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RESEARCH ARTICLE

Environmental performances of production and land application of sludge-based phosphate fertilizers—a life cycle assessment case study

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Abstract

Phosphorus (P) is a non-renewable resource extracted from phosphate rock to produce agricultural fertilizers. Since P is essential for life, it is important to preserve this resource and explore alternative sources of P to reduce its criticality. This study aimed to assess whether fertilizing with sludge-based phosphate fertilizers (SBPF) can be a suitable alternative to doing so with fertilizers produced from phosphate rock. Environmental impacts of production and land application of SBPF from four recovery processes were compared to those of two reference scenarios: triple super phosphate (TSP) and sewage sludge. To avoid bias when comparing scenarios, part of the environmental burden of wastewater treatment is allocated to sludge production. The CML-IA method was used to perform life cycle impact assessment. Results highlighted that production and land application of SBPF had higher environmental impacts than those of TSP due to the large amounts of energy and reactants needed to recover P, especially when sludge had a low P concentration. Certain environmental impacts of production and land application of sewage sludge were similar to those of SBPF. Sensitivity analysis conducted for cropping systems highlighted variability in potential application rates of sewage sludge or SBPF. Finally, because they contain lower contents of heavy metals than sewage sludge or TSP, SBPF are of great interest, but they require more mineral fertilizers to supplement their fertilization than sewage sludge. Thus, SBPF have advantages and disadvantages that need to be considered, since they may influence their use within fertilization practices.

Keywords Life cycle assessment · Phosphorus recovery · Sludge-based phosphate fertilizers · Mineral phosphate fertilizers · Struvite

Introduction

Phosphorus (P) is a critical element for living organisms that cannot be replaced by any other element in fertilizers. With the human population estimated to grow to 9 billion people by 2050, demand for food and consequently fertilizers will dramatically increase worldwide (Sorensen et al. 2015; Steen 2006), since food production monopolizes 90% of global demand for P (Cordell et al. 2009). P is extracted mainly from phosphate rock reserves (i.e., 148 million t per year). In 2016, 86% of these reserves were controlled by only six countries, making the availability of phosphate rock subject to high geopolitical risks (USGS 2018). Consequently, since the European Union's (EU) supply of phosphate rock depends completely on imports (reserves are not mined in the EU) and because of its economic importance, phosphate rock was added to the EU's list of critical raw materials in 2017 (European Commission 2017). The risk posed to the supply can be reduced, however, only by increasing the end-of-life

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recycling input rate from waste feedstock since there is no substitute for phosphate rock. Waste recycling is also one pillar of the circular economy concept and is integrated within the "Energy Transition for Green Growth Act" of the French Ministry of Environment, Energy and the Sea (MTES 2015).

There is thus a growing interest in exploring alternative sources of P, especially from waste. Humans excrete 98% of the P they ingest (Kalmykova et al. 2015). This P ends up in wastewater and sewage sludge, making it an attractive resource for P recovery. P is found in both mineral and organic forms in sludge, which can be applied directly to the soil as an organic fertilizer (Houot et al. 2014). Sludge application increases soil fertility because of the minerals and organic matter that sludge provides (Dodd and Sharpley 2015). While quantities of sewage sludge increase as population increases (Charlton et al. 2016), Kleemann et al. (2015) point out that no regulations limit the amount of P that can be applied from urban sludge. In vulnerable zones in the EU, the amount of sludge allowed to be applied is governed by the Nitrates Directive and calculated using nitrogen (N) as the element that limits application (here, "limiting element"). Since the amount of P is not regulated, it can be applied in excess (relative to plant requirements) and cause pollution (Amery and Schoumans 2014). In addition, the presence of heavy metals, such as copper, zinc, and cadmium, in sewage sludge is also a source of concern because plants can easily absorb them and thus pass them into human diets, posing public health problems (Houot et al. 2014; Lindberg et al. 2007). Some countries have already tightened regulations on fertilizer application to address this problem (Linderholm et al. 2012). Finally, land application of sludge also poses problems of social acceptance, mainly due to the odors emitted by sludge. Due to these constraints, new technologies have been developed to extract and recover this P in a form that can be manipulated easily as magnesium ammonium phosphate (struvite, $NH_4MgPO_4 \cdot 6H_2O$) or calcium sodium phosphates (Rhenania phosphate). Unlike Rhenania phosphate, struvite has the advantage of also providing N to crops in the form of ammonium. In addition, using these sludge-based phosphate fertilizers (SBPF) instead of sewage sludge in a fertilizing plan is made easier since they contain mainly P, unlike sludge, for which it is necessary to consider not only P but also N, potassium, and organic matter. In addition, a multicriteria assessment performed by Möller et al. (2018) highlighted benefits of SBPF (struvite and Rhenania phosphate), since they have higher P fertilizer value, lower risk of causing potentially toxic elements to accumulate, and lower risk of negative impacts from organic contaminants than dewatered sludge. According to previous studies, P is recovered mainly as struvite or Rhenania phosphate, and this recovery is more efficient before and after anaerobic digestion or from sewage sludge ashes (Egle et al. 2015, 2016). In addition, many studies describe struvite as a "slow-diffusion fertilizer" (Bashan and Bashan 2014; El Diwani et al. 2007; Rahman et al. 2014) that has the same fertility value as a conventional fertilizer (Montag et al. 2007). The minerals it releases gradually would not exceed plant requirements and would thus be less subject to leaching, runoff, and other indirect N losses than conventional fertilizers (Rahman et al. 2014). Use of struvite would also require less frequent application than those of conventional fertilizers (Münch and Barr 2001).

Some studies have assessed environmental impacts of struvite precipitation in wastewater treatment sludge, but few studies have considered environmental impacts of its application to land using life cycle assessment (LCA) (Sena and Hicks 2018). LCA is a framework for assessing environmental impacts based on international standards (ISO 2006a, b). Johansson et al. (2008) and Linderholm et al. (2012) assessed use of P recovered from sewage sludge as fertilizer on agricultural soils. Johansson et al. (2008) highlighted that supercritical water oxidation provided the best P-recovery option. Linderholm et al. (2012) highlighted that directly applying sewage sludge to soil was the option with the lowest energy use and greenhouse gas emissions due to the beneficial association with N in sludge. Bradford-Hartke et al. (2015) compared environmental benefits and impacts of recovering P as struvite from dewatering return liquor using four centralized and two decentralized systems. They estimated that operational savings and avoided fertilizer production offset the energy and chemical use required, resulting in positive environmental impacts of P recovery. In these comparative LCAs, sewage sludge was considered as waste; thus, no environmental burdens were allocated to its production.

However, sludge can no longer be considered as waste but rather must be considered as a coproduct of a wastewater treatment plant (WWTP), since sludge treatment is specifically designed to produce SBPF with high added value (Pradel et al. 2016). This assertion is also shared by several other researchers, who consider that the status of "waste" is subjective and questionable, especially when high nutrient- or energy-recovery potential is expected, and who question the "zero burden" assumption (Cleary 2010; Holden 2015; Oldfield and Holden 2014; Oldfield et al. 2018). Despite this assertion, most LCA studies investigating P recovery still consider sludge as waste, even when struvite is applied to agricultural land as fertilizer (Sena and Hicks 2018).

This study therefore aimed to assess whether fertilizing with struvite recovered from sludge can be a suitable alternative to crop fertilization with mineral fertilizers from phosphate rock. To do so, four fertilization scenarios, each using struvite recovered from a specific recovery process, were compared to two fertilization reference scenarios, one using sewage sludge and the other using triple super phosphate (TSP). To avoid bias when comparing scenarios, SBPF was considered a coproduct of the WWTP, and part of the environmental burden of wastewater treatment was allocated to sludge production.

Materials and methods

In LCA, the system to be studied (boundaries, functions, and the related functional unit) is first defined within the goal and scope of the study, as are the scenarios assessed and assumptions made. Then, raw material, energy, and emissions used by each process are described within the life cycle inventory (LCI) step. The third step, called life cycle impact assessment (LCIA), consists of estimating environmental impacts of the system by relating the inventory flows in the LCI to their respective environmental impacts. Finally, results are interpreted and sensitivity analyses are performed.

Goal and scope definition

Following a request from the French Agency for Biodiversity (AFB—Agence Française pour la Biodiversité), LCA was used to assess environmental impacts of fertilization practices with struvite obtained from different recovery processes compared to sewage sludge and TSP.

System boundaries

Since SBPF were compared to conventional fertilizers in a comparative LCA, upstream processes of sludge production had to be considered. Indeed, when considering sludge as a coproduct, the wastewater treatment line becomes a multifunctional process that provides both sludge and "clean water" that are given a second life. To address this multifunctional process, the environmental burdens of the wastewater treatment line (i.e., sewer network, pretreatment, primary and biological treatments) were allocated to sludge (hereafter, "sludge production") and included within the system boundaries. The system boundaries also included sludge treatment (i.e., thickening, anaerobic digestion, dewatering), P-recovery processes (i.e., struvite precipitation), struvite storage and application to land, as well as storage, land application, and incineration of sewage sludge (Fig. 1).

System boundaries of the reference scenario (i.e., production of TSP as mineral phosphate fertilizer) included phosphate rock extraction and beneficiation as well as production of phosphoric acid, commonly used in fertilizer production. For each process, mass balances of each nutrient (carbon, N, and P) in sludge were estimated and used to calculate emissions to air, water, and soil. Construction of buildings and equipment used for each unit process (hereafter, "infrastructure") were included within the system boundaries, while their dismantlement was not, following recommendations of Corominas et al. (2013).

