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Life cycle assessment of struvite recovery and wastewater sludge end-use: A Flemish illustration

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ABSTRACT

Phosphate rock (PR) has been designated as a Critical Raw Material in the European Union (EU). This has led to increased emphasis on alternative P recovery (APR) from secondary streams like wastewater sludge (WWS). However, WWS end-use is a contentious topic, and EU member states prefer different end-use pathways (land application/incineration/valorisation in cement kilns).

Previous Life Cycle Assessments (LCA) on APRs from WWS reached contrasting conclusions; while most considered WWS as *waste* and highlighted a net benefit relative to PR mining and beneficiation, others viewed WWS as a *resource* and highlighted a net burden of the treatment. We used a combined functional unit (that views WWS from a *waste* as well as a resource perspective) and applied it on a Flemish wastewater treatment plant (WWTP) with struvite recovery as APR technology. Firstly, a retrospective comparison was performed to measure the WWTP performance before and after struvite recovery and the analysis was complemented by uncertainty and global sensitivity analyses. The results showed struvite recovery provides marginal environmental benefits due to improved WWS dewatering and reduced polymer use. Secondly, a prospective LCA approach was performed to reflect policy changes regarding WWS end-use options in Flanders. Results indicated complete mono-incineration of WWS, ash processing to recover P and the subsequent land application appears to be less sustainable in terms of climate change, human toxicity, and terrestrial acidification relative to the status quo, i.e., co-incineration with municipal solid waste and valorisation at cement kilns. Impacts on fossil depletion, however, favour mono-incineration over the status quo.

1. Introduction

Phosphorous (P) plays an essential role in the metabolism of living organisms and is an irreplaceable macronutrient in food and feed production. Crops are supplemented with P fertilizer, which can be derived from primary (phosphate rock) and secondary sources (waste streams-wastewater sludge, manure) (Geissler et al., 2018).

Unlike Nitrogen or Potassium that is abundantly available, the supply of primary P is limited to ores sourced from sedimentary and igneous phosphate rock (PR) deposits (Geissler et al., 2019). Currently, the global anthropogenic P flow due to mining is 44 Mt P/ year of which 35% (15.5 Mt P) are lost during the beneficiation process. These beneficiation losses are almost three times higher than estimated annual P flows in municipal wastewater (around 4–5.3 Mt P per year). Despite these proportions, European countries promote efforts and research to the recycling of P from secondary streams such as municipal wastewater sludge (WWS) (Scholz and Hirth, 2015). There are two main reasons for this (i) high aquatic pollution due to high non-retrievable P losses (approximately 35%) and (ii) precautionary management of world food supply (Scholz and Wellmer, 2015). Furthermore, the European Union (EU) faces a geopolitical liability: while global P scarcity is unlikely to occur in the foreseeable future (Scholz and Hirth, 2015; Scholz and Wellmer, 2013), import-dependencies of the EU are at 92% (De Boer et al., 2018). Given the geopolitics surrounding PR, the EU included P in its list of Critical Raw Materials (CRM). The CRM is complementary to another EU agenda, i.e., fostering a circular economy for P. This has provided an impetus to implement alternative P recovery (APR) techniques from wastewater sludge.

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Struvite crystallization is one of the most frequently proposed APR techniques for use in full-scale wastewater treatment plants (WWTPs) in the EU (Kabbe and Remy, 2015), as it can recover approximately 10%–30% of the influent P (Egle et al., 2016). Struvite deposition is a natural occurrence in WWTPs, and clogs pipes via encrustation and scaling, resulting in high operating and maintenance costs (De Boer et al., 2018; Doyle and Parsons, 2002). To address these concerns, WWTPs are increasingly implementing intentional struvite recovery. It is recovered from either (i) digested WWS or (ii) the centrate formed after WWS dewatering (Huygens et al., 2019).

The technology analysed in this study is the Nutrient Recovery System (NuReSys®), a full-scale struvite recovery plant, installed at Aquafin WWTP in Leuven (capacity: 120,000 inhabitants; 36,000 m^3 wastewater inflow/ day). The struvite here is recovered from digested WWS, preceded by an Enhanced Biological Phosphorus Removal (EBPR) system. The NuReSys® concept has been explained by Marchi et al. (2020) and further information regarding the techno-economic aspects is available in Saerens et al. (2021) and Marchi et al. (2015).

While the solubility of struvite is lower than that of most mineral P fertilizers, it has shown to be an efficient fertilizer in plant growth trials (Möller et al., 2018; Vaneeckhaute et al., 2016). Also, struvite recovered from WWTPs has significantly lower Cadmium and other heavy metal concentrations when compared to synthetic P fertilizers (Egle et al., 2016; Kataki et al., 2016). Although there have been some questions regarding its market potential, especially amongst fertilizer companies (De Boer et al., 2018), struvite has achieved a secondary 'product' status, provided it complies with the minimum nutrient content, maximum limit values for inorganic contaminants, and biological pathogens (EC Regulation No 2019/ 1009).

Further downstream, almost two-thirds of residual P remains in the WWS, despite optimistic P recovery rates through struvite. There are different possibilities amongst EU member states for WWS end-use, such as land-spreading or incineration. In Belgium, WWS is regionally managed, with separate regulations governing Flanders, Wallonia, and the Brussels region. Belgium produces 1.03×10^9 kg dry matter (DM) of WWS annually (Kelessidis and Stasinakis, 2012). While more than 90% of WWS is incinerated in Flanders and the Brussels region, only 45% is incinerated in Wallonia, with the remainder being applied to the land. In Flanders, approximately two-thirds of WWS is co-incinerated with municipal solid waste, while the remaining one-third is dried and pelletized for use as an alternate fuel in cement kilns. By 2026, Flanders intends to implement mono-incineration for all WWS generated, with an emphasis on energy recovery via incineration and P recovery via ash processing (Aquafin 2020). With this plan in prospect, and the potential for struvite as a P fertilizer, as well as the operational benefits for the WWTP associated with its implementation (Saerens et al., 2021), an environmental impact assessment is needed, firstly, to evaluate possible benefits or drawbacks of P recovery in the form of struvite and secondly, to evaluate WWS end-use.

There have already been three comprehensive reviews that assessed the environmental impacts of APRs like struvite recovery and WWS end-use from municipal WWTPs (Ding et al., 2021; Lam et al., 2020; Sena and Hicks 2018). Around 65 peer-reviewed Life Cycle Assessments (LCA) revealed that 90% of the studies conferred WWS with a *waste* or *waste-to-product* status instead of a *product* status (Lam et al., 2020). Considering WWS as a *waste* effectively means that the environmental impacts from wastewater treatment are cut-off from the system boundaries because the sole function of a WWTP is to treat the influent wastewater. Therefore only impacts due to WWS treatment and APR are considered. On the other hand, from a *product* perspective, it is seen as a multi-functional system that produces clean water as well as P fertilizer and therefore the entire system boundary including wastewater treatment is considered.

