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Life Cycle Assessment of Phosphorus Sources from Phosphate ore and urban sinks:Sewage Sludge and MSW Incineration fly ash

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ABSTRACT: Urban sinks accumulate phosphorus and other elements and may serve as sources of secondary raw materials. This paper evaluates phosphorus sources based on their environmental impact. In a life cycle assessment (LCA) the conventional production was used as a yardstick against which to measure the performance of two recycling options: spreading of sewage sludge and phosphorus recovery from municipal solid waste incineration fly ash (MSWA). When compared as three gate-to-gate processes, the sludge spreading had the lowest potential environmental impact, except in the impact categories eco- and human toxicity. In the future, the sludge spreading could potentially outperform the conventional process also with regard to toxicity, provided its Hg and Cu content can be reduced. Phosphorus extracted from the MSWA had the highest impact, except in relation to eutrophication. The benefits of avoiding the conventional production were greater than the sludge recycling impacts for all categories except toxicity. When conventional production is substituted by the MSWA recycling, the eutrophication and land-use impacts are avoided, while the impacts in other categories are considerable. The development needs identified for this method include substitution of HCl, reduced water consumption, and reduction of the product's metal content. Solutions to all of these challenges have been proposed and are currently being tested.

Key words: Urban mining, Environmental impact, Life cycle assessment, Recycling

INTRODUCTION

Modern agriculture depends on the input of fertilizers for production of food, feed, fiber and biofuels. Nitrogen (N) and phosphorus (P) are both essential to life and non-substitutable, however P is also a non-renewable resource derived from rock deposits. We argue that local recycling of secondary phosphorus resources must be developed because of the strategic importance of fertilizers for food security, due to geopolitical risks, in order to mitigate the environmental impacts of fertilizer production, and in response to the declining quality of the remaining ore, something that further increases both the impacts and costs of conventional production methods.

The focus of this paper is on urban secondary resources. In industrialized countries, more than 2/3s of the population is urban and by 2050, 2/3ds of the global population will live in cities. The largest urban flows of phosphorus are related to food, but paper, wood, textiles and chemicals also contain phosphorus (Kalmykova *et al.*, 2012). Humans excrete 98% of the phosphorus consumed in food and this accumulates in sewage sludge. In the EU, 0.6 Kg P/cap per year is found in sludge (Dawson and Hilton, 2011). The simplest way to recycle phosphorus from sludge is to spread it on land (direct recycling) where organic materials such as P and N can be recycled.

Solid waste is an underestimated source of phosphorus. A study of phosphorus flows for the EU27, Japan, and a municipality in Sweden, recently showed that solid waste, and its incineration residues in particular, contains as much phosphorus as sewage sludge on an annual basis (Kalmykova *et al.*, 2012;

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Matsubae-Yokoyama *et al.,* 2009; Ott and Rechberger, 2012). Recently, a method for phosphorus extraction from MSW fly ash (MSWA) by leaching and precipitation procedures has been published by authors Kalmykova and Karlfeldt Fedje (2013).

In this paper we investigate the suitability of this newly developed method for recycling of urban phosphorus. We also examine the pre-requisites for direct recycling of sewage sludge and how these compare to conventional phosphorus production.

MATERIALS & METHODS

Life Cycle Assessment (LCA) is a standardized systemic and systematic method for assessment of the potential environmental impact of a product or process over its entire life cycle, i.e. from extraction of raw materials to waste management (ISO 14040, 1997; ISO 14041, 1998; ISO 14042, 2000).
The following options were compared:

- Baseline: conventional production of phosphorus fertilizer from phosphorus ore through the acid (wet) process, as this clearly dominates the market; no recycling, i.e. the phosphorus produced ends up in landfill.
- Option 1: recycling of phosphorus from MSWA by means of a recently developed process using acid leaching and CaO precipitation.
- Option 2: recycling of phosphorus through spreading of sewage sludge from municipal waste water treatment plants on arable land.

As this LCA is change-oriented, all processes that are identical to all three options have been excluded.