Function and functional unit

Since P has low mobility in the soil, it remains in the soil's solid phase and is gradually released depending on crop requirements. Since there is usually no advantage in staggering mineral P inputs, they are usually applied before cultivation (COMIFER 2017). Thus, unlike N, P is not applied to the crop every year and, in soils with enough P, phosphate fertilization can be planned at the scale of a crop rotation (ARVALIS 2013). We thus assumed that P was applied once per crop rotation and that it diffused progressively into the soil over several years.

The two-year "straw cereal–oilseed" crop rotation is the most common in France, covering 24% of the area planted in field crops, mainly with rapeseed as the oilseed throughout France (AGRESTE 2014). The three-year "rapeseed–wheat–winter barley" crop rotation is characteristic of conventional agriculture in the Centre region of France and covers at least

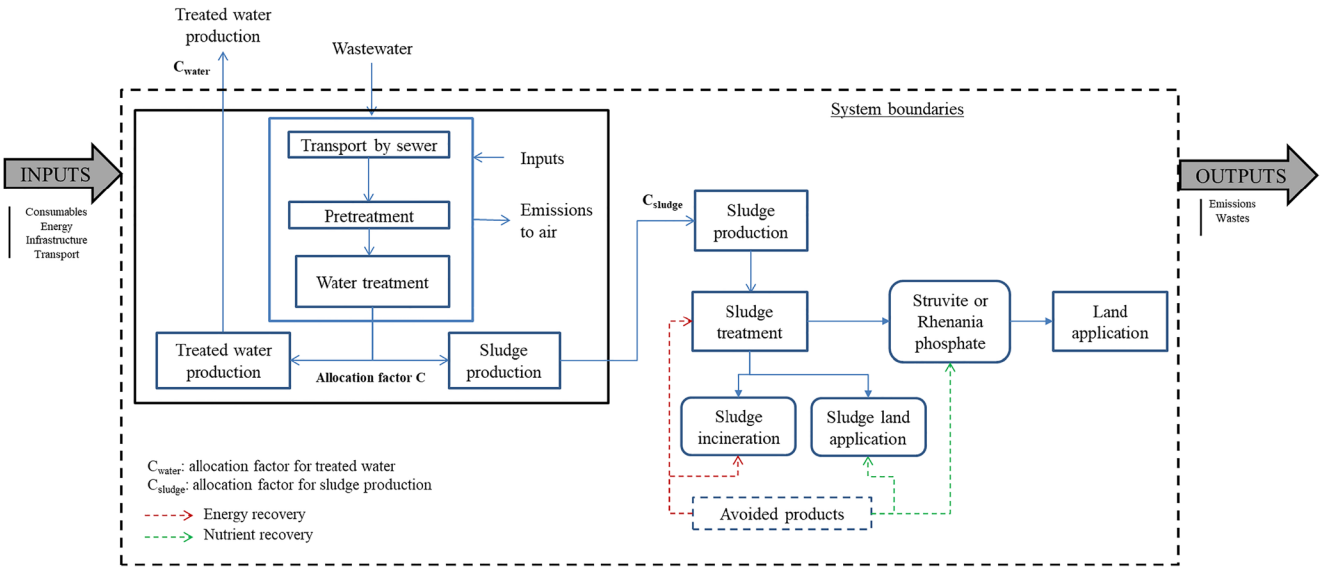

Fig. 1 System boundaries for the land application of SBPF

60% of its arable land (Ecophyto2018—Centre Region Fact Sheet). Since agricultural organizations recommend longer rotations and alternating winter and spring crops to break biological cycles of weeds, diseases, and pests, we assumed inclusion of a legume (pea) in the crop rotation (AGRESTE 2014). The function of the phosphate fertilizer in this study was thus to meet the P requirements of a four-year crop rotation (rapeseed–wheat–winter barley–pea) in France. Phosphate fertilization is performed "at the head of rotation," in which rapeseed requires the most P (COMIFER 2009). It can thus be assumed that P applied at rapeseed sowing will be used most effectively by plants at the scale of the crop rotation.

The functional unit was based on the amount of P needed to meet the crop rotation's P requirement, which was estimated from each crop's mean expected yield (2010–2014, France) and P content (Table 1). Because crops in the rotation were estimated to require 60.3 kg P ha^{-1}, the functional unit was defined as "production and land application of 60.3 kg ha^{-1} of P available for plants in mineral form for an entire crop rotation of rapeseed–wheat–winter barley–pea in France." The term "mineral form" refers to the mineral P in sludge, struvite, Rhenania phosphate, or TSP.

Four scenarios of production and land application of struvite from sludge according to four P-recovery processes were studied which are as follows: biological acidification before anaerobic digestion (S1-BioAcid), P crystallization using dewatering return liquor (S2-Crystal), and recovery from sewage sludge ashes or dewatering using the AshDec® process (S3-AshDec) or Gifhorn® process (S4-Gifhorn), respectively. Two reference scenarios ($Sref_{min}$ and $Sref_{sludge}$) were also used for comparison, providing the functional unit with TSP or sewage sludge, respectively (Fig. 2).

SBPF production and land application have additional functions. The first one is energy recovery, which occurs during anaerobic digestion and cogeneration of the resulting biogas and from incineration. The heat and electricity produced during these processes decrease the amount of external heat and electricity taken from the grid, either to maintain the temperature of the digester or because they provide a self-generated source of electricity. System expansion was used to model the energy that leaves the system as a coproduct, leading to avoided heat and electricity production. The second additional function is the production of sewage sludge with a low P content, which can be applied to land as an organic fertilizer. Like for heat and electricity, system expansion was used and avoided production of mineral fertilizers was assumed (Fig. 1).

Description of the scenarios

Each scenario included fertilizer production, sludge treatment, and sludge end-of-life (Fig. 2). The scenarios used in this study came from Pradel and Aissani (2019) but were extended to consider land application of SBPF. The first reference scenario ($Sref_{min}$) assumed production of TSP, which results from a reaction between 30% phosphate rock (from Morocco) and 70% phosphoric acid (by mass). After being mined, phosphate rock is processed (crushed, washed, dried, etc.), yielding a product containing 33% P_2O_5. Phosphoric acid is produced by dissolving phosphate rock with sulfuric acid using the dihydrate wet process. Phosphoric acid contains 50.7% of P_2O_5 per kg. Once processed, the final product (TSP) contains 48% P_2O_5 (Althaus et al. 2007; Nemecek and Kägi 2007).

The SBPF were assumed to be produced by a French WWTP with a capacity of 300,000 population equivalent. WWTP characteristics are provided in Section 8 of the Supporting Information (SI). Input P flows monitored in the wastewater treatment line in 2016 were used for this study (Irstea 2016). P flows from wastewater, sewage sludge treatment, and P-recovery processes were calculated using transfer coefficients (Doka 2007) and process performances provided by the WWTP owner (Tables SI-1 and SI-2). The wastewater treatment line is composed of pretreatment and primary treatment lines that produce primary sludge, followed by

Table 1 Amount of P required by crops for the entire crop rotation

Crop	Estimated yield[a] (t FM ha^{-1})	Dry matter (%)[b]	P_2O_5 content (kg t DM^{-1})[b]	P content (kg t DM^{-1})[c]	Total P required by crops[d] (kg ha^{-1})
Rapeseed	3.37	91	12.5	5.5	16.9
Wheat	6.88	85	6.5	2.8	16.4
Winter barley	6.34	85	6.5	2.8	15.1
Pea	3.95	86	8	3.5	11.9
Total					60.3

FM fresh matter, *DM* dry matter

[a] Mean yield calculated from French yields from 2010 to 2014 (data from http://www.fao.org/faostat/en/#data/QC)

[b] COMIFER (2009)