The main discussion points from these reviews on the current status of APR LCAs included (i) methodological inconsistencies, such as the multi-functionality conundrum for WWS (Lam et al., 2020), (ii) the need for a more focussed and detailed assessment of a full-scale struvite recovery system (Sena and Hicks, 2018), (iii) lack of consideration for the uncertainty of results in many studies (Lam et al.2020) and (iv) lack of transparency on the life cycle inventory (LCI), methodological assumptions and system boundaries (Lam et al.,2020).

We aim to present a transparent LCI and corroborate the LCA results of the LCA for struvite recovery and WWS end-use with uncertainty and global sensitivity analyses, which may be of interest to future LCA practitioners working on P recovery. Furthermore, the results from this study may inform policy making with regards to Flanders' management of WWS in the future. The specific objectives are:

- To compare the environmental impacts of a WWTP before and after struvite recovery in the environmental and legislative circumstances in Flanders.
- To compare the environmental performance of different WWS end-use scenarios, including valorisation at cement kilns and co-incineration with municipal solid waste, mono-incineration, and land application, all of which assume struvite recovery.

2. Material & methods

2.1. Modelling approach

To investigate the first objective, we took a retrospective approach to compare the existing WWTP with struvite recovery versus a point in the past, where the WWTP functioned without struvite recovery. As Ekvall et al. (2005) pointed out, a retrospective approach can be used to ascertain whether or not to become associated with a system, for instance, the product that is being investigated, which in our case is struvite recovery. Time does not affect the LCI modelling principles, so it is possible to either use an attributional or consequential perspective for retrospective LCAs (ILCD, 2010). We chose an attributional perspective since it can trace back impacts in the relative past, before product provision (Schaubroeck et al., 2021). Here, the product system is composed of all allocated shares of activities and uses average market suppliers.

To investigate the second objective, we chose a prospective approach to evaluate the future end-use options for WWS, assuming upstream struvite recovery. A consequential perspective is chosen in this case since it can best describe and estimate the consequences of a decision (Ekvall, 2019).

2.2. System boundaries

Multi-functionality in LCAs can be solved by either system expansion (SE) or allocation. For SE, the boundaries of the studied system are expanded to include the impacts of alternate production of exported functions (Ekvall and Finnveden, 2001). Or, in other words, it enables a fair comparison of a system with two outputs versus a system with one output, the boundaries of the latter system are "expanded" to add the product of interest.

The rationale for choosing SE is (i) it is recommended as a first-step option by ISO 14,044:2006 (ii) Allocation is still a contentious topic amongst the LCA community and (iii) SE is shown to be the only system model that consistently maintains mass balances of the resulting single-product systems for consequential LCAs (Schmidt and Weidema, 2007). We thus base our study on SE.

2.2.1. Objective 1

In circular economy LCAs, where materials are in a loop, it is important to devise a functional unit (FU) capable of accounting for the upstream impacts of waste treatment and further downstream impacts after the *waste* transitions into a useful *product*. Similarly in our study which is based on open-loop recycling, we propose a combined FU which views the system from both a product as well as a waste perspective. The combined FU chosen for Objective 1 is 1 kg of plant-available P (FU_1) recovered in the form of struvite, and treatment of its equivalent wastewater inflow, i.e. 3927m^3 (FU_2).

Fig. 1 illustrates the system boundaries for Objective 1. Scenario 1(a) compares the present-day impacts of the WWTP versus the environmental impacts before struvite recovery was implemented (Scenario 1 (b)). In scenario 1(a) the system is already multi-functional, i.e. the influent is treated and a 'useful' P product is generated. In 1(b) however, the system's normal function is to treat the influent. To ensure a fair comparison between 1(a) and 1(b), we expand the system by incorporating 1 kg of plant-available P in the form of synthetic fertilizers. For synthetic P, only the fertilizers manufactured through the sulphuric acid route are considered (Triple superphosphate (TSP) and Single superphosphate (SSP)). The system boundary for TSP and SSP includes mining and beneficiation, transport, and processing of PR to the end product.

According to the chosen attributional approach we conducted a system expansion using a marked mix of P fertilizers. The estimated market share of marketable-phosphate rock in Germany is 58% from Israel, 28% from Senegal and 14% from Morocco and the market shares for TSP and SSP (fertilizers manufactured through the sulphuric acid route) are 71% and 29%, respectively (Kraus et al., 2019). We assume the same market share for Belgium and the LCI for TSP, SSP, sulfuric acid production, and PR beneficiation process is based on Kraus et al. (2019), who updated the existing LCI in the ecoinvent database.

Various studies have proven the P efficacy of struvite and its plant availability (Bogdan et al., 2021; Egle et al., 2016; Saerens et al., 2021). Therefore, an important assumption in Objective 1 is that all the P in the struvite is considered plant available. Also, the infrastructure component, i.e., building or equipment and the sewer network have not been considered in the analysis.

2.2.2. Objective 2

Objective 2 is assessed through 3 sub-scenarios and follows a consequential approach. We assume that the WWTP is implemented with the NuReSys® struvite recovery system. The FU is 1 tonne of digested sludge (DM content 5%) (Fig. 2).

Scenario 2(a): Scenario 2(a) represents the status quo of WWS treatment in Flanders (330 kg for valorisation at cement kilns and 670 kg for co-incineration). While the DM content of dewatered WWS (30%) is sufficient for co-incineration, valorisation at cement kilns requires a higher dry matter content i.e., 70–80%. Therefore, an intermediate drying step has been considered for 330 kg of WWS. After drying, the WWS is transported to the cement kiln, where it is used as an alternate fuel for clinker production. The average transport distance to the cement industry is 110 km (Saerens, 2021).

The system boundary for alternative fuel use (in this case WWS) excludes the "production-to-gate" activities like cement milling, transport to end-users, and subsequent use since the environmental performance of these activities is unaffected by a shift in fuel type (Prosman and Sacchi, 2018). Here, the use of WWS as an alternative fuel for clinker production is the marginal treatment option for WWS and it substitutes conventional fuel, i.e., hard coal, based on its calorific value.

