Environmental assessment of phosphorus recovery from dairy sludge: A comparative LCA study

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Research Paper

Environmental assessment of phosphorus recovery from dairy sludge: A comparative LCA study

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ABSTRACT

Phosphate rock is a finite, non-renewable mineral resource that is used primarily in fertiliser production. The scarcity and the increasing demand for this finite material led the European Commission to include it in the critical raw material list in 2014. As a consequence, efforts have been directed towards enhancing material use efficiency, initiating recycling efforts, and formulating waste policies to mitigate the criticality of raw materials. Interest in the development of technologies for nutrient recovery from organic waste streams has increased in recent years, and dairy processing sludge (DPS) is a potential input waste stream. Although the recovery of P from DPS can contribute to more circular flows of nutrients in society, it has to be assessed whether there are also overall environmental gains. This paper reports on a life cycle assessment (LCA) of the environmental impacts of three scenarios for phosphorus (P) recovery involving hydrothermal carbonization (HTC) and struvite precipitation and a comparison to a reference drying scenario. HTC produces a solid fraction (hydrochar), and a liquid fraction (process water) and in one of the scenarios (Scenario 3), leaching the hydrochar for additional P recovery is considered. From the process water as well as from the hydrochar leachate, P is precipitated in the form of struvite. Scenarios 1 and 2 both consider HTC and struvite production with the only difference that the hydrochar is used as a fuel instead of as a fertilizer in the latter case, and Scenario 3 adds leaching of the hydrochar with subsequent struvite production and considers that hydrochar is used as a fuel. In the fourth (reference) scenario, dewatering and drying of DPS is considered. The recovered product use in agriculture was not assessed at this stage. The assessment of the emerging technologies in Scenarios 1–3 was done by studying the technologies in early stages of development but modelling them as more developed in the future. Additional functions beyond the functional unit of one kg of P recovered were handled through a system expansion by substitution approach. This way, the system was credited for calcium ammonium nitrate (CAN) production in all scenarios and for wood chips production in Scenarios 2 and 3. Looking at net outcomes for all scenarios, the life cycle impact indicator results for scenario 2 are lower than the other scenarios in several impact categories. Large gains in scenario 2 are related to the avoided production of wood chips.

1. Introduction

Several forms of phosphate exist in nature, but the form mainly used for fertiliser production is phosphate rock. This is a finite, non-renewable ore for which demand is expected to increase owing to an increasing global population. Phosphate rock deposits are not equally distributed throughout the globe, with two thirds of the world production controlled by Morocco, the USA and China (Reijnders, 2014). There is only one mine in Europe, which is in Finland. The lack of exploitable phosphate mineral reserves in Europe (Schröder et al., 2010), combined with the general scarcity and the increasing global demand for this material, poses a threat to European food security. This looming phosphorus (P) crisis led the European Commission to include this resource in the critical raw material list in 2014 (European Commission, 2020) and to push for the exploration of alternatives for fertiliser production. Therefore, European policies target the efficiency of material use and recycling, waste policies, and international cooperation to address raw materials' criticality. The recovery and safe reuse of P from food and municipal waste flows is prioritised through the Circular Economy Package (European Commission, 2016) and the Critical Raw Materials Act (European Commission, 2023).

A waste stream of considerable size that could potentially be utilized

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in this context is dairy waste. The dairy industry is the most economically important sector of the European agri-food industry, with an aggregate 160 million tonnes of milk produced annually (22 % of the world's total milk production) (EDA, 2018). The expansion of this food sector has been followed by an increase in amounts of dairy wastewater (DWW) and dairy processing sludge (DPS), which has therefore become an important waste flow in Europe (Carvalho et al., 2013). These wastewaters and sludges are recognized as potential feedstocks for fertilisers and soil amendments (Shi et al., 2021). In addition, DPS has the potential to be used as biofuel or as an additive in compost, or animal feed (Korsström & Lampi, 2001; Ryan & Walsh, 2016).

Currently, DPS is generally categorised as a biosolid in the EU (Pankakoski et al., 2000), and therefore can be spread as organic fertiliser on agricultural land (Ryan & Walsh, 2016). DPS contains a higher amount of P compared to biosolids obtained from sewage sludge (Shi et al., 2021). Ashekuzzaman et al. (2019) reported that DPS possesses a significant nutrient content in addition to having heavy metal content that is below EU limits. As a result of the Circular Economy Package, interest in the development of technologies for nutrient recovery from organic waste streams has increased. Such emerging technologies can combine DPS treatment with the recovery of P for use as fertiliser in agriculture.

To achieve the goal of P recovery in the dairy industry, the REFLOW European Training Network (ETN) has focused on developing and demonstrating processes for the recovery and use of P fertiliser products from DWW. The current study was done within the work in REFLOW and it focuses specifically on the combination of two technical methods: hydrothermal carbonisation (HTC) and struvite precipitation (REFLOW ETN, 2019).

The present paper is novel on account of its focus on the further treatment of DPS using HTC. HTC is one of the promising approaches for the thermochemical treatment of DPS, enabling the production of value-added fertiliser products (Khalaf et al., 2022) such as the recovery of P as struvite. The main advantage of HTC is the ability to process biomass with a high moisture content, without any pre-treatment. The HTC is a technique that operates between 180 °C and 250 °C to partially degrade the organic content of waste in the presence of water and produce a solid fraction (hydrochar), and a liquid fraction (process water) (Atallah et al., 2019). The hydrochar has a carbonaceous structure with coal-like properties. Experiments on the HTC treatment of DPS have been carried out at laboratory scale within REFLOW (Khalaf et al., 2022).

Struvite contains P and nitrogen (N) in forms efficiently available for plant nutrition (Daneshgar et al., 2018). Struvite precipitation from liquid organic waste flows may therefore generate a useful fertiliser product. The system considered within this study combines HTC processing with the recovery of P as struvite from the process water. It also investigates acid leaching of the generated hydrochar for the release of more P and additional struvite generation in a subsequent step. Presently, the hydrochar produced through the HTC of biomass, typically agricultural waste, is used as a soil amendment or fertiliser in agriculture, due to its potential to improve soil quality and enhance plant growth (Suarez et al., 2023). Within this study, the use of hydrochar as a bio-fertiliser is considered but also its use as a solid fuel (before or after leaching).

Although the recovery of P from DPS can contribute to more circular flows of nutrients in society, it has to be assessed whether there are also overall environmental gains. A common method for assessing the overall environmental impacts of such technologies is life cycle assessment (LCA) (Behjat et al., 2022). Moreover, one of the positive aspects of performing an LCA on processes and technologies that are in early stages of development, is the possibility to identify improvement opportunities in the process design and thereby enable process developers to choose a more beneficial track in the development process.