[c] Derived from P_2O_5 values

[d] Equals P content × DM percentage × estimated yield

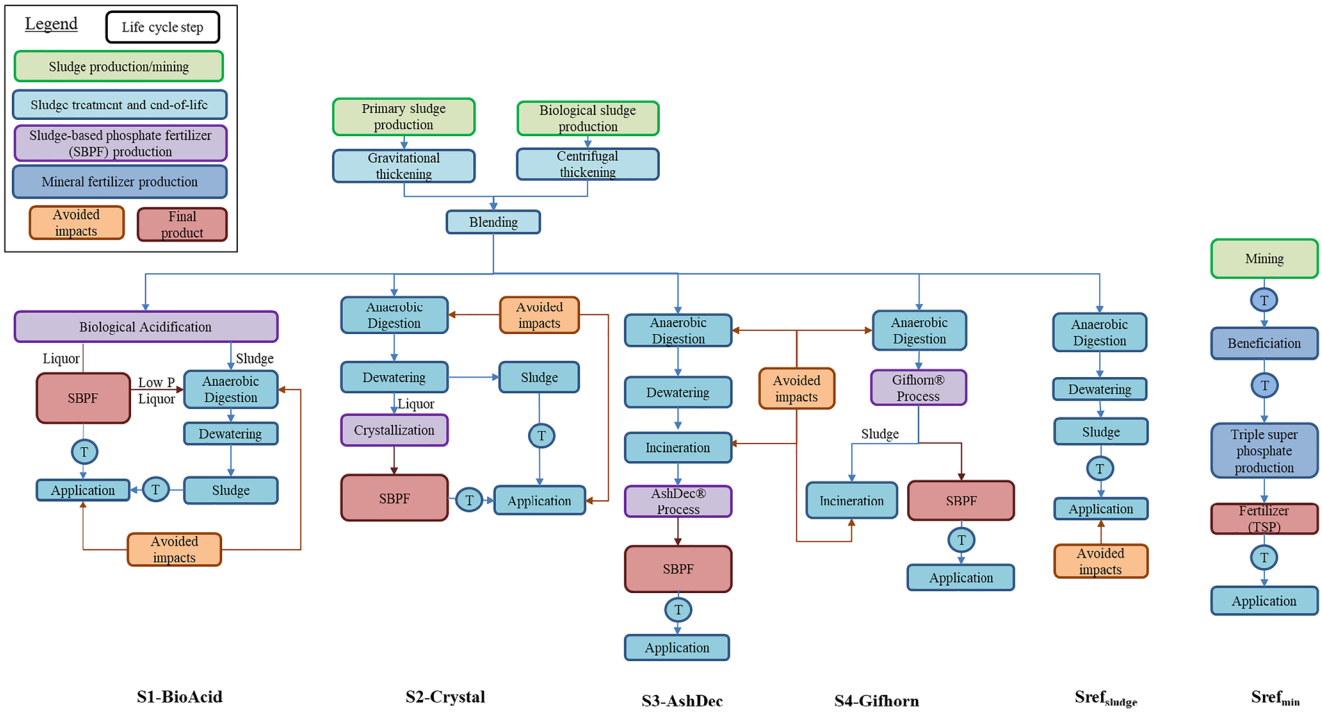

Fig. 2 Life cycle steps of SBPF scenarios (S1-BioAcid, S2-Crystal, S3-AshDec, S4-Gifhorn) and reference scenarios ($Sref_{min}$, $Sref_{sludge}$)

biological treatment lines that produce biological sludge; the two types of sludge are then thickened and blended at a 65:35 ratio, respectively, before entering an anaerobic digester (Fig. 3). Of the 83,220 kg of P that entered the WWTP, 52195 kg (63%) was recovered in sludge after wastewater treatment. The second reference scenario ($Sref_{sluge}$) assumed that no P is recovered: after anaerobic digestion, the sludge is dewatered and then stored and applied to land.

In S1-BioAcid, P is separated from organic matter using biological dissolution by acidification, which involves two types of microorganisms. An organic substrate (i.e., the fermentable fraction of organic waste) is transformed into volatile fatty acids (VFAs) by acidifying bacteria. These VFAs acidify the sludge and dissolve calcium and magnesium phosphates or P adsorbed on iron hydroxide. Then, under anaerobic conditions, polyphosphate-accumulating organisms (PAOs) that are naturally present in biological sludge use these VFAs to release P. The acidification does not inhibit P release by the PAOs and prevents macro-elements, such as calcium, magnesium, and iron from precipitating with the P. After biological acidification, polymers are added to improve liquid/solid separation during centrifugation. Finally, 60% of the P in sludge is recovered within the liquid fraction. The solid fraction of sludge is sent to anaerobic digestion, while the

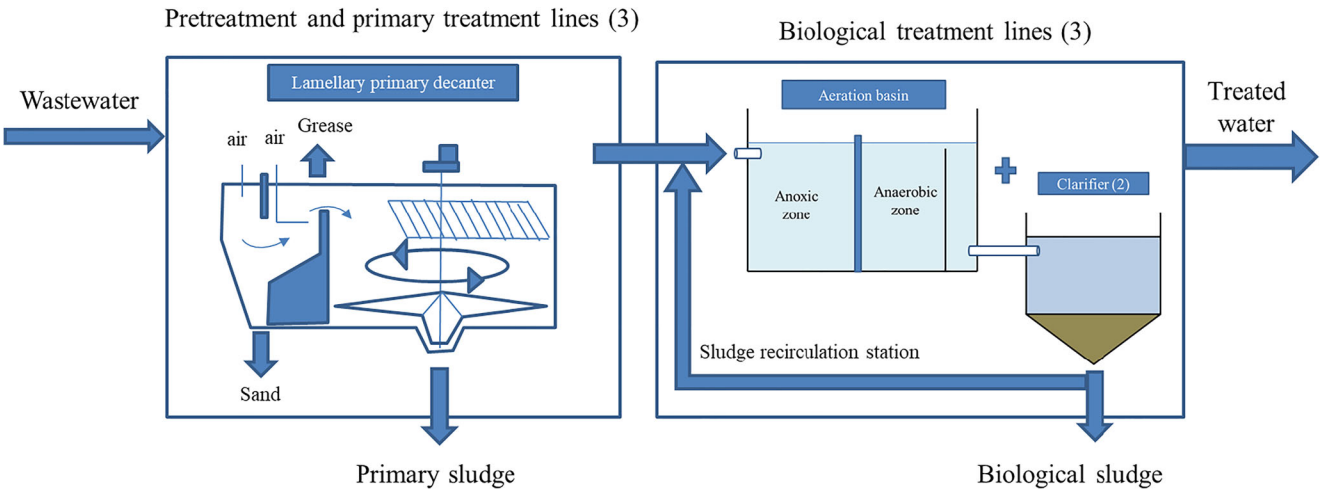

Fig. 3 Description of the wastewater-treatment line used to produce sludge (numbers in parentheses refer to the number of primary and biological treatment lines)

return liquor, high in solubilized P, is purified using a cationic resin. This resin captures excess iron to prevent formation of ferric phosphate during the crystallization step. The iron trapped in the resin is recovered using hydrochloric acid and reused by the WWTP. Magnesium chloride and sodium bicarbonate are added to produce struvite and increase the pH, respectively. The liquor remaining after crystallization is injected into the anaerobic digester. The SBPF, composed of 65% struvite and 35% calcium and magnesium carbonates, is then stored. After anaerobic digestion, the sludge is dewatered, stored, and applied to land. P recovery equals 27,423 kg (i.e., 53% and 33% of the P entering the sludge line and wastewater-treatment line, respectively).

In S2-Crystal, sludge is anaerobically digested and dewatered. P is crystallized in the dewatering return liquor using magnesium oxide. The final product is composed of 90% struvite. Like in S1-BioAcid, the remaining sludge, once dewatered, is stored and applied to land. P recovery equals 9639 kg (i.e., 18% and 12% of the P entering the sludge line and wastewater-treatment line, respectively).

In S3-AshDec, the AshDec® process is used to recover P (Jossa and Remy 2015). This process mixes pre-heated sewage sludge ashes with sodium sulfate and a reducing agent (e.g., dried sewage sludge). This mixture is heated using natural gas to 900–1000 °C in a rotary kiln for at least 20 min. During heating, P in ashes is transformed into $NaCaPO_4$, a plant-available mineral form. Volatile heavy metals (e.g., arsenic, cadmium, mercury, lead, zinc) are evaporated and removed with the gas phase. Off-gasses are also treated to decrease dust (since fly ash contains heavy metals) and combustion gasses to acceptable limits. The final product of the AshDec® process ($NaCaPO_4$) is similar to Rhenania phosphate. Besides the P product and off-gasses, the process generates no other waste. P recovery equals 47,622 kg (i.e., 91% and 57% of the P entering the sludge line and wastewater-treatment line, respectively).

In S4-Gifhorn, the Gifhorn® process is used to recover P (Jossa and Remy 2015). Unlike S1-BioAcid, digested sludge is acidified in a first reactor using sulfuric acid. The pH decreases to 4.5, which dissolves the phosphate chemically bound to the sludge into the liquor, as well as large amounts of metals (iron and heavy metals). To avoid transferring these metals to the final P product, Na_2S is added, and the dissolved metals are precipitated as sulfides. The pH is then raised to 5.6 by adding NaOH. The P-rich liquor and sludge are then separated in a centrifuge with added polymers. In a second reactor, a small dose of $Mg(OH)_2$ is used to precipitate P in the liquor and raise the pH to 9; if necessary, NaOH is added to control pH. The final product is composed of struvite, and the remaining sludge is incinerated. P recovery equals 30,197 kg (i.e., 58% and 36% of the P entering the sludge line and wastewater treatment line, respectively).

Life cycle inventory

LCI data for the reference scenario with mineral phosphate fertilizer ($Sref_{min}$) were those for TSP production from the ecoinvent v2.2 database ("RER: triple superphosphate, as P_2O_5, at regional storage") (Ecoinvent Professional Database 2007). LCIs for scenarios for SBPF (S1-BioAcid, S2-Crystal, S3-AshDec, S4-Gifhorn) and the sludge reference scenario ($Sref_{sludge}$) were modeled according to the French WWTP, with input data from the ecoinvent v2.2 database. LCI data for avoided fertilizers, modeled as ammonium nitrate, came from the ecoinvent v2.2 database ("RER: Ammonium nitrate, as N, at regional storage"). SBPF and sludge reference scenarios assumed 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). LCI input and output data are detailed in Section 6 of the Supporting Information.

LCI data used to allocate environmental burdens to sludge production

LCI data for the sewer network and wastewater treatment line came from technical documents provided by the WWTP owner. They were modeled according to Risch et al. (2015). Emissions from the wastewater treatment line were calculated according to carbon, N, and P mass balances using transfer coefficients from the literature and expert opinion (Doka 2007) (Fig. SI-1). The method developed by Pradel et al. (2018) was used to allocate burdens of the wastewater treatment line to primary and biological sludge production. Allocation factors (Table 2) were based on process-related (oxygen demand and alternation of aerobic and anoxic periods) and product-related (carbon and N content ratio, P assimilation and chemical precipitation) parameters. Detailed calculation of the allocation factors used is provided in the Supporting Information of a previous article (Pradel and Aissani 2019).

