and regionally transported liquid SS, rock phosphate, the ash-based RPF and TSP present low to medium FEDs.

Results for the GWP do not consider infrastructure's impacts but the co-products for the organic fertilisers (humus sequestration, N- or K-fertiliser effect). Highest GWP was found for composts as a consequence of N- and C-losses during composting (Figure 3). For the sensitivity analysis with data from Pardo et al. (2014) vs. Ecoinvent (2014), we found a nearly equal impact of the biogenic emissions on the GWP and hence no effect on varying results. However, the performance of organic fertilisers improves substantially with consideration of the substitution potential of co-products and even results in negative net emissions per kg P – except for standard composting. Especially due to the co-products' effect, the best net emission result was found for digestates. Furthermore, the effect of potential CH₄ leakage from the biogas process is almost offset by substituted emissions through the net energy gain. Contrarily, liquid digestates show a comparably high impact of transportation on the GWP due to the significantly lower P-content (i.e. the higher water content). Regionally transported (liquid) stabilised SS show a relatively low GWP. Reasons for the low environmental impacts of SS

are the comparably high P content in the sludge and the assumed relatively low transport distances for a regional application and mainly the substitution potential of co-products. The ultimate substitution potential depends upon which N fertiliser is replaced with the organic (P-) fertiliser, the long term nitrogen use efficiency of the N in the organic fertiliser and how successfully – depending on the humus saturation of the soil – C_{org} is sequestered in a long-term. As compared to organic RPFs such as digestates and especially composts, struvites or mineral P-fertilisers do not (or hardly) contain C, N or K anymore. Consequently, the organic fertilisers (with the exception of standard composting) are favourable in terms of most LCA results when they are related to their overall fertilising value.

Comparably low GWPs were also found for the conventional references rock phosphate and TSP. The latter has a slightly higher environmental impact due to the beneficiation processes for the raw material phosphate rocks. The analysed RPF based on SS is in advantage as compared to the struvites due to an efficient use of the by-product heat from mono-incineration of SS (for the drying of the fertiliser product). The GWP of struvites (especially for #2) is comparable to net emissions of the low emission compost. The two struvites based on SS need relatively high amounts of chemicals and energy embodied in these auxiliary materials (including their transport activities) as well as high energy demands for the precipitation process itself. Thus they show medium GWP results. The GWP for meat and bone meal is dominated by the fossil energy demand, which is mainly a consequence of energy needed for drying, extracting and especially for pressure sterilisation of the livestock material.

