

Environmental impacts of phosphorus recovery from municipal wastewater

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ABSTRACT

Phosphorus mining from phosphate rock is associated with economic as well as environmental concerns. Through phosphorus recovery from municipal wastewater, countries could decrease their dependency on the global phosphate rock market, however, conceivably leading to an increase in environmental impacts from fertilizer production. In this work 18 phosphorus recovery technologies are evaluated in terms of cumulative energy demand, global warming potential and acidification potential with the methodology of life cycle analysis. These indicators are then contrasted with other environmental criteria, i.e. recovery potential, heavy metal and organic micropollutant decontamination potential and fertilizer efficiency, to determine their overall environmental performance. The LCA shows that a broad spectrum of changes in gaseous emissions and energy demand can be expected through the implementation of P recovery from wastewater. Linkage to further environmental performance results exposes certain trade-offs for the different technologies. Recovery from the liquid phase has mostly positive or comparably little impacts on emissions and energy demand but the low recovery potential contradicts the demand for efficient recycling rates. For recovery from sewage sludge, those technologies that already are or are close to being applied full-scale, are associated with comparatively high emissions and energy demand. Recovery from sewage sludge ash shows varying results, partly revealing trade-offs between heavy metal decontamination, emissions and energy demand. Nevertheless, recovery from ash is correlated with the highest potential for an efficient recycling of phosphorus. Further research should include implications of local infrastructures and legal frameworks to determine economically and environmentally optimised P recovery and recycling concepts.

1. Introduction

Phosphorus (P), as an essential nutrient for all life, takes on a substantial and non-replaceable role in our environment. Nevertheless, current P use practices are accompanied by various environmental concerns, as mining of P from raw phosphate rock (PR) leads to emissions to the air and eutrophication of water bodies, land degradation through phosphogypsum stacks near the mining site (phosphoric acid production) and soil contamination through cadmium (Cd) and uranium (U) application with fertilizers (FEI, 2000; Silva and Kulay, 2003, 2005; Spiegel et al., 2003; Smidt et al., 2012; Hakkou et al., 2016; Kratz and Schnug, 2016). While these environmental concerns cannot be neglected, it were economic concerns, i.e. the increasing awareness of the concentrated PR-mining in only a handful of countries worldwide, the overall increasing demand for P and the fact that PR is a non-renewable resource, that led the European Commission to declare PR as a critical raw material in 2014 (EC, 2014).

Simultaneously, research, governments and industry recognised the

importance of another, for the major part unexploited P source: municipal wastewater. Municipal wastewater has the potential to substitute a significant portion of the demand for PR (Binder et al., 2009; Egle et al., 2014; Zoboli et al., 2016a,b) and therefore to increase circular economy while simultaneously reducing overall environmental impacts from current P use practices. Intensive research and innovation in recent years has led to the development of a broad spectrum of technologies for phosphorus recovery from wastewater. Their development was accompanied by comparative studies, dealing mainly with the technical and economic assessment of these technologies, in order to identify those that are technically applicable and can be considered market-feasible alternatives to PR-mining and conventional fertilizer production (Cornel and Schaum, 2009; LfU, 2015; Fux et al., 2015; Egle et al., 2015, 2016; Nätörp et al., 2017). However, to provide a more comprehensive picture for legislators dealing with how future P-recycling can be best put into action, knowledge as to how different technologies could impact the environment is an additional prerequisite. Bearing in mind the environmental impacts from

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conventional fertilizer production, such assessments are also indispensable to avoid the replacement of one environmental problem (e.g. land degradation through PR mining) with another (e.g. increased energy demand for P recovery).

Environmental impact assessment related to P recovery from municipal wastewater and sewage sludge treatment and recycling has been covered by some studies in the past. Johansson et al. (2008) studied sewage sludge recycling options, namely use for restoration, composting, hygienisation and agricultural use and also one super-critical oxidation P recovery technology (AcquaReci®). Of special importance for this work is that (i) they found the consideration of environmental savings for PR-derived fertilizer replacement to have large impacts on the results, and (ii) the magnitude of these savings depends on the amount but, more importantly, also on the fertilizer that is assumed to be replaced. Further, the P recovery technology AquaReci® was correlated with environmental savings due to the simultaneous possibility for energy recovery, and due to the stabilisation of the sludge through total destruction of organic material. Major environmental impacts were however derived from the use of magnesium oxide, a first indication that indirect effects through chemical use can be expected to play a major role in this work. Buonocore et al. (2016) also found different circularity patterns of reuse in WWTP (especially co-digestion of sludge with reuse of effluent water for fertirrigation of energy crops) to decrease overall environmental impacts of WWTPs, e.g. effects on climate change, fossil depletion, eutrophication, human toxicity and terrestrial acidification. However, they doubted that the use of eutrophication and human toxicity indicators are appropriate tools to evaluate potential impacts in these categories as local conditions are highly relevant, but cannot be accounted for in the assessment. Lederer and Rechberger (2010) attributed a thermo-chemical treatment process that recovers P from sewage sludge ash (SSA) as more effective in terms of P recycling compared to agricultural sludge application and mono-incineration with soil application of the residual ash. Correlated advantages were organic micropollutant (OM) and heavy metal removal, however, with low additional emissions but higher energy requirements, revealing certain trade-offs between different environmental criteria in the case of P recovery. Compared to a conventional mineral P fertilizer, findings from Linderholm et al. (2012) indicate a higher energy demand and lower greenhouse gas emissions through struvite precipitation of P and higher energy demand and emissions through recovery from SSA with the AshDec® process. They therefore detected large differences in environmental effects from those technologies especially due to varying chemical use and energy requirements.

Nevertheless, none of the presented studies deal with comparing the broad spectrum of currently available P recovery technologies. In this context, the profound study done by Remy and Jossa (2015) inside the EUs P-REX project has played a particularly significant role in the life cycle assessment (LCA) of P recovery technologies and was beneficial for comparison and plausibilisation of the results in this work. Though a similar assessment approach was used by Remy and Jossa as in the here presented work, various modelling choices in their study (e.g. use of sludge from a reference WWTP of 1,000,000 PE, focus on the sludge treatment line with only simplified accounting for altered return loads to the WWTP) do not consider all impacts of material flows changes on the whole wastewater treatment process and do not reflect more medium-scale WWTPs (as mainly present e.g. in Austria) to a satisfying extent. In addition, the here presented work provides the final piece to a comprehensive study on P recovery from wastewater. Previously published work dealt with the quantification of unexploited phosphorus flows (Egle et al., 2014), an overview of the technologies that were assessed (Egle et al., 2015) and an integrated comparative technological, environmental and economic assessment to determine optimal recovery concepts and technologies (Egle et al., 2016). In Egle et al. (2016) a first set of environmental criteria seen as directly relevant to phosphorus recycling and soil conservation (i.e. nutrient, heavy metal and organic micropollutant content, direct heavy metal emissions

particularly to soil, recovery rate and solubility/plant availability) was addressed by using material flow analysis (MFA), the damage unit (DU) method, the reference soil method and knowledge taken from a literature review. This work complements these criteria by using LCA to analyse further impacts (i.e. gaseous emissions and energy demand) that are not in direct relation to the agricultural application of products, but provide additional information for legislators and decision makers on global and national relevant aspects. In this context, this work will refrain from using a fully aggregated impact factor through choosing weighting coefficients for the single impact factors, since (i) only some environmental impacts are calculated via LCA and (ii) it is preferred to rather provide a set of results, where trade-offs between different criteria are fully disclosed, than an incomprehensible absolute value. The same approach was taken by Remy and Jossa (2015), therefore applying the same allows for an enhanced comparison between two studies that can be seen as complementary in each other as they look at this topic from two different contexts and scales.

As P recovery technologies are implemented at, or in succession to WWTPs and can have beneficial or unfavourable impacts on their environmental footprint (e.g. reduction energy demand, de/increase of chemical use), this study aims at analysing these impacts in relation to a defined reference WWTP. In addition, putting into place a successful circular economy concept for P will replace a share of the demand for PR-derived fertilizer and therefore reduce the overall environmental impact of PR-derived fertilizer production, which can be accounted for as credits. Finally, to provide the adequate perspective with P being a major, non-substitutable nutrient for all human life, this work will compare the environmental impacts from P-recycling to our overall impacts per capita.