The functional unit was set to 1 tonne 100% phosphate (P_2O_5) produced or recycled. Only midpoint characterization has been applied and the following impact categories were chosen: abiotic depletion, global warming, acidification, eutrophication and toxicity. CML 2001 was used for characterization, except for toxicity, which was characterized using the USETox model (Rosenbaum *et al.,* 2008). The options studied have different geographic boundaries. The conventional production is assumed to take place in Morocco, as this is the world's largest producer and exporter of phosphorus rock and phosphoric acid. The two recycling options are assumed to be carried out locally, in this case in Gothenburg, Sweden. As the study is a change-oriented LCA, the time frame is prospective. Where a specific time frame was needed for data collection, 1 year was used as boundary. For the impact categories in CML 2001, the default time frame was adopted. The focus was on the operation of the processes. Infrastructure and personnel requirements were not considered. The processes and emissions of municipal solid waste incineration and waste water treatment were excluded. The system boundary is shown in Fig.1. Data was mostly taken from the SimaPro software integrated database, including for the entire process of phosphorus mining and production. Additional data from various literature sources or personal contacts were retrieved for the recycling options. See Yu (2012) for further details. The sensitivity of the results was examined by comparing the outcomes when using different data sources, estimation methods and characterization methods.

RESULTS & DISCUSSION

About 90% of the phosphate rock worldwide is processed by the so called wet-chemical or acid route to produce phosphate fertilizers. This process was used as a "conventional" P production, to be compared to the recycling options. For the detailed description of the process requirements and the impacts see another part of this study (Kalmykova *et al.,* 2014b). The data for phosphorus production by the acid route was extracted from the Ecoinvent database (Ecoinvent Centre, 2007). The data was used as unit processes (rather than as system processes), i.e. emissions and resource inputs were extracted from the database for each process step included. Fertilizers in the database are reported according to specific types, e.g. monoammonia phosphate (MAP), diammonia phosphate (DAP) and triple superphosphate (TSP). These three fertilizers account for 95% of the phosphoric acid production and their annual production volume was used to calculate the proportions for 1 tonne of phosphorus fertilizer (Table 1). Consequently, conventional production was calculated as follows; 1 tonne of phosphate fertilizer consists of 45% of DAP, 49% of MAP and 7% of TSP. Other types of fertilizers were neglected, while contaminants in the fertilizers were included (Petersen *et al.,* 2009).

The major phosphorus export countries are the US, Russia, China and Morocco, which together make up 68% of the total export (Scholz and Wellmer, 2013). The transport from production to consumption was calculated by multiplying the port to port distances from the export countries to Sweden with the percentage of the respective export volume, followed by adding to the weighted distance of 5688km.