To provide input to environmental assessment and to guide technical development towards environmental sustainability for the considered technologies, three research questions were formulated:

1. What are the environmental hot-spots in the life cycle of P fertilizer recovered from DPS by means of HTC and struvite precipitation and what are influencing factors and improvement opportunities?
2. How do different possible system configurations of HTC and struvite production compare environmentally?
3. How do the environmental impacts compare to those related to the direct use of DPS in agriculture after dewatering and drying?

LCAs have been employed previously to evaluate the environmental consequences of dairy production and the recovery of P from various wastewaters. However, no full LCA investigation into P recovery from dairy wastewater has been published at this point (Behjat et al., 2022). With this study, we aim to contribute to the evaluation of the environmental performance of DPS treatment with P recovery at an early stage, to provide input to further development and upscaling.

2. Materials and method

2.1. Description of the investigated technologies

2.1.1. Feedstock and HTC treatment

The feedstock considered in this study is the DPS used in the laboratory experiment described by Khalaf et al. (2022) and Numviyimana et al. (2022), which came from Arrabawn Company, located near Limerick, Ireland. The company has 400 employees and supplies milk, butter and cream using raw milk produced by 1000 farmers (Arrabawn, 2023) This DPS has a solid content of 21 % and a P concentration of 57 mg/kg, as reported by Khalaf et al. (2022).

Khalaf et al. (2022) reported that the highest hydrochar yield of 61 % of dry mass, coupled with a maximum retention of P in the hydrochar (80 %), was achieved under the specific operating conditions of pH = 2.25 and T = 180 °C for a reaction time of two hours, in a batch volume of one litre. To achieve the desired pH, sulfuric acid (H_2SO_4) was used (Khalaf et al., 2022). Following the reaction, the reactor was left to cool down at room temperature, and the gases were then released into the fume hood (Khalaf et al., 2022). After cooling, the solid hydrochar was separated from the process water by filtration. The solid fraction was further subjected to drying at 105 °C for 24 h, while the process water was collected for P recovery through the formation of struvite.

2.1.2. Struvite production

The struvite recovery process consists of two steps, both conducted at room temperature. The first is focused on reducing the concentration of struvite precipitation inhibitors in the process water, i.e., iron (Fe) and calcium (Ca) (Yan & Shih, 2016). To achieve this, oxalic acid was added (Elomaa et al., 2019), and the reaction proceeded for half an hour (Numviyimana et al., 2022). Following the reaction, the supernatant was filtered from a cake rich in Fe and Ca. The supernatant was used in the second step for struvite precipitation. In their study, Numviyimana et al. (2022) reported that recovery of 99.96 % of P was achieved under the specific operating conditions of pH = 9 and salt dosage of 1.73:1.14:1 for $Mg:NH_4^+:PO_4^{3-}$ mole ratio (Numviyimana et al., 2022). Sodium hydroxide (NaOH) is used to achieve the desired pH. Magnesium chloride ($MgCl_2$), ammonium chloride (NH_4Cl) or phosphoric acid (H_3PO_4) were used to reach the desired composition for optimal struvite precipitation. Depending on the quantity of Mg, NH_4^+, and PO_4^{3-} in the supernatant, two of these three reagents are used. The reaction proceeded for a duration of one hour (Numviyimana et al., 2022). Following the second reaction, crystals of struvite were separated from the liquid phase by filtration. Both the oxalate cake and the struvite were dried at 105 °C for 24 h.

2.1.3. Leaching

The produced hydrochar can be used as a bio-fertiliser, contributing to enhanced crop growth and improved soil quality, as shown by Azzaz et al. (2020). However, in the study conducted by Khomenko et al. (2023), the hydrochar exhibited a lower release rate for P than struvite, biochar (from DPS pyrolysis) and DPS. Therefore, an alternative way of valorising the P in the hydrochar might be needed. Several studies have investigated the effectiveness of P recovery through acid leaching from the hydrochar (Marin-Batista et al., 2020; Oliver-Tomas et al., 2019). The leaching process considered in the present study was the same as assessed and described by Mannarino et al. (2022). The hydrochar post HTC then undergoes homogenization in a grinder before being introduced into a mixer, where nitric acid (HNO_3) and water are added to facilitate P leaching from the solid fraction into the liquid phase (Mannarino et al., 2022). The use of HNO_3 resulted in the successful recovery of 78 % of the P initially retained within the hydrochar (Mannarino et al., 2022). Following the acid leaching, the leachate is used for struvite recovery and the remaining hydrochar is dried at 105 °C for 24 h. Drying the hydrochar removes moisture and helps to prevent the material from decomposing or undergoing further chemical changes.

2.1.4. DPS drying

DPS can also be used directly as a biosolid fertiliser and spread on agricultural land (Ashekuzzaman et al., 2019, 2021b; Ryan & Walsh, 2016). In this case, DPS undergoes only two process steps: dewatering, and subsequent drying at 105 °C for a duration of 24 h. The DPS considered in this study already has a total solids content of 21 % (Khalaf et al., 2022); the preceding thickening process was not included, which is the same assumption as for all considered technological set-ups.

2.1.5. Upscaling

The scale that was considered for the scaled-up model was a batch of feedstock mass of 10 tonnes of DPS. This is the average amount of DPS produced per month per dairy in Ireland (Arrabawn, 2023; Ashekuzzaman et al., 2019), and also the input feedstock to produce approximately two tonnes of struvite daily (Ostara Pearl, 2018). Since it is not economically reasonable to run the system only once per month, it was assumed that DPS would be collected from multiple dairies to produce 2 tonnes of struvite every day.

For the scaled-up model, the HTC as well as the struvite production will be performed in a continuous plant. A pressure belt filter was considered for the separation of hydrochar from the process water as well as from acid leachate, and for separating struvite inhibitors and struvite from the liquid phases, for the scaled-up model. This type of filter was also considered for the additional dewatering of the untreated DPS. Belt filter was considered for its low operating costs and equipment durability (Kamboj et al., 2023).

The cake with struvite inhibitors was not considered to be used, nor treated, in the scaled-up model. It is assumed that the cake can in fact be used in production of the nanomaterial siderite, or coagulants (Numviyimana et al., 2022), and therefore a life cycle system cut-off was deemed appropriate.

Energy consumption data for the scaled-up model were calculated based on Piccinno et al. (2016), which provides a procedure for simulating industrial scale production when only data for smaller scale or even lab scale are available. More details are provided in the life cycle inventory analysis section (2.2.3).