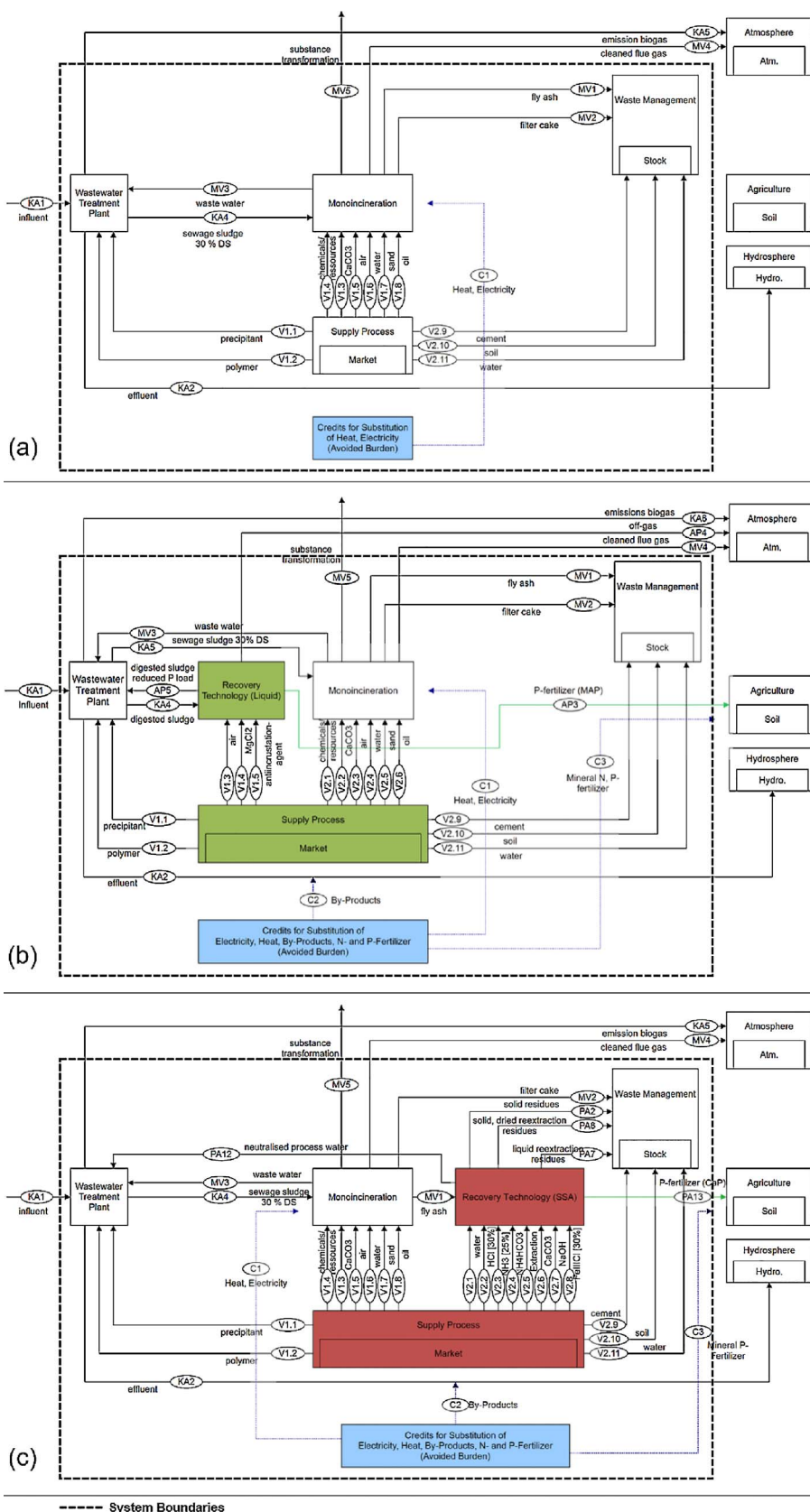
2. Materials and methods

The potential emissions and energy demand brought forth by the different P recovery technologies are analysed through life cycle assessment (LCA; ISO standard 14040, 2006). LCA is a widely used methodology to quantify environmental impacts of a technology, in this case for the recovery of a P material. Through adequate system boundaries, LCA supports the incorporation of all related impacts not only on-site but also preceding and succeeding a technology (e.g. utility production, waste disposal). This enables the comparison of environmental impacts from fairly different technologies and material uses, as is the case in this study. In accordance with ISO 14040, the steps undertaken in this study are defining system boundaries, the functional unit and environmental indicators, setting up a life cycle inventory of related material flows, performing the impact assessment and lastly, interpreting the results.

2.1. System and boundary definition

To analyse additional or reduced environmental impacts of P recovery from wastewater, the status quo is defined first. For this purpose, a typical WWTP with a pollution load of 100,000 population equivalents (PE) (corresponding to a P_{tot} load of 65,700 kg a⁻¹), a mono-incineration plant for sewage sludge and a waste management process for treatment and disposal of occurring wastes are chosen as a reference system (see detailed descriptions of the reference system in previous work by Egle et al. (2015, 2016) and for chosen resource demand in Tables A2–A4). The only adaptation made in comparison to previous work was to change the co-incineration to mono-incineration for the reference system, as to avoid alteration of the results due to the impact of a change in energy yield. This impact should of course not be neglected for optimising national sludge disposal and P recovery concepts when co-incineration is part of the current treatment schemes. Final receiving compartments for this reference system are the processes of waste management, soil/agriculture, the atmosphere and the hydro-sphere, each with a stock function. The system is structured as a

Fig. 1. Exemplary process schemes and system boundaries of (a) the reference WWTP, (b) with implemented P recovery from a process recovering from the liquid phase (AirPrex™) and (c) with a SSA process (PASCH).



modular system with defined reference processes and detailed subprocesses (see Fig. 1(a)).

The reference system then is adapted to account for changes brought forth by the technologies and new processes are added. Direct

effects (increase/decrease of resources and energy demand) of a technology but also indirect environmental effects from production of utilities or from disposal of wastes are considered in the adapted system. Technology construction cannot be accounted for due to a lack of data

Table 1
Selected P recovery technologies from the liquid phase (digester supernatant/dissolved P in anaerobic sludge/effluent), sewage sludge and sewage sludge ash.

Phase	Technology	Technical principle	Product	Technology status
Liquid phase	REM-NUT [®]	Ion exchange, precipitation	CaP	Pilot plant
	Ostara Pearl [®]	Crystallisation	Struvite	Full-scale
	PRISA	Precipitation/crystallisation	Struvite	Pilot plant
	P-RoC	Crystallisation	CaP	Pilot plant
	AirPrex [®]	Precipitation/crystallisation	Struvite	Full-scale
	DHV Crystallactor [®]	Crystallisation	CaP	Full-scale
Sewage sludge (SS)	Gifhorn	Wet-chemical extraction, sulfidic precipitation of interfering ions, precipitation	Struvite	Full-scale
	Stuttgart	Wet-chemical extraction, complexation of interfering ions, precipitation	Struvite	Pilot plant
	MEPHREC [®]	Metallurgic smelt-gassing process	P-rich slag	Large scale pilot plant
	Aqua Reci [®]	Supercritical water oxidation, acidic/alkaline leaching, precipitation	CaP	Pilot plant
	PHOXNAN	Wet-oxidation, precipitation	Struvite	Pilot plant
Sewage sludge ash (SSA)	AshDec [®]	Thermo-chemical, heavy metal depollution	Depolluted ash	Pilot plant
	LEACHPHOS [®]	Acidic wet-chemical leaching	CaP	Pilot plant
	PASCH	Acidic wet-chemical leaching, liquid–liquid removal of heavy metals	CaP	Laboratory scale
	RecoPhos [®]	Acidic wet-chemical extraction	Mineral fertilizer	Industrial scale
	Fertilizer Industry	Acidic wet-chemical extraction	SSP	Industrial scale
	EcoPhos [®]	Acidic wet-chemical leaching, heavy metal removal through ion-exchange	Phosphoric acid	Full-scale
	Thermphos [®]	Thermo-electric	P4	Industrial scale

on resource demand, however, previous studies showed only minor relevance of the construction phase in wastewater treatment (Lundin et al., 2000; Machado et al., 2007).

System boundaries of the reference system and examples for adapted systems under study are given in Fig. 1, showing the aggregated systems for (a) the reference situation, (b) a recovery process from the liquid phase and (c) for recovery from SSA. The system boundaries include the WWTP process, the implementation of the recovery technologies, the mono-incineration of sludge, the supply of chemicals and resources, the final disposal of wastes in the waste management sector, the substitution of energy and resources and transport of sludge, wastes (fly ash, filter cake, technology wastes) and the recovered material to agriculture. Output flows are emissions to the atmosphere, hydrosphere and application of fertilizer on agricultural soils. Out of scope of this study are substance transformations/emissions/leaching in the compartments of atmosphere, hydrosphere and during/after fertilizer application in agriculture.