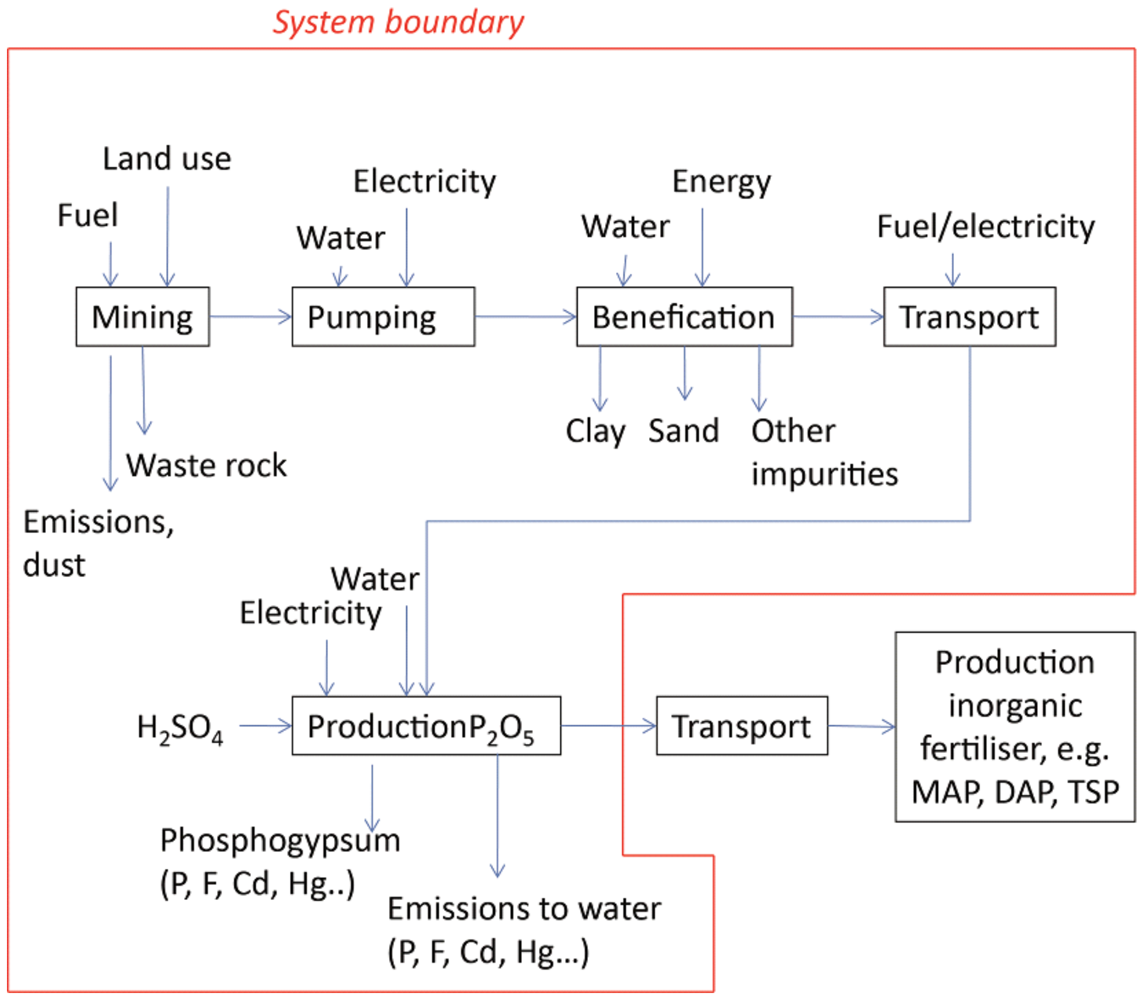

Fig.1. System boundary, unitless

Table 1. Annual production of common N and P fertilizers and their precursors, phosphoric acid and ammonia, in 2009, in metric tonnes (IFA, 2011)

Product	Annual production [t P_2O_5]	Product	Annual production [t N]
MAP	15446	AS	4106
DAP	14221	CAN	3665
TSP	2075	Urea	67761
		AN	13995
Phosphoric acid	33588	NH_3	125297

The phosphorus content in the sewage sludge is 3% (7% as P_2O_5). This means that to recycle 1 tonne of P_2O_5, 14 tonnes of sludge has to be spread onto the land. According to estimates by (Tideström *et al.*, 2009), the sludge is transported on average 15 km from the treatment plant to the nearest arable land. The heavy metal and organic pollutants content of the sludge was accounted for as soil input (Pettersson and Wahlberg, 2010).

When the sludge is added to the soil, nitrogen is also recycled. The credit for recycling nitrogen has been taken into account. The annual production volumes of four common fertilizers were used to calculate the constituents of 1 tonne N fertilizer (Table 1). The total volume amounted to 70% of the total ammonia production but was assumed to be 100% of the market, as the remaining fraction of ammonia use was diverse. Based on this assumption, 1 tonne of N fertilizer consists of 75.7% Urea, 15.6% ammonia nitrate (AN), 4.6% ammonia sulphate (AS), and 4.1% calcium ammonia nitrate (CAN).

For all the studied options, the application of the fertilizers is regarded as processing and the emissions and energy consumption used come from the database.

The material needed to recover 1 tonne P_2O_5 from MSW fly ash and the waste products generated are listed in another part of this study (Kalmykova et. al. 2014). Within the LCA, a choice was made for the HCl source with the lowest possible impact, in particular, "36% HCl in H_2O from reacting propylene and chlorine". Electricity is required for the slurry agitation in a reaction chamber and for the anaerobic digester in the waste water treatment. An electricity consumption of 0.008kWh/m^3 for a cylindrical tank with turbine mixer was assumed (Tchobanoglous *et al.*, 2004). The air emission is assumed to be negligible. Although the remaining leachate may be subject to further recovery processes for other resources, in particular metals, in this study the leachate is assumed to be waste requiring treatment for heavy metals removal. The data for heavy metals removal from leachate was estimated from a similar industrial process using precipitation and absorption and with an electricity consumption of 0.33kWh/m^3. Although the remaining waste ash may be used as a construction material, in this study it was assumed that this waste product was transported to landfill. The distance to the landfill in the studied case is 10 km.