Global coal consumption is estimated to have fallen by 7% (500 million tonnes) between 2018 and 2020. Assuming global economic recovery after the COVID-19 pandemic, the International Energy Agency (IEA) forecasts a 2.6% increase in coal demand in 2021, mainly led by China, India, and the ASEAN countries. Trends, however, show a decline in demand for coal in the EU (Arnold et al., 2020), which leads to a drop in coal prices. During a decreasing market trend, most coal producers are unlikely to invest in new technology, and therefore the affected

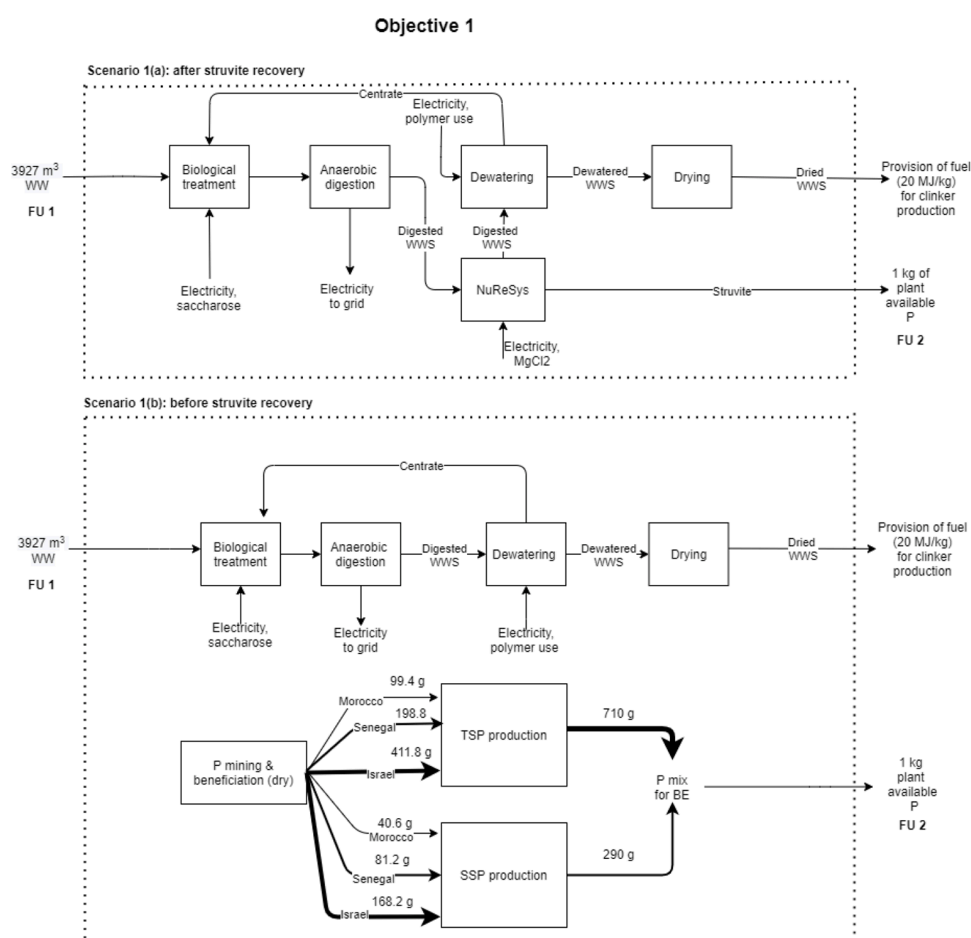


Fig. 1. System boundary diagram for Objective 1. FU_1 and FU_2 represent the combined functional unit.

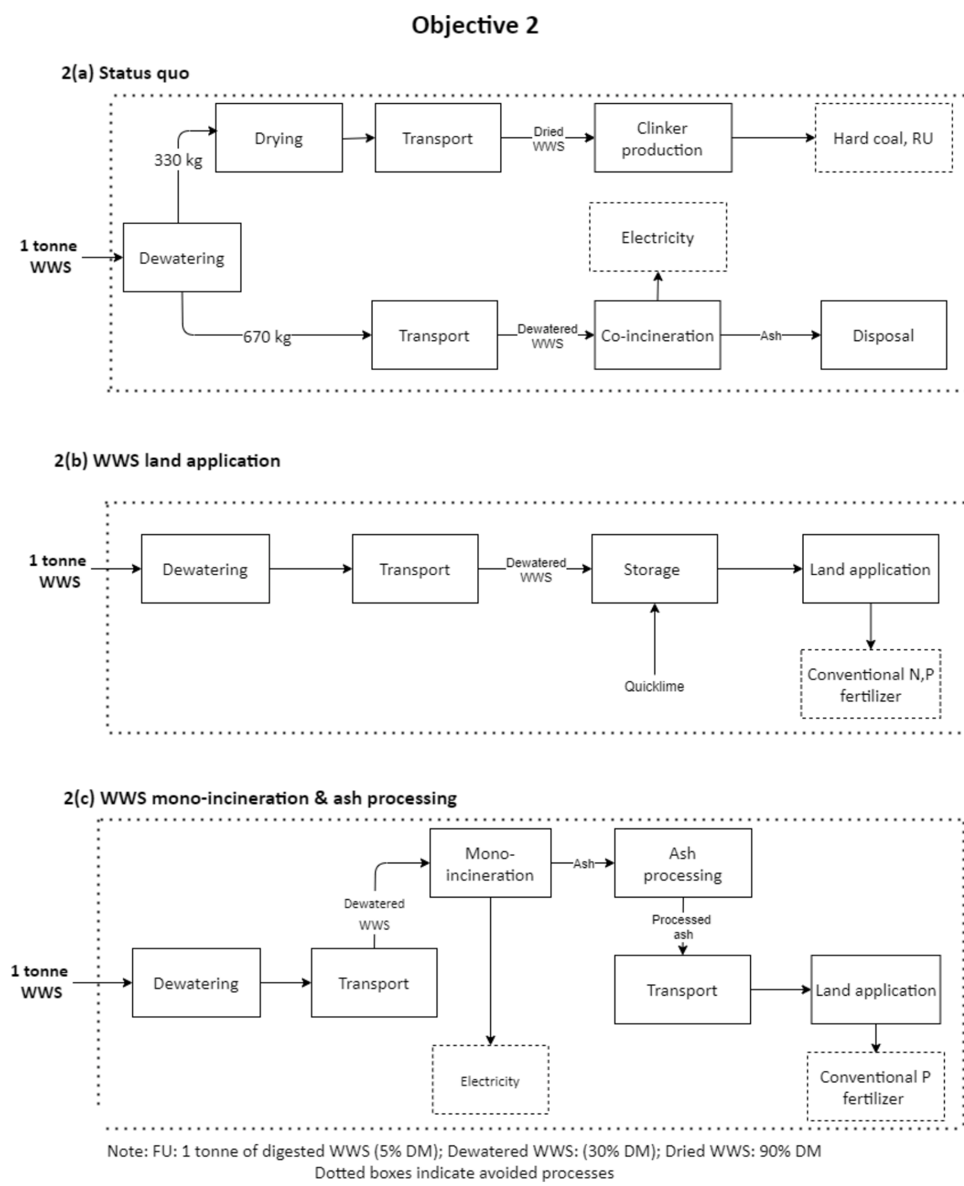


Fig. 2. System boundary diagram for Objective 2. FU is 1 tonne of wastewater sludge to be managed.

marginal producers of coal are the least competitive (Weidema, 2014). These are typically the deep and underground mines that have higher costs. In our case, we assume that the marginal coal producer is Russia since European coal imports are mainly sourced either from the Russian Federation or Colombia (1472 kt and 15kt, respectively) (IEA, 2019). Since the ecoinvent database does not have a process for Colombian coal, Russia was selected as the marginal producer.