Production of net energy and resources (i.e. electricity, heat, by-products, P-fertilizer, N-fertilizer) is accounted for by taking the avoided burden approach, assigning energy or emission credits to the studied systems for substitution of these products. For further details how credits are calculated see Section 2.6.

2.2. Functional unit

In regards to the reference system, the extent of additional or reduced environmental impacts at the WWTP will depend largely on the treated amounts of wastewater. Emissions and energy demand of P-recycling are therefore related to the treatment of 1 population equivalent and year (1 PE⁻¹ a⁻¹). Population equivalent is a common indicator to describe the organic pollution load of a WWTP and in general has a good correlation with P loads. Through a conversion of PE to inhabitants by the factor of 0.5, the environmental impacts of the recovery technologies are comparable to total annual emissions and energy demand per inhabitant in Austria.

As the used quantities of the different recovered materials will predominantly be determined by their P content, technologies will be further compared based on a functional unit of 1 kg of recovered P (1 kg P_{rec}⁻¹).

2.3. Environmental indicators

The environmental indicators considered in this work are cumulative energy demand (CED), global warming potential (GWP) and

acidification potential (AP).

CED (total, expressed as kWh) is an indicator to determine the energy requirements during the entire life cycle of a product (VDI, 2012). The CED is calculated by taking into account the direct energy demand of the recovery technologies (e.g. electricity, gas) and the indirect energy demand needed to produce the necessary resources for the technology.

GWP (gaseous emissions of CO₂, CH₄ and N₂O, expressed as CO₂ equivalents (CO₂e) is chosen due to the fact that significant differences in CO₂e between the considered P recovery technologies are expected, as they differ in type and amount of used chemicals and in their use of primary fuels for combustion. In addition, emissions from WWTPs have been discussed largely in recent years (Lorenzo-Toja et al., 2016; Ahn et al., 2010; Foley et al., 2010) and the relevance of a further P recovery step therefore should be analysed. GWP was calculated with emission characterisation factors of IPCC 2007 (Solomon et al., 2007).

AP (gaseous emissions of SO₂, NO_x, HCl and NH₃ expressed as SO₂ equivalents (SO₂e) has direct impacts on soil acidity and is therefore indirectly linked to agriculture and P fertilizers. It is chosen as some technologies use large amounts of chemicals, which through their production evoke high emissions (Egle et al., 2016) and thus might cause relevant differences. AP is calculated by the problem oriented approach baseline CML 2001, including fate perspective and for average European total values (Huijbregts, 1999; de Bruijn et al., 2001). GWP and AP are calculated by summing up direct and indirect gaseous emissions. As an example, the incineration of heating oil causes direct emissions, while indirect emissions derive from the production of heating oil.

2.4. Data

A total of 18 technologies have been selected, aiming at a high coverage of the principal technical approaches applicable for P recovery (Table 1). To facilitate comparison, the chosen technologies are grouped by their point of application: liquid phase, sewage sludge or SSA. Ideally, technologies have been chosen due to a high level of development and the availability of a thorough data basis for analysis. However, processes only implemented at pilot-scale and in one case lab-scale (PASCH) have been selected too, to include technical approaches that would not have been considered otherwise. A thorough description of the technologies is given in Egle et al. (2015). It is stressed by the authors that the field of phosphorus recovery remains a highly dynamic field with few full-scale references at different sites, rarely tested under varying conditions (e.g. sludge composition). The presented results

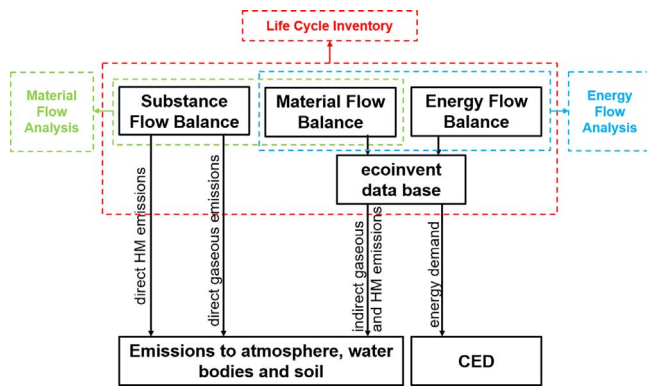


Fig. 2. Approach to establish the life cycle inventory.

therefore capture only a snapshot in time, conveying the current developments and providing an overview of the principles and the potential magnitude of their ecological consequences. Data gathered in Egle et al. (2014, 2015, 2016) and Remy and Jossa (2015) are used as a basis for the environmental impact assessment (see full disclosure of references and uncertainties in Table A6). The chosen values for energy and resource demand as well as material outputs of the technologies can be found in Tables A7–A9.

2.5. Life cycle inventory (LCI)

The LCI is developed by combining different methodologies as demonstrated in Fig. 2. The methodology of material flow analysis (MFA; Brunner and Rechberger, 2016) is used to set up the material and substance flow balances. Input of raw materials and chemicals as well as output of P rich materials, waste by-products and other direct gaseous emissions to environmental compartments are correlated in the respective MFA models. CO₂ emissions from the treatment of sewage sludge are not taken into account as carbon in sewage sludge is generally considered being of biogenic origin, thus CO₂ emissions treated as climate neutral. Nevertheless, it needs to be addressed that recent research has indicated that a non negligible part of organic carbon in wastewater is of fossil origin (7–14% of total organic carbon, Law et al., 2013; 25% of DOC, Griffith et al., 2009), therefore impacts on GWP from e.g. sewage sludge incineration are likely underestimated.

As mentioned before, the results and evaluation of direct heavy metal emissions were previously published by Egle et al. (2016). Parts of the results are recalled in the discussion to provide a final overview of the environmental aspects assessed throughout this work.

External energy sources and also the energy content which is included in materials as e.g. sewage sludge are quantified by the methodology of energy flow analysis (EFA; Suh, 2005). For processes that enable the recovery of energy from sewage sludge incineration, syngas, biogas, etc. these amounts are positively credited or if incineration is averted considered as energy losses. Transport of sewage sludge, ash or residues is further considered in the energy flow analysis (assumptions taken for the transport process see supplementary data Tables A3 and A4).

Indirect heavy metal and gaseous emissions and indirect energy demand of the resources are calculated by taking their mass or energy flow as determined with MFA/EFA and correlating it to emissions and energy demand per mass or energy unit as defined in the life cycle data base ecoinvent (Wernet et al., 2016) version 3.3. ecoinvent considers all emissions and the whole energy demand of a process chain to generate a material up to e.g. its placement on the market. The chosen reference processes from the ecoinvent database for the different resources and energy carriers are given in Table A12. Results of indirect heavy metal emissions from the production of resources are not specifically addressed in this work since they are, for the most part, insignificant in comparison to both direct and indirect heavy metal emissions originating from wastewater treatment and the mono-incineration of the sludge.

2.6. Impacts on the reference system and credits for production of resources and energy

For the comparison of environmental impacts of P recovery in relation to the reference system, impacts on the wastewater treatment and sludge disposal are accounted for in the form of a reduction or increase of the resources (chemicals) and energy needed for the treatment process, indirectly resulting in a change of CED, GWP and AP. Further, credits of CO₂e, SO₂e and kWh are granted for the substitution of resources and energy that are produced by the different technologies. A summary of the changes and the credits that are accounted for is given in Table 2.

Effects from the reduction of nutrient (N, P) return loads in the WWTPs excess water after dewatering of the sludge are twofold. For the reduction of P, a reduced demand for flocculating agents for P-removal can be assumed. For the reduction of N, less oxygen demand and hence aeration demand in the biological treatment steps for N nitrification/removal is needed (calculation factors see Table A10). Changes on sludge volume/mass considered in this study are calculated based on mass balances (reduction dry mass from flocculating agent input, changes in organic carbon content due to reduction anaerobic

Table 2
Accounted effects for impacts on WWTP process and credited products for resource production.

Impact on the reference system	Accounted effect
Reduction P-backflow	Reduction flocculating agent demand
Reduction N-backflow	Reduction energy demand for aeration
Increase/reduction anorganic sludge dry mass	Increase/reduction sludge transport volumes
Increase/reduction organic sludge dry mass	Increase/reduction in sludge transport volumes, increase/reduction resource demand for and of energy gains from mono-incineration
Increase/reduction sludge dewaterability	Increase/reduction in sludge transport volumes, increase/reduction resource demand for and of energy gains from mono-incineration
Credited (by-)products	Chosen reference product from ecoinvent (Wernet et al., 2016) version 3.3
Phosphorus fertilizer	Market for phosphate fertiliser, as P2O5 [GLO]
Phosphoric acid	Market for phosphoric acid, fertiliser grade, without water, in 70% solution state [GLO]
White phosphorus (P ₄)	Market for phosphorus, white, liquid [GLO]
CaCl	Calcium chloride to generic market for de-icer [GLO]
FeCl	Iron (III) chloride, without water, in 40% solution state [CH]
Electricity	Market for electricity, high voltage [AT]
Heat	Market for heat, district or industrial, natural gas [Europe without Switzerland]

digestion) or based on data from technology providers (e.g. increased dewaterability of sludge in AirPrex[®] process). The considered effects are changes in transport volumes and resource demand and energy gains in mono-incineration. Compiled changes from the technologies are given in Table A11.