LCI data for sludge treatment

Performances of sludge treatment processes and assumptions used to calculate emissions from the sludge treatment mass balance are shown in Table SI-1. Data for gravitational thickening came from Gourdet et al. (2017) for methane (CH_4), nitrous oxide (N_2O), and carbon dioxide (CO_2) emission factors. No emissions were modeled for centrifugal thickening, which uses polymers composed of 50% nitric acid and 50% acrylonitrile.

Mesophilic anaerobic digestion was modeled, with biogas loss (due to uncontrolled leakage, which emits CH_4 and biogenic CO_2 to air) estimated at 10% (upper value from IPCC (2006b)). The biogas not lost is used to cogenerate electricity,

Table 2 Allocation factors used to allocate burdens of life cycle inventory (LCI) data to production of sludge and treated water (adapted from Pradel and Aissani 2019)

LCI data	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
Carbon dioxide	0	0.45	0.55
Nitrogen gas	0	0.37	0.63
Nitrous oxide	0	0.37	0.63

and it was assumed that all CH_4 in the biogas is transformed into CO_2. Biogas combustion generates several other emissions, such as N oxides (NO_x) and sulfur oxides (SO_x) (INERIS 2002; RDC Environnement 2007). Cogeneration also generates additional emissions, such as NO_x, SO_x, and carbon monoxide, from incomplete combustion of biogas and non-CH_4 volatile organic compounds. Polymers used during press-filter dewatering are composed of 50% nitric acid and 50% acrylonitrile as reactants.

Dewatered low-P sludge was assumed to be stored for six months in a concrete area in the WWTP in S1-BioAcid and S2-Crystal but incinerated in S3-AshDec and S4-Gifhorn. LCI data for emissions from sludge storage (i.e., ammonia (NH_3), N_2O, CH_4, and CO_2) came from Gourdet et al. (2017), while those from incineration came from Jossa and Remy (2015).

LCI data for SBPF production

Data for biological acidification, which came from an experimental pilot (Daumer 2015), were used to model the P-recovery process at the scale of the WWTP. Reactant use and energy consumption in the WWTP were assumed to be proportional to those in the pilot (M.L. Daumer, pers. comm.). Since emissions were not measured in the pilot; however, it was not possible to include emissions in the LCI for biological acidification. The remaining emissions of chlorine from adding magnesium chloride to precipitate P were included. Data for struvite crystallization in S2-Crystal came from an industrial process in another French WWTP. Data for struvite precipitation using the Gifhorn® or AshDec® process came from Jossa and Remy (2015). Process performances for each scenario are summarized in Table SI-2.

LCI data for transport and land application

After storage, sludge and SBPF are transported and applied to land. In S1-BioAcid and S2-Crystal, both SBPF and low-P sludge are applied to land. The amount of each product needed to reach the functional unit was calculated as a function of its P content. In the other scenarios, P is provided only by the SBPF (S3-AshDec, S4-Gifhorn), sludge ($Sref_{sludge}$), or mineral fertilizer ($Sref_{min}$) (Table 3).

SBPF and sludge were assumed to be transported in two steps: transported 40 km (Linderholm et al. 2012) by tractor-trailer (7.5–16 t) from the WWTP to intermediate storage and then loaded in a spreader and transported 4 km to the field (Pradel 2016). The loading was considered negligible (Pradel 2010). Fuel consumption, heavy metal emissions due to tire abrasion, and fuel combustion were included in the LCI and are detailed in Section 3 of the Supporting Information.

N and P emissions were calculated according to IPCC (2006a) and Nemecek and Kägi (2007), respectively. Emissions of heavy metals in sludge were included, but since no data on them were available, values from Houot et al. (2014) were used. Emission factors, heavy metal content, and agronomic characteristics for each fertilizing product are reported in Section 3 of the Supporting Information.

Table 3 Origin and amount of P (kg) needed to reach the functional unit by scenario

Scenario	P from mineral fertilizer	P from sludge	P from sludge-based fertilizer
S1-BioAcid	–	26.4	33.9
S2-Crystal	–	48.3	12.0
S3-AshDec	–	–	60.3
S4-Gifhorn	–	–	60.3
$Sref_{sludge}$	–	60.3	–
$Sref_{min}$	60.3	–	–

LCI data for avoided products

The avoided conventional fertilizer was assumed to be ammonium nitrate (35% N) whose amount was based on the amount and bioavailability of N in struvite. The bioavailability of N in synthetic mineral fertilizers is estimated at 100% (Bisinella de Faria et al. 2015), while that in struvite is subject to discussion and the topic of several studies, including pot experiments over several months or years. These studies show that plants absorb the ammonium in struvite as much as the nitrate in conventional fertilizers (i.e., 100%) (Bradford-Hartke et al. 2015; Cabeza et al. 2011; Egle et al. 2016; Rahman et al. 2014; Thibodeau et al. 2014; Zapka and Muskolus 2015). In contrast, since some of the N in sludge is organic, only 50% of it was estimated to become bioavailable for crops (Pradel 2016). Avoided ammonium nitrate impacts were considered for both struvite and sludge in S1-BioAcid and for struvite in S4-Gifhorn. Since struvite supplies almost no N, avoided ammonium nitrate impacts were considered only for sludge in S2-Crystal. Since the final product of the AshDec® process contains no N, no substitution was assumed in the S3-AshDec scenario. Avoided heat and electricity were modeled as the "heat, natural gas, at boiler modulating < 100 kW" and "electricity, production mix FR" ecoinvent V2.2 processes, respectively, for anaerobic digestion in each scenario and incineration in the S3-AshDec and S4-Gifhorn scenarios.

Life cycle impact assessment

Scenarios were modeled using GaBi® v6 LCA software to ensure energy and mass balances. GaBi can also trace mass and energy flows and perform parameterized modeling. Details of the scenario modeling with GaBi® are provided in Section 6.7 of the Supporting Information. Potential environmental impacts of each scenario were estimated using the most recent characterization factors (January 2016) of the CML-IA method (Guinée et al. 2002), developed by the Leiden Institute of Environmental Sciences (Centrum voor Milieuwetenschappen—CML). Eleven impact categories were included which are as follows: mineral reserve-based resource depletion (AD element), fossil energy resource depletion (AD fossil), acidification (Acid), eutrophication (Eutro), climate change including biogenic emissions (CC), freshwater aquatic ecotoxicity (FAET), marine aquatic ecotoxicity (MAET), terrestrial ecotoxicity (TET), human toxicity (HT), ozone depletion (OD), and photochemical oxidation (POC).

Sensitivity analysis

Sensitivity analysis is often performed in LCA to highlight the robustness of results and their sensitivity to data, assumptions, and models.

Sensitivity analysis to system modeling

A first sensitivity analysis was conducted to highlight the variability caused by the system modeling (reference flow, allocation procedure). To do so, results on net impacts were assessed for S1-BioAcid and S2-Crystal alternative scenarios for which only SBPF were used to reach the functional unit. Sensitivity analyses that considered the sludge as a waste or not (i.e., by allocating environmental burdens of sludge production to it or not) and inclusion of capital goods within the system boundaries (i.e., by considering different lengths of sewer network) are available in a previous article (Pradel and Aissani 2019).

Sensitivity analysis to the choice of cropping system and heavy metals in fertilizers

A second sensitivity analysis was conducted to highlight the variability caused by the choice of cropping system. Because SBPF provide much P and little N to crops, it is easily to calculate P application rates for them. In contrast, land application of sewage sludge supplies crops with P, N, and organic matter, which makes it more difficult to calculate application rates. These fertilizers differ mainly their ability to provide required nutrients to crops and their heavy metal content. To explore variability due to these differences, sensitivity analysis was performed concerning (a) the limiting element and its influence on the need for additional mineral fertilizers to meet crop requirements and (b) the heavy metal content of these fertilizers compared to those of the French legislation.

Description of the cropping systems used in the sensitivity analysis The sensitivity analysis was based on eight cropping systems observed on French sites (Mt1, Mt2, Kl1, Kl2, Kl3, Ms1, Pt1, and Cx1), each with a specific crop rotation and a land application period (Table 4). These cropping systems cover a wide range of French soils, weather, and agronomic conditions (Pradel et al. 2013). The optimal application rate of sewage sludge or struvite was based on their agronomic characteristics (Tables SI-5, SI-6, and SI-7) and determined for each cropping system, according to plant nutrient balances and French legislation, especially that dealing with heavy metal thresholds. N and P balances were calculated according to the requirement of the receiving crop(s) and the entire crop rotation, respectively (Table 4).