2.2. Environmental assessment

The environmental assessment was performed considering the relevant standard of ISO 14040 for LCA (International Organization for Standardization, 2006). Specifically, this study evaluated and compared, through LCA, the environmental performance of three alternative HTC system configurations as well as a drying scenario for DPS treatment:

- **Scenario 1** – HTC + struvite production: produces dried hydrochar and struvite, both intended to be used as fertiliser.
- **Scenario 2** – HTC + struvite production: produces dried hydrochar and struvite, intended to be used as fuel and fertiliser, respectively.
- **Scenario 3** – HTC + leaching + struvite production: produces two different product flows of struvite, and dried hydrochar low in P, intended to be used as fertilisers and fuel, respectively.
- **Scenario 4** – (reference scenario) dewatering and drying of the DPS intended to be used as fertiliser.

Note that scenarios 1 and 2 have the exact same HTC and struvite production set-up but that the dried hydrochar has a different use.

2.2.1. Goal, scope, and system boundaries of the study

Defining the goal and scope of a study is the first step when conducting an LCA, to clarify the purpose of the LCA study. The goal of this LCA study is to inform design optimisation by identifying hot-spots in three alternative HTC system configurations, and to compare their overall performance also with a scenario that involves only drying of DPS. The intended audience of this work is primarily stakeholders involved in the relevant dairy waste treatment and fertilizer industry, who want information on process design priorities. Fig. 1 shows the four studied scenarios.

The HTC process, struvite precipitation and hydrochar leaching are the core technical processes that are combined in the three alternative HTC system configurations. In the fourth scenario, dewatering and drying of DPS are the two core processes. All core processes (foreground system) are fully included within the system boundaries, as are the production processes of chemicals and energy (background system). Impacts caused by the construction of equipment and facilities are not included within the environmental analysis because they are generally considered low; for example, the construction of a wastewater treatment plant has been shown to have 3–7 % of the total impact (from construction and operation) (Schulz et al., 2012).

Due to the focus on HTC and struvite production technologies, the upstream dairy wastewater treatment (DWWT) and the downstream application of the recovered products as fertilisers on agricultural land were not included within the system boundaries, other than when substitutions were needed – see the next section. The DWWT is excluded because it is the same for all the four scenarios and would not add useful information in this comparative study. Future studies could investigate the downstream agricultural activities as these are not exactly the same for the different fertiliser products from the different scenarios; these were not considered here due to lack of data on the actual performance of the products at this early stage of technology development. Because of the omission of agricultural activities in the system, impacts related to potential toxic elements in the DPS or in hydrochar, such as heavy metals, are not captured in the assessment.

The functional unit (FU), the "reference flow to which all other modelled flows of the system are related" (Baumann & Tillman, 2004), has to be carefully chosen to represent the function of compared scenarios in the most meaningful way. To enable an understanding of the environmental impacts from the processes involved in the three HTC configurations and compare their performance with the DPS drying scenario, the FU selected was the recovery of 1 kg of P (or rather, 1 kg of P in total in all product flows intended for use as fertiliser). This FU is meaningful and practical because it corresponds to a common unit of measure in the context of P management. It makes it easy for the targeted stakeholders to understand what is being measured and compared for the P recovery. For operators responsible for DWWT and sludge management that intend to discover opportunities for process optimization, an alternative FU based on the amount of DPS feedstock would also make sense, but this would make less sense to fertilizer industry.

2.2.2. System expansion

This study considered not only the environmental impacts from direct emissions and from production and consumption of chemicals and energy, but also the benefits derived from resource recovery, such as

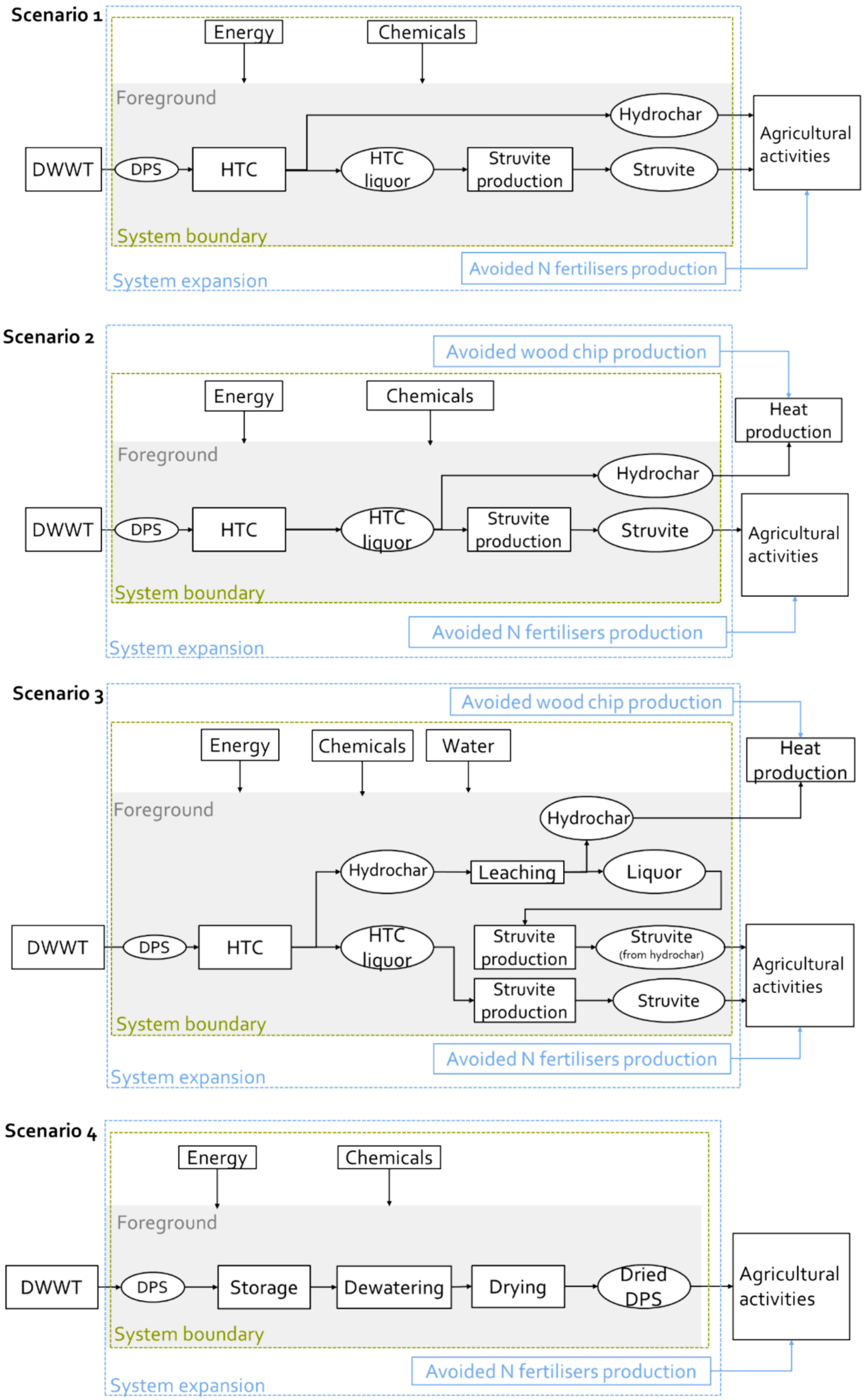

Fig. 1. System boundaries of **Scenario 1** (the HTC + Struvite production scenario); **Scenario 2** (the HTC + Struvite production scenario, with hydrochar as solid fuel product); **Scenario 3** (the HTC + Leaching + Struvite production scenario); and **Scenario 4** (the reference scenario – dewatering and drying) considered for the LCA study.