Credits for (by-)products from the processes are considered by the 'avoided burden approach', assigning a 100% replacement of virgin materials with recycled products (equal exchange of products based reference product and unit, e.g. 1 kg of P in mineral fertilizer with 1 kg of P_{rec}). It is pointed out by Johansson et al. (2008) that credits for P-fertilizer replacement depend largely on the type of fertilizer that is assumed to be replaced. However, in the case of P recovery from wastewater, the materials produced vary highly, and it is not clear which type of fertilizer is going to be replaced by them. Therefore, in this work credits are assumed from theecoinvent process 'market for phosphate fertiliser' which is a reference process representing a consumption mixture of different phosphorus fertilizers (with SSP production accounting for around half of the mass mixture). This is with the exemption of the EcoPhos[®] and ThermPhos[®] technologies, where credits are assigned for the replacement of phosphoric acid and white elementary phosphorus respectively.

2.7. Uncertainty management

The quality of data used in this study differs strongly. Data gaps of certain technologies are e.g. addressed by using knowledge on basic chemical principles or data from other similar technologies, thereby introducing a higher uncertainty for those technologies. Further, the determination of additional benefits up- and downstream of the technologies' implementation point can be challenging at times. In some cases this results in higher uncertainties concerning the extent of those benefits. To tackle these uncertainties, a qualitative approach is used to determine the uncertainty of the data depending on the credibility of the source (see Egle et al., 2016). A description of the chosen uncertainty characterisation of the data sources is given in Table A5. Assigned uncertainties for both the overall data basis as well as for each material or energy flow are given in Tables A6–A9.

One further drawback of this assessment is represented by the still missing evaluation or limited plant availability of some recovered P-materials (see Egle et al., 2016). Although it is deemed possible that further processing of the materials will be necessary before their use in agriculture, these are yet unknown and therefore cannot be accounted for.

As a consequence of the uncertainty management, results are discussed in regard to the reliability of the applied data and the uncertainty of systemic effects.

3. Results

In the following section results for the different environmental indicators are presented and individual contributions of different aspects and processes involved in the recovery are shown in detail. Further, net values, which considering both negative as well as positive impacts in comparison to the reference system, are given for each technology.

3.1. Cumulative energy demand

CED of the reference system amounts to 20 kWh PE⁻¹ a⁻¹ (Fig. 3). Changes in CED due to implementation of P recovery vary highly between the technologies. Most technologies that recover from the liquid phase via precipitation show a decrease in CED due to a low energy and resource demand and benefits through P (all) and N (REM-NUT[®], Ostara Pearl[®], AirPrex[®]) substitution, as well as beneficial impacts on the WWTP. These technologies can mainly be implemented at WWTPs with P removal by luxury uptake (BioP), where P backflow after anaerobic digestion and dewatering of the sludge is considerably higher than at

WWTPs with chemical P-removal. Therefore, removing the excess P from the WWTP will evoke positive benefits of reduced need for flocculating agents and further, when N is removed as well, a reduction in the aeration demand needed for N removal can be achieved. One exception is the crystallisation process DHV Crystallactor[®], for which CED is increased compared to the reference system as the anaerobic digestion of the reference system has to be replaced with aerobic treatment, therefore reducing the energy production through biogas valorisation at the WWTP (accounted for as energy losses).

All technologies recovering from sewage sludge show a considerable increase in CED compared to the reference system. The wet-chemical extraction processes Stuttgart and Gifhorn have a high need for chemicals which are partly energy-intensive in their production too (citric acid, sodium hydroxide and sulphuric acid). The metallurgic Mephrec[®] process gains credits for the substitution of energy as well as a reduced transport demand for sludge, since sludge is mineralised on-site and the need for mono-incineration ceases to exist. Nevertheless, the additional demand for resources (coke, dolomite and oxygen) offsets those credits. The super-critical-wet-oxidation process AquaReCi[®] is attested with the highest CED despite the fact that comparatively high credits can be obtained through the autothermic production and valorisation of heat. The demand for gas, electricity and liquid oxygen is attributed with higher CED than can be credited for by resource substitution. In comparison, the CED of the wet-oxidation process PHOXNAN is mainly dominated by its demand for oxygen, whereas substitution of energy cannot offset the demand.

Other than for sewage sludge, technologies that recover P from SSA show only a medium increase or even decrease in CED compared to the reference system. Credits for P fertilizer replacement are high for all SSA processes as recovery rates are generally above ~80% of the WWTPs inlet. Both thermo-chemical AshDec[®] processes, the acidic wet-chemical leaching process LEACHPHOS[®] and the direct integration into the fertilizer industry with wet-chemical extraction show only a low change in CED as the additional demand for chemicals and energy is offset by the P fertilizer credits. PASCH (wet-chemical leaching) has a high demand for the chemicals citric acid and ammonia, leading to an increase in CED. In comparison, RecoPhos[®] (wet-chemical extraction) uses high amounts of chemicals too (phosphoric acid), however, the P contained in the phosphoric acid stays in the product and can therefore be credited with P fertilizer substitution as well. The EcoPhos[®] (phosphoric acid production, wet-chemical leaching) can save considerable amounts of energy, however, only if the additional by-products of the process CaCl₂ (de-icing agent) and FeCl₃ can be valorised. For the thermo-electric ThermPhos[®] process CED is lower compared to the reference system, as the credits given for P₄ production offsets the electricity and chemical demand (mainly for clay) for the process with SSA use.

3.2. Global warming potential

GWP of the reference system amounts to 9 kg CO₂e PE⁻¹ a⁻¹ (Fig. 4). The ranking of the technologies for GWP is for the most part similar to the changes in CED. With the exception of P-RoC and the DHV Crystallactor[®], all processes that recover from the liquid phase fare better in terms of total GWP than the reference system. The DHV Crystallactor[®] is attributed with higher emissions due to the increase of sludge volumes to the mono-incineration plant and therefore higher emissions from incineration. Also, resource use in upstream processes of the WWTP (e.g. acetic acid needed as a feeding agent for bacteria to force a release of the former biologically bound P), but also of the use of lime as a precipitant instead of magnesium chloride increase its impact further.

Both Gifhorn and Stuttgart will increase GWP through high CO₂e emissions from the production of the used chemicals. The processes that take care of sludge mineralisation while replacing the need for mono-incineration of the sludge (MEPHREC[®], AquaReCi[®], PHOXNAN) would