Impact of fertilizer application rate according to the limiting element Sustainable fertilization practices need to supply sufficient nutrients to crops. Since fertilizers (e.g., sewage sludge) may contain several nutrients, application rates were calculated based on the limiting element (i.e., the element that first met crop requirements). The remaining elements were supplied by specific additional mineral fertilizers. The optimal

Table 4 Description of the cropping systems studied

French site	Crop rotation	Application period	Receiving crop(s)	N requirement of receiving crop(s) (kg N ha^{-1})	P requirement of the crop rotation (kg P_2O_5 ha^{-1})
Montoldre (Mt1)	RS, WW, WB	Late July	WB	115	132
Montoldre (Mt2)	RS, WW, WB	August	RS	206	132
Kerlavic (Kl1)	WW, CS	Late February	MS	48	102
Kerlavic (Kl2)	WW, CS	March	WW	150	102
Kerlavic (Kl3)	TG, maize, WW	Late February	TG followed by maize	28	140
Mons (Ms1)	SB, WW, WB	Late July	SB	148	128
Poitou (Pt1)	RS, WW, sunflower, WW	August	RS	185	122
Pays de Caux (Cx1)	FF, NCC, SB, WW, WB	August	NCC followed by SB	114	130

RS rapeseed, *WW* winter wheat, *WB* winter barley, *MS* maize silage, *TG* temporary grassland, *SB* sugar beet, *NCC* N catch crop, *FF* fiber flax

application rate was obtained by dividing the amount of N or P_2O_5 needed to meet crop requirements for N or P, respectively, by the amount of each that was bioavailable in the fertilizer and then choosing the smaller of the two application rates. These application rates were calculated for the following three kinds of fertilizers: (a) sewage sludge with low-P content from S1-BioAcid and S2-Crystal, (b) sewage sludge from $Sref_{sludge}$, and (c) SBPF from S1-BioAcid, S2-Crystal, S3-AshDec, and S4-Gifhorn.

Results and discussion

Environmental impacts of SBPF, sewage sludge, and mineral phosphate fertilizer

The $Sref_{min}$ scenario had slightly lower impacts than the SBPF scenarios, except for MAET, which equaled those of $Sref_{sludge}$ and S2-Crystal (Fig. 4a). $Sref_{sludge}$ and S2-Crystal had similar impacts for AD element, AD fossil, OD, and POC. S1-

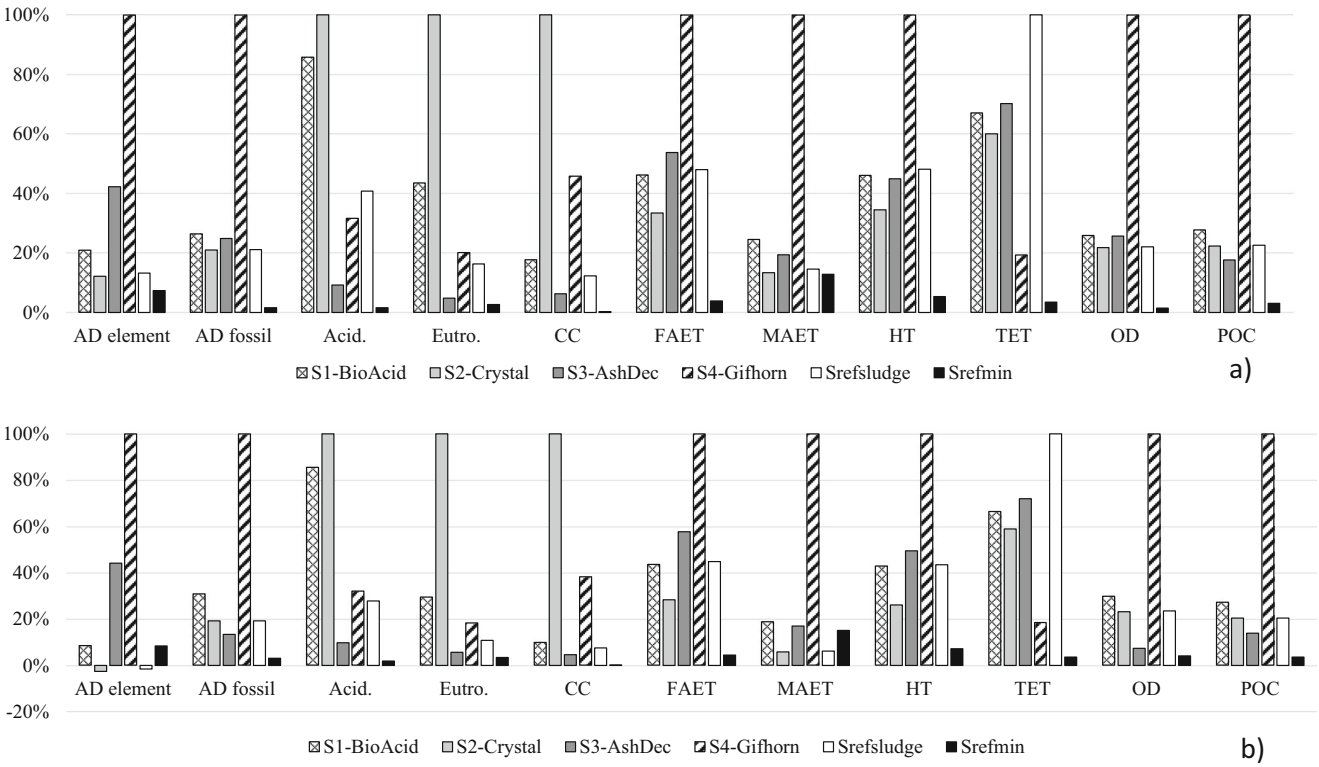

Fig. 4 Relative (**a**) gross impacts and (**b**) net impacts (gross impacts minus avoided impacts) of each scenario for each impact category (AD element, mineral reserve-based resource depletion; AD fossil, fossil energy resource depletion; Acid., acidification; Eutro., eutrophication; CC, climate change including biogenic emissions; FAET, freshwater aquatic ecotoxicity; MAET, marine aquatic ecotoxicity; TET, terrestrial ecotoxicity; HT, human toxicity; OD, ozone depletion; POC, photochemical oxidation)

BioAcid, S3-AshDec, and S4-Gifhorn had the highest impacts for these categories (except for S3-AshDec, for which POC was lower than that of $Sref_{sludge}$). AD element was caused mainly by sludge production from wastewater treatment and SBPF production, while AD fossil and OD were caused mainly by sludge production from wastewater treatment (Fig. 5). The use of reactants in SBPF scenarios also contributed greatly to AD element. The mineral resources depleted were sodium chloride, used to produce NaOH (S1-BioAcid, S4-Gifhorn) and sodium sulfate (S3-AshDec). Regardless of the scenario, AD fossil was caused mainly by production of the bitumen used to build the sewer network.

For Acid, S1-BioAcid, S2-Crystal, and $Sref_{sludge}$ had higher impacts than S3-AshDec and S4-Gifhorn due to NH_3 emissions during storage and application and a higher N content in sludge and struvite. For Eutro and CC, all SBPF scenarios except S3-AshDec had higher impacts than $Sref_{sludge}$. Like for Acid, storage and application were the main contributors to Eutro (due to NH_3, nitrate, and N_2O emissions) and CC (due to N_2O emissions). Biogas leakage during anaerobic digestion in all scenarios also contributed to CC since CO_2 and CH_4 were emitted. In addition, use of large amounts of reactants contributed greatly to Acid and CC for S4-Gifhorn, unlike for the other SBPF scenarios (Fig. 5).

For FAET and HT, S2-Crystal had the lowest impacts and S4-Gifhorn the highest impacts, while S3-AshDec had higher and lower impacts than $Sref_{sludge}$, respectively. For TET, all SBPF scenarios had lower environmental impacts than $Sref_{sludge}$. Ecotoxicity and toxicity impacts can be explained by three main causes. First, direct emissions of heavy metals to agricultural soils contributed to FAET (due to copper and nickel), TET (due to chromium and mercury), and HT (due to chromium, lead, and nickel) in all scenarios except S4-Gifhorn and $Sref_{min}$. These direct emissions during land application were also the main contributor to HT. Electricity used for biological sludge production and bitumen used in the sewer network also contributed to FAET, MAET, and TET in almost all scenarios except $Sref_{min}$. Finally, reactants used in the Gifhorn® process, such as sodium persulfate and magnesium, contributed to its FAET, MAET, and TET impacts, while NaOH used in S1-BioAcid during crystallization contributed to its FAET and MAET impacts.

Considering net impacts, however, avoided fertilizers counterbalanced gross impacts and even resulted in negative results for S2-Crystal and $Sref_{sludge}$ for AD element (Fig. 4b). Nonetheless, the rank of impacts by scenario did not change. Net impacts for AD element, AD fossil, FAET, MAET, HT, OD, and POC were highest for S4-Gifhorn; for Acid, Eutro, and CC for S2-Crystal and for TET for $Sref_{sludge}$. Avoided impacts were driven by the following (Fig. 5):

i. avoided land application of mineral fertilizers for AD element, ecotoxicity, and toxicity impacts;
ii. avoided production of energy as heat and electricity depend on the scenario (Fig. SI-2);
iii. avoided N emissions for Acid, Eutro, and CC in scenarios with land application (i.e., S1-BioAcid and S2-Crystal);
iv. avoided energy for CC in S4-Gifhorn (i.e., avoided CO_2 emissions).

Sensitivity of results to system modeling

Results were also driven by how the system was modeled to reach the functional unit and by the allocation procedures used to model the system.