energy from hydrochar combustion, and nutrient values beyond P. This must be done to ensure that only the function expressed by the functional unit is performed by each of the compared systems. Additional functions of the systems were therefore handled through a "system expansion by substitution" approach. This is a common approach in similar studies and it means that the alternative production is added in the LCA model and the systems are credited for the related impacts in a way that makes them functionally equivalent (Heimersson et al., 2019). The system expansions are shown graphically in Fig. 1.

According to Khalaf et al. (2022) and Liu et al. (2023), the higher heating value (HHV) of hydrochar before and after leaching is 15.5 MJ/kg and 18.1 MJ/kg respectively, which is in our study considered to displace wood chips (when hydrochar is used as a fuel in Scenarios 2 and 3) on an equivalent basis of energy content. The HHV of wood chips is 16.2 MJ/kg (wood density = 0.701 kg/m^3) (ToolBox, 2003). The selection of the HHV over the lower heating value (LHV) was primarily driven by data availability considerations. However, the HHV and LHV for hydrochar essentially represent very similar fundamental energy content (Fiori et al., 2014). The HHV is typically favoured in energy calculations because it includes the latent heat of evaporation. When employing LHV, an assumption is made that all the generated vapor is immediately expelled as exhaust without any condensation. Conversely, opting for HHV implies the assumption that there is a presence of condensate and that it is of importance to account for the energy content of the vapor that undergoes condensation within the unit. Data for the production process of substituted wood chips were taken from Sphera (Sphera, 2023).

The exploitation of the P content in the struvite, hydrochar and sludge can reduce the need to produce other fertilisers. The recovered products are assumed to be used in agricultural activities, exploiting their fertiliser potential, but without including the agricultural activities as such in the study. Using a functional unit of 1 kg of recovered P (and thereby implicitly rescaling all systems based on the P content in products) creates a need, however, to capture differences between the systems in terms of other fertilising effects represented by the products. This was done for N, when deemed relevant. The avoided fertiliser used for the system expansion is calcium ammonium nitrate (CAN; 27 % N). Other contents in the dried DPS and hydrochar, such as carbon content or micronutrients, were, however, not accounted for. It has to be remembered that P and N content in product flows does not always originate from only the DPS, which is further discussed later.

More information about the calculations for the system expansions is reported in the supplementary material (SM).

2.2.3. Life cycle inventory

Data and information about the HTC reactor, leaching, and struvite recovery from process water as well as from hydrochar leachate were collected from experimental laboratory tests and in some cases from literature. A summary of the data inventory for the scale of 10 tonnes of DPS input per month and the data sources is found in Table 1. To make it easier to interpret results that are reported later for the FU of 1 kg of P recovered, P content is shown in Table 1. Further, N content is shown to aid in understanding the influence of N credited in the system expansions.

As part of the inventory work, a mass balance was conducted for the feedstock (10 tonnes of DPS), as well as for chemicals and water. Additionally, a substance flow analysis for the total P content was performed. The overall mass and P balances were performed using spreadsheets for all the input and output information for all systems, both to generate some of the needed inventory data and to check the consistency of some other inventory data. The input concentration of P was the same for all the scenarios in the mass balance calculations, while the P accumulated in the effluents and in the recovered products vary (see Table 1). Since, the quality of the effluents aligns with EU discharge limits, no further treatment was considered in our case.

The upscaling of the process set-up was done for the same scale: a feedstock of 10 tonnes of DPS. Thermal energy, used in drying and for the HTC processes, and electric energy used for pumps and stirrers, were calculated based on Piccinno et al. (2016). The mass of the used chemicals was mostly rescaled based on experimental data. When no experimental data was available, process performance was estimated using literature data. More details are reported in the SM.

Data for the background system and for substitutions, i.e., energy, chemicals and avoided production of CAN and wood chips, were retrieved from the Ecoinvent (https://www.ecoinvent.org) and Sphera (https://www.gabisoftware.com) databases, for EU-25 conditions. Regarding the production of $MgCl_2$ and oxalic acid, no processes currently exist in these databases. Instead, an estimation based on the production of NaCl was used for the $MgCl_2$, as in Raymond Alena et al. (2021) and Högstrand et al. (2023), and acetic acid for the oxalic acid since these two acids can be produced through similar chemical processes (Tan et al., 2012).

2.2.4. Life cycle impact assessment

In an earlier study that reviewed available information in published LCA studies in this field, it was observed that three environmental impact categories were the most common: cumulative energy demand (CED), climate impact based on global warming potential (GWP), and acidification based on acidification potential (AP) (Behjat et al., 2022). It was also pointed out that even though eutrophication is a main driver behind installing P recovery from wastewater treatment, it was only rarely assessed as an impact category in LCA of such technologies. Other recent work that built on and extended the mentioned review (Behjat et al., 2024) suggested a longer list based on environmental impact categories that could be related to general input and output flows for P recovery from dairy wastewater, as well as the priorities of potential decision-makers in technology development in such contexts. Given this input, the CML 2016 (CML-IA Characterisation Factors, 2016) method, or rather, 10 of its 17 impact categories, was selected for the LCIA: global warming potential (GWP 100 years) in kg CO_2 eq.; acidification potential (AP) in kg SO_2 eq., photochemical ozone creation potential (POCP) in kg ethene eq.; eutrophication potential (EP) in kg phosphate eq.; freshwater aquatic ecotoxicity potential (FAETP) in kg dichlorobenzene (DCB) eq., marine aquatic ecotoxicity potential (MAETP) in kg DCB eq., terrestrial ecotoxicity potential (TETP) in kg DCB eq.; human toxicity potential (HTP) in kg DCB eq.; abiotic depletion potential (ADP) for elements in kg Sb eq., and for fossil resources in MJ. The impact calculation was performed using the "LCA for Experts“ software, commonly known as GaBi (https://www.gabisoftware.com).