The inventory data have been translated to characterization results and normalized to the world 1990 for the categories characterized using the CML method . The y-axis shows the value for each impact category, normalized to the world 1990 levels (normalization method integrated in CML 2001). For toxicity indicators, the USETox method was applied and the results for human toxicity show the potential number of disease cases caused by the processes. The ecotoxicity result shows the Potentially Affected Fraction i.e. the proportion of the total number of species in an ecosystem that would (potentially) be affected by the stressor.

In order to quantify and compare the environmental impacts of the investigated phosphorus sources, conventional production, recovery from MSWA, and sludge spreading have been cut off from the life cycle and considered as gate-to-gate processes. In this way, the processes can be compared to each other as independent processes. The results are shown in Figure 4. Sludge recycling has the lowest environmental impact, as it only requires machinery to transport and spread. Only the human non-caner toxicity and ecotoxicity impacts are significantly higher than for the conventional production, due to the heavy metals and other toxins content.

The recovery process causes the greatest impacts, except in relation to eutrophication and cancer risks (Fig. 2). The abiotic depletion impact is due to the HCl demand that requires consumption of oil. Burning of coal in the CaO production contributes to the global warming potential and acidification (SO_2 emissions).

In order to evaluate whether urban phosphorus ought to be recycled from an environmental perspective, the impacts of recycling were compared to the benefits of avoiding the conventional phosphorus production method (Fig. 3). As the impacts from production are equal in both the baseline and alternative scenarios, these were omitted. Positive values imply that recycling has a greater impact than conventional production, and avoided impacts are denoted by negative values. Neither of the recycling processes had any impact on eutrophication, and sludge spreading avoids the global warming potential. The impacts concerning toxicity are however considerable due to the heavy metal content of the recycled phosphorus products. The metals that contribute most to the toxicity of the phosphorus recycled from MSWA are As and Zn, followed by V, Pb, and Sn. For sludge, Hg and Cu are the main sources of toxicity. The recycling from MSWA avoided the impact categories urban land occupation and natural land transformation (land-use, not shown).

Results using different sources of data, and different estimation and characterization methods have been compared to identify significant factors and avoid uncertainties caused by arbitrary, or partially arbitrary, choices.

First of all, survey results (CML2001 and USETox) for conventional phosphorus production using the data from the software integrated database and from the literature were compared (Davis, 1999; EFMA, 2000; EC, 2007; Hocking, 2005). When using literature data, a much greater impact was predicted for the abiotic depletion (73% greater) and global

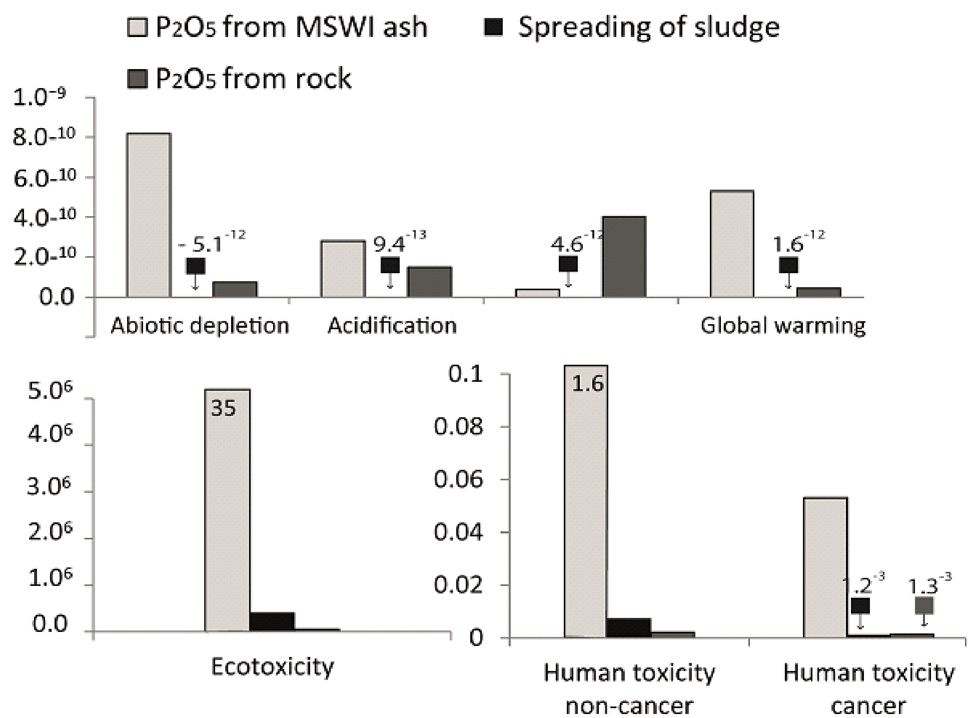

Fig. 2. Impacts of production and recycling excluding incineration and wastewater treatment. The unit for the toxicity-related impacts is "potentially affected cases" for people and "Potentially Affected Fraction" (PAF) of species for other living organisms. For other impacts, the unit is the "fraction of the entire global impact"