For the remaining two-thirds, the WWS is dewatered and transported to a co-incineration facility processing municipal solid waste and WWS. The distance to the co-incineration facility is unknown, and hence we consider a default distance of 100 km to end-of-life treatment (Zampori and Pant, 2019). The electricity (gross electric efficiency: 28.51%) and heat (gross thermal efficiency: 28.51%) produced from co-incineration are fed into the Belgian national grid, for which equivalent credits are included. We consider landfilling of ash produced from co-incineration since it has a lower P concentration and potentially a higher amount of impurities and contaminants (Huygens et al., 2019).

Scenario 2(b): Legislation in Flanders prohibits direct WWS land application, but it is common practice in other parts of Belgium (Wallonia) as well as other parts throughout Europe. Therefore, Scenario 2(b) represents a hypothetical 'what-if' situation where Flemish WWS is transported and land applied in Wallonia, assuming no legal constraints for inter-regional transport of WWS within Belgium. For transportation, a distance of 500 km is considered.

When applied on land, P in WWS is either organically bound or in mineral form. It is common practice to apply WWS for several years of crop requirement through a single spreading. Amongst the peer-reviewed APR LCAs, most assume a direct 1:1 substitution between P in secondary P products (such as WWS) and synthetic P fertilizers. However, this may be overoptimistic for many scarcely available P products such as sewage sludge and ash. To estimate the substitution rates of secondary P product application, Hoeve et al. (2018) developed a P life cycle inventory model (P-LCI). For our study, we applied the P-LCI model to estimate the substitution, plant uptake and losses of WWS and processed ash (Scenario 2(c)). Further information regarding P-LCI and the model calculations are presented in *'A-Supplementary information, Section 1'*.

The fate of heavy metals (Pb, Cd, Cu, Hg, Ni, Zn) from WWS land application is inventoried as direct emissions to the soil (Niero et al., 2014).

Scenario 2(c): 2(c) represents the future scenario in Flanders. To reflect this situation as closely as possible, mono-incineration of dewatered WWS is considered, followed by sodium sulphate processing of the ash. Credits due to electricity generated from mono-incineration as well as the substituted P fertilizer (due to processed ash application) are included. The ash substitution is calculated by the P-LCI model.

Scenario 2(c) would typically fall under the purview of a prospective or an ex-ante situation wherein, the future is assessed through a range of possible scenarios that define the space in which a particular technology would operate (Cucurachi et al., 2018). In our case, we consider the predominant management pathway of WWS in the future to be mono-incineration. Furthermore, the marginal electricity mix linked to mono-incineration is likely to change. A common approach in ex-ante LCAs involves forecasting the future energy mixes of a particular region and linking them to the foreground system. We believe this is particularly pertinent to Belgium since there is a planned phase-out of 5.94 GW nuclear generation capacity (36% of total mix) until 2025.

Total consumption in 2020 stood at 81 TWh and the mixes are detailed in *'A-Supplementary information, Table A.4'*. The Belgian electricity mix in the ecoinvent 3.6 database (Substitution, consequential, long-term) is based on the future electricity market compositions forecasted by the European Commission and the IEA (Ecoinvent 2021). However, there are discrepancies between the IEA forecast and the alternative forecasts made by the Belgian Federal Planning Bureau (FPB) (Buyle et al. 2019). The FPB forecast alternatives include varying levels of GHG reductions (27% for Alt1, 32% for Alt2, and 35% for Alt3) prompted by the non-Emission trading scheme (non-ETS) sector in 2030 compared to 2005. The non-ETS sector includes buildings and transport.

Until 2030, the deficit created by nuclear phase-out in Belgium and reduction in coal results in market delimitation for the electricity mix. This can be addressed by expanding the market and including cross-border exchanges (CBE), i.e. imports and exports. The criteria to include countries involved in CBE is determined using a ratio of trade flow compared to the total production volume of the market, and an arbitrary ratio of 3% is considered (Buyle et al. 2019). We use the same rationale and include Germany, Netherlands, and France for CBE.

In our study, we used the projections made by the FPB, to calculate the percentage of CBE, based on the representation of technologies, market mechanisms, and policy instruments. More information on the calculation is available in 'A-Supplementary information, Section 2'

2.3. Life cycle inventory

The life cycle inventory is explained in two segments. The foreground systems for Objective 1 was built from primary data available from Aquafin Inc and the background processes were linked using the ecoinvent database (version 3.6) (Wernet et al., 2016). For Objective 2, the LCI was built from literature and the ecoinvent database. The geographical context is set to Flanders and the LCA model's response is measured through uncertainty and global sensitivity analyses. The key exchanges are presented in *'A-Supplementary information, Section 3'*

The LCI was modelled using a combination of the Activity Browser (Steubing et al., 2020) and Brightway2 (Mutel, 2017).

2.4. Life cycle impact assessment method (LCIA)

For our analysis, we chose the ReCiPe LCIA method at the midpoint level (Huijbregts et al., 2017). The following impact categories are relevant for LCAs related to wastewater treatment (Niero et al., 2014; Renou et al., 2008):

- Climate change potential - in kg CO_2 equivalent (eq),
- Fossil depletion potential - in kg oil eq,
- Human toxicity potential - in kg 1,4 dichlorobenzene (DB) eq,
- Terrestrial, freshwater, and marine ecotoxicity potential - in kg 1,4 DB eq,
- Freshwater eutrophication potential - in kg P eq,
- Marine eutrophication potential - in kg N eq.

2.5. Uncertainty and sensitivity analysis

To address uncertainty in LCA, International Organisation for Standardisation (ISO) and Joint Research Centre (JRC) recommend a Monte Carlo (MC) simulation be carried out to support a rigorous conclusion. Typically, the number of MC runs chosen in LCAs is arbitrary and varies between 1000 and 10,000, but we considered 1000 runs.

In what is an extension of MC simulation, a global sensitivity analysis (GSA) determines how much the uncertainty of each input parameter can contribute to the variance of the output uncertainty. There are several methods to compute GSA for LCA (Groen et al., 2017), but for our study, we adopt the Sobol indices method using the Saltelli approach (Saltelli et al., 2019) and the Borgonovo approach (Borgonovo and Peccati, 2007).

The Saltelli method estimates sensitivity indices, namely the first-order sensitivity index (S_1) and the total-effect index (ST) based on relative variances. For instance, the total effect of factor 1 on the output variance of a model is given in the following equation

$$ST_1 = S_1 + S_{12} + S_{13} + S_{123} \tag{1}$$

where S_1 describes the effect of factor 1 on the model output whereas S_{12} describes the second-order sensitivity indices, i.e., interactions between factor 1 and other factors (2 or 3) that contribute to the model variance.

