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**Environmental impacts of phosphorus recovery from a “product” Life
Cycle Assessment perspective: allocating burdens of wastewater treatment
in the production of sludge-based phosphate fertilizers**

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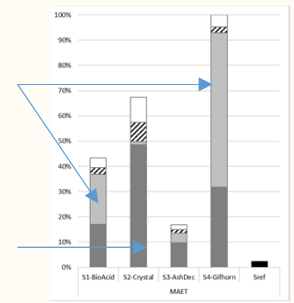
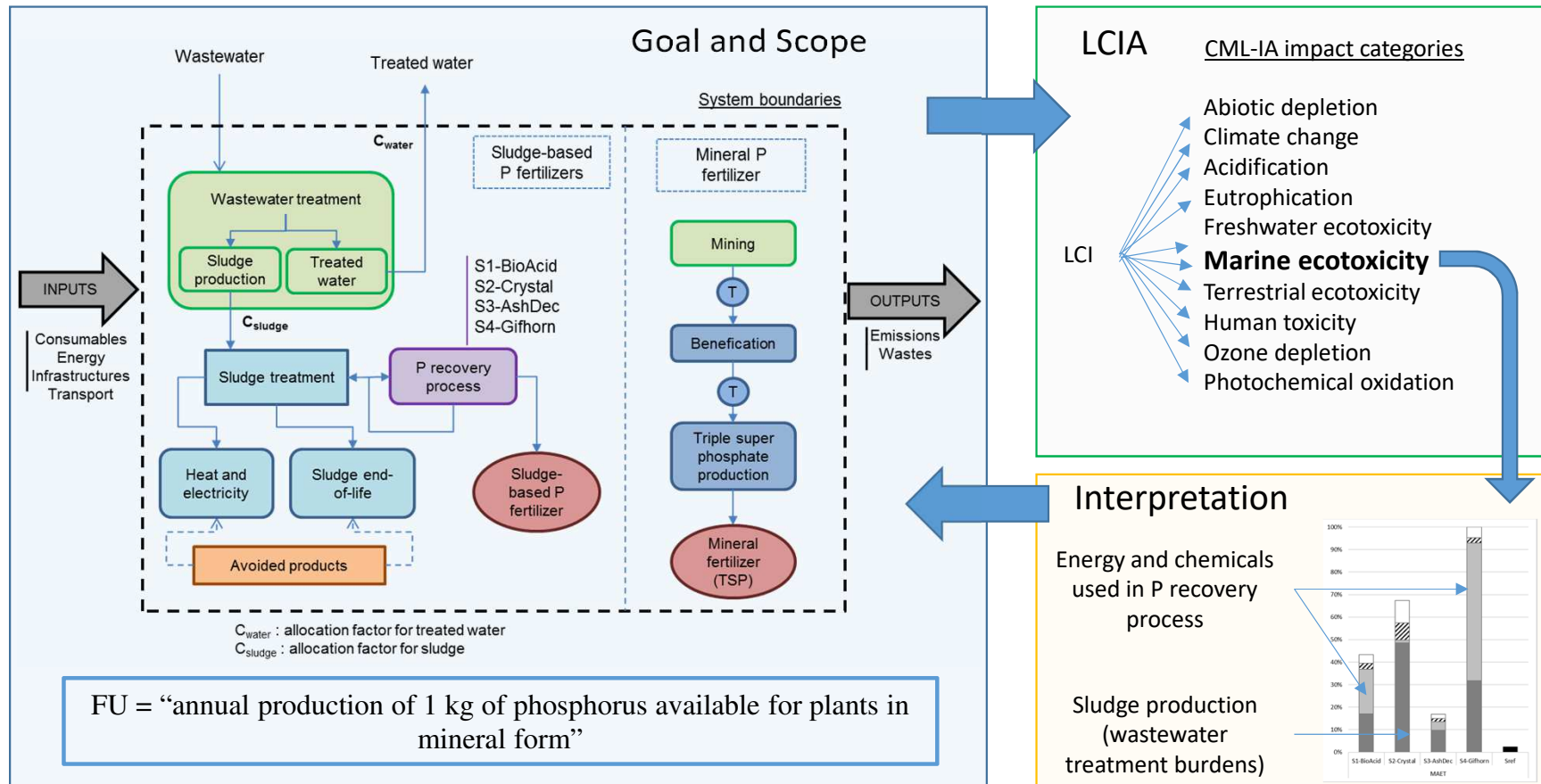
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Highlights

- LCA is used to compare mineral and sludge-based phosphate fertilizer production.
- Sludge production environmental burdens are included in the fertilizer life cycle.
- Mineral fertilizer has less environmental impacts than sludge-based fertilizer.
- Reasons are limited P yields, low P content and high need of energy and reactants.
- P resource depletion is of great concern and needs to be better integrated in LCIA.



Abstract

Since phosphorus (P) is a non-renewable element essential for life, it is extremely important to explore any potential supply of P, including that recovered from human excreta and urban wastewater. This study aimed to assess, using Life Cycle Assessment (LCA), whether recovering dissipated P by producing sludge-based phosphate fertilizer can be a suitable method to reduce P depletion. Environmental impacts of four scenarios of production of sludge-based phosphate fertilizers were compared to those of production of triple super phosphate, a mineral phosphate fertilizer used as a reference scenario. The novelty of this study was to estimate environmental impacts of sludge-based phosphate fertilizer production using a “product” LCA perspective instead of a “waste” LCA perspective. Consequently, upstream production of sludge was considered by allocating part of the environmental burdens of wastewater treatment to sludge production. Life Cycle Impact Assessment was performed using the CML-IA characterization method. Results indicated that sludge-based phosphate fertilizers appeared less environmentally friendly than mineral phosphate fertilizers, due to the contribution of the upstream burden of sludge production and P recovery. Finally, although P recovery helps preserve the mineral P resource, the overall assessment remains unfavorable for sludge-based products due to the low yields of P recovery, low P concentration of the sludge and the large amounts of energy and reactants needed to recover the P.

Keywords

Life Cycle Assessment, phosphorus recovery, sludge-based phosphate fertilizers, mineral phosphate fertilizers, allocation, product LCA

1. Introduction

Phosphorus (P) is an essential resource since it is vital for the development of plants, animals and humans. P is also a key component of mineral fertilizers, since ca. 148 million t of phosphate rock are used per year, and 90% of global demand for P is for food production (Cordell et al., 2009). With the rapid growth of world population (estimated at 9 billion people by 2050), increasing demand for food and therefore for fertilizers is expected worldwide (Sorensen et al., 2015; Steen, 2006). However, P is a non-renewable resource that cannot be replaced by another element in fertilizers. P in mineral form can be found highly concentrated in reserves of phosphate rocks. These rocks are found almost worldwide, but ca. 86% of them were controlled by only six countries in 2016 (Morocco/Western Sahara (71.4%), China (4.7%), Algeria (3.1%), Syria (2.6%), Brazil (2.4%), and South Africa (2.1%)); thus, their availability is subject to high geopolitical risks (USGS, 2018). Moreover, P extraction from phosphate rocks is projected to peak around 2030. Afterwards, extraction will decrease, and global reserves should start to run out within 75-100 years, exhausting reserves of phosphate rocks by the end during the 22nd century (Rosemarin et al., 2009). In addition, the quality of phosphate rock will decline, increasing its price drastically. In the past 20 years, its price increased by 273% due to the increasing costs of extraction, processing and shipping (The World Bank, 2017). One direct impact will be an increase in the cost of producing food (Cordell et al., 2009; Rosemarin et al., 2009).

In 2017, phosphate rocks and P were added to the European Union's (EU) list of critical raw materials (European Commission, 2017). A raw material is considered critical when its supply risk and economic importance exceed a given threshold. The EU supply of P and phosphate rock depends completely on imports since they are not produced or mined, respectively, in the EU. Supply risk can be reduced by increasing the end-of-life recycling input rate (EOL-RIR) and the substitution potential (i.e. the ability to replace a critical raw material with a non-critical one). Since there is no substitute for these materials, supply risk can be reduced only by increasing the EOL-RIR of the ratio of recycling from waste feedstock to EU demand for a given raw material, the latter equal to primary and secondary material supply inputs to the EU. The EOL-RIR is estimated at 17% for phosphate rock and equals zero for P.

It is therefore extremely important to explore any potential supply of P given these constraints. P can be recovered or reused from several sources, including human excreta. Nearly 98% of ingested P ends up in wastewater and accumulates in sewage sludge (Kalmykova et al., 2015), making it an attractive resource for P recovery. Sludge can contain both mineral and organic P and be spread directly on soil as an organic fertilizer (Houot et al., 2014). Due to several constraints (presence of heavy metals and organic pollutants, social acceptability, etc.), however,

new technologies have been developed to extract and recover this dissipated P. Sludge-based phosphate fertilizers can be used safely on agricultural soils. According to Egle et al. (2015, 2016), the most efficient P recovery technologies occur before and after anaerobic digestion and from sewage sludge ashes, mainly in the form of magnesium ammonium phosphate (struvite, $\text{NH}_4\text{MgPO}_4 \cdot 6\text{H}_2\text{O}$) or calcium phosphates.

One unsolved question remains the overall environmental impacts of recovering this dissipated P compared to extracting phosphate from rocks. Some studies have assessed environmental impacts of sludge used as phosphate fertilizer using Life Cycle Assessment (LCA) (Sena and Hicks, 2018). Johansson et al. (2008) and Linderholm et al. (2012) compared four alternative options for handling sludge, with use of its P as fertilizer on agricultural soils. Bradford-Hartke et al. (2015) compared environmental benefits and burdens of recovering P as struvite from dewatering return liquors in four centralized and two decentralized systems. In these comparative LCAs, recovering P from sludge was seen more as an alternative waste treatment than as sludge-based fertilizer production; thus, sludge was considered to have no environmental burdens. In this context, using supercritical water oxidation to recover P appeared to be the best option for Johansson et al. (2008). In contrast, direct spreading of sewage sludge on soil was the option with the lowest energy use and greenhouse gas emissions for Linderholm et al. (2012), due to the beneficial association with nitrogen in sludge. For Bradford-Hartke et al. (2015), recovering P using struvite precipitation resulted in positive environmental impacts due to energy and chemical use being offset by operational savings and avoided fertilizer production.

The critical review of Pradel et al. (2016), however, emphasized that if sludge treatment is specifically designed to produce sludge-based fertilizers with high added value, sludge can no longer be considered as waste but rather as a coproduct of the wastewater treatment plant (WWTP). This assertion was shared by several authors who also started to question the “zero burden assumption” (Cleary, 2010; Holden; Oldfield and Holden, 2014; Oldfield et al., 2018) by considering that the status of “waste” is subjective and questionable, especially if it has a high nutrient or energy-recovery potential. However, most LCA studies dealing with P recovery still consider sludge as a waste (Sena and Hicks, 2018). To raise awareness of the need to consider upstream production of waste-based products in LCA, we performed LCA of a case study of sludge-based fertilizer production.

This study aimed to assess, using a “product” LCA perspective, whether recovering dissipated P by producing sludge-based phosphate fertilizer can be a suitable alternative to producing mineral fertilizers from phosphate rocks. To reach this goal, four scenarios of the production of sludge-based phosphate fertilizers were compared to the production of phosphate fertilizer from phosphate rocks. The goal and scope definition, inventory data and characterization method used for each scenario are presented in the material and methods section. Four points are

then discussed in the results: the consequence of allocating part of the environmental burdens of wastewater treatment to sludge production, the unequal balance between consumed resources and recovered P, the ability to reduce P depletion if the recovery rate of diffuse P is improved and the difficulty assessing P resource depletion in LCA characterization methods.