3. Results and discussion

3.1. Phosphorus recovery rates

Before presenting results from the LCA, it is useful to first highlight the varying levels of P recovery for the different scenarios. Results are compiled in Table 2 (more details about the composition of the hydrocar can be found in Kwapinska et al. (2023)). The final row in Table 2 is the scaling basis for the FU in the LCA results presented in section 3.2.

It is worth noting that 151 kg of the 343 kg P delivered by the first struvite production process (scenarios 1–3) is P that was added as phosphoric acid, and this P does therefore not originate from the DPS itself. It can be useful to think about these types of processes not purely as P recovery processes from a particular source, but rather attempts to integrate fertiliser production with P recovery in various circular economy manifestations. It is clear that scenario 4 can lead to the highest level of P recovery from the DPS itself if the P in the dried DPS can be fully valorised, and that when hydrochar is used as a fuel without preceding P leaching (scenario 2), the highest overall losses of P can be expected.

However, although scenario 4 can lead to the recovery of a high level of P, the direct use of DPS in the soil as fertiliser raises concerns about the nutrient availability and potential impact on soil and crop yield (Ashekuzzaman et al., 2021a). Many countries have different regulations governing the application of sludge to agricultural land to protect water quality and human health, which can pose a limitation to the use of untreated DPS. Furthermore, although struvite recovery might not recover the largest amount of P, it generates a more versatile and valuable P product. Economy or versatility was not considered in this study but should be explored in future work.

This study intends to inform technical development for P recovery technologies foreseen to enter the market. The results support decision-making by quantifying and assessing the potential environmental impacts of the systems, enabling technology developers to understand trade-offs between technical, economic and environmental performance when evaluated together with other information.

Table 1
Quantity of input and output flows as well as the information source for each investigated process.

High Data specificity Low

Technology	Mass and Energy flows		Expert estimation	Scaled up based on Piccinno et al. (2016)	Calculation based on literature data (reference)		P	N
HTC	***Input***	Feedstock (DSP)	10000 kg				572 kg	515 kg
		Thermal energy		2425 kWh				
		Electric energy (pumps)		0.43 kWh				
		Electric energy (stirrer)		0.01 kWh				
		Electric energy (filter)	0.75 kWh					
		H_2SO_4	1012 kg					
	Output	Gaseous emissions*			631 kg	Atallah et al., 2019		
		Hydrochar (dried)	1266 kg				303 kg	245 kg
		Process water	6162 kg				269 kg	270 kg
Leaching	***Input***	Hydrochar (dried)	1266 kg				303 kg	245 kg
		Thermal energy		549 kWh				
		Electric energy			92 kWh	Mannarino et al., 2022		
		HNO_3			321 kg	Mannarino et al., 2022		71 kg
		Water			1330 kg	Mannarino et al., 2022		
	Output	Hydrochar (dried after leaching)			840 kg	Mannarino et al., 2022	67 kg	283 kg
		Leachate			1866 kg	Mannarino et al., 2022	236 kg	33 kg
Struvite production	***Input***	Process water	6162 kg				269 kg	270 kg
		Thermal energy		50 kWh				
		Electric energy (pumps)		0.76 kWh				
		Electric energy (stirrer)		0.12 kWh				
		Electric energy (filter)	1.50 kWh					
		Oxalic acid	200 kg					
		H_3PO_4	562 kg				151 kg	
		$MgCl_2$	1823 kg					
		NaOH	803 kg					
	Output	Effluents	5348 kg				77 kg	125 kg
		Cake	621 kg				0.05 kg	0.75 kg
		Struvite	2788 kg				343 kg	145 kg
Struvite production *Struvite recovery from hydrochar leachate; only for Scenario 3*	***Input***	Leachate			1866 kg	Mannarino et al., 2022	236 kg	33 kg
		Thermal energy		23 kWh				
		Electric energy (pumps)		0.26 kWh				
		Electric energy (stirrer)		0.03 kWh				
		Electric energy (filter)	1.50 kWh					
		Oxalic acid	63 kg					
		$MgCl_2$	226 kg					
		NaOH	253 kg					
		NH_4Cl	324 kg					109 kg
	Output	Effluents	920 kg				67 kg	71 kg
		Cake	196 kg				0.05 kg	0.08 kg
		Struvite	1371 kg				169 kg	71 kg

(*continued on next page*)

Table 1 (*continued*)

			High	Data specificity		Low		
Technology	Mass and Energy flows		Expert estimation	Scaled up based on Piccinno et al. (2016)	Calculation based on literature data (reference)		P	N
Sludge Dewatering + Drying	**_Input_**	Feedstock (DSP)	10000 kg				572 kg	515 kg
		Thermal energy		79 kWh				
		Electric energy (pumps)		0.31 kWh				
		Electric energy (filter)	0.75 kWh					
	Output	Dried sludge			5250 kg	Khalaf et al., 2022	501 kg	180 kg
		Effluents			3437 kg	Khalaf et al., 2022	71 kg	317 kg
		Gaseous emissions	1313 kg					18 kg

[*] Additional information on the gas composition of these emissions is reported in the SM.

Table 2
Mass of phosphorus contained in all products recovered from 10 tonnes of DPS treated for the four different scenarios.

	Scenario 1	Scenario 2	Scenario 3	Scenario 4
P in struvite – *first batch* (kg)	343	343	343	
P in struvite – *second batch only for scenario 3* (kg)			169	
P in hydrochar (kg)	303			
P in dried sludge (kg)				501
Tot P recovered (kg/10 tonnes of DPS)	646	343	512	501

3.2. Overall environmental impact results

The study aimed to analyse the processes involved in the three HTC configurations and the reference drying scenario, for each environmental impact category. The main purpose was to provide an understanding of what influences the environmental impact of each scenario and what should be considered in further technical development work. Here, graphical results are provided as absolute values per 1 kg of recovered P, see Fig. 2. Note that this FU scales all impacts to the varying levels of P in different product flows (and P originates from both DPS and phosphoric acid added in Scenarios 1–3, as discussed in section 3.1). The section of the bars above zero are the impacts related to the different processes in each scenario and the section below zero represents the avoided impacts related to the substitutions. Detailed quantitative results are presented in the SM.

An initial observation is that the drying scenario (scenario 4) generally exhibits low environmental impacts, except when it comes to EP, which is mainly due to the loss of P in the effluent during the dewatering process, but still lower than for scenarios 2 and 3. For several impact categories, the net results for scenario 4 is negative owing to the gains from replacing CAN production due to the N content in the sludge that is considered to be valorised in agriculture.