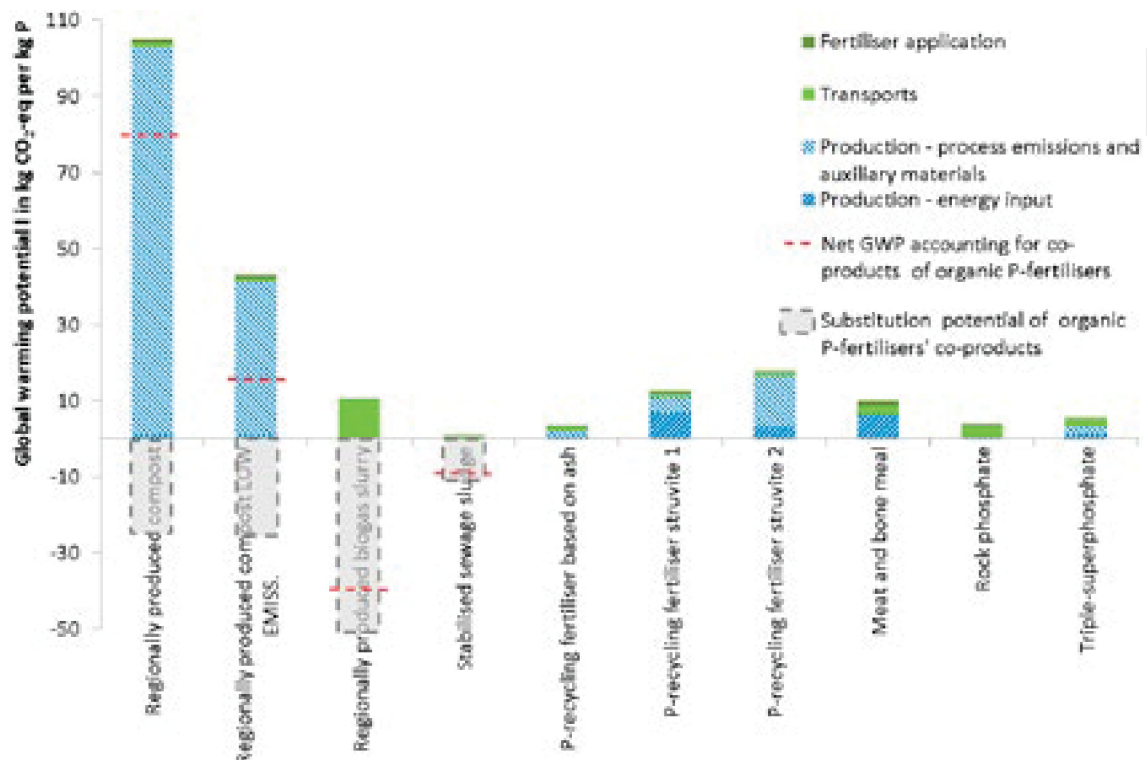


Figure 3. Global warming potential (GWP, greenhouse gas potential) in kg CO₂-eq per kg of P without infrastructure emissions but with co-products from the organic fertilisers (humus sequestration, N- or K-fertilising) taken into account.

P in ash-based RPFs as well as in rock phosphate is hardly available for plants, especially in alkaline soils. Thus, a good LCA result can be paired with a trade-off concerning the quality of the RPF and vice versa. The LCA results represent one important contribution to analyse strengths and weaknesses of P-fertilisers and recycling pathways but they have to be accompanied by other indicators such as plant availability, the effectively recyclable proportion of limited nutrients and the risk assessment on accumulation of contaminants (see chapter after the next).

Relative advantages and disadvantages of specific fertilisers for all five LCA-indicators:

Figure 4 presents an overview of all indicators analysed in this study, i.e. ADP, EP and AP in addition to the FED and the GWP as relative advantage between the best/lowest result per indicator (0) and the worst/highest result per indicator (1). A high column indicates high environmental impacts. Whereas fertilisers based on phosphate rock have a high ADP, struvite partially a high FED and EP and compost very high AP, EP and GWP, the RPFs based on SS show lower values in all categories. The low emission compost and the stabilised SS present moderate to good overall results. The best relative results for all indicators were found for biogas digestate (see Fig. 4).

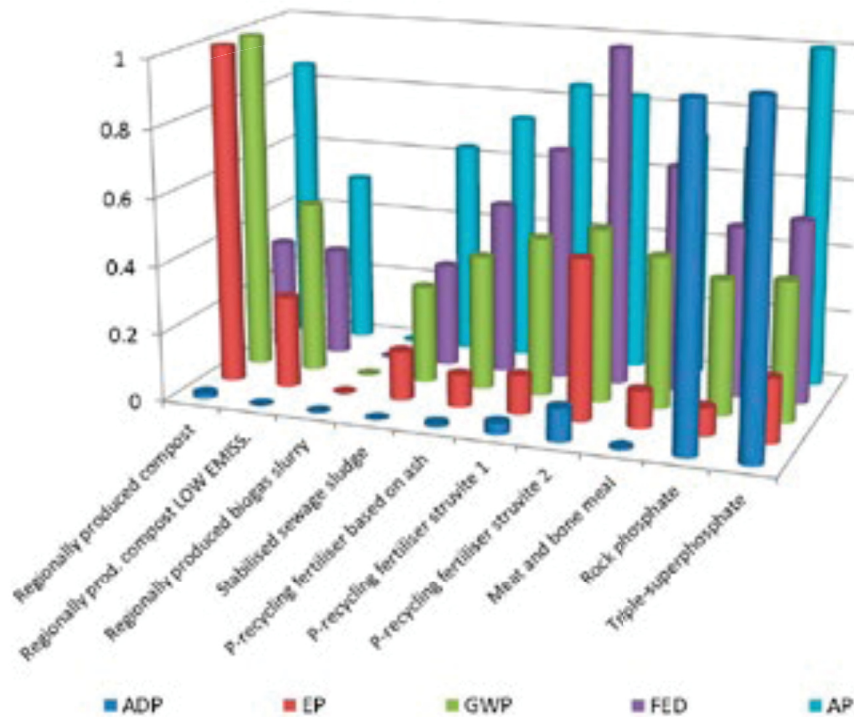


Figure 4. Relative values per kg P for abiotic resources depletion (ADP; in kg Sb-eq), fossil energy depletion (FED; in MJ fossil primary energy), global warming potential (GWP; in kg CO₂-eq), acidification potential (AP; in kg SO₂-eq) and eutrophication potential (EP; in kg phosphate-eq) from selected P-fertilisers between the best/lowest result per indicator (0) and the worst/highest result per indicator (1).