2. Materials and methods

LCA is a four-step procedure based on international standards (ISO, 2006a; ISO, 2006b). The first step consists of defining the goal and scope of the study, i.e., the system to be studied, its boundaries, functions, and the related functional unit (the reference all the inventory data are related to), the allocation methods used and the assumptions made. The second step is the Life Cycle Inventory (LCI) during which all the inputs (raw material, energy) and outputs (emissions) related to each process in the system are listed and considered. The third step is Life Cycle Impact Assessment (LCIA), which relates the inputs and outputs of the system to their environmental impacts. In the final step, the results are interpreted according to the goal, the system boundaries and the assumptions made.

2.1. Goal and scope definition

LCA was used to assess the production of phosphate fertilizer from diffuse and concentrated sources of P (i.e. sewage sludge from WWTPs and phosphate rocks, respectively). The system boundaries, the functional unit and the scenario studied are described in the following subsections.

2.1.1. System boundaries

As highlighted by Pradel et al. (2016), “waste” sludge is moving from a “waste-to-product” or a “product” when wastewater and sludge treatment are designed to produce a product with high added value (e.g. fertilizers). This paradigm shift implies considering processes upstream of sludge production when sludge-based fertilizers are compared to conventional fertilizers in comparative LCA. Sludge is the natural result of cleaning wastewater in WWTPs. Considering sludge as a product means that the water treatment line is a multifunctional process that provides two coproducts that are given a second life: sludge and “clean water”. Consequently, the environmental burdens of the water treatment line need to be allocated between these two coproducts, for example by using the allocation factor developed by Pradel et al. (2018).

The system boundaries for sludge-based phosphate fertilizers therefore include the environmental burdens of the water treatment line allocated to sludge (hereafter referred to as “sludge production”) and the environmental burdens of sludge treatment. Sludge production from water treatment includes transport of dissolved P in the wastewater by the sewer network to the WWTP, pretreatment, and primary and biological treatments, the latter two which produce primary and biological sludge, respectively. Sludge treatment is a chain of multiple steps that depend on the scenario considered: thickening, anaerobic digestion, dewatering, struvite precipitation, storage and incineration (Fig. 1).

The system boundaries for mineral phosphate fertilizer refer to the production of triple super phosphate (TSP) and include phosphate rock extraction and beneficiation as well as production of phosphoric acid, commonly used in fertilizer manufacturing.

For each process, inflows and outflows of the nitrogen, carbon and P mass balance were quantified, as were the system’s inputs (consumption of energy, reactants and fuel) and outputs (emissions to air, water and soil). Infrastructure resources were also considered; they referred to the construction of buildings and equipment used for each process unit. Dismantlement of infrastructure was not included.

2.1.2. Functional unit

The primary function of the studied systems is production of phosphate fertilizer from diffuse and concentrated sources of P. The functional unit was therefore defined as “annual production of 1 kg of P available for plants in mineral form”. The mineral form of the functional unit refers to struvite, “Rhenania phosphate” or TSP.

Environmental impacts of producing sludge-based phosphate fertilizer were estimated in four scenarios that differed in the method used to recover diffuse P: before anaerobic digestion using biological acidification (S1-BioAcid), after dewatering using P crystallization of the return liquor (S2-Crystal), after sludge incineration using the AshDec® process (S3-AshDec) and during dewatering using the Gifhorn® process (S4-Gifhorn). These scenarios were compared to a reference scenario (Sref) providing 1 kg of P as TSP using a concentrated source of P.

Sludge-based phosphate fertilizer production has additional functions defined by energy and nutrient recovery. The first is production of heat and electricity either from anaerobic digestion of sludge and cogeneration of the resulting biogas and from incineration. Some of the heat and electricity are used respectively to maintain the digester at its operating temperature and provide a self-generated source of electricity, thus decreasing the amount

of external heat and electricity taken from the grid. The remaining heat and electricity not used to produce fertilizer are coproducts of the WWTP. The second additional function is the production of an organic fertilizer – the remaining sludge – which is low in P but rich in nitrogen and can be spread on soil. These additional functions were managed using system expansion, which led us to include avoided products (Fig. 1).

2.1.3. Description of the scenarios

Each scenario, including fertilizer production, sludge treatment and its end-of-life, is presented in Fig. 2. TSP (Sref) results from the reaction of phosphate rock (from Morocco) with phosphoric acid. In the final product, the P must be supplied as 30% rock and 70% acid. After mining, phosphate rock is processed (crushed, washed, dried, etc.) and supplied as dry rock with 33% P₂O₅. Phosphoric acid is produced using the dihydrate wet process method, which dissolves phosphate rock with sulfuric acid, and is composed of 50.7% of P₂O₅ per kg of acid produced. Once processed, TSP reaches a composition of 48% P₂O₅ (Althaus et al., 2007; Nemecek and Kägi, 2007). As usual in LCA, phosphate rock was considered a natural resource. Only the environmental impacts of its extraction and the subsequent processes up to the production of the TSP were considered.

The sludge-based phosphate fertilizers are produced from a French WWTP with a capacity of 300 000 population equivalent. The wastewater treatment line is composed of three structures combining pretreatment and primary treatment. The pretreatment is composed of a grit chamber and oil separator, while the primary treatment is composed of a lamellar primary decanter, operating without reactants or sludge recirculation. During primary treatment, particulate organic carbon is recovered in the form of primary sludge. Part of the biodegradable and inert organic particulate nitrogen as well as part of the organic P ends up in the primary sludge. The wastewater is then sent to three biological treatment lines. Each line is composed of a biological basin, a degassing structure, two clarifiers and a sludge recirculation station. The biological basin has both anoxic and aerobic zones so that nitrogen can be removed by nitrification and denitrification and P assimilated by the biomass (i.e. the biological sludge). After being thickened by gravity and centrifugal thickeners, primary and biological sludge are blended at a 65:35 ratio, respectively, before entering the anaerobic digester. The four scenarios for sludge-based phosphate fertilizer production were then assessed.

In the first scenario (S1-BioAcid), biological dissolution by acidification is used to separate P from organic matter. P dissolution during biological acidification involves two types of microorganisms. Acidifying bacteria first transform a substrate such as the fermentable fraction of organic waste into volatile fatty acids (VFAs), which

acidify the sludge, dissolving some of the mineral P (calcium and magnesium phosphate or adsorbed on iron hydroxide). Polyphosphate Accumulating Organisms (PAOs) found in the biological sludge will then use VFAs to release P under anaerobic conditions. The acidification during this process is sufficient to maintain the P release and prevent P from precipitating with Ca, Mg or Fe and high enough not to inhibit the P release by the PAOs. Once biological acidification is complete, 60% of the P in sludge is recovered within the liquid fraction after adding polymers to improve liquid/solid separation during centrifugation. The solid fraction of sludge is sent to anaerobic digestion while the return liquor, rich in solubilized P, needs to be purified. The liquor is purified using a cationic resin that captures excess Fe and thus prevents it from interacting with P during the struvite crystallization step. The resin is regenerated using hydrochloric acid, and the recovered Fe is reused within the WWTP. Struvite is then crystallized using magnesium chloride ($MgCl_2$) and sodium bicarbonate to increase the pH. The liquor from crystallization is injected into the anaerobic digester, while the sludge-based phosphate fertilizer obtained is stored. The fertilizer is composed of 65% struvite and 35% calcium and magnesium carbonates.

In the second scenario (S2-Crystal), the sludge is digested and then dewatered. The P is recovered from the return liquor of the dewatering process using crystallization with a magnesium oxide reactant. The fertilizer obtained is composed of 90% struvite.

In the third scenario (S3-AshDec), P is recovered using the AshDec® process (Jossa and Remy, 2015), in which pre-heated ashes (from previous sludge incineration) are mixed with alkali additives ($NaSO_4$) and a reducing agent (e.g. dried sewage sludge) before entering a rotary kiln. In the rotary kiln, the mixture is heated to 900-1000°C for at least 20 min using natural gas as fuel for the kiln. During the process, phosphate phases present in the ash are transformed into plant-available forms (mostly $NaCaPO_4$), whereas volatile heavy metals (As, Cd, Hg, Pb, Zn) are evaporated in the reducing atmosphere and are thus partially removed via the gas phase. Off-gases must be further treated to decrease dust (since fly ash contains heavy metals) and combustion gases to acceptable limits. The final product of the AshDec® process is similar to “Rhenania phosphate”, which was produced as plant fertilizer in the 20th century. Besides the P product and off-gases, no further waste is generated in the process.

The fourth scenario (S4-Gifhorn) is modeled according to the Gifhorn® process (Jossa and Remy, 2015). Digested sludge is directly acidified to pH 4.5 by adding sulfuric acid in a first reactor, thus dissolving the phosphate that is chemically bound to the sludge into the liquor. At this pH, large amounts of metals (Fe and heavy metals) are mobilized in the liquor. To prevent the transfer of Fe and heavy metals to the final P product, dissolved metals are precipitated as sulfides in a second step by adding Na_2S . NaOH is also added to raise the pH to 5.6. Leached sludge

is then dewatered in a centrifuge with added polymers, so that P-rich liquor and sludge are separated. In a second reactor, precipitation of the P product in liquor is initiated by a small dose of $Mg(OH)_2$ to supply additional Mg and raise the pH to 9, eventually adding NaOH to control pH. Due to the composition of the liquor in the Gifhorn® process, the P product is precipitated and recovered as struvite.

2.2. Life Cycle Inventory

The mineral phosphate fertilizer scenario (Sref) is the LCI for TSP production from the ecoinvent v2.2 database (RER: triple superphosphate, as P_2O_5 , at regional storage) (ecoinvent, 2007). Sludge-based phosphate fertilizer scenarios were modeled based on a French WWTP. The main characteristics of the WWTP are shown in Table 1. LCI input data for sludge-based fertilizer scenarios were modeled using the ecoinvent v2.2 database. LCI data of avoided fertilizers come from ecoinvent v2.2 database and are modeled as ammonium nitrate (RER: Ammonium nitrate, as N, at regional storage) and triple superphosphate (RER: triple superphosphate, as P_2O_5 , at regional storage). Flowcharts are described in detail in Supporting Information 1 (SI1). Sludge-based fertilizer scenarios assume a 30-year lifetime for wastewater and sludge treatment infrastructure and a 50-year lifetime for the sewer network according to Risch et al. (2015) (Supporting Information 2 (SI2)). LCI input and output data are summarized in Tables 2 and 3, respectively.

2.2.1. LCI data used to allocate environmental burdens to sludge production

LCI data used to assess sludge production are provided in this section. LCI data from sewer network and wastewater treatment line as well as the allocation factor used to allocate the LCI data to sludge production are provided in the following subsections.

2.2.1.1. LCI data for the sewer network and wastewater treatment line

LCI data for the sewer network were provided by the WWTP owner. LCI data for the sewer network are modeled according to Risch et al. (2015). LCI data of the wastewater treatment line come from technical documents provided by the WWTP owner. LCI output data for the wastewater treatment line are composed of emissions, which are calculated according to nitrogen, carbon and P mass balances using transfer coefficients from the literature and expert opinion (Doka, 2007). Details of mass balance calculations are shown in Fig. SI2-1 in the SI2.