Although agricultural considerations are in most respects neglected in this study, it is nevertheless important to reflect on the implications of some choices made. P and N availabilities in struvite, in hydrochar and in CAN, respectively, are not fully comparable, but the nutrient availability of the recovered products and replaced conventional fertiliser were here considered the same, which is of course an oversimplification. As already mentioned, hydrochar has been shown to exhibit a low release rate for P. Comparing the struvite with CAN, the first is a slow-release fertiliser; the nutrients are gradually released over time as they are dissolved in soil moisture (Talboys et al., 2016; Valle et al., 2022). However, rapid release of nutrients can increase the risk of nutrient loss to water bodies and to air, if not applied in a way that they can be efficiently used by the plants.

Looking at net outcomes for all scenarios, scenario 2 performs better than the drying scenario in several impact categories; exceptions are AP, EP, FAETP and ADP (fossil). Large gains in scenario 2 are related to the substitution for avoided production of wood chips when hydrochar is used as a fuel. Scenario 3 also produces hydrochar that can be used to replace wood chips, but the additional environmental pressure related to the production of chemicals used for leaching counteracts this, as does the here assumed reduction of the mass of hydrochar used as a fuel due to leaching and the division of the gain over the larger amounts of P recovered. Scenarios 1 and 2 share the exact same core processes but scenario 2 has a better environmental performance for most impact categories due to the use of hydrochar for energy recovery instead of agricultural use.

Given the important effect of the energy recovery and how this replaces wood chips production, it is important to reflect on the theoretical assumptions made. One potential limitation is the use of the HHV for the hydrochar. This HHV can potentially vary, depending on the specific properties and composition of the hydrochar, as well as the experimental conditions of the leaching process. The liquid–solid ratio, acid concentration, and contact time are the acid leaching factors that have a significant effect on the HHV (Liu et al., 2023) and consequently influencing what is relevant to assume in the substitutions. This should be explored when actual upscaling is considered.

Fig. 2 also indicates the overall contribution from chemicals and energy production in the different steps of each scenario. Impacts from chemicals (see all legends marked Chemicals production in Fig. 2) are always more important than impacts from energy (see legends marked Thermal energy or Electricity in Fig. 2) although GWP, MAETP and ADP (fossil) have important contributions from energy. For scenarios 1–3, the environmental burdens due to chemicals production for struvite production and the HTC process are considerable while the chemicals production needed for leaching in scenario 3 has a smaller impact. The production of the chemicals used in struvite production, in fact, causes the main contribution to all the environmental impact categories in scenario 3, except for the EP, see more details in Section 3.3.

Emissions to air during the HTC process influence the GWP results but not the other impact categories, see more details in Section 3.3.

3.3. Contribution analysis – The importance of chemicals production

Graphical results in Fig. 3 show relative values for the contributions of each process in the three HTC system configurations but without the system expansions to allow for a detailed analysis of process-related impacts. Scenarios 1 and 2 are therefore the same since they only differ in their system expansions. Note that the scaling to the P content in

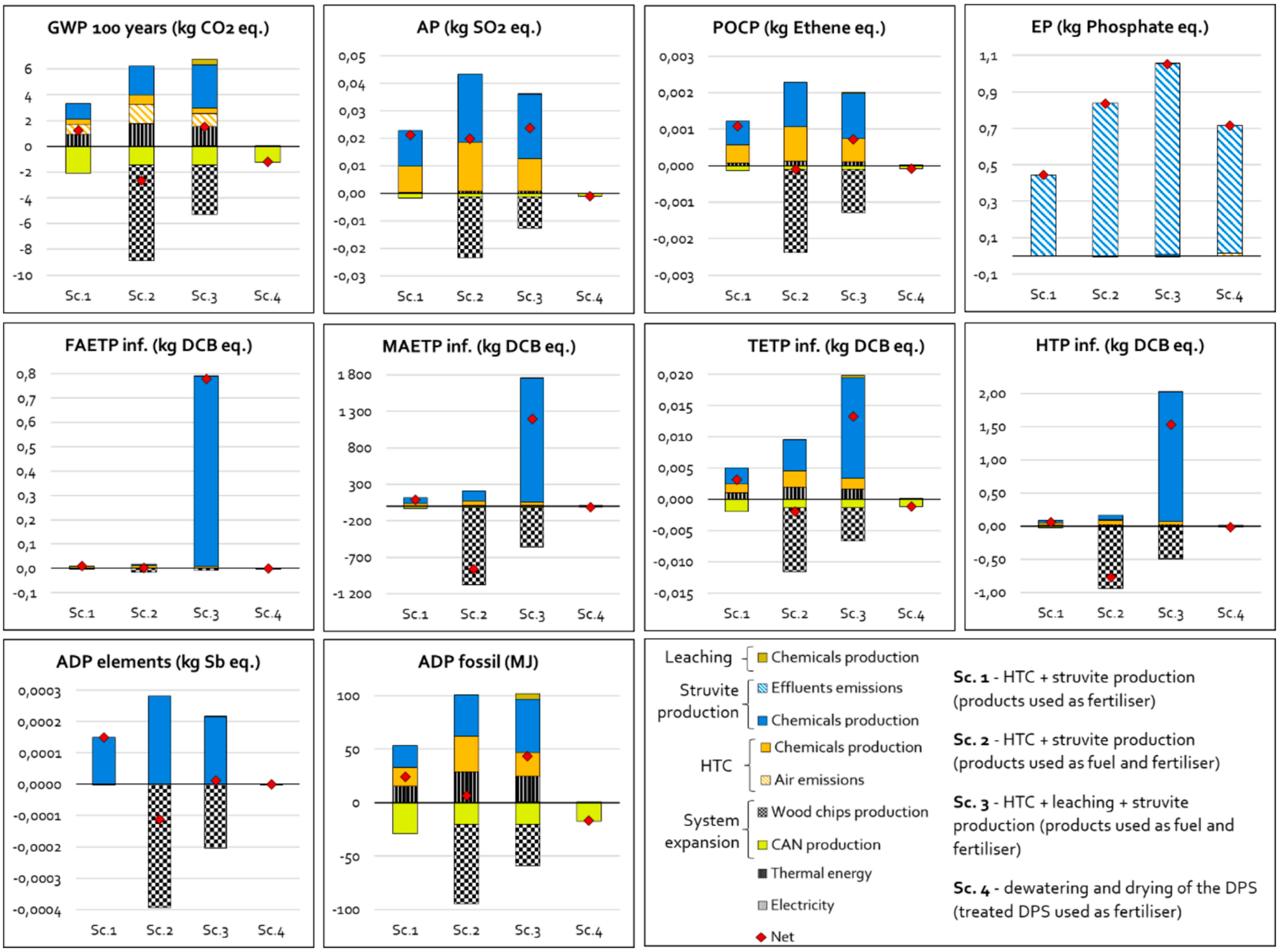

Fig. 2. Life cycle environmental impacts for the three HTC system scenarios (Sc.1, Sc.2, Sc.3) and the drying scenario (Sc.4) for each of the investigated impact categories per 1 kg of recovered P, divided into activities. GWP = global warming potential, AP = acidification potential, POCP = photochemical ozone creation potential, EP = eutrophication potential, FAETP = freshwater aquatic ecotoxicity potential, MAETP = marine aquatic ecotoxicity potential, TETP = terrestrial ecotoxicity potential, HTP = human toxicity potential, ADP = abiotic depletion potential.

the products is not seen in Fig. 3. More details are provided in Fig. 3 than in Fig. 2 and colours are coded to reveal which core process is responsible and the patterns show whether this is via direct emissions, input chemicals or energy.