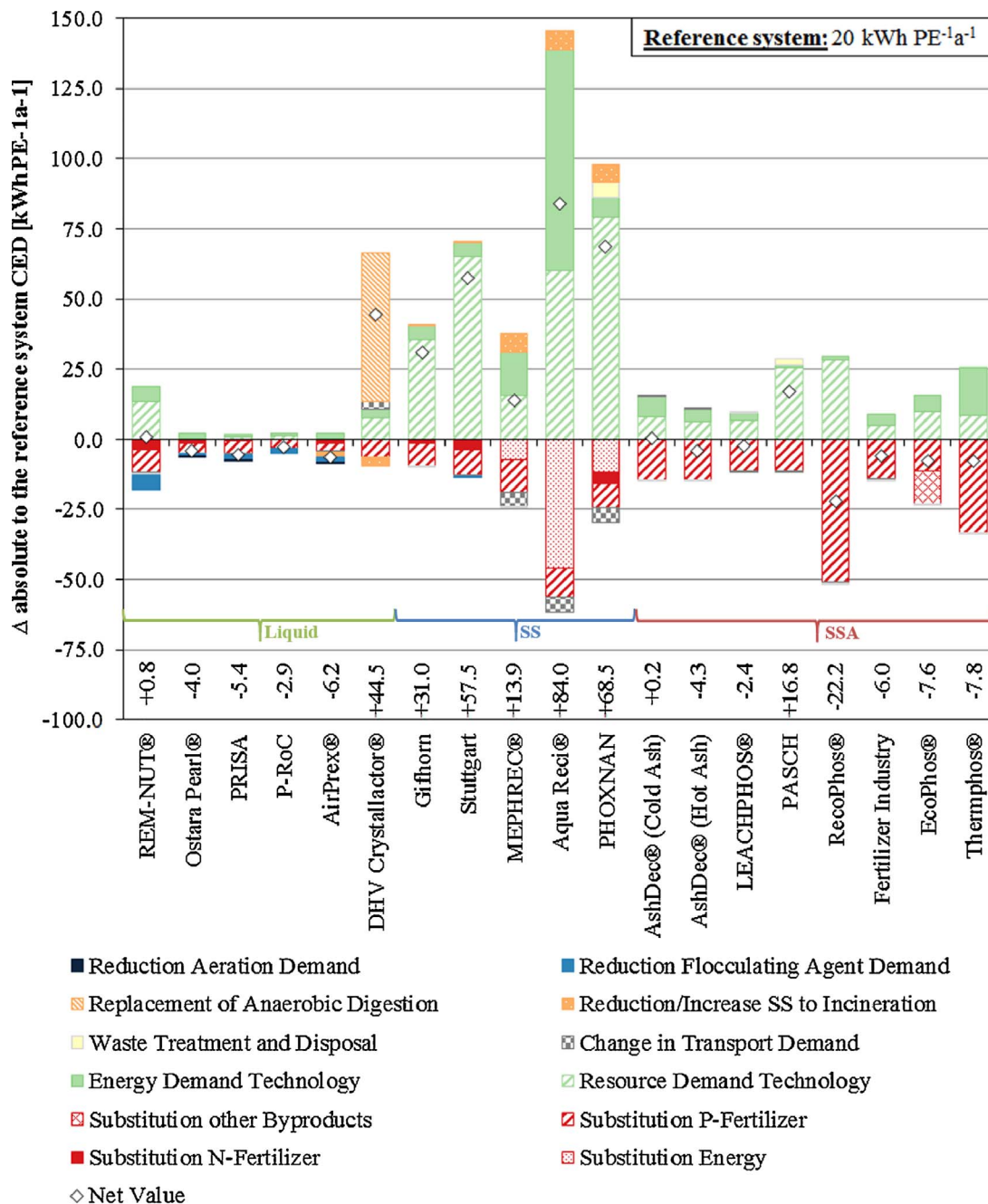


Fig. 3. Relative changes in cumulative energy demand (kWh) in relation to the reference system.

contribute to an increase in GWP due to their high demand for resources and/or fossil energy in the form of coke (MEPHREC®) or natural gas (AquaReci®).

SSA de-pollution with AshDec® results in lower GWP due to fertilizer credits and the operation with hot ash fares slightly better due to a reduced energy demand for heating. Emissions of LeachPhos® stem mainly from the use of sulphuric acid in the process, however, considering the credits for P substitution, the implementation of the technology would evoke an equal GWP as the reference process. As with CED, PASCH increases GWP due to its chemical use. For RecoPhos®, again, phosphoric acid use is correlated with an increase in GWP, however, the credits from the additional P in combination with the P from the SSA offsets this increase. With the direct integration of the SSA into the fertilizer industry, or through recovery with EcoPhos® or ThermPhos® overall GWP can be improved.

3.3. Acidification potential

AP of the reference system amounts to 42 g SO₂e PE⁻¹ a⁻¹ (Fig. 5). Like with GWP, the general ranking of the technologies is quite similar as for CED. With recovery from the liquid phase, an improvement of the AP by ~20% can be achieved compared to the reference system. Again, this is with the exception of the DHV Crystallactor® for which emissions from mono-incineration are increased through an increase in sludge production. All technologies that recover from sewage sludge, except MEPHREC®, are attributed with a considerable increase in AP by ~80 to ~170% compared to the reference system. In most cases this is due to resource use in the processes (i.e. mostly sulphuric acid and lye for Gifhorn, sulphuric acid, lye and citric acid for Stuttgart, oxygen for AquaReci® and sulphuric acid and oxygen for PHOXNAN). For MEPHREC® and AquaReci® an additional increase in AP results from direct emissions of the technologies. SO₂e emissions from most of the

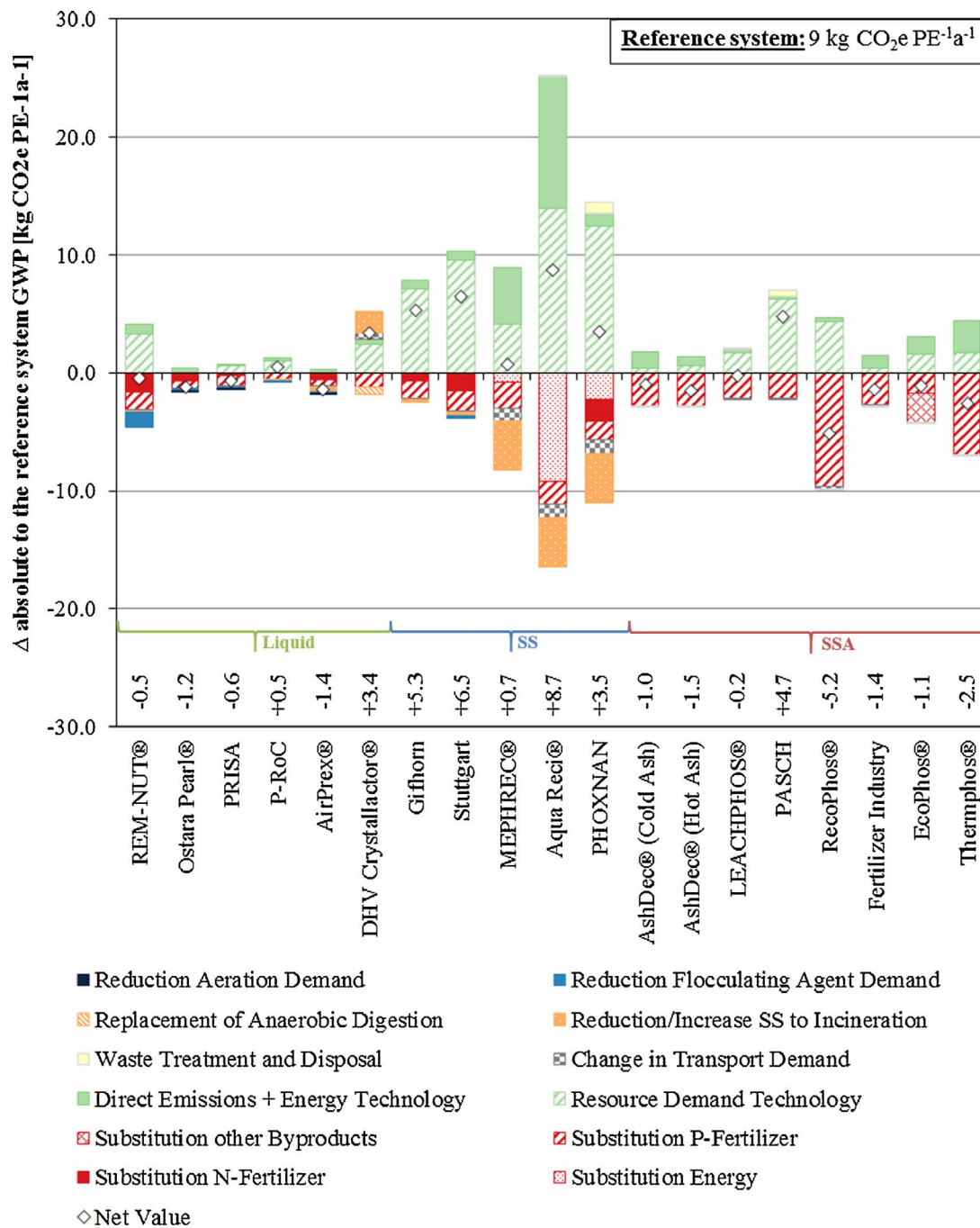


Fig. 4. Relative changes in global warming potential (CO₂e) in relation to the reference system.

technologies that recover from SSA are low and high credits can be attributed due to the substitution of mineral P fertilizer, resulting in a negative net AP for AshDec®, LEACHPHOS®, the direct integration into the fertilizer industry, RecoPhos®, as well as for EcoPhos® and ThermPhos®.

3.4. Energy demand, emissions and recovery rate

Fig. 6 shows the changes of CED, GWP and AP in relation to the P recovery rate of the different technologies. As seen in the results before, technologies that recover from the liquid phase in general reduce CED, GWP and AP, however, with only a low potential for actual recovery of P. Technologies that recover from SSA on the other hand predominantly show a similar beneficial reduction for the studied indicators combined

with a comparatively high potential for P recovery of 70–90% of P at the WWTP's inlet. If recovery is applied to sewage sludge, around 40% – max. 70% of P can be recovered with the given technologies, however, at the cost of increases in CED, GWP and AP.