2.2.1.2. Allocation factors

Burdens of the wastewater treatment line are allocated to both primary and biological sludge production according to their respective allocation factors according to the method of Pradel et al. (2018), which defines allocation factors based on process- and product-related parameters that reflect technological performances of the wastewater treatment line. Calculation follows a four-step procedure: (1) identify and (2) quantify process- and product-related parameters and their combination to define an overall allocation factor, (3) construct a corrective inventory matrix to adjust the overall allocation factor for LCI flows that are specific to certain processes and (4) mathematically model the final corrected allocation factor. Detailed calculation of the allocation factors used is provided in SI1. A summary of these allocation factors is shown in Table 4.

2.2.2. LCI data for sludge treatment

LCI data used for sludge treatment are provided for each process involved in the following subsection. Sludge treatment process performances and assumptions used to calculate emissions from the sludge treatment mass balance are shown in Table SI2-1 in SI2.

2.2.2.1. Thickening

Methane (CH₄), nitrous oxide (N₂O) and carbon dioxide (CO₂) emission factors for gravitational thickening come from Gourdet et al. (2017). No emissions are modeled for centrifugal thickening, which uses polymers composed of 50% nitric acid and 50% acrylonitrile as reactants.

2.2.2.2. Anaerobic digestion

Mesophilic anaerobic digestion (37°C) is assumed. Grease from pretreatment is used as an input for anaerobic digestion. Biogas lost (due to leakage, which emits CH₄ and biogenic CO₂ to air) is estimated as 10% (upper value from IPCC 2006). All “net biogas” (i.e. that which is not lost) is cogenerated. It is assumed that all of the CH₄ in the biogas is transformed into CO₂ and then emitted into the air along with nitrogen oxides (NO_x) and sulfur oxides (SO_x) generated by biogas combustion (INERIS, 2002; RDC Environnement, 2007). This process has additional emissions, including NO_x, SO_x, carbon monoxide (CO) and non-CH₄ volatile organic compounds.

2.2.2.3. Dewatering

Dewatering is performed using a press filter. Sludge is conditioned with polymers composed of 50% nitric acid and 50% acrylonitrile as reactants. Ammonium emissions for dewatering come from Gourdet et al. (2017).

2.2.2.4. Sludge end-of-life

In S1-BioAcid and S2-Crystal, the dewatered low-P sludge is stored for six months in a concrete area in the WWTP. During storage, sludge emits ammonia (NH_3), N_2O , CH_4 and CO_2 . Emission factors for storage come from Gourdet et al. (2017). After storage, the sludge is transported and spread on soil. Nitrogenous emissions are calculated according to the IPCC (2006), while P emissions are calculated according to Nemecek and Kägi (2007). Heavy metals in sludge are included, but since no data on them were available, values from French legislation on sludge spreading are used (Journal Officiel, 1998). In S3-AshDec and S4-Gifhorn, the dewatered low-P sludge is incinerated. Data for incineration in both scenarios come from Jossa and Remy (2015).

2.2.3. LCI data for sludge-based fertilizer production

Data for biological acidification come from an experimental pilot (Daumer, 2015). According to expert opinion, it is assumed that the experimental data are sufficiently representative to be used at a WWTP scale. Reactant use and energy consumption in the pilot are assumed to be proportional to those in the experiment. Since no emissions were measured in the experimental pilot, however, it was not possible to model emissions in the LCI for this process. The remaining emissions of Cl from adding MgCl_2 to precipitate P are included. According to Yoshida et al. (2015), since Cl is highly soluble, it is removed mainly in the return liquor and thus in the wastewater. Since Cl is not affected by wastewater treatment, all of it is released to the environment. However, no characterization factor for Cl is available in the characterization method CML-IA.

Data for struvite crystallization in S2-Crystal come from an industrial process in another French WWTP. Data from struvite precipitation using the Gifhorn® process in S4-Gifhorn and the AshDec® process in S3-AshDec come from the P-Rex project (Jossa and Remy, 2015). Potential P recovery for each scenario is summarized in Table 5. Process performances for each scenario are summarized in Table SI2-2 in SI2.

2.3. Life Cycle Impact Assessment

Once the LCIs were completed, the scenarios were modeled using GaBi® v6 LCA software because it allows energy and mass balances to be balanced. It can also trace mass and energy flows and perform parameterized modeling. Potential environmental impacts of each scenario were assessed using the midpoint characterization method CML-IA (Guinée et al., 2002), developed by the Leiden Institute of Science and Environment (Centrum voor Milieuwetenschappen – CML), because it was the only one that included P depletion within the abiotic depletion impact category when the study was conducted.

2.4. Sensitivity analysis

Based on the relative contribution of sludge production to impacts, a sensitivity analysis was performed to highlight how impacts change when the centralized WWTP collects wastewater with a shorter sewer network on a smaller catchment area. The sewer network modeled is composed of 92 km of wastewater network and 160 km of mixed sewer network (wastewater + rainwater). The length of the sewer network can vary greatly among WWTPs, making it difficult to set lower and upper values for it. Consequently, we based our sensitivity analysis on an extreme lower value: one linear meter (1m) of sewer network. While unrealistic, assuming only 1 m can assess whether the decrease in impacts of sludge production could ever be low enough to make impacts of sludge-based phosphate fertilizer production lower than those of mineral fertilizer. The sensitivity analysis was performed for S2-Crystal, because of the high contribution of sludge production to its gross impacts, and S3-AshDec, because it had lower impact for most categories than the other sludge-based scenarios.

Allocating part of the environmental burdens of wastewater treatment to sludge production also highlights differences between impacts when producing phosphate fertilizer from diffuse vs. concentrated P resources. To examine these differences, we calculated three mean ratios of gross impacts of different combinations of processes of sludge-based phosphate fertilizer production to those of TSP production (Sref):

- Ratio 1 (R1): impact of P recovery process/impact of TSP production
- Ratio 2 (R2): (impact of sludge production process + impact of P recovery process)/impact of TSP production. The inclusion of the sludge production process in R2 highlights which impacts it increases and by how much.
- Ratio 3 (R3): impact of the entire life cycle of sludge-based fertilizer (with or without impact of the sludge production process)/impact of TSP production.

3. Results and discussion

3.1. Environmental impacts of sludge-based phosphate fertilizers compared to those of mineral phosphate fertilizer

Results are expressed per kg of P produced as struvite (S1-BioAcid, S2-Crystal, S4-Gifhorn), “Rhenania phosphate” (S3-AshDec) or TSP (Sref). Gross impacts and net impacts (gross impacts minus avoided impacts) are shown in Figs. 3a and 3b, respectively. When comparing gross impacts of the production of 1 kg of P from diffuse

sources to those of concentrated sources, the sludge-based fertilizer scenarios have higher environmental impacts in all categories than Sref (Fig. 3a). Scenario S2-Crystal has the highest impacts except for mineral resource depletion (AD elements) and marine aquatic ecotoxicity (MAET), for which S4-Gifhorn has the highest impact. Despite using large amounts of reactants, the highest P recovery for S3-AshDec (57%) gives this scenario lower impacts, except for AD elements, than the other sludge-based scenarios. Impacts of S1-BioAcid are 30-50% of those of the scenario with the highest impacts, except for eutrophication and climate change, for which its impacts are 15% and 5% of the highest impacts, respectively.

S2-Crystal has most of the highest impacts mainly because it has (i) the lowest percentage of recovered P (12%), and (ii) a large amount of sludge to be treated and spread, causing high acidification, eutrophication and climate change impacts (due to high nitrogenous emissions) and ecotoxicity impacts (due to heavy metal emissions to agricultural soil). However, considering net impacts, avoided fertilizers counterbalance gross impacts and even result in negative results for S2-Crystal for AD elements and MAET (Fig. 3b). It also changes the ranking of scenarios for the depletion of fossil energy resources (AD fossil) and photochemical oxidation, making S4-Gifhorn the scenario with the highest net impacts for these four categories. Despite the avoided impacts, the ranking of scenarios for the other impact categories remains unchanged.

According to contribution analysis, AD elements is caused mainly by sludge-based fertilizer production, while AD fossil and ozone depletion (OD) are caused mainly by sludge production from wastewater treatment (Fig. 4). The processes used to precipitate dissipated P (biological acidification, crystallization, Gifhorn® and AshDec® processes) need large amounts of reactants, which contributes greatly to AD elements. The mineral resources depleted are sodium chloride, used to produce NaOH (S1-AcidBio, S4-Gifhorn) and sodium sulfate (S3-AshDec). Regardless of the scenario, AD fossil is caused mainly by production of the bitumen used to build the sewer network.

Nitrogenous emissions during sludge dewatering, storage and spreading are the main contributors to acidification (NH₃), eutrophication (NH₃, NO₃, N₂O) and climate change (N₂O) for S1-AcidBio, S2-Crystal and S3-AshDec. Biogas leakage during anaerobic digestion in all scenarios also contributes to climate change since CO₂ and CH₄ are emitted. Sludge production contributes to eutrophication in S4-Gifhorn and S3-AshDec due to the bitumen used in the sewer network. The Gifhorn® process used in S4-Gifhorn needs reactants such as sodium persulfate, sulfuric acid and magnesium, which contribute to acidification, eutrophication and climate change impacts. Ecotoxicity and toxicity impacts can be explained by three main causes:

- i. Direct emissions: Direct heavy metal emissions to agricultural soils contribute to freshwater aquatic toxicity (Cu, Ni), terrestrial ecotoxicity (Cr, Hg) and human toxicity (Cr, Pb, Ni) in scenarios in which sludge is spread on soil (S1-AcidBio and S2-Crystal). Emission of Hg to the air during incineration also contributes to terrestrial ecotoxicity in S3-AshDec.
- ii. Sludge production: Electricity used for biological sludge production and bitumen used in the sewer network contribute to freshwater and marine aquatic ecotoxicities in almost all scenarios.
- iii. Use of reactants: Sodium persulfate and magnesium reactants used in the Gifhorn® process contribute to all ecotoxicity and toxicity impacts in S4-Gifhorn. NaOH used during crystallization contributes to freshwater and marine aquatic ecotoxicities in S1-AcidBio.

For abiotic resource depletion, ecotoxicity and toxicity impacts, avoided impacts are driven by avoided spreading of mineral fertilizers and avoided production of energy as heat and electricity depending on the scenario (results not shown). For acidification, eutrophication and climate change, avoided impacts are driven by avoided nitrogenous emissions in scenarios with spreading on soil (S1-AcidBio and S2-Crystal) and by avoided energy for climate change in S4-Gifhorn (avoided CO₂ emissions).

3.2. Consequence of allocating part of the environmental burdens of wastewater treatment to sludge production

In this study, we consider sludge-based fertilizers as products that are intentionally produced by the WWTP owner. As a consequence, a “product” LCA was conducted instead of a “waste” LCA and part of the environmental burdens of wastewater treatment were allocated to sludge production. Following this approach is thus a proactive process, since it allows for more accurate assessment of environmental impacts of sludge-based phosphate fertilizers when compared to those of mineral fertilizers. Moreover, this approach explicitly considers environmental performances of innovative technologies used to concentrate diffuse P in wastewater.

The contribution of sludge production to gross impacts varies greatly among scenarios and impact categories. Sludge production contributes more to AD fossil and OD than sludge treatment or valorization, regardless of the sludge-based scenario, and contributes the least to eutrophication and climate change in scenarios with sludge spreading on soil (S1-AcidBio and S2-Crystal) (Fig. 4). Sludge production contributes greatly to freshwater and marine aquatic ecotoxicities and human toxicity in all scenarios except S1-AcidBio.