As already mentioned above, most impacts are due to the production of chemicals (single colour parts in the bars in Fig. 3), mainly those used during the struvite production, except for EP and FAETP for scenarios 1 and 2, for which the shares of impacts related to thermal energy are considerable. The production of thermal energy used for the HTC process has a strong influence on the EP, FAETP and ADP (fossil), and is more pronounced for scenarios 1 and 2 since the additional chemical use in scenario 3 seems to overshadow many other impacts, but as said before, EP is an exception. Production of electricity has a minor impact in all scenarios, since it was used only for the pumps, stirrers and filters.

For potential process development, this indicates the need to use chemicals efficiently and to select, when possible, chemicals with a moderate environmental impact.

The production of the H_2SO_4 used in the HTC process contributes in important ways to many impact categories (exceptions are EP and ADP elements), especially for scenarios 1 and 2. For ADP (elements), NaCl and H_3PO_4 used for the struvite precipitation have the largest contributions for all three scenarios. H_3PO_4 production, in fact, is significantly responsible also for the AP and POCP results. In scenarios 1 and 2, the chemicals used for the struvite production do not have the same significant effect as in scenario 3.

Scenario 3, compared to the other scenarios, used a further chemical, NH_4Cl, in the second struvite precipitation, after leaching, to add additional N needed for the struvite. The production of this chemical has a considerable influence on the four toxicity-related impact categories (FAETP, TETP, MAETP, and HTP). The selection of this particular chemical therefore strongly influences the environmental impact results. In fact, for some impact categories, this chemical has a higher impact than all other chemicals together. Other choices could be explored further.

In our study, the lack of data for some specific chemicals considered in the experiments is a potential limitation. In particular, the replacement of oxalic acid with acetic acid; these two acids, despite similarities in their production processes, use different raw material, oxidation procedure, and reaction conditions. Likewise, the processes involved in the production of $MgCl_2$ and NaCl are very similar. However, although they share similarities due to their extraction from naturally occurring brine sources and subsequent evaporation and crystallization, they are distinct salts with different chemical compositions that may influence industrial scale outcomes. This should therefore be explored more if actual upscaling is considered.

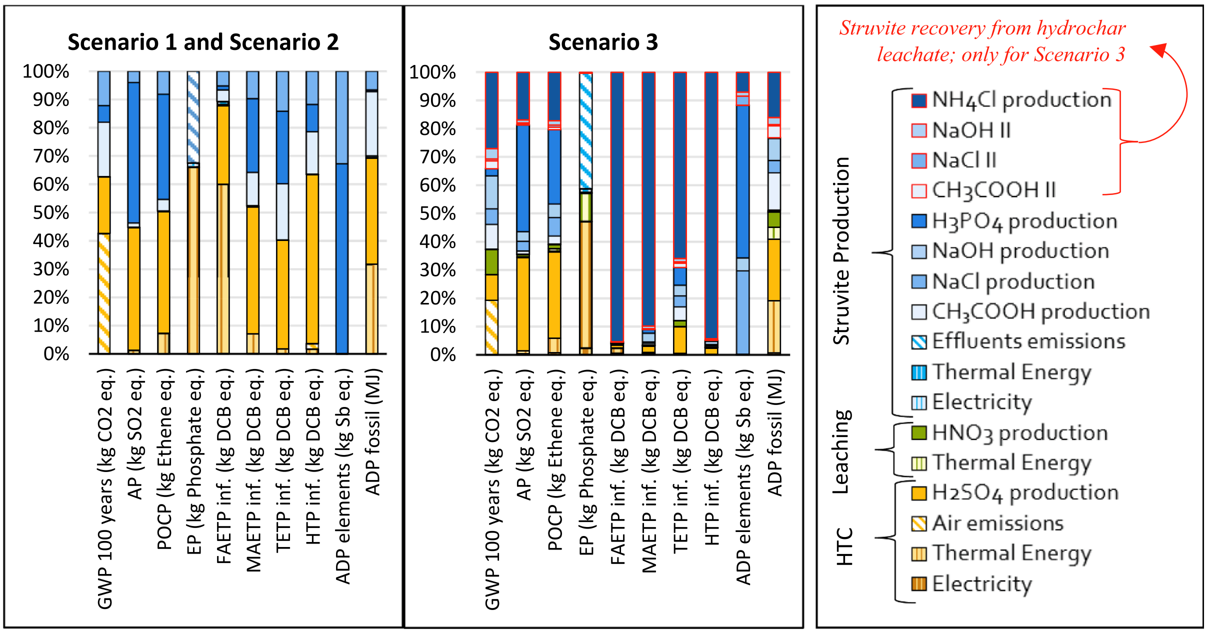

Fig. 3. Contribution analysis for processes involved in the three HTC system configurations. GWP = global warming potential, AP = acidification potential, POCP = photochemical ozone creation potential, EP = eutrophication potential, FAETP = freshwater aquatic ecotoxicity potential, MAETP = marine aquatic ecotoxicity potential, TETP = terrestrial ecotoxicity potential, HTP = human toxicity potential, ADP = abiotic depletion potential.

3.4. *Sensitivity analysis for HTC processing conditions*

Given the early stage of technical development for the HTC process, it is important to consider how the choice of the specific experimental conditions of the HTC process could influence the impact assessment results. Indeed, the hydrochar yield and its P content both vary depending on the temperature (T) and pH (Khalaf et al., 2022). Various experiments have been carried out in the laboratory to determine the optimal experimental conditions for a high concentration of P in the process water. The HTC conditions considered in this study (pH = 2.25 and T = 180 °C), as assessed above, release 47 % of P from the DPS to the process water. If the experiment had been conducted under different experimental conditions (e.g. pH = 4.6 and T = 220 °C), the amount of P released to the process water would be expected to be lower (20 % in the experiments) but so would the acid consumption, but heating needs are higher. A sensitivity analysis was performed to reveal the sensitivity to altered processing conditions. A comparison of some of the environmental impact results for the scenarios for the two different operating conditions are reported in Fig. 4 (impact categories that exhibit interesting differences are shown; more results are reported in the SM).