Reference flows used to reach the functional unit

For S3-AshDec and S4-Gifhorn, reference flows were calculated using only SBPF. Only 58% of P was

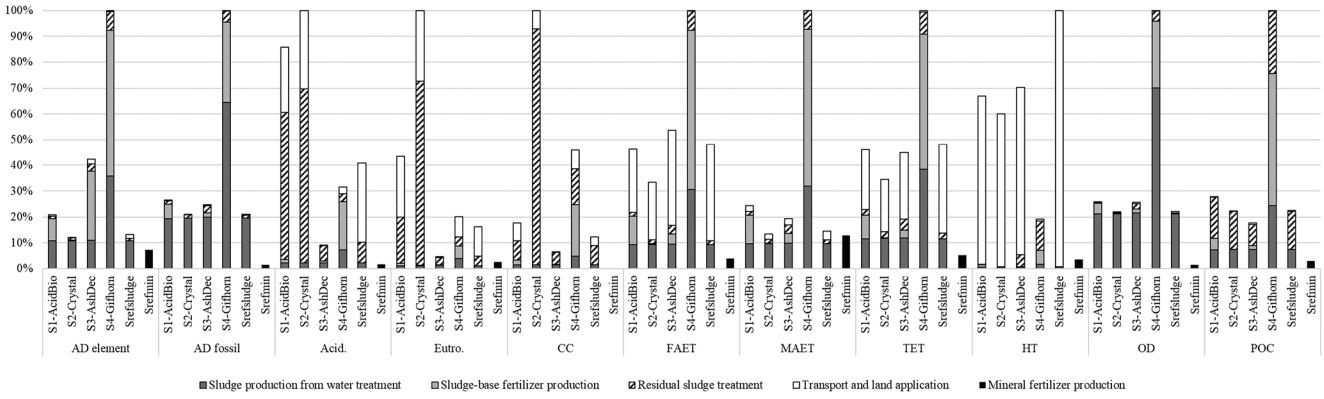

Fig. 5 Contribution analysis of processes to relative gross impacts of each scenario for each impact category (AD element, mineral reserve-based resource depletion; AD fossil, fossil energy resource depletion; Acid., acidification; Eutro., eutrophication; CC, climate change including biogenic emissions; FAET, freshwater aquatic ecotoxicity; MAET, marine aquatic ecotoxicity; TET, terrestrial ecotoxicity; HT, human toxicity; OD, ozone depletion; POC, photochemical oxidation)

recovered from sludge in S4-Gifhorn despite using large amounts of reactants, while 91% was recovered from sludge ashes in S3-AshDec with the use of small amounts of reactants. S4-Gifhorn thus had higher impacts than S3-AshDec because more sludge was needed to produce enough SBPF to reach the functional unit (see Section 6.7 in the Supporting Information). In contrast, for S1-BioAcid and S2-Crystal, reference flows were calculated for both struvite and low-P sludge application based on their P contents. SBPF provided only 20% of the P applied in S2-Crystal but 56% in S1-BioAcid. Thus, using some of the P in the low-P sludge to reach the functional unit decreased the impacts of these two scenarios.

Comparing the alternative scenarios for S1-BioACid and S2-Crystal to the other original scenarios (S3-AshDec, S4-Gifhorn and the reference scenarios), the rank of impacts by scenario did not change for AD element, Acid, Eutro or CC, although the differences between scenarios increased due to greater emissions during sludge end-of-life (i.e., more sludge applied) (Fig. 6). The rank of impacts by scenario was modified slightly for S2-Crystal (Alt.) and S1-BioAcid (Alt.) for FAET, MAET, TET, HT, OD, and POC. S4-Gifhorn remained the worst scenario for AD fossil and MAET despite the increase of S1-BioAcid (Alt) and S2-Crystal (Alt) for these two impact categories. The main reason for differences in impact due to the modeling was the small amount of P recovered in S2-Crystal. More sludge was needed to reach the functional unit, leading to higher impacts (see Section 6.7 in the Supporting Information).

Allocation procedures

WWTPs are increasingly seen as complex systems with two functions, "waste treatment" and "nutrient and energy production." Modeling WWTPs in LCA can therefore result in several types of models depending on the objective of the study, the functional unit, and how a WWTP's multifunctionality is considered. Currently, the most common functions associated with WWTPs are as follows:

- to treat water, commonly using "m^3 of treated water" as the functional unit;
- to treat water and recover nutrients and energy, also using "m^3 of treated water" as the functional unit, but including nutrient and energy recovery as additional functions using system expansion

According to Schrijvers (2017), WWTP modeling in attributional LCA is process-oriented, and a consistent modeling approach consists of system expansion (partial or complete), which is in line with how LCA are modeled currently. With the development of effective nutrient-recovery processes, WWTP can also produce products with high economic value such as fertilizers or biomethane. Thus, demand for SBPF or biomethane from WWTPs may occur in the future. Consequently, LCA of such products can no longer be process-oriented but must become product-oriented. According to Schrijvers (2017), a consistent way to model this type of LCA consists of using allocation among coproducts.

However, the transition from process-oriented to product-oriented LCA for SBPF production has rarely been studied in the literature. First, few LCA are performed using an output-

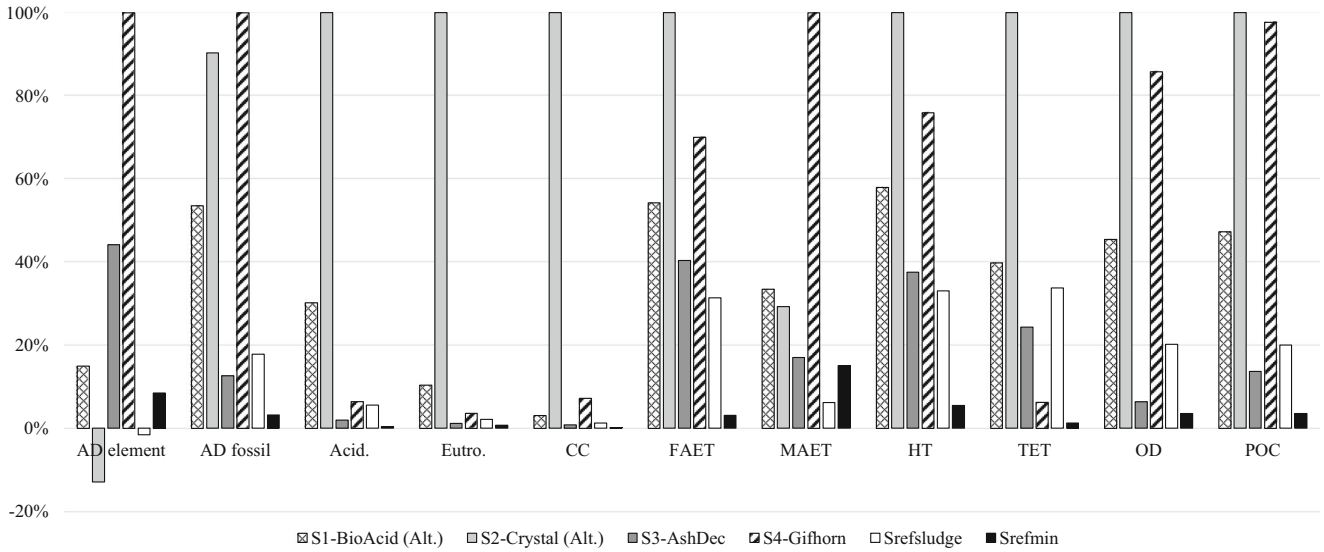

Fig. 6 Relative net impacts of two alternative scenarios (S1-BioAcid and S2-Crystal) and four original scenarios (S3-AshDec, S4-Gifhorn, $Sref_{min}$ and $Sref_{sludge}$) for each impact category (AD element, mineral reserve-based resource depletion; AD fossil, fossil energy resource depletion; Acid., acidification; Eutro., eutrophication; CC, climate change including biogenic emissions; FAET, freshwater aquatic ecotoxicity; MAET, marine aquatic ecotoxicity; TET, terrestrial ecotoxicity; HT, human toxicity; OD, ozone depletion; POC, photochemical oxidation)

oriented functional unit, such as the amounts of fertilizers produced as a function of their P contents. Bradford-Hartke et al. (2015) and Linderholm et al. (2012) compared P recovery from WWTPs using kg of P recovered as a functional unit but used system expansion because they modeled the system as a process-oriented LCA. To our knowledge, the present study is the first to perform an LCA of SBPF production from a product-oriented perspective. However, using allocation to address the multifunctionality of WWTPs using this type of LCA is challenging, since several sub-processes of WWTPs have multiple functions, such as wastewater treatment (production of sludge and "clean water"), anaerobic digestion (production of low-P sludge, struvite, and heat/energy), and incineration (production of Rhenania phosphate and heat/energy). Although we allocated environmental burdens between sludge and treated water, this approach was only partial, since other multifunctional sub-processes were not allocated, due to methodological difficulties. Therefore, in addition to allocation, we used system expansion to consider benefits of heat/energy recovery and low-P sludge production.

Drivers of improvements in environmental impacts of SBPF

Based on these results, several drivers were identified to decrease environmental impacts of SBPF. First, SBPF scenarios had higher impacts than phosphate fertilizers from mining since more resources were needed to concentrate and recover diffuse P. Indeed, the low efficiency of P recovery combined with use of large amounts of reactants to concentrate diffuse P from wastewater sludge increased impacts of SBPF.

The amounts of electricity, natural gas, reactants, and infrastructure used to recover P from sludge and then treat the low-P sludge were much higher than those for $Sref_{min}$, since phosphate rock contains high concentrations of P (Table 5). S1-BioAcid and S4-Gifhorn used 20 times as much electricity as $Sref_{min}$ and 3 times as much as $Sref_{sludge}$. S2-Crystal and S3-AshDec, however, used approximately as many inputs as $Sref_{sludge}$. Natural gas in S1-BioAcid and S3-AshDec was used mainly during incineration, which was considered a P-recovery process in S3-AshDec and sludge end-of-life in S4-Gifhorn. SBPF also required much more infrastructure than $Sref_{min}$. The reactants were used mainly to recover P, except in S2-Crystal, which used slightly more reactant than $Sref_{sludge}$. Diffuse P recovery used more reactants than $Sref_{min}$. 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.