Of sludge production’s gross impacts, the sewer network contributes 45-56% for freshwater and marine terrestrial ecotoxicities and human toxicity, 63% for climate change and 75-96% for the other categories. Fig. 5 shows

relative gross and net impacts of each scenario when considering the real sewer network and one km of sewer network compared to the reference scenario in the sensitivity analysis. The length of wastewater and mixed sewer network needed to equal the impacts of Sref for each impact category was also calculated (Table SI2-3). When considering gross impacts when the sewer network decreases to 1 km, only AD fossil and OD decrease greatly, but S2-Crystal and S3-AshDec still have higher impacts than Sref. In contrast, for net impacts, avoided impacts greatly decrease four of the impacts (AD elements, AD fossil, MAET, OD) for S2-Crystal and two of the impacts (AD fossil, OD) for S3-AshDec. These results suggest that if improvements must be made to decrease environmental impacts of sludge production, they cannot be made to the sewer network, which has little room for improvement.

The magnitude of difference in environmental impacts between sludge-based fertilizer and TSP production (R1 ratio) varies among impact categories and scenarios (i.e. the technologies used to precipitate diffuse P) (Table 6). For instance, technologies using small amounts of reactants, such as S2-Crystal can have similar impact (1-3 times as high) as TSP production, regardless of the impact category. In contrast, for technologies using large amounts of reactants, such as S4-Gifhorn, sludge-based fertilizer production is ca. 10-557 times as high as that of TSP production among impact categories. The differences between R1 and R2 for S3-AshDec are smaller than those for the other three sludge-based scenarios. The inclusion of sludge treatment and end-of-life processes in R3 (i.e. the entire life cycle of the sludge-based product), make R3 differ the most from R1. The largest R3 was obtained for S2-Crystal for climate change (3 to ca. 14 000 times as high), while the smallest R3 was obtained for S3-AshDec for eutrophication (1-9 times as high). Excluding sludge production from R3, however, emphasizes how much it contributes to mineral and fossil abiotic depletion, MAET and OD.

Producing phosphate fertilizer from diffuse P therefore has higher impacts than doing so from phosphate mining, but to varying degrees. When P recovery efficiency is maximized and residual sludge valorized by incinerating it to recover energy (S3-Ashdec), producing phosphate fertilizer from diffuse P has impacts ca. 7-400 times as high as those of mining. When efficiency of P recovery is minimized and residual sludge is spread on soil (S2-Crystal), producing phosphate fertilizer from diffuse P has impacts ca. 25-14 000 as high as those of mining.

To our knowledge, this study is the first to estimate environmental impacts of sludge-based phosphate fertilizers from a diffuse P resource from a “product” LCA perspective. According to the review of Sena and Hicks (2018), several authors define a functional unit that focuses on using struvite precipitation to recover or remove P from the waste stream. Consequently, they do not consider upstream burdens of wastewater treatment when calculating environmental impacts of struvite production, thus conducting their studies from a “waste” LCA perspective. Some of them clearly use the “zero burden assumption” for their P recovery processes, since the recovery is based on

waste feedstock (Hörtnhuber et al., 2017). Other authors, such as Linderholm et al. (2012), compare fertilization with P recovered from a WWTP to that with mineral fertilizers such as TSP. For P mining, they consider the entire chain from extraction of the mineral to spreading, but for P recovery from a WWTP, they consider only P recovery and spreading. Their system boundaries exclude the wastewater treatment line and sludge treatment. Their study estimates that struvite precipitation has slightly less climate change impact than that of mineral fertilizers, which disagrees with results of the present study (in which the climate change impact of P recovery is 3-557 times higher than that of the entire production process of TSP) (R1 in Table 6).

Most of these authors consider that benefits of struvite precipitation offset impacts of producing conventional fertilizers. Their studies generally highlight net environmental benefits of struvite precipitation compared to a reference scenario, as in Johansson et al. (2008) and Bradford-Hartke et al. (2015). The latter compare P recovery in centralized and decentralized WWTPs and focus the system boundaries solely on P recovery and spreading. They highlight that struvite precipitation from dewatering liquid at centralized WWTPs results in net environmental benefits in most categories; however, they do not consider upstream burdens of the WWTPs.

3.3. Identifying room for improvement for decreasing environmental impacts of sludge-based phosphate fertilizers

Several factors can explain the environmental impacts of sludge-based phosphate fertilizers. First, they have higher environmental impacts than phosphate fertilizers from mining since more resources are needed to recover diffuse P. Second, low P recovery efficiency combined with use of large amounts of reactants to recover diffuse P from wastewater sludge increases impacts of sludge-based fertilizers. Last, P recovery technology have to be efficient enough to produce sludge-based fertilizers with the same fertilizing value as phosphate fertilizer from mining (e.g. TSP).

3.3.1. Imbalance between consumed resources and recovered P for sludge-based phosphate fertilizers

Table 7 shows the amounts of electricity, heat, reactants and infrastructure used to recover 1 kg of P from sludge or phosphate rocks. These values include the inputs used in the P recovery processes and those used for sludge treatment and end-of-life. Since phosphate rocks contain high concentrations of P, the amounts of inputs used to recover this P are drastically smaller than those used to recover P from sludge. The reactants used during the entire life cycle are used mainly to recover P. Except for S2-Crystal, diffuse P recovery uses larger amounts of reactants than those used for TSP. The amount of electricity used to recover P is also greater than that of TSP, either for P

recovery only or the entire life cycle. The infrastructure involved in the sludge-based scenarios (i.e. the WWTP) is also larger, by a factor of 10 000, than that involved in TSP production. This factor drops to 1 000 when considering only the infrastructure of the P recovery process.

The greatest differences are observed for emissions generated during P recovery. Only heavy metals are emitted during TSP production, while CH₄, CO₂ and nitrogenous emissions (from sludge treatment and end-of-life) are emitted in sludge-based scenarios. When considering emissions during P recovery, N₂O, CO and Hg are emitted during the AshDec® process. NH₃ is emitted during struvite precipitation from dewatering for S2-Crystal and S4-Gifhorn, but these emissions remain higher than those of TSP production. These results agree with those of Bradford-Hartke et al. (2015), who highlighted that P recovery does not necessarily have net environmental benefits, since the technology used does not necessarily offset the resources consumed in the process.

3.3.2. Increasing diffuse P recovery rate using smaller amounts of reactants

The wastewater entering the WWTP studied has an influent mass flow of 83 220 kg of P per year that can be recovered. Wastewater treatment captures 63% of the P in sludge. However, the amount of P in the final product varies among scenarios. The P recovered in the final product varies from 18-91% of the P in sludge after wastewater treatment but only 12-57% of the total P entering the WWTP (Table 5). Each scenario needs different amounts of reactants to recover 1 kg of P (Table 8).

Two factors can explain the low P recovery rate in S2-Crystal. First, only 20% of the P in sludge is solubilized in a mineral form after anaerobic digestion and dewatering and then recovered in the return liquor. Second, 90% of this P is recovered as struvite by crystallization. In S2-Crystal, the lower the P capture rate, the less reactant is used to precipitate P as struvite. In S4-Gifhorn, the recovery rate of P from sludge is 58% due to using large amounts of reactants (Table 8). In comparison, S1-BioAcid has a recovery rate of 53% using smaller amounts of reactants since a biological process acidifies the sludge. However, the AshDec® process has the highest recovery rate, since 91% of P is recovered from sludge ashes with the use of small amounts of chemical reactants.

To decrease environmental impacts of sludge-based fertilizers, it is important to have a high P recovery rate while using the smallest amounts of reactants possible. Chemical reactants are generally used to acidify the media, which solubilizes the P in sludge (S4-Gifhorn). An alternative to chemical acidification is to increase biological acidification by using an organic substrate to develop acidifying bacteria (Capdevielle et al., 2016; Guilayn et al., 2017), as in S1-BioAcid. Another solution is to combine several struvite precipitation processes at different steps

of sludge treatment (e.g. during anaerobic digestion and from ashes) so that all P from wastewater sludge can be recovered. Municipal wastewater represents a useful P reserve. If the P recovery percentage is maximized (i.e. 70% of P recovered from wastewater to sludge and then 91% P recovered from sludge to sludge-based fertilizer), it has the potential to meet ca. 40% of the annual P demand in France, estimated to be 191 677 t (UNIFA, 2018). This is the twice the potential estimated by the P-Rex project (i.e. 20% of the P demand in Europe).

Finally, although P can be recovered from centralized WWTPs, other substrates can be used directly, such as urine in decentralized WWTPs. Indeed, urine represents less than 1% of total influent wastewater but 50% of its P load (Wilsenach and van Loosdrecht, 2006). Separating urine streams would therefore increase P concentration, decrease the volume of water to treat and simplify centralized WWTPs (Sena and Hicks, 2018). Due to infrastructure constraints such as the installation of new pipe networks or toilet replacement on a large scale, these decentralized systems are uncommon. However, if LCA of struvite production from urine were to be conducted from a “product” LCA perspective, new allocation factors would need to be created to allocate environmental burdens of the decentralized WWTPs between the urine and the other wastewater streams (i.e. graywater and brownwater) transported by sewer to centralized WWTPs. The environmental burdens to consider would be mainly the infrastructure of the source-separation systems (e.g. new pipe networks, urine diverting toilets).

3.3.3. Providing a sludge-based fertilizer with the same fertilizing value as phosphate rock fertilizers

The scenarios in this study assume that sludge-based and phosphate rock mineral fertilizers have the same fertilizing value (i.e. P is 100% available for plants). Fertilizing with sludge-based phosphate fertilizers can be an interesting alternative to phosphate rock fertilizers and socially more acceptable than fertilizing with sewage sludge. Indeed, struvite is described as a “slow-diffusion fertilizer” in many studies (de-Bashan and Bashan, 2004; El Diwani et al., 2007; Rahman et al., 2014; Talboys et al., 2016). The fertilizing value of struvite is the same as that of phosphate rock fertilizer (Montag et al., 2007). However, the minerals in struvite are gradually released and would not be in excess; thus, they are less subject to leaching, runoff and other indirect P losses than phosphate rock fertilizers (Rahman et al., 2014). Struvite can also be applied less frequently than conventional fertilizers (Münch and Barr, 2001). Further studies are therefore needed to compare environmental impacts of sludge-based and phosphate rock fertilizers from an agronomic perspective by considering their effective fertilizing values.

3.4. Assessing P resource depletion with existing LCA characterization methods

When this study was conducted, P resource depletion could be assessed in LCA only with two characterization methods, one of them CML-IA. In it, the P resource is converted to antimony equivalents (Sbeq) using a characterization factor of $5.52 \text{ E-}06 \text{ kg Sbeq}$ when “ultimate reserves” are assumed (CML-UR, baseline method). This characterization factor is below the median ($1.91 \text{ E-}05$) of characterization factors of 174 substances in CML-IA. CML-IA can also calculate resource depletion assuming other types of reserves such as “reserve base” (CML-RB) and “economic reserves” (CML-ER) (non-baseline methods). The ILCD Handbook recommends the baseline method, but results provided by CML-IA can change greatly depending on the type of reserves assumed (Fig. 6). Comparison of the scenarios using the CML-IA baseline and non-baseline methods highlights an increasing contribution of P depletion to the AD elements impact in Sref, in this order: CML-UR (18%), CML-RB (41%) and CML-ER (53%). Sludge-based scenarios deplete other resources, such as barium sulfate, sodium sulfate, gypsum and sodium chloride, but not P (Fig. 6). Nevertheless, these results do not highlight benefits of P recovery in sludge-based scenarios, which consume other critical resources. Some of the non-recovered P ends up in the sludge, which can be either incinerated or spread on soil. If incinerated, the remaining P is lost (as in S4-Gifhorn) or recovered (as in S3-AshDec). If spread, the sludge can replace phosphate fertilizer from phosphate rock and thus help decrease P depletion. Thus, this study should be expanded to estimate the magnitude of potential of P savings. This will be the focus of our next study, which will assess fertilization practices with sludge-based and phosphate rock fertilizers.