Due to diminished amounts of total P recovered in the three new HTC scenarios, the environmental impacts associated with the same FU of 1 kg of P recovered under the new experimental conditions are more pronounced compared to the impacts observed in the initial experiments. This affects impacts as well as the savings in the substitutions. Looking at the net outcomes for all impact categories, with the exception

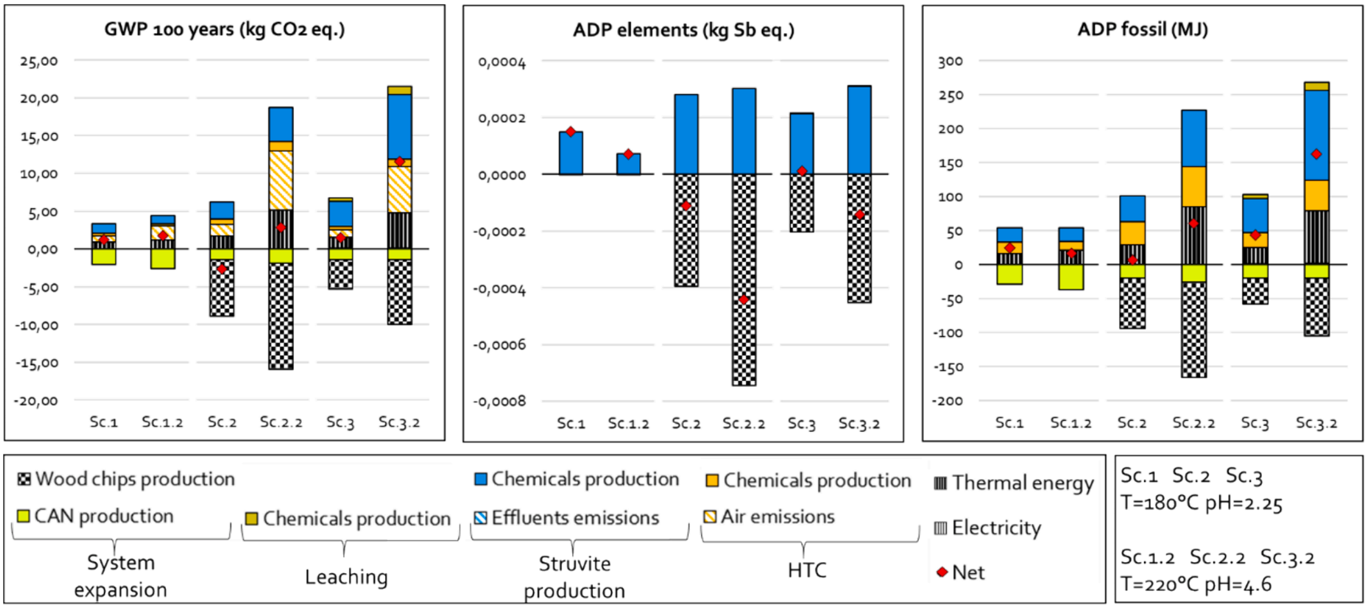

Fig. 4. Global warming potential (GWP) and abiotic depletion potential (ADP) results for the HTC system configurations if the initial experimental conditions (T = 180 °C and pH = 2.5) of the HTC change to T = 220 °C and pH = 4.6.

of GWP, the LCIA indicators for scenario 1.2 are lower than in scenario 1; indicators for scenario 2.2 are lower than for scenario 2 only for ADP (elements), POCP, AP, and HTP; scenario 3.2, however, has lower indicator results than scenario 3 only for ADP (elements) (see SM for more details). Furthermore, the P content in the process water produced under the new experimental conditions is reduced. This reduction leads to a higher consumption of H_3PO_4 and NH_4Cl for the salt dosage of 1.73:1.14:1 for the Mg:NH_4^+:PO_4^{3-} mole ratio during the struvite production process, which results in an increase of environmental impacts. For ADP (elements), scenarios 2.2 and 2.3 perform better, mostly related to the increased capacity to avoid production of wood chips per kg of P recovered. For GWP and ADP (fossil), however, the net outcomes is worse because of the chemical production used for the struvite precipitation and thermal energy production (see Fig. 4). Future efforts should be put into selecting optimal processing conditions. The current study and its underlying experiments can help in this matter but should also consider the actual DPS available and the local context. Other than temperature and pH in HTC processing, the initial chemical-physical characteristics of the DPS will influence the hydrochar yield and its P content.

4. Conclusions and recommendations

Performing life-cycle environmental assessment of a combination of processes that does not yet exist is challenging, but worthwhile to preempt design problems. Scaling up of the considered technologies was performed to provide the data inventory for LCA of alternative scenarios involving DPS treatment and P recovery. This article therefore provides an example of how the lack of specific information at the early stages of process design can be overcome by combining experimental and literature data in an upscaling exercise involving mass balance calculations.

In the present study, the environmental impacts of the experimental HTC and struvite recovery technologies were determined by LCA, and the identified hot-spots were compared to only drying of the DPS. LCA showed to be a useful tool for evaluating the environmental impacts of the described systems and comparing them based on the recovered P amounts in the product flows.

The drying scenario (scenario 4) often performs well, but in many cases, scenario 2 performs even better due to the large gains from using the hydrochar as a fuel and replacing the burning of wood chips. Using additional chemicals to recover additional P from hydrochar (scenario 3) does not seem to pay off in our comparison.

The main hotspots are related to chemicals productions, in particular NH_4Cl and H_2SO_4. Hence, an exploration of alternative input chemicals is recommended before full-scale implementation of the HTC and struvite recovery technologies. Further, ways to mitigate P emissions to the effluent and NH_3 emissions to air should be investigated. Given the early stage of many of the core technologies evaluated, there may be large uncertainties in the inventory data that should be checked before implementation. It was demonstrated that by changing the experimental conditions of the HTC, the environmental impacts change in important ways, in the specific case shown for the worse.

Beyond the environmental consequences discussed here there can be other advantages to treating DPS with HTC, that concern the stability and uniformity of agricultural byproducts, which may simplify storage, transport and use of the recovered. Further research is warranted to quantify such potential benefits.

CRediT authorship contribution statement

Marta Behjat: Writing – review & editing, Writing – original draft, Software, Investigation, Data curation, Conceptualization. **Magdalena Svanström:** Writing – review & editing, Supervision, Funding acquisition, Conceptualization. **Gregory Peters:** Writing – review & editing, Supervision, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary material

Supplementary material to this article can be found online at https://doi.org/10.1016/j.wasman.2024.06.011.

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