The Borgonovo method estimates the delta index (δ), where δ represents the normalized shift in the distribution of model output, provoked by the input parameter. The value of delta lies between 0 and 1, and 0 would imply that the output uncertainty is independent of the input parameter (Eq. (2)).

$$\delta = \frac{1}{2} E_{x_i} \left[\int_{D_Y} \left| f_Y(y) - f_{Y|X_i}(y) \right| d_y \right] \tag{2}$$

Where $f_Y(y)$ is the output distribution and $f_{Y|Xi}$ (y) is the conditional output distribution y for an input variable x_i and E_{x_i} is the expected value of input variable x_i.

The SAlib library in python and Activity Browser was used to perform the GSA. The selected impact categories for the GSA included climate change, freshwater ecotoxicity and human toxicity potentials.

3. Results

3.1. Objective 1: overall results and contribution analysis

Potential impacts in selected impact categories are reported in this section, while the rest are available in *'D- Objective 1 results'*.

The violin plots in Fig. 3 depict the overall impacts while the heatmaps define impact contributions, where positive values represent environmental burdens and negative values environmental benefits. Both the violin plots and the heatmaps for each impact category are split into two subplots, with the first one comparing Scenarios 1(a) and 1(b), and the second ('diff') highlighting the difference between 1(a) and 1(b). The violin plots show a probability distribution using kernel density estimates (KDE) of overall impacts from the MC simulation results. KDE is a non-parametric method to estimate the probability distribution of a finite data set, or in other words, the KDE attempts to infer characteristics of a population, based on a finite data set. In this case, the finite data set refers to results from each MC run.

The results indicate that Scenario 1(a) i.e., after-struvite recovery, has slightly lower impacts than Scenario 1(b) i.e. before-struvite recovery for all impact categories except freshwater eutrophication, which is similar.

3.1.1. Climate change and fossil fuel depletion potential

The climate change potential for 1(a) [median: 455 kg CO_2-eq] is normally distributed, while 1(b) [median: 476 kg CO_2-eq] appears to be

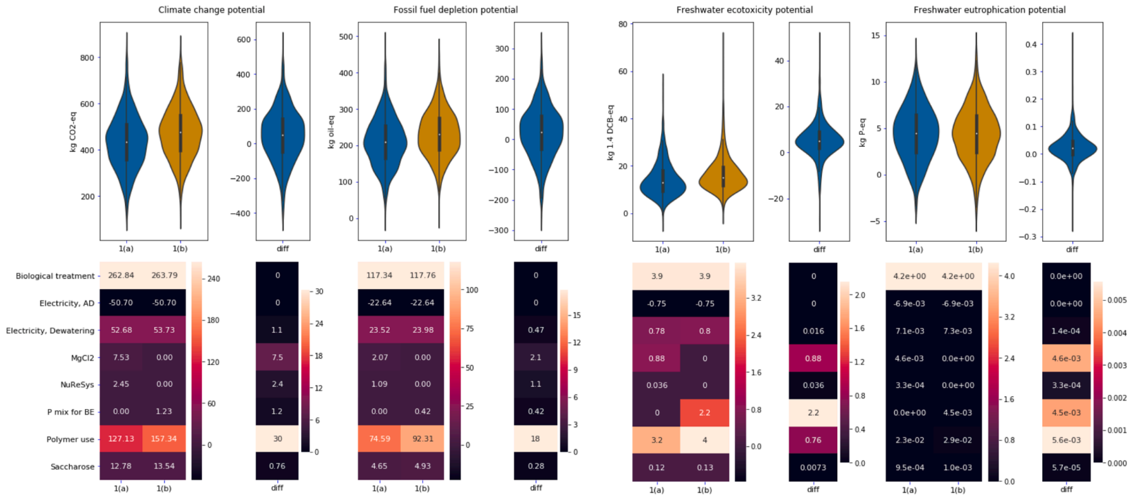


Fig. 3. Objective 1- Overall impacts (violin plots) and process contributions (heat map) 1(a) represents one FU in a scenario with struvite recovery, 1(b) one FU in a scenario without struvite recovery and 'diff' represents the difference between 1(a) and 1(b). White dots in the violin plots represent the median values, boxes represent the interquartile range; violins represent probability distributions using kernel density estimation on either side.

normally distributed albeit with a long tail, implying a high occurrence of extreme values. A deeper analysis of the individual contributions revealed a negligible difference in impacts from biological treatment and anaerobic digestion between both scenarios. Leaving these impacts aside, the major impact contribution in Scenario 1(a) is the struvite recovery step (NuReSys® and $MgCl_2$ usage), which contributes to around 10 kg CO_2-eq. This is roughly 8 times higher than the corresponding conventional P fertilizer imports (1.2 kg CO_2-eq) in Scenario 1 (b). However, these impacts are offset by the polymer (polyacrylamide) use in Scenario 1(b) which is 21% higher (157 kg CO_2), when compared to 1(a) (127 kg CO_2-eq). The upstream impacts from polyacrylamide manufacturing are due to the ammoxidation process used to produce acrylonitrile (79.8%).

Given the close proximity of medians for 1(a) [455 kg CO_2-eq] and 1 (b) [476 kg CO_2-eq], the difference ('diff') between both is expected to be close to 0. However, the violin plot for 'diff' produces a median of 48 kg CO_2-eq and is characterised by a wide range, implying high uncertainty. This is mainly a consequence of polymer usage, whose input in the WWTP has high variance. The variable polymer usage subsequently impacts the climate change potential score, shifting the median value away from 0. This evidence is further corroborated by the Borgonovo (δ) and the first-order Sobol (S_1) indices for Scenario 1(a) and 1(b). As shown in Table 1, the most important contribution to variance in the climate change potential for both 1(a) and 1(b) is the parameter polymer use [δ = 0.49, S_1 =0.73 for 1(a); δ = 0.54, S_1 =0.79 for 1(b)]. Other sensitive parameters on the output uncertainty include the activities under dewatering and biological treatment.

The trend for fossil fuel depletion potential, like climate change, shows an occurrence of extreme values for 1(a), 1(b) and 'diff'. While 1 (a) appears to be normally distributed [median: 217 kg oil-eq], 1(b) shows a plateaued peak [median: 232 kg oil-eq], implying an equal probability of values over the interquartile range. The 'diff' for fossil fuel depletion potential too, exhibits a wide range, and similar to climate change potential this can mainly be attributed to polymer usage. Thus, the WWTP shows a reduction in climate change and fossil depletion potential after the implementation of struvite recovery.

Table 1
Borgonovo Delta (δ) indices and first-order Sobol (S_1) indices for climate change and freshwater ecotoxicity potential (Objective 1).