4. Conclusion

The results highlight that producing 1 kg of P from phosphate rock has lower environmental impacts than producing 1 kg of P from wastewater sludge. The low yields of P recovery associated with a low P concentration of sludge and need for large amounts of energy and reactants to recover P are responsible for the higher environmental impacts of sludge-based scenarios. However, their environmental impacts could be decreased if a good compromise is found between P recovery efficiency and the reactants needed by the P recovery technologies. The advantage of allocating part of the environmental burdens to sludge production is double. First, it improves knowledge about how the wastewater treatment system (e.g. technology used, P recovery efficiency) contributes to environmental impacts of sludge-based phosphate fertilizers. Second, and as a consequence, these overall impacts can be decreased by decreasing those of their production processes (e.g. precipitating diffuse P into a value-added fertilizer). Results highlighted that there is little potential to reduce environmental impacts of sludge

production. Another solution, not studied in this study but one that could be an interesting option for P recovery, would be to recover P during struvite precipitation from urine in decentralized systems. To do so, however, new allocation factors need to be calculated to represent the urine source-separation infrastructure within the system boundaries. Finally, results highlighted that production of sludge-based phosphate fertilizers depletes other resources that may be more critical than P. While P recovery initially aimed to reduce depletion of mineral P, resource criticality, not only P depletion, has become a great concern and needs to be better integrated in LCIA.

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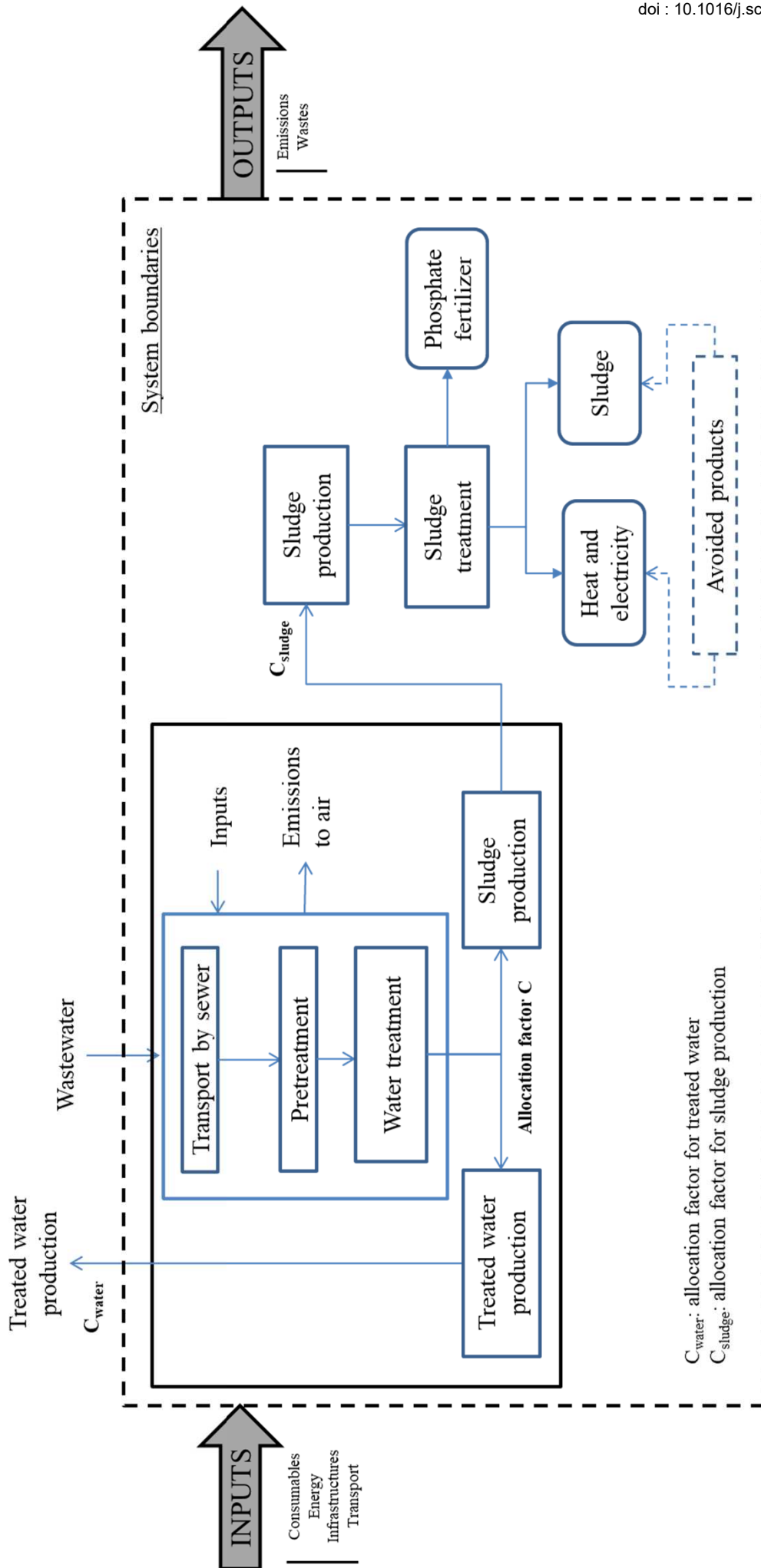


Fig 1. System boundaries of the LCA of sludge-based phosphate fertilizers

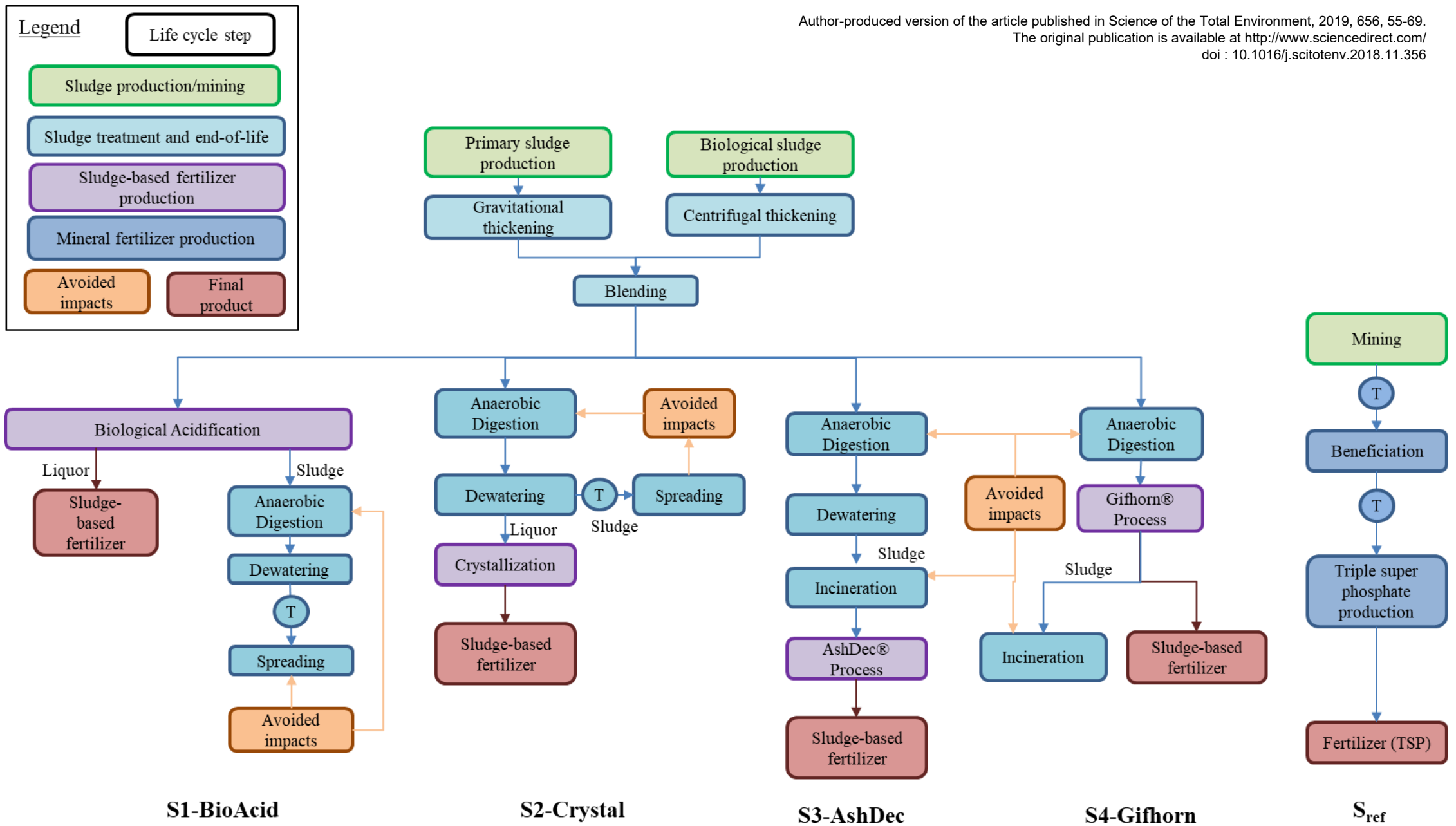


Fig 2. Description of scenarios for production of sludge-based phosphate fertilizers and mineral phosphate fertilizer (S_{ref}, the reference scenario)