3.5. Relevance of emissions

To assess the total relevance of environmental impacts from P recovery, results are further compared to mean emissions and energy demand of an Austrian inhabitant (Table 3). Annual Austrian emissions with GWP and AP conform to 10,000 kg CO₂e inh⁻¹ a⁻¹ (UBA, 2013) and 30,100 g SO₂e inh⁻¹ a⁻¹ (UBA, 2015). Corresponding emissions of the reference system are 9 kg CO₂e inh⁻¹ a⁻¹ and 42 g SO₂e inh⁻¹ a⁻¹, amounting to a share of total emissions of 0.19%

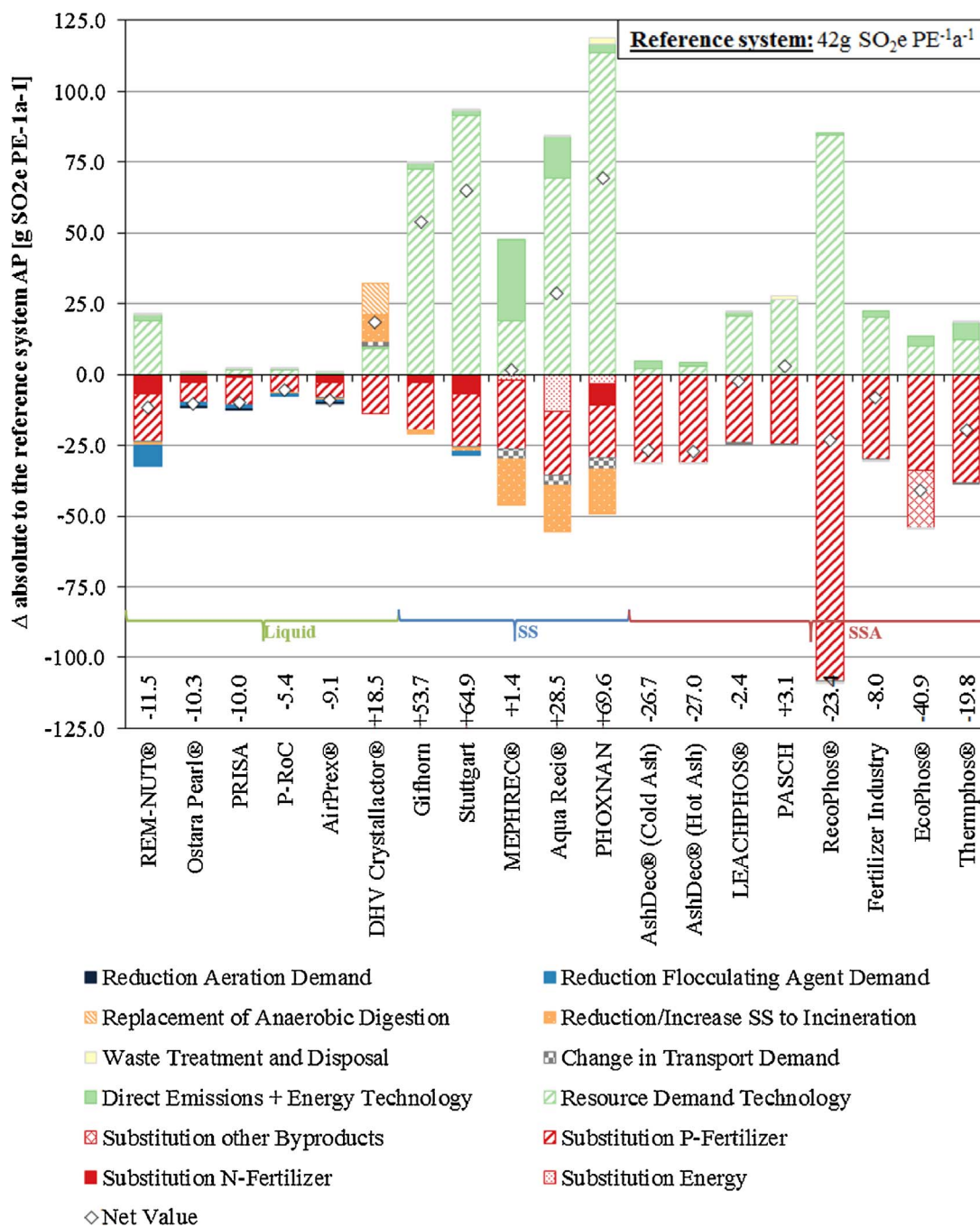


Fig. 5. Relative changes in acidification potential (SO₂e) in relation to the reference system.

and 0.27%, respectively.

To assess the relevance of CED, total energy demand per Austrian inhabitant is calculated from gross inland consumption of energy in kg of oil equivalents (EUROSTAT, 2015). In 2013, per capita consumption was 3990 kg oil equivalents or 46,300 kWh inh⁻¹ a⁻¹. Corresponding CED of the reference WWTP is 20 kWh inh⁻¹ a⁻¹ amounting to a share of total demand of 0.09%. It needs to be added that while the reference system considers mono-incineration of sewage sludge, currently only around half of the Austrian sewage sludge is incinerated (Überreiter et al., 2016) while none is mono-incinerated, and therefore actual share of the conventional wastewater treatment is likely to be different.

On basis of the reference system, implementing P-recovery at or in succession to WWTPs corresponds to changes of total Austrian values in the range of -0.1% to +0.4% for CED, -0.1% to +0.2% for GWP and -0.3% to +0.5% for AP. For most of the technologies savings can be

acquired for at least one (REM-NUT®, P-RoC®, AshDec® Cold Ash) or all of the shown impact categories (Ostara Pearl®, PRISA, AirPrex®, AshDec® Hot Ash, LEACHPHOS®, RecoPhos®, direct integration fertilizer industry, EcoPhos®, Thermphos®). On the other hand, impact of single technologies on CED, GWP and AP is considerably higher than others, as is the case for DHV Crystallactor®, Gifhorn, Stuttgart, AquaReci® and PHOXNAN.

4. Discussion

4.1. Uncertainty and limitations

Even though MFA and EFA are methodologies that enable a robust and consistent quantification of life cycle inventory data, certain limitations arise at multiple levels of this study. For some of the considered