Sewage sludge represents a useful P reserve, since maximizing the P-recovery rate has the potential to meet ca. 40% of the annual P demand in France, estimated to be 191,677 t (UNIFA 2018). However, in the present study, the P recovered in the final product varied from 12 to 57% of the P entering the WWTP, depending on the scenario. Indeed, S2-Crystal had the lowest P-recovery rate and used the lowest amounts of reactants. In this scenario, only 20% of the P in sludge was solubilized in mineral form after anaerobic digestion and dewatering and then recovered in the return liquor; however, 90% of this P was recovered as struvite by crystallization. S4-Gifhorn had a 58% P-recovery rate due to its use of large amounts of reactants. In comparison, S1-BioAcid had a P-recovery rate of 53% using smaller amounts of reactants, since a biological process acidified the sludge. However, S3-AshDec had the highest amount of P recovered per kg of reactant (91% from sludge ashes).

Since impacts of P recovery were associated mainly with chemical reactants and energy use, it is necessary to increase the P-recovery rate by using the smallest amounts of reactants possible to decrease environmental impacts of SBPF. Traditionally, chemical reactants are used to acidify the media and solubilize the P in sludge. A more environmentally friendly alternative is to replace chemical acidification with biological acidification, such as in S1-BioAcid, which uses organic substrates to develop acidifying bacteria (Braak et al. 2016; Guilayn et al. 2017). Another solution is to combine several struvite precipitation processes at different steps of sludge treatment (e.g., during anaerobic digestion, from ashes) to recover the maximum amount of P with technologies that use as little reactant as possible.

Table 5 Total use of inputs and infrastructure for P recovery and sludge treatment ("Total") and the percentage of these totals used for P recovery only, by scenario

Input	S1-BioAcid		S2-Crystal		S3-AshDec		S4-Gifhorn		$Sref_{sludge}$	$Sref_{min}$
	Total	%	Total	%	Total	%	Total	%		
Electricity (kWh)	1833	60%	846	14%	920	19%	2000	42%	731	104
Natural gas (kWh)	–	–	–	–	55	100%	138	0%	–	56
Reactants (kg)	576	95%	42	36%	811	97%	4540	92%	27	174
Infrastructure (p)	2.89E−04	29%	2.07E−04	20%	3.46E−04	39%	1.20E−03	36%	2.07E−04	1.15E−07

Table 6 Fertilizer application rate (t/ha) and the limiting element (LE) (N or P) for each cropping system

Cropping system	Sewage sludge with low P content				Sewage sludge		SBPF							
	S1-BioAcid	LE	S2-Crystal	LE	$Sref_{sludge}$	LE	S1-BioAcid	LE	S2-Crystal	LE	S3-AshDec	LE	S4-Gifhorn	LE
Mt1	13.03	N	7.36	N	10.83	P	0.70	P	0.21	P	1.59	P	0.47	P
Mt2	21.78	P	8.07	P	10.83	P	0.70	P	0.21	P	1.59	P	0.47	P
Kl1	5.42	N	3.06	N	5.31	N	0.54	P	0.16	P	1.23	P	0.36	P
Kl2	16.85	P	6.25	P	8.38	P	0.54	P	0.16	P	1.23	P	0.36	P
Kl3	3.13	N	1.77	N	3.07	N	0.70	N	0.22	P	1.69	P	0.31	N
Ms1	16.71	N	7.84	P	10.52	P	0.68	P	0.20	P	1.54	P	0.46	P
Pt1	20.25	P	7.50	P	10.07	P	0.65	P	0.19	P	1.48	P	0.44	P
Cx1	12.87	N	7.27	N	10.73	P	0.69	P	0.20	P	1.57	P	0.47	P

Nonetheless, due to the lack of specific data for the entire life cycle, these results were obtained using input data of variable quality, since some processes have not yet been performed at full scale and rely on pilot or even experimental input data. This strong assumption implies that these data must be validated in full-scale plants to enable final analysis of environmental impacts of these processes. Similarly, $Sref_{min}$ was modeled based on TSP data from ecoinvent that have not been updated since 2007 and therefore do not consider the most recent technological developments in TSP production.

Sensitivity analysis to the choice of cropping system and heavy metals in fertilizers

Sensitivity analysis of SBPF and sewage sludge application rates

For most cropping systems, the optimal application rate depended on crop requirements for P (Table 6). P was the limiting element for each fertilizer in Mt2, Kl2, Ms1 (except for S1-BioAcid low-P sludge), and Pt1. For the other cropping systems, P was the limiting element for sewage sludge and SBPF (Mt1, Cx1), for all SBPF (Kl1), or for the SBPF from S2-Crystal, S3-AshDec, and S4-Gifhorn (Kl3). To provide all nutrients needed by the cropping system, mineral fertilizers were applied to meet the need for N or P in the form of ammonium nitrate or TSP, respectively (Table 7).

The amount of ammonium nitrate applied to meet crop N needs varied from 2.2 to 614.6 kg depending on the cropping system and the fertilizer applied. Little N supplementation (S1-BioAcid for low-P sludge in Kl2) meant that the fertilizer was well adapted to the cropping system. In contrast, the S3-AshDec SBPF in Mt2 seemed unsuitable, since high N supplementation was required. The amount of TSP applied to meet crop P needs varied from 21.1 to 318.0 kg depending on the cropping system and the fertilizer applied. The range was smaller than that for ammonium nitrate, and the same cropping system (Kl3), for which N was the limiting element, had the minimum and maximum TSP application rates. Unlike sewage sludge, SBPF usually needed to be supplemented with mineral N fertilizer (all cropping systems except Kl3 for S1-BioAcid and S4-Gifhorn). In contrast, application

Table 7 Application rates (kg/ha) of mineral fertilizer (ammonium nitrate (AN) (33% N) and triple super-phosphate (TSP) (48% P_2O_5) needed to compensate crop needs for each cropping system

Cropping system	Sewage sludge with low P content				Sewage sludge		SBPF							
	S1-BioAcid		S2-Crystal		$Sref_{sludge}$		S1-BioAcid		S2-Crystal		S3-AshDec		S4-Gifhorn	
	AN	TSP[a]	AN	TSP	AN	TSP	AN	TSP	AN	TSP	AN	TSP	AN	TSP
Mt1	–	139.02	–	30.47	51.97	–	261.78	–	313.87	–	344.48	–	219.60	–
Mt2	38.84	–	236.89	–	322.12	–	531.93	–	584.02	–	614.63	–	489.75	–
Kl1	–	181.65	–	136.50	–	98.25	79.30	–	119.60	–	143.28	–	46.66	–
Kl2	2.24	–	155.48	–	221.43	–	383.77	–	424.08	–	447.76	–	351.14	–
Kl3	–	317.98	–	291.88	–	269.77	–	21.12	50.31	–	82.84	–	-	138.21
Ms1	–	70.66	74.82	–	157.62	–	361.45	–	412.06	–	441.79	–	320.48	–
Pt1	16.96	–	201.07	–	280.30	–	475.36	–	523.79	–	552.24	–	436.15	–
Cx1	–	138.13	–	30.91	50.66	–	258.41	–	309.99	–	340.30	–	216.65	–

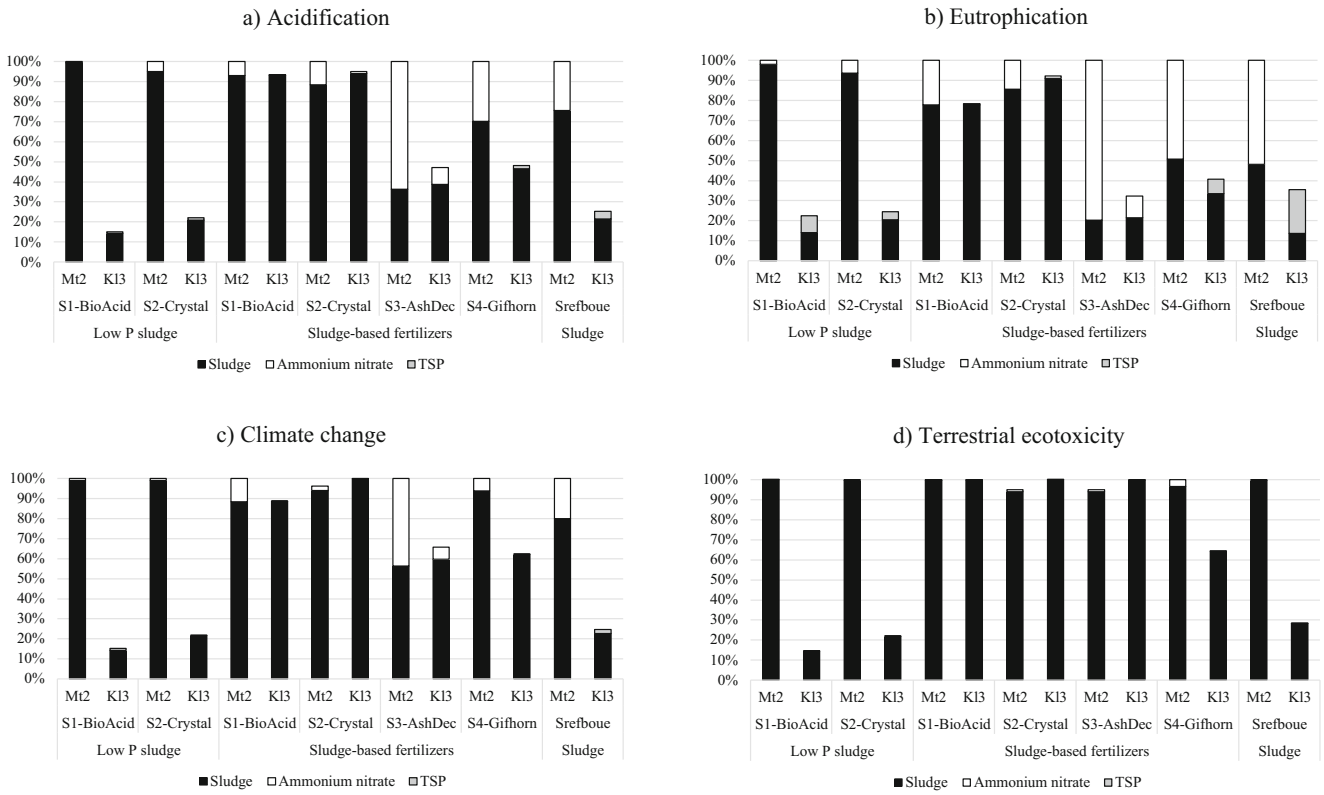

Fig. 7 Relative impacts of fertilization practices for a P-limited cropping system (Mt2) and a N-limited cropping system for four impact categories using low P-sludge (S1-BioAcid and S2-Crystal), sludge-based fertilizer (from each scenario studied) and sewage sludge ($Sref_{sludge}$).