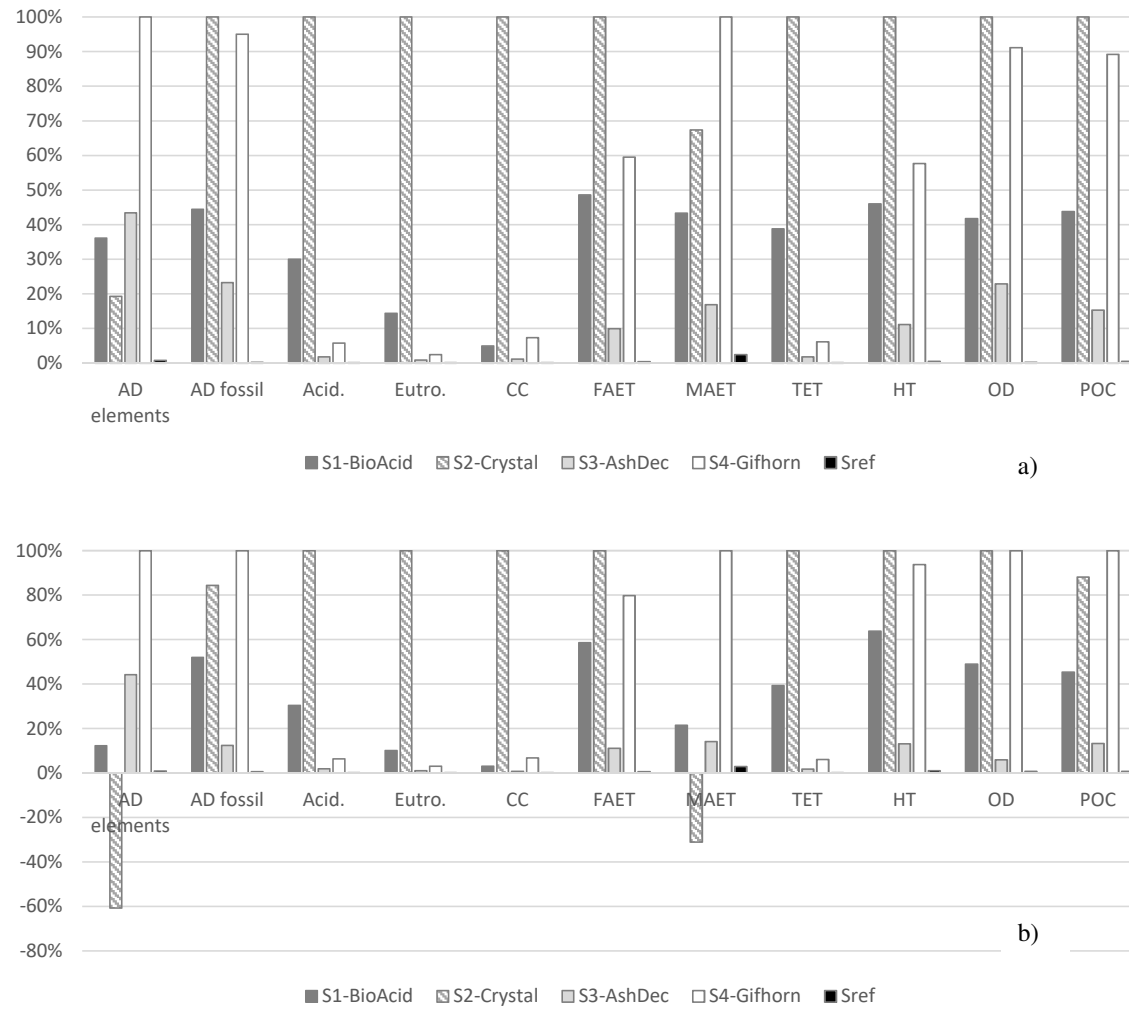


Fig 3. Relative (a) gross impacts and (b) net impacts (gross impacts minus avoided impacts) for each CML-IA impact category for each scenario (AD elements: mineral resource depletion, AD fossil: fossil energy resource depletion, Acid.: acidification, Eutro.: eutrophication, CC: Climate change, FAET: freshwater aquatic ecotoxicity, MAET: Marine aquatic ecotoxicity, TET: terrestrial ecotoxicity, HT: Human toxicity, OD: Ozone depletion, POC: photochemical oxidation)

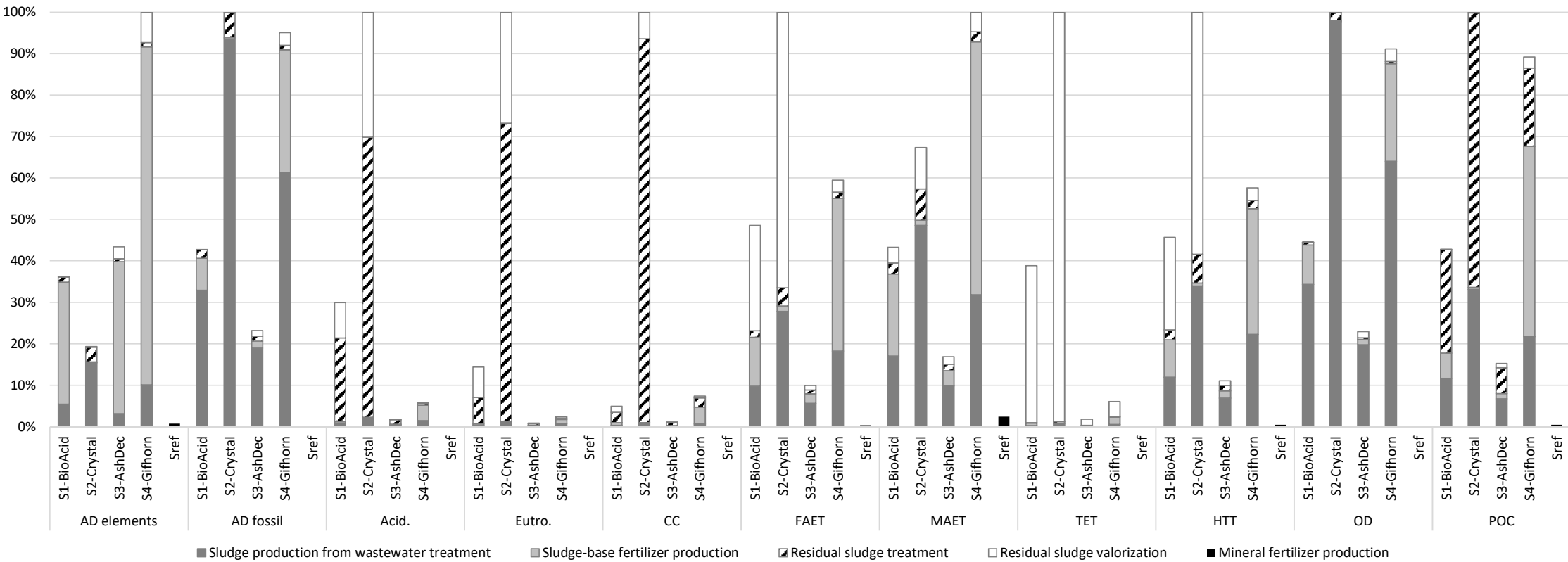


Fig 4. Contribution analysis of processes to relative gross impacts of each scenario for each CML-IA impact category (AD elements: mineral resource depletion, AD fossil: fossil energy resource depletion, Acid.: acidification, Eutro.: eutrophication, CC: Climate change, FAET: freshwater aquatic ecotoxicity, MAET: Marine aquatic ecotoxicity, TET: terrestrial ecotoxicity, HT: Human toxicity, OD: Ozone depletion, POC: photochemical oxidation)

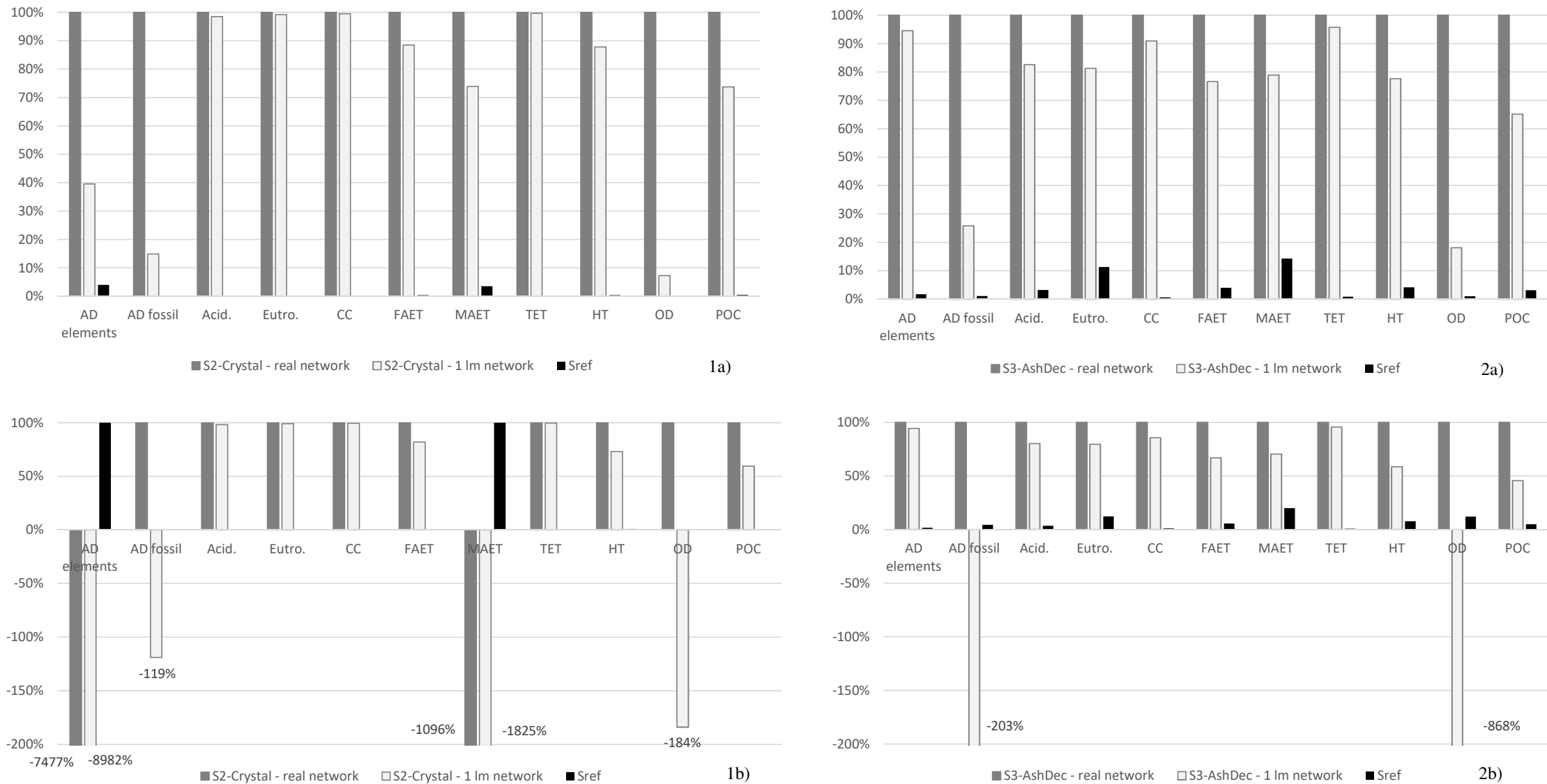


Fig 5. Sensitivity analysis of relative (a) gross impacts and (b) net impacts (gross impacts minus avoided impacts) in scenarios (1) S2-Crystal and (2) S3-AshDec assuming the real sewer network (252 km) or an extreme lower value of 1 linear meter (1m) of sewer network, compared to those of the reference scenario (Sref)