If the infrastructure is taken into account, the relative comparison would also show a slightly different picture. The most important change would be found for the indicators FED and ADP: after inclusion of infrastructure, highest values would result for recycled fertilisers (struvites or ash-based

RPF) due to their impacts of the chemical inputs and the highly technical production plants. However, even if the more comprehensive analysis on ADP including infrastructural processes shows a better result for the phosphate rock-based fertilisers, RPFs should be further developed and used before the fossil P deposits are depleted.

Results on the risk assessment: PTE accumulation in soil depended mostly on pollutant-to-nutrient ratios in fertiliser and the behaviour of the respective PTE in soil. Fertilisers with low P concentration in dry weight, like composts, had a high PTE accumulation in soil. The risk assessment calculation indicates the highest PTE accumulation rate for a soil with pH 7 and a low precipitation excess ($F = 0.1 \text{ m yr}^{-1}$), followed by scenario pH 7 $F 0.3 \text{ m yr}^{-1}$ and pH 5 $F 0.1 \text{ m yr}^{-1}$. The smallest accumulation potential showed a soil with pH 5 0.3 m yr^{-1} . This means, that PTEs in a soil with low pH and a high precipitation excess are leached to the water body. Exceedance of thresholds was found only for Cr, the other five PTEs remained below the threshold. The average European soil background concentration of Cr was already higher than the proposed threshold (244 kg ha^{-1}). Although in pH 5 scenarios a Cr depletion was found after 200 years, the threshold was not reached. In scenarios with pH 7, an accumulation was found for the composts (328 kg ha^{-1}), cattle manure and the digestate (OF), other fertilisers remained close to soil background concentration (308 kg ha^{-1}). Cd showed the highest accumulation potential in soil pH 7 for TSP (1.56 kg ha^{-1}) followed by composts and rock phosphate, cattle manure (1.23 kg ha^{-1}), while the other fertilizer were similar to the background concentration of 0.9 kg ha^{-1} . Cu was depleted or close to soil background concentration (56 kg ha^{-1}) with TSP, struvite and rock phosphate, meat and bone meal OF and increased with digestates, ashes, cattle manure, SS, untreated SS-ash and composts (88 kg ha^{-1}). For Ni we found a strong depletion in scenario pH 5 $F 0.3 \text{ m yr}^{-1}$ to approx. 17 kg ha^{-1} for all fertilisers, in scenario pH 5 0.1 m yr^{-1} between 71 to 61 kg ha^{-1} . Ni accumulated with compost fertilisation in soil pH 7, other fertilisers were close to soil background concentration (121 kg ha^{-1}). For Pb, no big difference was between the scenarios; green waste compost had again the highest accumulation potential (136 kg ha^{-1}), followed by compost catering waste, compost biowaste, digestate OF (111 kg ha^{-1}), SS ash, cattle manure and SS. Struvite, ashes, meat and bone meal OF, TSP and rock phosphate was close to the soil background concentration of 105 kg ha^{-1} . Zn was depleted in scenario pH 5 $F 0.3 \text{ m yr}^{-1}$. For soils with pH 5 and $F 0.1 \text{ m yr}^{-1}$, an accumulation for composts was found. In soil pH 7 all fertilisers led to Zn accumulation, especially composts (287 kg ha^{-1}), followed by SS ash, digestate OF, SS, most SS ash-based RPFs, cattle manure and digestate catering waste. Close to soil background concentration (221 kg ha^{-1}) remained meat- and bone meal OF, struvite, one SS ash-based RPF, TSP and rock phosphate.

For POPs no accumulation in soil was found, the calculated values after 200 years were lower than the soil background concentration.

4. Conclusions

In order preserve the essential element P for future agriculture, it is important to close nutrient cycles with recycled P-fertilisers. This needs the further development and improvement of suitable recycled P-fertilisers as well as an appropriate political framework (including farmers' associations guidelines, etc.) and their broad application in practical agriculture. From an LCA perspective, the different fertilisers show specific advantages and disadvantages, also depending on the system boundaries and the method of the assessment. Furthermore, a number of trade-offs between different indicators' results, including results from the risk assessment and other critical factors, was found.

However, it can be noted that – in the light of the results of this contribution – SS and its products should be used on behalf of the needs of future generations. SS with low pollutant contents could eventually be used directly, especially if it is not used for foods that are produced directly for human consumption. For SS with higher contamination levels, various processing options exist, which lead to a highly valuable products at equitable costs and equitable environmental effects (struvite or ashes for application in alkaline and acidic soils, respectively). Additionally, other treatment methods for organic wastes, e.g. anaerobic digestion to be used for energy-dense substrates and low emission composting on woody materials or the use of meat and bone meal, are recommended from the LCA perspective on behalf of P-supply for future generations.