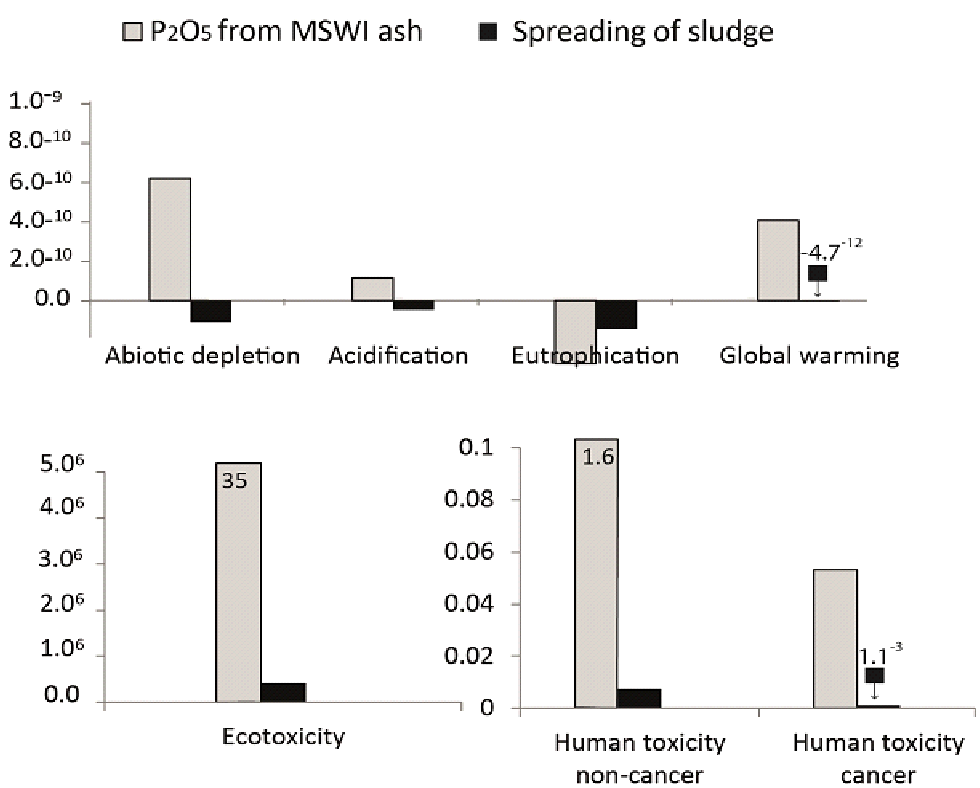

Fig. 3. Impact of recycling of phosphorus including the benefits of avoiding virgin phosphorus use. The unit for the toxicity-related impacts is "potentially affected cases" for people and "Potentially Affected Fraction" (PAF) of species for other living organisms. For other impacts, the unit is the "fraction of the entire global impact"

warming (87% greater). However, the eutrophication and acidification impacts were 71% and 43% lower, respectively. The toxicity results only varied by 3-9%, either up or down. Preference was given to the results obtained with data from the software integrated database, as this source is more extensive than the alternative sources and covers most of the material and energy flows, and also for the purpose of ensuring consistency with and comparability to other studies.

In comparisons of different characterization methods, including ReCiPe, TRACI, USEtox and CML, the results were consistent for most of the categories. In particular, for all the methods, the recovery process showed the greatest impact, apart from for a number of uncommon categories such as land occupation and smog.