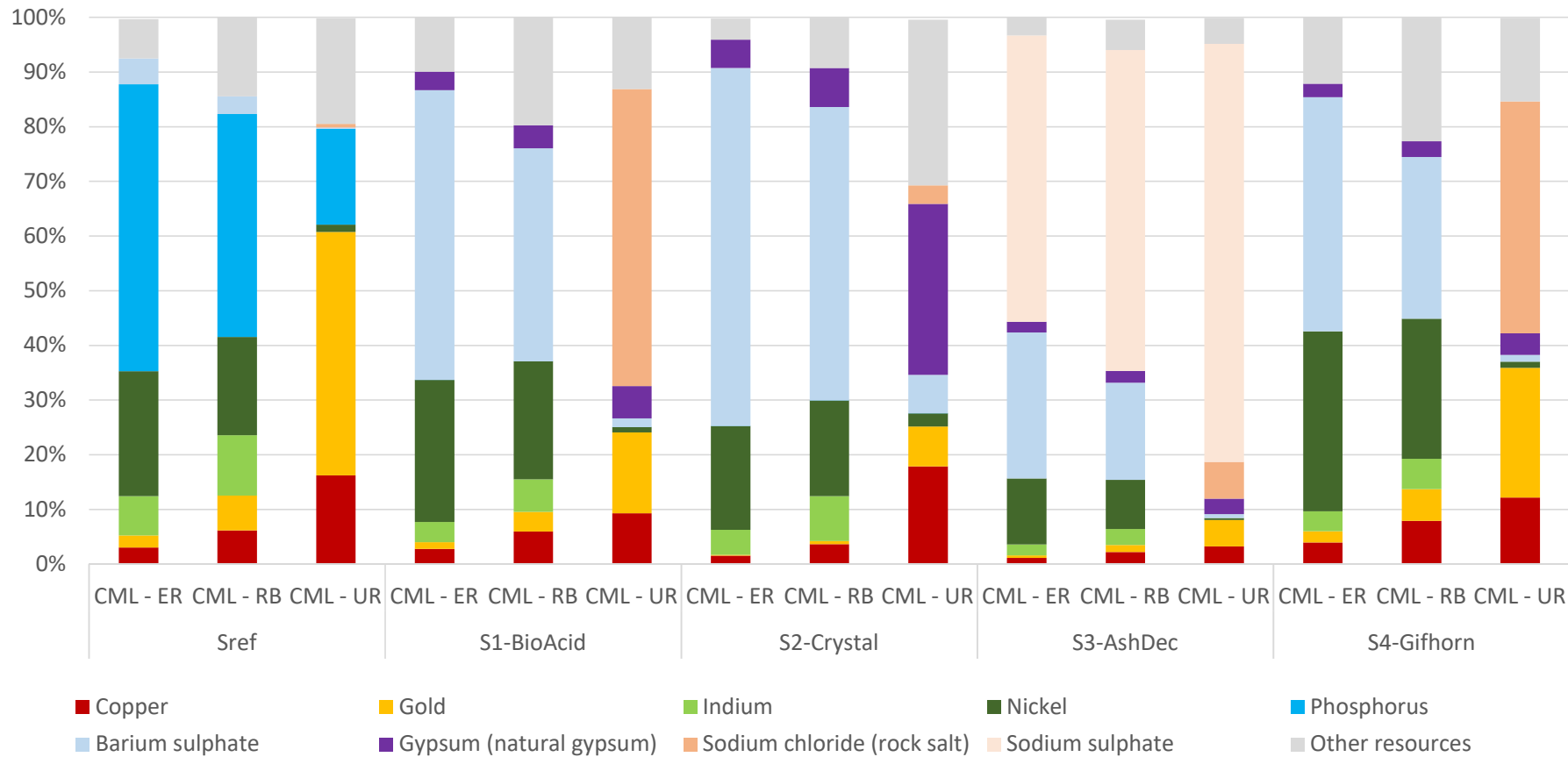


Fig 6. Contribution of mineral resources to the abiotic depletion impact of the CML-IA characterization method in the sludge-based phosphate fertilizer scenarios (S1-S4) and mineral phosphate fertilizer scenario (Sref) according to the reserves assumed: ultimate (CML-UR), reserve based (CML-RB) or economic reserve (CML-ER)

Table 1. Average influent, average effluent and general characteristics for the wastewater treatment plant (WWTP)

		WWTP
General data		
	Amount of wastewater treated	38 975 m ³ .d ⁻¹
	Person Equivalent	300 000
	Sludge production from water treatment (on a dry matter basis)	Primary sludge: 5.58 t.d ⁻¹ Biological sludge: 3.57 t.d ⁻¹
	Sludge siccidity when entering sludge treatment	Primary sludge: 5% Biological sludge: 1%
	Biogas production from sludge digestion	2 161 Nm ³ .d ⁻¹
	Final product	Struvite or “Rhenania phosphate”
	Amount of phosphorus recovered in final product	S1-BioAcid: 75.13 kg.d ⁻¹ S2-Crystal: 26.41 kg.d ⁻¹ S3-AshDec: 83.73 kg.d ⁻¹ S4-Gifhorn: 130.47 kg.d ⁻¹
Influent characteristics		
	Chemical Oxygen Demand (COD)	475.46 mg.l ⁻¹
	Total Kjeldahl Nitrogen (TKN)	44.59 mg.l ⁻¹
	Total Phosphorus (TP)	5.85 mg.l ⁻¹
	Total Suspended Solids (TSS)	221.60 mg.l ⁻¹
	Total Organic Carbon (TOC) ^a	125.12 mg.l ⁻¹
Effluent characteristics		
	Chemical Oxygen Demand (COD)	27.99 mg.l ⁻¹
	Total Kjeldahl Nitrogen (TKN)	6.63 mg.l ⁻¹
	Total Phosphorus (TP)	2.18 mg.l ⁻¹
	Total Suspended Solids (TSS)	9.06 mg.l ⁻¹
	Total Organic Carbon (TOC) ^a	9.03 mg.l ⁻¹

^a TOC is calculated based on the ratio COD:TOC, which is set at 3.8 for influent and 3.1 for effluent (Rocher et al., 2016)

Table 2. Life cycle inventory input data for the sludge-based phosphate fertilizer scenarios (S1, S2, S3 and S4) – Values are presented per functional unit: 1 kg of P produced.

Inputs	Unit	S1-BioAcid ^c	S2-Crystal	S3-AshDec ^d	S4-Gifhorn ^d
Sewer network^{a,e}	p ^b	$2.07E-06 \times (C1+C2)$	$2.07E-06 \times (C1+C2)$	$4.20E-07 \times (C1+C2)$	$6.62E-07 \times (C1+C2)$
Pretreatment^{a,e}	p	$3.65E-05 \times (C1+C2)$	$1.04E-04 \times (C1+C2)$	$2.10E-05 \times (C1+C2)$	$3.31E-05 \times (C1+C2)$
Wastewater treatment line^{c,e}					
Primary treatment infrastructure resources ^a	p	$1.22E-06 \times (C1+C2)$	$3.46E-06 \times (C1+C2)$	$7.00E-07 \times (C1+C2)$	$1.10E-06 \times (C1+C2)$
Secondary treatment infrastructure resources ^a	p	$1.22E-06 \times (C1+C2)$	$3.46E-06 \times (C1+C2)$	$7.00E-07 \times (C1+C2)$	$1.10E-06 \times (C1+C2)$
Electricity	kWh	$1.74E+02 \times (C1+C2)$	$4.95E+02 \times (C1+C2)$	$1.00E+02 \times (C1+C2)$	$1.58E+02 \times (C1+C2)$
Sludge treatment line					
Thickening					
Infrastructure resources	p	1.22E-06	3.46E-06	7.00E-07	1.10E-06
Electricity	kWh	1.02E+01	2.91E+01	5.89E+00	9.29E+00
Polymers	kg	9.54E-02	2.71E-01	5.49E-02	8.67E-02
Truck transport	t.km	9.54E-03	2.71E-02	5.49E-03	8.67E-03
Rail transport	t.km	5.73E-02	1.63E-01	3.30E-02	5.20E-02
Anaerobic digestion					
Infrastructure resources	p	1.22E-06	3.46E-06	7.00E-07	1.10E-06
Electricity	kWh	1.72E+01	4.89E+01	9.90E+00	1.56E+01
Grease (from primary treatment)	kg	6.89E+00	1.96E+01	3.97E+00	6.26E+00
Soft salt	kg	1.58E-03	4.50E-03	9.10E-04	1.44E-03
Truck transport	t.km	7.90E-05	2.25E-04	4.55E-05	7.18E-05
Dewatering					
Infrastructure resources	p	1.22E-06	3.46E-06	7.00E-07	-
Electricity	kWh	1.13E+01	3.20E+01	6.48E+00	-
Polymers	kg	6.84E-01	1.95E+00	3.94E-01	-
Rail transport	t.km	4.10E-01	1.17E+00	2.36E-01	-
Truck transport	t.km	6.84E-02	1.95E-01	3.94E-02	-
Struvite precipitation ^e					
Infrastructure resources	p	1.22E-06	3.46E-06	7.00E-07	1.10E-06
Electricity	kWh	1.17E+01	9.56E+00	2.82E+00	6.86E+00
H ₂ SO ₄	kg	-	-	-	1.11E+01
Na ₂ S	kg	-	-	1.00E+01	1.19E+01
NaOH	kg	1.10E+01	-	-	8.00E+00
Mg(OH) ₂ or MgCl ₂ or Ca(OH) ₂	kg	1.55E+00	1.24E+00	5.43E-01	7.66E-01
Polymers	kg	-	-	-	6.27E-01
Tap water	m ³	1.77E-03	2.42E-01	-	-
Resin	kg	2.65E+00	-	-	-
HCl	kg	1.99E-04	-	-	-
Rail transport	t.km	5.41E+00	-	2.12E+00	1.94E+01
Truck transport	t.km	1.52E+00	7.28E-01	1.03E+00	3.24E+00
Incineration (end-of-life)					
Infrastructure resources	p	-	-	7.00E-07	1.10E-06
Electricity	kWh	-	-	1.51E+01	1.86E+01
Heat	kWh	-	-	3.27E+00	4.04E+00

	Sand	kg	-	-	4.58E-02	5.65E-02
	Coke	kg	-	-	1.96E-02	2.42E-02
	Lime	kg	-	-	3.27E-01	4.04E-01
	NaOH (30%)	kg	-	-	1.08E+00	1.33E+00
	NH ₃ (25%)	kg	-	-	7.92E-01	9.77E-01
	Rail transport	t.km	-	-	4.70E-01	5.79E-01
	Truck transport > 32t	t.km	-	-	2.00E-01	2.46E-01
	Truck transport > 16t	t.km	-	-	2.59E-02	3.19E-02
	Barge transport	t.km	-	-	5.60E-02	6.90E-02
Transport (sludge end-of-life)						
	Truck transport 7.5t	t.km	3.12E-02	1.61E-01	-	-
	Tractor	kg	6.14E-04	2.18E-03	-	-
	Diesel	kg	1.04E-02	3.71E-02	-	-
	Spreader	kg	1.16E-03	4.13E-03	-	-
Spreading (sludge end-of-life)						
	Tractor	kg	4.70E-04	1.67E-03	-	-
	Diesel	kg	6.16E-03	2.18E-02	-	-
	Spreader	kg	3.49E-03	1.24E-02	-	-
<hr/>						
Avoided products						
Anaerobic digestion						
	Electricity	kWh	6.09E+01	1.59E+02	3.22E+01	5.07E+01
	Heat	kWh	1.01E+02	2.65E+02	5.36E+01	8.46E+01
Incineration (end-of-life)						
	Electricity	kWh	-	-	2.83E+01	3.49E+01
	Heat	kWh	-	-	1.15E+02	1.42E+02
Spreading (end-of-life)						
	Tractor	kg	5.78E-03	2.26E-02	-	-
	Spreader	kg	3.81E-03	1.49E-02	-	-
	Diesel	kg	8.85E-02	3.46E-01	-	-
	Truck transport 16t	t.km	7.87E-01	3.08E+00	-	-
	Rail transport	t.km	4.72E+00	1.85E+01	-	-
	Triple superphosphate (45% P ₂ O ₅) as P ₂ O ₅	kg	1.78E+00	9.24E+00	-	-
	Ammonitrate (35% N) as N	kg	5.72E+00	2.14E+01	-	-

^a Sewer network, pretreatment and infrastructures resources for water and sludge treatment are detailed in Supporting Information 2

^b Refers to the infrastructure unit for each process

^c Data from Irstea (2016)

^d Data from Jossa and Remy (2015)

^e Allocation factors C1 and C2 are used to allocate impacts of the sewer network, pretreatment and the water treatment line to sludge production (“×” is the multiplication sign).