Climate change potential							
Scenario 1(a)				**Scenario 1(b)**			
Activity	***GSA parameter***	***δ***	***S_1***	***Activity***	***GSA parameter***	***δ***	***S_1***
Dewatering	Polymer usage	0.49 ±0.01	0.72 ±0.03	Dewatering	Polymer usage	0.54 ±0.02	0.79±0.03
Biological treatment	Electricity	0.08 ±0.01	0.06 ±0.03	Biological treatment	Electricity, natural gas, combined cycle power plant	0.08 ±0.02	0.06±0.02
NuReSys®	Market for sodium chloride, powder	0.07 ±0.03	0.01 ±0.03	Dewatering	Ammonia production, steam reforming, liquid	0.05 ±0.01	0.007 ±0.011
Dewatering	Market for chemical factory	0.06 ±0.02	0.01 ±0.01	Dewatering	Transport, freight, lorry	0.05 ±0.01	0.007 ±0.01
Biological treatment	Saccharose use	0.06 ±0.01	0.02 ±0.01	Biological Treatment	Saccharose use	0.04 ±0.01	0.02±0.01
Freshwater ecotoxicity potential							
Scenario 1(a)				**Scenario 1(b)**			
Activity	***GSA parameter***	***δ***	***S_1***	***Activity***	***GSA parameter***	***δ***	***S_1***
Dewatering	Polymer usage	0.36 ±0.04	0.60 ±0.07	Dewatering	Polymer usage	0.39 ±0.03	0.66±0.05
Dewatering	Sulfidic tailings, from copper mine operation	0.10 ±0.03	0.09 ±0.07	P mix for BE	Chemical factory, organics	0.10 ±0.03	0.05±0.05
Dewatering	Steel low-alloyed, market for steel	0.07 ±0.03	0.07 ±0.06	P mix for BE	SSP production	0.07 ±0.02	0.005 ±0.017
Dewatering	HCl production, for polyaluminium chloride	0.07 ±0.03	0.08 ±0.02	P mix for BE	TSP production	0.07 ±0.01	0.015 ±0.01

δ represents Delta lies between 0 and 1 and it is zero when the model output is independent of the parameter.
"S1″ is the Sobol first-order indices: it measures the contribution to the output variance by a single model input alone.

3.1.2. Freshwater ecotoxicity and eutrophication potential

Freshwater ecotoxicity potential was lower for Scenario 1(a) [median: 13.15 kg DCB-eq] than Scenario 1(b) [median: 15.34 kg DCB-eq]. The violin plots indicate that the potential of 1(b) is associated with larger uncertainty than 1(a). Based on the GSA (Table 1), the most sensitive parameters for 1(a) were associated with the dewatering activity, with polymer usage contributing most to the output uncertainty [$\delta = 0.36$, $S_1 = 0.60$]. For 1(b), the sensitive parameters were associated with the dewatering activity as well as the P mix. The median [5.14 kg DCB-eq] for 'diff' shifts away from 0 due to the aforementioned uncertainties of polymer usage and P mix.

The heatmaps show that the potential impacts from freshwater ecotoxicity due to the P fertilizer mix is 2.3 kg DCB-eq. Further analysis upstream shows that the impacts from the P mix are due to TSP (52%) and SSP (48%) manufacturing. As for other activities, polymer usage contributed to a 19% increase in freshwater ecotoxicity potential in 1(b) (3.95 kg DCB-eq) over 1(a) (3.19 kg DCB-eq).

For freshwater eutrophication potential, the overall impacts of 1(a) and 1(b) are similar and are to a large extent influenced by effluent discharge from the WWTP. The violin plot for the 'diff' shows the median [0.02 kg-P eq] is close to 0 but is still influenced by extreme values. This can mostly be attributed to the struvite recovery process. Thus, implementing struvite recovery at a WWTP may possibly reduce potential freshwater ecotoxicity impacts, whereas there is no significant difference for eutrophication potential.

3.2. Objective 2: Overall results and contribution analysis

3.2.1. Climate change and fossil fuel depletion potential

Fig. 4 indicates that Scenario 2(b), i.e., land application of WWS performs better than the status quo-Scenario 2(a) (Clinker production and co-incineration), and the future perspective Scenario 2(c) (mono-incineration). Emissions due to WWS storage (62 kg CO_2-eq) and quicklime production for WWS stabilization during storage (17 kg CO_2-eq), contribute to more than 90% of the burdens in 2(b). However, these burdens are offset by major CO_2 emission savings from avoided N fertilizer (−56 kg CO_2-eq) and avoided P fertilizer (−15 kg CO_2-eq). This benefit due to avoided N fertilizer is not obtained in Scenario 2(a) and 2(c) because in WWS incineration most N is lost. In terms of avoided P fertilizer, 2(b) results in greater CO_2 savings (−15 kg CO_2 eq) due to the higher P content per FU, as opposed to 2(c) (−2.6 kg CO2 eq), where P is possibly lost during incineration.

In Scenario 2(a), the major burdens are from WWS drying (18 kg CO_2-eq), which is due to the use of natural gas. Almost 85% of these emissions are offset by the avoided import of hard coal from Russia (−13 kg CO_2-eq) and the co-incineration process, which feeds electricity and heat into the grid (−2.4 kg CO_2-eq).

In Scenario 2(c), more than 90% of the climate change potential stem from mono-incineration itself, associated with high N_2O emissions. This causes a 10-fold increase in impacts relative to the status quo, i.e. 2(a). Also, ash processing (6.5 kg CO_2-eq) results in a net positive impact (3.9 kg CO_2-eq). This is after taking into account the avoided conventional P fertilizer use (−2.6 kg CO_2-eq) due to processed ash. The GSA on climate change potential for Objective 2 can be seen in Table 2. The Sobol indices showed transportation was the most sensitive parameter on the output uncertainty for climate change in 2(a), whereas in 2(b) and 2(c) it was WWS storage and mono-incineration respectively.

Managing WWS via clinker production and co-incineration has the highest fossil depletion potential. This is mostly attributed to WWS drying, which consumes 6.9 kg of oil-eq. Thus mono-incineration is preferable for WWS end-use due to its low fossil depletion potential.