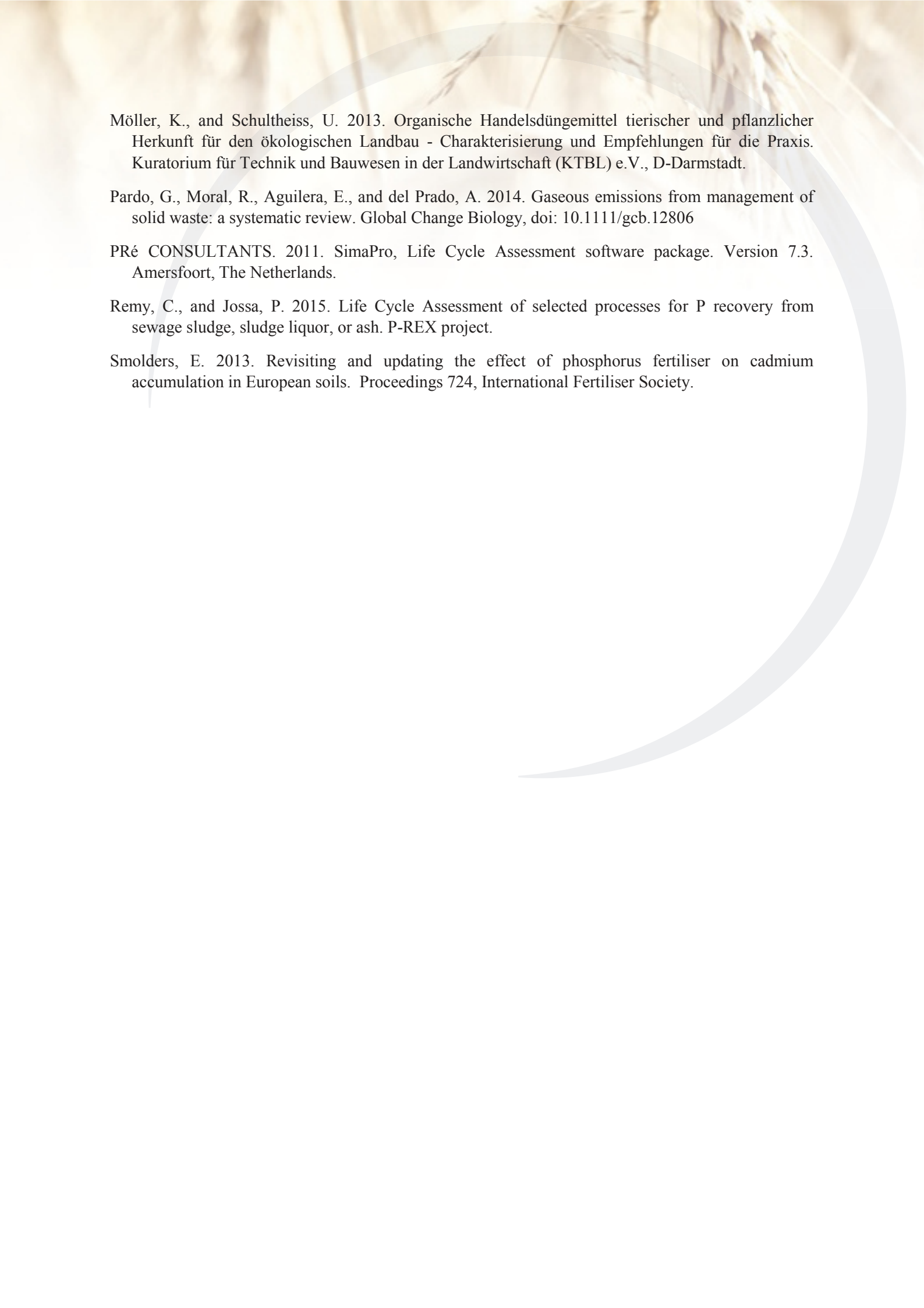
The risk of the soil contamination with PTEs increased with increasing soil pH, decreasing precipitation excess and increasing pollutant-to-nutrient ratios in fertilisers. In general, for typical fertiliser application rates a low accumulation potential of toxic elements was found, especially for the recycled P fertilisers based on ashes, struvite and meat and bone meal. But even if the accumulation in soil is little, it should be considered that PTEs applied to the soil are partially transferred to other environmental compartments like water or dust where they may cause further negative environmental impacts. The POPs did not accumulate in soil and values after 200 years of fertiliser application remained below the soil background concentration.

5. Acknowledgements

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