A choice had to be made in relation to the metal removal efficiency from the waste leachate in the recovery process. One option was to use the threshold values for effluents that the facility has to comply with; the other was to use the 99.5% removal efficiency typically obtained in other processes. The removal efficiency depends on conditions such as the initial concentration and choice of removal agents. The threshold values are local for the region and may not be appropriate beyond the boundary, however, the thresholds used are low (Cd – 0.5 µg/L; Hg – 1.0 µg/L; Pb, Cr and Ni 50 µg/L; Zn and Cu 0.5 mg/L). The characterization results of the two estimation methods were different for human toxicity (52% higher for the threshold method), fresh water aquatic toxicity (25% lower for the threshold method) and marine aquatic toxicity (13% lower for the threshold method). The threshold method was chosen because of its higher human toxicity impact and because the removal efficiency had not been investigated for this particular water.

The recovery from MSWA was shown to have a greater environmental impact than both conventional production and sludge spreading. However, the developed process is a first laboratory experiment and can be adjusted using the obtained LCA results as guidance. The high impact of the recovery process is due to the following: the large input of ash (inherently very basic) which in turn requires a large volume of acid for pH control, the large volume of waste leachate containing heavy metals, and heavy metal impurities in the final phosphorus product. The consumed acid in particular is a major contributor to abiotic depletion, which is much higher than for other processes. On the other hand, according to the ongoing process development, the HCl requirement could potentially be reduced by four fifths by pre-treatment of the ash with the acidic process waste at the MSW incineration plant. In addition, the $CaCO_3$, NaOH and TMT currently used at the plant for process waste purification and pH adjustment could be saved, offering both economic and environmental advantages. The metal content of both the waste leachate and the final phosphorus product needs to be reduced. Current method development aims to enable metals to be recovered from the ash prior to the phosphorus recovery. This would produce additional resources, reduce leachate and final product metal content to trace levels, as well as reduce the acid requirement during leaching.This modification also allows phosphorus extraction to be carried out in one step and avoids the intermediate Fe-rich precipitate. Consequently, the modified method substitutes clean water and chemicals (HCl) for an existing industrial waste product of suitable properties and one of the process wastes (Fe-precipitate) is also avoided.

Sludge spreading performed better in all categories except for toxicity, where it scored higher than the conventional production but lower than the recovery process. This result coincides with the focus of the debate on whether to use sludge as fertilizer. Due to the nature of the LCA methodology, the results can only be used to compare the different alternatives to each other. Other assessments, e.g. Risk Assessment, should be used to investigate whether sludge spreading on arable land is harmful to humans and ecosystems. We expect, however, that sludge spreading as a phosphorus resource will have a lower overall impact than phosphorus from conventional production in the near future. This is mostly due to the considerable improvement in sludge quality resulting from the implementation of the REWAQ certification and upstream pollution control, but also to the fact that the quality of the mined phosphate is declining, with lower phosphorus content and more impurities.

It is probable that the impact of conventional production on abiotic depletion is actually greater. The awareness of phosphate rock depletion has only increased recently and there is still a large quantity of lower grade phosphate rock that could be used, although the environmental impacts would be higher. These factors are not yet accounted for in the characterization methods.

CONCLUSION

Two recycling opportunities from urban sinks of phosphorus were investigated: spreading of sewage sludge and the novel method for phosphorus recovery from MSW incineration fly ash (MSWA). In one analysis, the recycling options and the conventional production method were compared as three gate-to-gate processes by LCA, and in another, the impact of recycling was compared to the potential benefits of avoiding the conventional production route.

Overall, spreading of sludge has lower impacts than the conventional process, apart from in relation to toxicity, due to its heavy metal content. The benefit of avoiding conventional production is greater than the impacts of sludge recycling in all categories, except toxicity. A reduction in the Hg and Cu content of the sludge would make this an environmentally preferable substitute for the rock-derived phosphorus. It should be noted that the impact of potential organic pollutants could not be assessed.

Compared to the other two options, phosphorus recycling from MSWA reduces only the eutrophication and land-use impacts, while the impacts in the rest of the investigated categories are higher. It should be noted that this method is at a lab bench stage of development and therefore not directly comparable to the conventional phosphate production method, which has been optimized for several decades. Important method development needs were, however, identified and solutions suggested. In particular, the metal content of the final product has to be reduced, and so must the resource requirements. The ongoing method development substituted clean water and HCl for an existing industrial waste product of suitable properties, as this would eliminate its abiotic depletion impact. Moreover, metals were extracted prior to the phosphorus, to produce an additional type of resource, reduce the phosphorus product metal content, and avoid one of the process wastes.

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