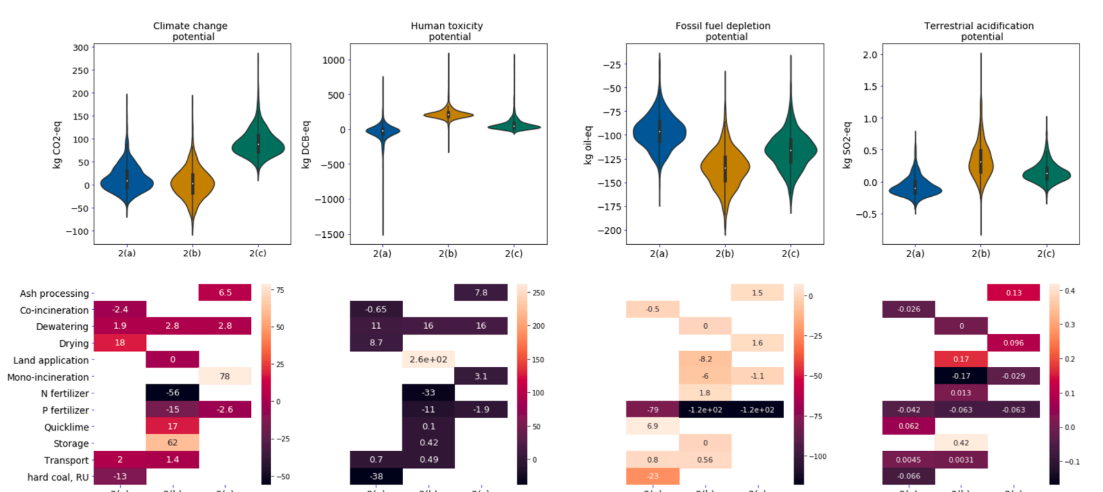
3.2.2. Human toxicity potential

Human toxicity potential for 2(b) is the highest (210 kg 1.4 DCB-eq) followed by 2(c) and 2(a). This is mainly attributed to the heavy metal emissions to the soil following WWS application. In 2(c), the major burdens are from WWS dewatering and ash processing, which contribute to ~95% of the total human toxicity potential. The avoided import of hard coal from Russia contributes to major human toxicity potential savings, ranking co-incineration and clinker production as the best alternative for WWS management.

The GSA for human toxicity potential (Table 2) showed that avoided coal import from Russia was the most sensitive parameter on the output uncertainty in 2(a) ($S_1 = 0.82$), whereas in 2(b) and 2(c), it is WWS storage-land application ($S_1 = 0.93$) and ash processing (1) respectively.

3.2.3. Terrestrial acidification potential

For terrestrial acidification, the violin plot for 2(b), characterized by



	Scenario 2(a)	Scenario 2(b)	Scenario 2(c)
Climate change potential (kg CO_2 -eq)	8.77	2.78	**87.35**
Human toxicity potential (kg 1.4 DCB -eq)	-22	**210**	43
Fossil fuel depletion potential (kg oil -eq)	**-96.40**	-134.33	-115.98
Terrestrial acidification potential (kg SO_2 -eq)	-0.10	**0.30**	0.12

Fig. 4. Objective 2-Overall impacts (violin plots) and process contributions (heat map) for 1 tonne of WWS; 2(a) represents status quo, i.e. WWS for co-incineration and use as an alternate fuel for clinker production, 2(b) represents WWS land application and 2(c) represents future perspective, i.e. complete mono-incineration and ash processing of WWS. White dots in the violin plot represent the median values, boxes represent the interquartile range; violins represent the kernel density plots on either side.

Table 2
First order Sobol indices for climate change and freshwater ecotoxicity potential (Objective 2).

Climate change potential								
Scenario 2(a)			**Scenario 2(b)**			**Scenario 2(c)**		
Activity	***GSA Parameter***	***S_1***	***Activity***	***GSA Parameter***	***S_1***	***Activity***	***GSA Parameter***	***S_1***
Transport	Transport to cement kiln	0.41±0.22	Storage and land application	WWS storage and land application	0.94±0.33	Mono-incineration	Mono-incineration	1.008±0.36
Transport	Transport to co-incineration facility	0.30±0.21	Storage	Quick lime	9e-05±0.001	Ash processing	Ash processing	0.001±0.02
Clinker production	Avoided coal	0.10±0.11						
Co-incineration	Co-incineration	0.04±0.04						
Drying	Drying	0.11±0.12						
Human toxicity potential								
Scenario 2(a)			**Scenario 2(b)**			**Scenario 2(c)**		
Activity	***GSA Parameter***	***S_1***	***Activity***	***GSA Parameter***	***S_1***	***Activity***	***Parameter***	***S_1***
Clinker production	Hard coal, Russia	0.82±0.26	Storage and land application	WWS storage and land application	0.93±0.62	Ash processing	Ash processing	1.03±0.45
Transport	Transport to co-incineration facility	0.07±0.06				Mono-incineration	Mono-incineration	0.056± 0.11
Transport	Transport to cement kiln	0.015±0.06						
Co-incineration	Co-incineration	0.003±0.01						
Drying	Drying	0.02±0.04						

The cut-off for GSA parameters is based on the first-order Sobol indices (S1), which sum up to 1.

a long tail, shows high uncertainty when compared to 2(a) and 2(c). This is caused by input uncertainties due to WWS storage and land application. The major acidification potential for 2(b) arises due to NH_3 emissions from WWS storage as well as avoided N fertilizer production. 2(c) suggests more than a two-fold reduction in impacts relative to 2(b), but the ash processing step results in more than 90% of its emissions. This can be attributed to the upstream impacts from sulphur dioxide production to produce sodium sulphate that is necessary for ash processing. Co-incineration and clinker production has the least impact, ranking it as the best alternative.

4. Discussion

The retrospective analysis of the environmental impacts of struvite recovery in a WWTP indicates that struvite recovery slightly alleviates the WWTP's environmental burdens post-implementation due to decreased polymer usage and energy demand for dewatering. It was also evident from the GSA that polymer use, i.e. polyacrylamide was the most sensitive parameter on the overall environmental impacts. Thus, in future studies, alternative polymers and their potential to change a WWTP's performance could be assessed. Peer-reviewed studies indicate the use of biopolymers (chitosan, cellulose alginates etc.) as possible eco-friendly substitutes, but research regarding their use on an industrial scale is still ongoing (Pandey, 2020). Furthermore, increasing the influent wastewater (current recovery is 5–6% of influent P) could not only offer a synthetic P fertilizer substitute but also allows for reduction of the P load on the centrate, thereby reducing electricity consumption (due to decreased aeration) and use of saccharose in the biological treatment step.