of low-P sludge created a need for either P, which had to be met by a small (Mt1, Cx1) or large (Kl3) amount of mineral phosphate fertilizer, or N (Mt2, Kl2, and Pt1).

LCA results for Acid, Eutro, CC, and TET for a P-limited cropping system (Mt2) and N-limited cropping system (Kl3) showed large differences depending on the fertilizer used (Figs. 7 and SI-3). Fertilization practices for Mt2 tended to have larger impacts than those for Kl3. Between cropping systems, low-P sludge from S1-BioAcid and S2-Crystal differed the most in impact, while SBPF from S1-BioAcid and S2-Crystal differed the least, regardless of the impact category. Ammonium nitrate fertilizers used as a supplement in P-limited cropping systems contributed 5–80% of Acid, Eutro, and CC impacts. In contrast, TSP fertilizers used in N-limited cropping system contributed only 2–22%, depending on the impact category.

To ensure sustainable fertilization practices, fertilizers must be chosen carefully based on their nutrient contents to meet crop requirements as much as possible. From an agronomic viewpoint, sewage sludge has greater value than SBPF and single-nutrient fertilizers since it provides both N and P. It also supplies organic matter, which can improve soil fertility, but

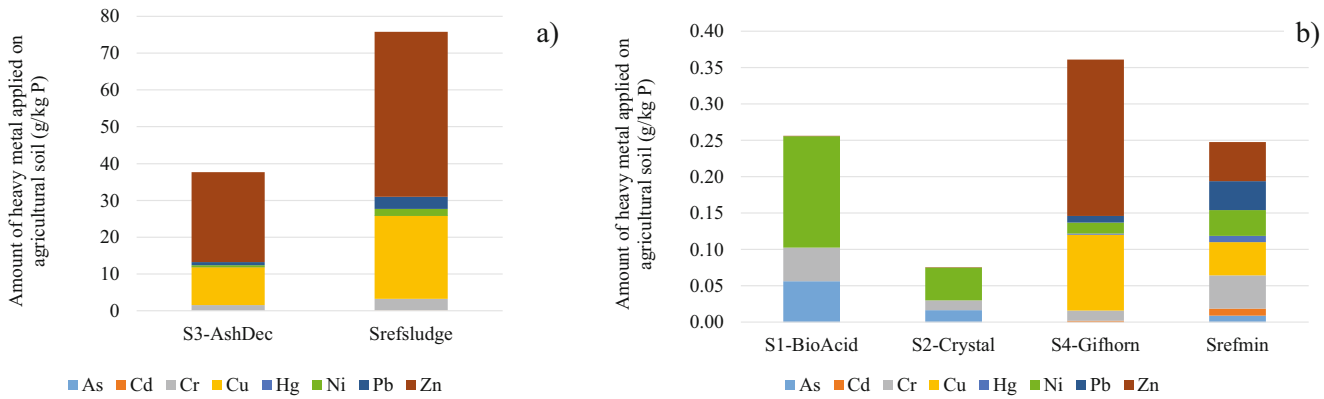

Fig. 8 Amounts of heavy metals per kg of P applied to land from (**a**) sewage sludge and SBPF (S3-AshDec) and (**b**) TSP ($Sref_{min}$) and SBPF (S1-BioAcid, S2-Crystal, S4-Gifhorn)

Table 8 Impacts for freshwater ecotoxicity (FAET), marine aquatic ecotoxicity (MAET), terrestrial ecotoxicity (TET) and human toxicity (HT) for scenarios with land application of sludge based on the heavy metal contents used in this study and those of upper thresholds ("Limit") for land application of sludge allowed by French legislation

Category	S1-BioAcid			S2-Crystal			$S_{refsludge}$		
	Present study	Limit	Times higher	Present study	Limit	Times higher	Present study	Limit	Times higher
FAET	1.14E+03	3.16E+03	2.8	7.41E+02	2.61E+03	3.5	1.17E+03	4.29E+03	3.7
MAET	1.55E+06	2.30E+06	1.5	4.80E+05	1.17E+06	2.4	5.11E+05	1.66E+06	3.2
TET	9.96E+02	1.76E+04	17.7	8.85E+02	1.62E+04	18.3	1.50E+03	2.71E+04	18.1
HT	2.23E+03	2.28E+04	10.2	1.36E+03	2.03E+04	14.9	2.26E+03	3.40E+04	15

this aspect was not assessed in this study and is difficult to assess using LCA in general.

Impact of fertilization practices according to heavy metal content

Heavy metal contamination may remain an environmental problem. Land application of sewage sludge is often criticized because its high heavy metal content could pollute agricultural soils. The S1-BioAcid and S2-Crystal SBPF were estimated to have the same heavy metal content, since no data were available for the former. According to analyses, S2-Crystal SBPF contained mainly arsenic (22%), chromium (18%), and nickel (60%). The S3-AshDec and S4-Gifhorn SBPF and sewage sludge in the reference scenario ($Sref_{sludge}$) had similar heavy metal contents: 59–65% zinc, 27–30% copper, and less than 4% each of lead, nickel, and chromium. In comparison, TSP had the largest diversity of heavy metals: 22% zinc, 18% each of chromium and copper, 16% lead, 14% nickel, and 4% each of arsenic, cadmium, and mercury.

However, per kg of P applied to land, sewage sludge ($Sref_{sludge}$) and S3-AshDec SBPF had the highest heavy metal contents (i.e., 76 and 38 g per kg P, respectively) (Fig. 8). Other fertilizers, including TSP, supplied few heavy metals to agricultural soils (0.08–0.36 g per kg of P applied).

Heavy metal contents in low-P sludge from Houot et al. (2014) used in this study for S1-BioAcid and S2-Crystal were much lower than the upper thresholds in French legislation for application of sludge to land (Journal Officiel 1998): 10 g t DM^{-1} for cadmium and mercury, 1000 g t DM^{-1} for chromium and copper, 200 g t DM^{-1} for nickel, 800 g t DM^{-1} for lead, and 3000 g t DM^{-1} for zinc.

When heavy metal contents were set to these upper thresholds, potential ecotoxicity and toxicity impacts are at least 1.5 (MAET for S1-AcidBio) to 18.3 (TET for S2-Crystal) times as high (Table 8). Overall, heavy metals applied to land with sludge influence HT and TET more than FAET and MAET. Thus, it is important to include heavy metal emissions when estimating impacts of applying sludge to land and to use realistic values (e.g., from physico-chemical analysis of sludge) rather than arbitrary values, such as those from legislation.

Conclusion

This study aimed to assess whether fertilizing with struvite or Rhenania phosphate recovered from sludge can be a suitable alternative to crop fertilization with mineral fertilizers from phosphate rock. To reach this goal, four fertilization scenarios were compared, each using struvite or Rhenania phosphate recovered from a specific recovery process, to two fertilizing reference scenarios, one using sewage sludge and the other using TSP. The results highlight that production and land

application of SBPF have higher environmental impacts than that of TSP, but that of sewage sludge can equal impacts of SBPF, depending on the impact category. The low yields of P recovery from sludge with a low P content and the need for large amounts of energy and reactants to recover P are responsible for the higher environmental impacts of SBPF scenarios. Their environmental impacts could be decreased, however, if a good compromise is found between P-recovery efficiency and the reactants needed to concentrate the P.

Sensitivity analysis highlights the importance of how the system is modeled and the assumptions made when defining system boundaries. Indeed, considering only land application of SBPF, especially if low P recovery is assumed, may drastically increase environmental impacts of production and land application of SBPF scenarios compared to the reference ones. Environmental impacts were assessed for a four-year crop rotation, but the sensitivity analysis performed for different cropping system highlights the variability in potential application rates using sewage sludge or SBPF. Therefore, it is important to perform sensitivity analysis or assess a wide range of cropping systems when estimating environmental impacts of land application of fertilizers.

Finally, SBPF remain of great interest since, except for those produced from P recovery from ashes, they contain much lower heavy metal contents than sewage sludge or TSP. This advantage is mitigated, however, by the need to supplement SBPF with mineral fertilizers. Thus, SBPF have advantages and disadvantages that need to be considered, since they may influence their use within sustainable fertilization practices.

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