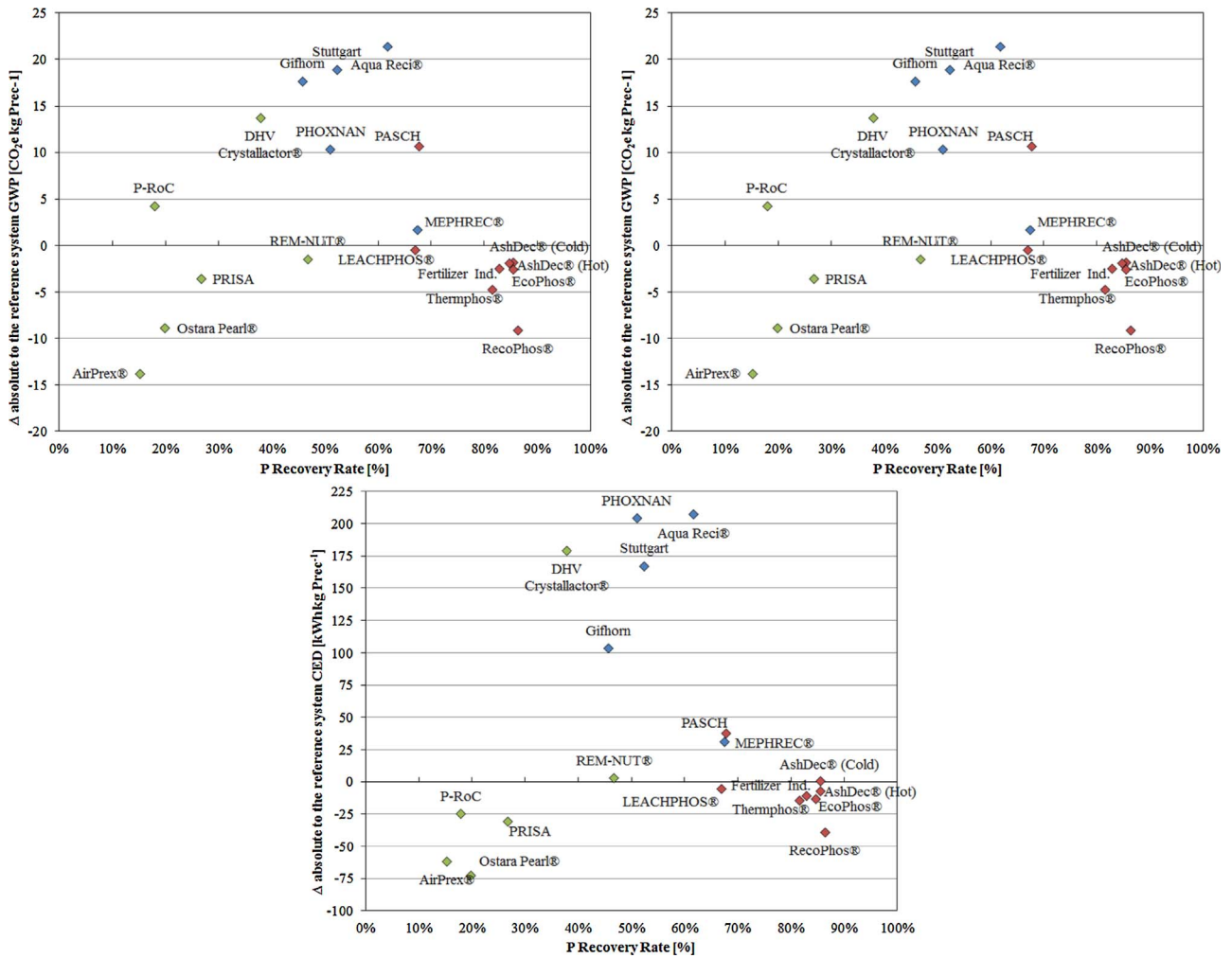


Fig. 6. Relative changes in cumulative energy demand (kWh), global warming potential (CO₂e) and acidification potential (SO₂e) in relation to the potential for phosphorus recovery (in %).

Table 3

Relative changes of energy demand and CO₂e/SO₂e emissions through implementation of P recovery per Austrian inhabitant and year.

Technology	Change of CED [%]	Change of GWP [%]	Change of AP [%]
REM-NUT [®]	+0.003	-0.009	-0.074
Ostara Pearl [®]	-0.017	-0.023	-0.067
PRISA	-0.023	-0.013	-0.064
P-RoC [®]	-0.013	+0.010	-0.035
AirPrex	-0.027	-0.028	-0.058
DHV Crystalactor [®]	+0.192	+0.068	+0.119
Gifhorn process	+0.134	+0.106	+0.346
Stuttgart process	+0.249	+0.130	+0.417
MEPHREC [®]	+0.060	+0.015	+0.009
AquaReCi [®]	+0.363	+0.173	+0.183
PHOXNAN	+0.296	+0.069	+0.448
AshDec [®] Cold Ash	+0.001	-0.021	-0.171
AshDec [®] Hot Ash	-0.018	-0.029	-0.174
LEACHPHOS [®]	-0.010	-0.005	-0.016
PASCH	+0.073	+0.095	+0.020
RecoPhos [®]	-0.096	-0.104	-0.150
Fertilizer Industry	-0.026	-0.027	-0.052
EcoPhos [®]	-0.033	-0.022	-0.263
Thermphos [®]	-0.034	-0.051	-0.127
MIN	-0.096	-0.104	-0.263
MAX	+0.363	+0.173	+0.448
Reference values	10,000 [kg CO ₂ e inh ⁻¹ a ⁻¹]	31,100 [g SO ₂ e inh ⁻¹ a ⁻¹]	46,300 [kWh inh ⁻¹ a ⁻¹]

Table 4
Overview of selected results of the environmental assessment.

Technology	Recovered material	Recovery potential in % of WWTP-influent	Decontamination potential		Relative fertilizer efficiency		Environmental indicators			
			Heavy metals	OM	Acidic soil	Alkaline soil	Damage unit	CED	GWP	AP
REM-NUT [®]	MAP	45–60%	++	(+)	100%	75%	0.010	4%	–5%	–27%
Ostara Pearl [®]	MAP	10–max. 25%	/	(+)	100%	75%	0.011	–20%	–13%	–24%
PRISA	MAP	10–max. 25%	/	(+)	100%	75%	0.072	–27%	–7%	–24%
P-RoC [®]	CaP/MAP	10–max. 25%	/	(+)	100%	75%	0.007	–15%	5%	–13%
AirPrex [®]	MAP	10–max. 25%	/	(+)	100%	75%	0.015	–31%	–15%	–21%
DHV Crystalactor [®]	CaP	10–max. 25%	/	(+)	75%	50%	0.018	225%	37%	44%
Sewage sludge	Sludge	90%		(-)	50–90%	70%	0.499	n.a	n.a	n.a
Gifhorn process	MAP/CaP/ FeP	35–55%	++	(o)	100%	75%	0.004	156%	57%	127%
Stuttgart process	MAP/CaP/ FeP	35–55%	++	(o)	100%	75%	0.033	290%	70%	154%
MEPHREC [®]	P-rich slag	~70%	+	(+)	0%	75%	0.105	70%	8%	3%
AquaReci [®]	CaP/FeP	~60%	++	(+)	75%	50%	0.016	423%	93%	67%
PHOXNAN	MAP	~40–50%	++	(o)	100%	75%	0.004	345%	37%	165%
Sewage sludge ash	Ash	87%		(-)	25–50%	20%	0.352	n.a	n.a	n.a
AshDec [®] Cold Ash	Depolluted ash	~85%	+/-	(+)	90%	0%	0.052	1%	–11%	–63%
AshDec [®] Hot Ash	Depolluted ash	~85%	+/-	(+)	90%	0%	0.052	–21%	–16%	–64%
PASCH	CaP	~60–70%	++	(+)	75%	50%	0.025	85%	51%	7%
LEACHPHOS [®]	CaP	~60–70%	+/-	(+)	100%	75%	0.131	–12%	–2%	–6%
RecoPhos [®]	Mineral fertilizer	~85%	-	(+)	100%	100%	0.158	–112%	–56%	–55%
Fertilizer industry	Mineral fertilizer	~85%	-	(+)	100%	75%	0.352	–31%	–15%	–19%
EcoPhos [®]	Phosphoric acid	~85%	++	(+)	100%	100%	0.002	–38%	–12%	–97%
Thermphos [®]	P ₄	~85%	++	(+)	-	-	-	–39%	–27%	–47%
Single super phosphate	Mineral fertilizer	-	-	-	100%	100%	0.225	-	-	-

n.a = not analysed.

Organic micropollutants

(+) no organic micropollutants; (o) organic micropollutants significantly reduced; (-) no reduction of organic micropollutants.

Heavy metals

++ higher than 80% for all HM; + higher than 40% for all HM; +/- inconsistent reduction depending on HM; - no reduction; / no reduction needed.

technologies, overall uncertainty on the input data is rated as medium to high (REM-NUT[®], DHV Crystalactor[®], AquaReci[®], MEPHREC[®], EcoPhos[®] and Thermphos[®]; see Table 4) and most of them have not been implemented and tested in full-scale yet (see Section 2.4). Therefore, a final full-scale quantification of energy and resource demand is still needed for most of the technologies. Furthermore, not all of the technologies produce ready-to-market products (see Egle et al., 2016) and additional steps might be necessary (e.g. leaching, granulating, mixing with other nutrients), potentially leading to higher emissions and energy demand.

This study reveals that highest increases in CED, GWP and AP are predominantly correlated with an excessive use of resources and chemicals and only in some cases with a high energy demand. On the other hand, decreases found for those indicators can mainly be attributed to the substitution of P fertilizers and of energy. Uncertainty in background data for these categories therefore has the potential to alter the results substantially. This is of special relevance since data on environmental impacts of fertilizer production stem from rather old data (Nemecek and Kägi (2007) based on Davis and Haglund (1999) in turn based on EFMA (1995), Patyk and Reinhardt (1997) and Kongshaug (1998)). Phosphate rock mining has seen increases in efficiency (Steiner et al., 2015) and the reference process ‘market for phosphate fertiliser’ chosen from theecoinvent database is a reference consumption mix of a variety of P fertilizers (with SSP share around 50%), not necessarily reflecting (i) the Austrian consumption mix and (ii) the actual products that could be replaced by the recovered P materials (e.g. a small fraction of organic P fertilisers is included in the consumption mix as well). For the technologies EcoPhos[®] and Thermphos[®] this was accounted for and product credits were replaced with values for phosphoric acid and P₄ markets, respectively.

In addition, some chemicals used for the technologies or in WWTPs are not part of the LCA databases inventory, hence similar chemicals had to be chosen for the assessment, potentially with different but unknown ecological backpacks. Other chemicals (e.g., in some cases, iron chloride precipitants in the WWTP process) are by-products/wastes

from industrial processes. LCA databases include only raw materials which are specially produced to fulfil a purpose, therefore ecological backpacks from by-products/wastes could be lower.