As observed by Pradel et al. (2016) and Lam et al. (2020), LCAs that viewed WWS from a waste perspective favoured APR over conventional PR, mostly because of the zero burden assumption. Studies that used the zero burden assumption did not account for the upstream impacts (for instance, biological treatment, digestion of WWS) leading up to struvite recovery. Thus, we chose to compare our results with studies that considered a product perspective. Most studies that considered WWS from a product perspective (i.e. a product-based FU, for example, provision of 1 kg of plant-available P as fertilizer) either compared struvite recovery versus synthetic fertilizer or other secondary P recovery processes. Linderholm et al. (2012) observed that struvite recovery had a lower climate change potential compared to synthetic P fertilizer. Amann et al. (2018) performed a study similar to ours, and compared struvite recovery (Gifhorn and Stuttgart process) from WWS versus a reference system without nutrient recovery. Their results also observed lower climate change potential compared to the reference. While the impacts on acidification potential were insignificant in ours, the impacts from acidification potential in their study were higher, mostly due to the use of chemicals (sulphuric acid, lye and citric acid). Tonini et al. (2019), who evaluated struvite recovery versus rock phosphate, observed lower impacts for climate change, terrestrial acidification, ecotoxicity and human toxicity potential for struvite recovery. While most of these studies used system expansion and favoured struvite recovery, Pradel and Aissani (2019) had contrasting views. They argued that if a critical material like P can be recovered from WWS, then it has a value and is essentially a co-product resulting from a multi-functional system i.e. a WWTP producing treated water and WWS (Pradel et al., 2016). Since WWS production is indivisible to wastewater treatment, an allocation factor (45% of the burdens to WWS management and 55% to WWT) was proposed to solve for multi-functionality (Pradel et al., 2018). After applying the allocation factor to WWS, the results highlighted that WWS- based P fertiliser like struvite appeared less environmentally friendly than synthetic P fertiliser. This was attributed to limited P yields, low P content and high energy demand for recovering P from WWS (Pradel and Aissani 2019).

Although there is a sound argument for WWS to be considered as a useful product (Pradel et al., 2016) and struvite recovery LCAs to be modelled as such, we still believe an absolute comparison of struvite versus synthetic P fertilizer is unreasonable since the environmental benefits are inevitably skewed in favour of the latter. This is because, in our study, the system boundaries for struvite recovery included burdens from the wastewater treatment, which in itself was roughly 200 times higher than the processes involved in synthetic P fertilizer manufacture (mining and beneficiation). Therefore, if there is a case to be built for circular economy and P recovery through secondary products, then the LCA should be based on measuring the relative difference in potential impacts between alternatives (in our case, before versus after struvite recovery). This is justified because even if struvite is not recovered, the wastewater would still have to be treated.

Furthermore, using an allocation factor, in our opinion, remains contentious for the following reasons: (i) Allocation is not recommended as a first-step approach to solving multi-functionality and its use may not be applicable for consequential LCAs, (ii) There is still no consensus regarding the accounting for the end-of-life for products through allocation, especially in an "open-loop recycling" scenario and (iii) Most importantly, however, the primary function of a WWTP is treating wastewater and it has to be treated irrespective of whether or not struvite is recovered in the process. It does not make sense to argue that only 45% has to be treated because a useful product must be made in the process.

An alternative to allocation factors is to possibly implement the circular footprint formula recommended by the Product Environment Footprint guideline (Zampori and Pant, 2019) to address open-loop recycling. But, Schrijvers et al. (2021) identified that, at the moment, the circular footprint formula does not account for a consequential perspective, but has the potential to do so if it incorporated market effects.

With regards to the WWS handling, the future perspective (Scenario 2c), i.e., mono-incineration of WWS and ash processing showed lower fossil depletion potential relative to the status quo (Scenario 2a), i.e. co-incineration with municipal solid waste and valorisation at cement kilns. However, for climate change potential, the future perspective is worse than the status quo and this can mostly be attributed to the N_2O emissions from mono-incineration. An important assumption here was that the WWTP had already been implemented with struvite recovery and how this interacts with the downstream treatment system depends on the WWS end-use options. Since only 5–6% of the influent P is recovered through struvite, the residual P remains in the WWS. If the WWS is directly land-applied, as is the case for a large fraction of the sludge in many European countries, then the rationale of branding struvite crystallization as a P recovery step is questionable, since all the residual P would be recycled anyway, although in a less plant available form. However, direct land application (Scenario 2b) has a significantly higher human toxicity potential compared to Scenario 2(a) and 2(c), thereby vindicating Flanders' and other countries' decisions on prohibiting land application of WWS. The high human toxicity potential from land application relative to other WWS end-use options that was observed in our study was also similar to Bradford-Hartke et al. (2015); Yoshida et al. (2018), whereas Heimersson et al. (2014) identified that the human toxicity results depended on the impact assessment method used. In the case of Scenario 2(a) struvite recovery seems beneficial, since all the residual P is lost during clinker production. Whereas, for Scenario 2(c), struvite recovery is beneficial since it recovers P and improves the dewaterability and the resultant dry matter content of WWS is conducive for mono-incineration.

As evidenced by other studies (Linderholm et al., 2012; Tonini et al., 2019) and reflected by ours, mono-incineration and ash processing are characterised by high climate change potential. Therefore, policymakers and technology providers could focus on N_2O emission control during WWS mono-incineration. Possible solutions include flue gas treatment technologies (selective catalytic and selective non-catalytic reduction), flue gas recirculation, flame cooling, staged air combustion, and low oxygen dilution combustion (Liang et al., 2021).

This study did not include the possible benefits of struvite recovery on the improved life span of mechanical components due to reduced encrustation and scaling. Furthermore, there is no dedicated ecoinvent process for mono-incineration of WWS and hence the LCI was obtained from Tonini et al., (2019), but without data uncertainty.

5. Conclusion

The current LCA indicated that struvite recovery improved the environmental performance of a WWTP in Flanders. The hotspot analysis, complemented by uncertainty and global sensitivity analyses identified that, albeit marginal, reduced polymer use, improved dewaterability, and avoided imports of synthetic P fertilizer resulted in a net benefit to the system as a consequence of the struvite precipitation. To further enhance the sustainability of WWTPs, plant operators may wish to focus on optimising polymer usage and identifying sustainable substitutes. Additionally, future research could examine the effects of encrustation and scaling on infrastructure components, both prior to and following struvite recovery.

The prospective scenario for WWS in 2026 - in which all WWS in Flanders is mono-incinerated - performs worse in all impact categories except fossil depletion potential than the status quo, in which one-third of WWS is valorised in cement kilns and the remaining two-thirds is co-incinerated. Additionally, the hotspot and global sensitivity analyses revealed that GHG emissions from mono-incineration contributed significantly to the climate change impacts in the prospective scenario. As a result, Flemish policymakers and plant operators should look for ways to improve the flue gas treatment process or other operational aspects of mono-incineration. After mono-incineration, ash processing to recover P (using sodium sulphate) results in net positive impacts and can be considered a sustainable method of closing the P loop.

Appendices

A-Supplementary information
B-Mass flows
C-LCI for Objective 1 and Objective 2
D-Objective 1 results
E- Electricity mix results
F-Objective 2 results
G-GSA results
H-GSA code example

CRediT authorship contribution statement

Rahul Ravi: Writing – review & editing, Conceptualization, Methodology. **Miriam Beyers:** Writing – review & editing, Conceptualization, Methodology. **Sander Bruun:** Writing – review & editing, Supervision. **Erik Meers:** Writing – review & editing, Supervision, Project administration, Funding acquisition.

Declaration of Competing Interest

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

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Supplementary materials

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