Table 3. Life cycle inventory output data for the sludge-based phosphate fertilizer scenarios (S1, S2, S3 and S4)
 – Values are presented per functional unit: 1 kg of P produced

Outputs	Unit	S1-BioAcid	S2-Crystal	S4-AshDec	S3-Gifhorn
Products					
Struvite	kg	7.91E+00	3.57E+00	-	8.20E+00
“Rhenanite phosphate”	kg	-	-	2.77E+01	-
Wastewater treatment line – Emissions to air^{a,c}					
CO ₂	kg	2.18E+01 × (C1+C2)	6.19E+01 × (C1+C2)	1.25E+01 × (C1+C2)	1.98E+01 × (C1+C2)
N ₂ O	kg	2.72E-02 × (C1+C2)	7.74E-02 × (C1+C2)	1.57E-02 × (C1+C2)	2.47E-02 × (C1+C2)
N ₂	kg	2.06E+00 × (C1+C2)	5.87E+00 × (C1+C2)	1.19E+00 × (C1+C2)	1.87E+00 × (C1+C2)
Sludge treatment line – Emissions to air					
Thickening ^b					
CH ₄	kg	2.17E-01	6.16E-01	1.25E-01	1.97E-01
CO ₂	kg	5.97E-01	1.70E+00	3.44E-01	5.43E-01
N ₂ O	kg	1.29E-01	3.68E-01	7.44E-02	1.17E-01
Anaerobic digestion – biogas leakage					
CH ₄	kg	1.76E+00	4.63E+00	9.38E-01	1.48E+00
CO ₂	kg	2.61E+00	6.88E+00	1.39E+00	2.20E+00
N ₂	kg	6.17E-02	2.17E-01	4.40E-02	6.94E-02
Anaerobic digestion – cogeneration					
CO ₂	kg	6.70E+01	1.77E+02	3.58E+01	5.64E+01
CO	kg	5.08E-02	1.33E-01	2.68E-02	4.23E-02
NO _x	kg	4.67E-02	1.22E-01	2.47E-02	3.89E-02
VOC	kg	1.00E-02	2.62E-02	5.30E-03	8.36E-03
SO _x	kg	1.82E-02	4.75E-02	9.61E-03	1.51E-02
Dewatering and struvite precipitation ^b					
NH ₃	kg	1.92E-01	8.36E-01	1.69E-01	2.67E-01
Incineration (end-of-life)					
SO ₂	kg	-	-	3.99E-03	4.92E-03
NO _x	kg	-	-	1.59E-02	1.96E-02
NH ₃	kg	-	-	9.82E-04	1.21E-03
CO	kg	-	-	3.99E-03	4.92E-03
Dust	kg	-	-	7.85E-04	9.69E-04
HCl	kg	-	-	1.64E-03	2.02E-03
10% Hg	kg	-	-	3.73E-05	4.60E-05
N ₂ O	kg	-	-	6.48E-02	7.99E-02
Gypsum	kg	-	-	2.75E-03	3.39E-03
Truck transport > 16t	t.km	-	-	1.10E-04	1.36E-04
Storage ^b (end-of-life)					
CH ₄	kg	3.59E+00	9.48E+00	-	-
CO ₂	kg	9.91E+00	2.61E+01	-	-
N ₂ O	kg	1.05E-01	3.66E-01	-	-
NH ₃	kg	1.22E-01	4.35E-01	-	-
Spreading (end-of-life)					
NH ₃	kg	1.48E+00	5.70E+00	-	-
N ₂ O	kg	6.78E-01	3.20E+00	-	-

Cd	g	1.11E-01	3.15E-01	-	-
Cr	g	3.69E+00	1.05E+01	-	-
Cu	g	2.59E+01	7.35E+01	-	-
Hg	g	6.65E-02	1.89E-01	-	-
Ni	g	2.22E+00	6.30E+00	-	-
Pb	g	3.69E+00	1.05E+01	-	-
Zn	g	5.17E+01	1.47E+02	-	-
NO ₃ ⁻	kg	8.08E+00	3.12E+01	-	-
P	kg	4.43E-03	2.69E-02	-	-

Avoided products

Spreading (end-of-life)

P	kg	4.76E-03	2.46E-02	-	-
NO ₃ ⁻	kg	1.72E+00	6.41E+00	-	-
N ₂ O	kg	6.40E-01	2.80E+00	-	-
NH ₃	kg	7.39E-01	2.62E+00	-	-
As	mg	7.85E+00	4.06E+01	-	-
Cd	mg	7.85E+00	4.06E+01	-	-
Cr	mg	3.93E+01	2.03E+02	-	-
Cu	mg	3.93E+01	2.03E+02	-	-
Hg	mg	7.49E+00	3.88E+01	-	-
Ni	mg	3.03E+01	1.57E+02	-	-
Pb	mg	3.39E+01	1.75E+02	-	-
Zn	mg	4.64E+01	2.40E+02	-	-

^a Emissions were calculated from transfer coefficients, provided in Supporting Information 2

^b Emission factors for thickening, dewatering and storage came from Gourdet et al. (2017)

^c. Allocation factors C1 and C2 are used to allocate impacts of the sewer network, pretreatment and the water treatment line to sludge production (“×” is the multiplication sign).

Table 4. Allocation factors used to allocate burdens of inventory data to sludge and treated water production

Life Cycle Inventory data concerned by the allocation factors	Primary sludge (C1)	Biological sludge (C2)	Treated Water
Sewer network	0.58	0.18	0.24
Pretreatment	0.58	0.18	0.24
Wastewater treatment			
Primary treatment infrastructure	0.58	0.18	0.24
Secondary treatment infrastructure	0	0.42	0.58
Electricity	0	0.42	0.58
CO ₂	0	0.45	0.55
N ₂	0	0.37	0.63
N ₂ O	0	0.37	0.63

Table 5. Phosphorus recovery potential for sludge-based phosphate fertilizer scenarios

	S1-BioAcid	S2-Crystal	S3-AshDec	S4-Gifhorn
P entering the wastewater treatment plant (WWTP) (kg)	83 220	83 220	83 220	83 200
P entering the sludge line (kg)	52 195	52 195	52 195	52 195
% of P recovered in sludge after water treatment	63%	63%	63%	63%
P recovered as sludge-based phosphate fertilizer (kg)	27 423	9 639	47 622	30 197
% of P recovered / P in sludge after water treatment	53%	18%	91%	58%
% of P recovered / P entering the WWTP	33%	12%	57%	36%

Table 6. Mean ratios of gross impacts of sludge-based phosphate fertilizer production based on (R1) sludge-based fertilizer production process only, (R2) sludge production and sludge-based fertilizer production processes or (R3) the entire life cycle of sludge-based fertilizer (with or without sludge production) to impacts of triple superphosphate production

Impact category	S1-AcidBio				S2-Crystal				S3-AshDec				S4-Gifhorn			
	R1	R2	R3		R1	R2	R3		R1	R2	R3		R1	R2	R3	
			W/O sludge prod.	With sludge prod.			W/O sludge prod.	With sludge prod.			W/O sludge prod.	With sludge prod.			W/O sludge prod.	With sludge prod.
Abiotic Depletion (mineral resources)	38	45	40	47	0	20	5	25	47	51	52	56	105	118	116	129
Abiotic Depletion (fossil energy resources)	31	161	39	170	2	373	26	397	7	82	17	92	118	361	134	377
Acidification	7	20	502	515	1	39	1 686	1 724	2	10	23	31	64	89	74	99
Eutrophication	3	8	146	150	0	13	1 032	1 045	1	3	6	9	10	18	17	26
Climate change	89	136	630	676	3	135	13 571	13 703	7	33	127	154	557	644	922	1 009
Freshwater Aquatic Ecotoxicity	30	54	98	122	3	73	182	252	6	20	11	25	93	139	104	150
Marine Aquatic Ecotoxicity	8	15	11	18	1	21	8	28	2	6	3	7	25	38	28	41
Terrestrial Ecotoxicity	35	53	2 398	2 415	1	51	6 172	6 222	5	15	100	110	111	144	346	379
Human Toxicity	20	45	73	99	2	75	143	217	4	19	9	24	66	114	77	125
Ozone depletion	40	187	43	190	1	418	10	427	100	374	116	389	6	90	13	98
Photochemical oxidation	12	36	64	88	1	69	137	205	94	139	138	183	3	16	18	31

Table 7. Life cycle inventory of sludge-based fertilizer scenarios compared to triple super phosphate (TSP) for the production of 1 kg of recovered phosphorus

Values per FU ^a	Unit	S1-BioAcid	S2-Crystal	S3-AshDec	S4-Gifhorn	TSP
		Inputs				
Electricity	kWh	123.36 (11.67)	327.30 (9.56)	82.19 (2.82)	116.63 (6.86)	1.72
Heat	kWh	-	-	3.27 (-)	4.04 (-)	0.93
Reactant	kg	15.95 (15.17)	3.47 (1.24)	13.30 (10.58)	35.29 (32.41)	2.89
Transport	t.km	7.50 (6.93)	2.44 (0.73)	4.21 (3.15)	23.67 (22.69)	31.87
Infrastructure	p	3.49E-05 (1.22E-06)	9.63E-05 (3.46E-06)	2.02E-05 (7.00E-07)	3.07E-05 (1.10E-06)	1.90E-09
Outputs (emissions)						
CO ₂	kg	80.00 (-)	213.26 (-)	43.17 (-)	68.08 (-)	-
CH ₄	kg	5.56 (-)	14.73 (-)	1.06 (-)	1.68 (-)	-
NH ₃	kg	1.79 (-)	6.97 (0.84)	0.17 (-)	0.27 (0.27)	-
NO ₃	kg	8.08 (-)	31.19 (-)	-	-	-
N ₂ O	kg	0.92 (-)	3.96 (-)	0.14 (0.06)	0.21 (-)	-
CO	kg	0.05 (-)	0.13 (-)	0.03 (0.03)	0.05 (-)	-
NO _x	kg	0.05 (-)	0.12 (-)	0.04 (0.016)	0.06 (-)	-
VOC	kg	0.01 (-)	0.03 (-)	0.01 (-)	0.01 (-)	-
SO _x	kg	0.02 (-)	0.05 (-)	0.01 (-)	0.02 (-)	-
Heavy metals	g	87.34 (-)	248.26 (-)	3.73E-05 (3.73E-05)	4.60E-05 (-)	2.71E-07

^a Values in parentheses refer to the input used for the phosphorus recovery process only

Table 8. Amounts of reactants needed to recover 1 kg of phosphorus in each scenario

Sref	S1-AcidBio	S2-Crystal	S3-AshDec	S4-Gifhorn
2.2 kg phosphoric acid	11 kg NaOH	1.2 kg MgO	10 kg Na ₂ S	8 kg NaOH
0.7 kg phosphate rock	1.6 kg MgCl ₂ 2.6 kg resin		0.54 kg Ca(OH) ₂	11 kg H ₂ SO ₄ 0.63 kg polymers 12 kg Na ₂ S 0.77 kg Mg(OH) ₂