Lastly, results of this work also reflect certain limitations that arise through the need to compare technologies based on a reference system. While this cannot be avoided when doing a holistic but generalised assessment, it has to be kept in mind that results will differ given the preconditions present in site specific cases (e.g. possibility to use excess heat, use of by-products from other processes, current sludge treatment and disposal routes). Especially in the case of wastewater treatment, where treatment plant schemes (aerobic vs. anaerobic) and sludge disposal routes (agriculture, composting, incineration) vary significantly, this poses a drawback of this evaluation.

4.2. Trade-offs in environmental impacts of the technologies

To fully assess the environmental performance of the different technologies, the environmental criteria analysed and previously published by Egle et al. (2016) are included for a final evaluation. Therefore an overview of selected results is given in Table 4 to establish trade-offs between different environmental performance indicators.

While most technologies that recover P from the liquid phase show a reduction or comparably low additional emissions and energy demand, and also provide highly plant available products with a low heavy metal and organic micropollutant content, their implementation is restricted to WWTPs with enhanced biological phosphorus removal (EBPR) and their recovery potential is limited to a maximum of 25% of the P in the WWTP's inlet. The DHC Crystalactor[®], admittedly with a high uncertainty, stands out negatively amongst those technologies, as fertilizer efficiency of CaP is less than that of MAP products and GWP, AP and CED are considerably higher (partly due to the unfavourable comparison to the reference system with anaerobic instead of aerobic stabilisation).

Gifhorn and Stuttgart both yield highly plant available products with a low pollutant content, but this hardly offsets the fact that their

recovery potential lies in the medium range and that they have amongst the highest GWP, AP and CED due to their intense chemical use. MEPHREC[®] shows the highest recovery rate of the sewage sludge technologies (~70%), but this is combined with a low plant availability of the P slag, especially on acidic soil. Values for CED, GWP and AP are also the lowest amongst the technologies that recover from sludge, nevertheless the currently ongoing trials in a large scale pilot plant will reveal if this optimised energy exploitation from syngas can be achieved and how the technology fares in terms of direct emissions to the atmosphere. For AquaReci[®], pollutant removal is high but the high CED and uncertainty regarding direct emissions and no full-scale experiences, as well as the medium recovery rate of around 60%, are major drawbacks of the technology. In addition, there has been no further innovation for this process in recent years. The evaluation of PHOXNAN states that CED, GWP and AP would be higher than those of the reference system, also with only medium recovery rates in the range of 40–50%. Even though general uncertainty of the data is low, just like AquaReci[®], PHOXNAN has seen no improvement in the last years and there is no full scale implementation to draw verified results from.

All technologies recovering from ash have high recovery rates up to ~90% of the WWTP influent, as well as no organic micropollutants in the products, as they are destroyed during sludge incineration. Both AshDec[®] decontamination approaches show a decrease in GWP and AP and steady or decreasing values for CED compared to the reference system. This is combined with a good removal of most heavy metals, however, the product shows no fertilizer efficiency on alkaline soil. Through LEACHPHOS[®] small gains can be achieved for all impact categories, however, at the expense of heavy metal removal. PASCH performs poor in comparison, since it is correlated with higher gaseous emissions and energy demand, as well as a low fertilizer efficiency, but heavy metal removal is improved due to an additional step of solvent-extraction. The lack of heavy metal removal makes RecoPhos[®] questionable in terms of soil emissions, nevertheless, due to the combination of recovered P and phosphoric acid, high credits for fertilizer replacement can be achieved and the product shows the same fertilizer efficiency as SSP. The direct integration into the fertilizer industry would lead to a reduction in GWP, AP and CED compared to the reference system, and high amounts of P could be recovered with a high plant availability. Nevertheless, as with RecoPhos[®], no heavy metals are removed in this process. Through the utilisation of ion exchangers, EcoPhos[®] can achieve a good heavy metal removal and the phosphoric acid can further be used in the fertilizer industry, all combined with a theoretic reduction of CED, GWP and AP, if a by-product valorisation can be realised. Thermphos[®] cannot be compared with the other processes on the basis of fertilizer efficiency and emissions to soils as its product P₄ is of high purity and used only in technical applications. Efforts in terms of energy demand and thus also emissions are however, lower in comparison to traditional P₄ production, as PR mining and its correlated resource and energy demand is substituted by use of SSA.

5. Conclusions

This work analysed environmental impacts from P recovery technologies to complement the integrated and comparative assessment by Egle et al. (2015, 2016) on P recovery from wastewater. Analysis showed that a broad spectrum of changes in cumulative energy demand, global warming potential and acidification potential can be expected through the implementation of P recovery from wastewater. Compared to the chosen reference system (WWTP with mono-incineration of sludge as well as disposal of all wastes) de- and increases vary between –110 to +420% for CED, –60 to +90% for GWP and –100 to +170% for AP, reflecting the great differences in technological principles applied. General trends, but no absolute distinction in results regarding the point of implementation (liquid, sewage sludge or SSA) could be observed, as the studied environmental impacts were found to also highly depend on specific technology parameters, e.g. the

used chemicals. Nevertheless, impacts with recovery from the liquid phase or SSA are for the most part lower than from sewage sludge.

Qualitative uncertainty assessment showed that even though a sound methodology was applied, reliability of results might vary depending on data availability and quality, technological principles applied, whether up- and downstream process effects can be quantified, how underlying chemicals and production processes are assessed in the chosen life cycle data base and how and which credits are attributed for the substitution of energy and resources. Since boundary conditions for P recovery (Remy and Jossa, 2015; Egle et al., 2016; Zhou et al., 2016) and environmental impacts from WWTPs and sludge disposal concepts vary corresponding to the present systems (Lundin et al., 2000; Foley et al., 2010; Lederer and Rechberger, 2010; Yoshida et al., 2013; Buonocore et al., 2016; Lorenzo-Toja et al., 2016), results from this LCA study cannot be universally applied, but rather convey an order of magnitude of potential environmental impacts from P recovery and point out specific drawbacks or advantages of the different technologies.

In this context, key take-aways of this work are twofold. First, a large variance in emissions and energy demand was shown for the different P recovery technologies. Comparison to Austrian emission and energy data made it evident that impact of P recovery from wastewater on these assessed environmental factors is low or even beneficial for recovery with many of the analysed technologies, with median decreases of CED, GWP and AP of 0.01%, 0.01% and 0.05%, respectively. Regardless, some technologies show a significant increase compared to the reference system and their future implementation should therefore be avoided unless further innovation improves their environmental performance.

Second, combining the LCA findings with other environmental criteria further puts the results of the different technologies into perspective. While technologies recovering from the liquid phase have beneficial or comparably little impacts on CED, GWP and AP, their low recovery potential contradicts the demand for efficient recycling rates. For recovery from sewage sludge, those technologies that already are or are close to being applied full-scale, are associated with comparatively high additional CED, GWP and AP, therefore making their application undesirable in environmental terms. Technologies designed to recover from SSA show varying results, partly revealing trade-offs between heavy metal decontamination, emissions and energy demand. Nevertheless, regarding environmental terms, recovery from SSA seems most promising as it enables a high recycling rate, heavy metal decontamination is possible, organic micropollutants are non-present and positive effects in terms of a reduction of gaseous emissions and energy demand can be achieved. In exchange for these efforts, a nationwide sewage sludge mono-incineration concept and subsequent recovery from SSA could substitute around 40% of the annual Austrian mineral P use (Egle et al., 2014, 2016), set the concept of circular economy into force and decrease countries' vulnerability to the highly dynamic global phosphate rock market (Zoboli et al., 2016b). Nevertheless, a full country-wide analysis of the environmental effects from P recovery implementation would further have to look at impacts brought forth by the change from co-incineration to mono-incineration of sewage sludge and the potential increase in sewage sludge transport demand to mono-incineration facilities.

Not all environmental impacts were taken into account in this study and other work indicates further benefits from P recovery, i.e. reduction in eutrophication potential (reduced phosphate rock mining and therefore lower P water emissions from mining (Remy and Jossa, 2015), mitigation of Cd and U input into agricultural soils (Bigalke et al., 2016), reduction of heavy metal input compared to conventional agricultural sewage sludge application (Lederer and Rechberger, 2010), and decreased nitrogen emissions for technologies recovering also nitrogen (Johansson et al., 2008). In addition, unquantifiable environmental damages from land degradation through mining of phosphate rock reserves speak further for sustainable P use and recycling concepts.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at <https://doi.org/10.1016/j.resconrec.2017.11.002>.

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