



# Life cycle assessment of digestate post-treatment and utilization

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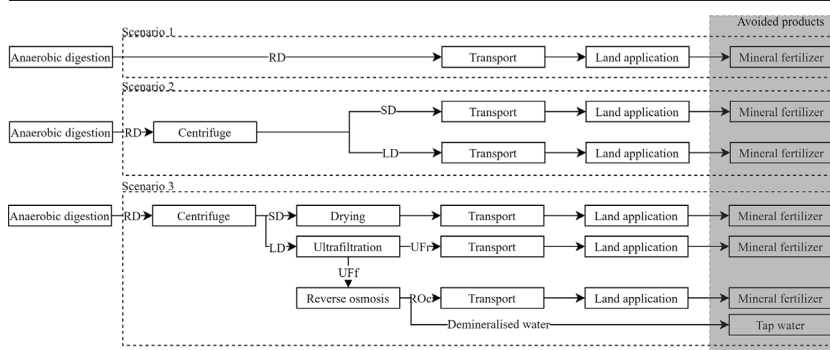
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## HIGHLIGHTS

- Digestate post-treatment can contribute to optimized circular resource management.
- Three digestate post-treatment scenarios are evaluated with life cycle assessment.
- Digestate post-treatment enhances the concentration of citrate soluble P.
- Transport of dried digestate is a preferred solution when local use is not feasible.
- Different quality of digestate products can answer diverse agronomic needs.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Three digestate utilization scenarios for bio-fertilizer production are evaluated with life cycle assessment. The aim is to determine the environmental performance of the digestate post-treatment with the goal to decrease the loss of nitrogen and phosphorus, support circular nutrient management, and increase the substitution of mineral fertilizers. The functional unit (FU) of the study is the utilization of 1 kg dry matter raw digestate, in three scenario designs. Scenario 1 (S1) describes a system where the raw digestate is directly spread on soil. In scenario 2 (S2) the raw digestate is processed by centrifugation with two recovered phases (liquid and solid digestate), which are spread on agricultural soil. In scenario 3 (S3) a more advanced post-treatment system is modelled, where the raw digestate is phase separated with centrifugation followed by drying of the solid digestate and further processing of the liquid digestate with a membrane filtration and a reverse osmosis unit. The studied scenarios show a global warming potential ranging from  $-0.14$  (S3) to  $-0.36$  (S1) kg CO<sub>2</sub> eq per FU. The fossil resource depletion per FU was decreased in scenario 1 ( $-0.053$  kg oil eq) and scenario 2 ( $-0.049$  kg oil eq) but increased in scenario 3 ( $0.002$  kg oil eq). The terrestrial acidification potential ranges from  $0.09$  (S3) to  $0.18$  (S1) kg SO<sub>2</sub> per FU. The digestate post-treatment is a sustainable solution able to tackle the problem of excess nutrients and their management in agricultural areas. It could replace conventional nitrogen removal processes (aerobic biological treatment) by a valorization chain keeping the nutrients in closed loop.

## 1. Introduction

Biomass valorization and bio-product optimization have received a lot of attention at the European and national levels (European Commission, 2012, 2018; The Danish Government, 2013). Considering the growing energy and fertilizer needs (Eurostat, 2021), anaerobic digestion (AD) proved to be an efficient and cost-effective technology for organic waste treatment,

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able to reduce greenhouse gas emissions from waste management activities (Niero et al., 2014; Rajendran and Murthy, 2019) and contribute to resource recovery (Cheong et al., 2020) and circular economy (Wainaina et al., 2020). The process of AD generates biogas, which is used for bio-energy production, and digestate, the residue of the technology, which has fertilizing potential and can be applied on agricultural soil (Tampio et al., 2016).

The digestate quality depends on the origin of the organic waste and the post-treatment technology (Angouria-Tsorochidou and Thomsen, 2021). It contains organic matter, fertilizing elements, and - potentially - micro-pollutants, such as cadmium (Pizzol et al., 2014). To fully utilize the fertilizing potential of the digestate, while avoiding the risk for soil contamination, there is the need to develop innovative nutrient management strategies capable of isolating the nutrients intended for valorization while removing or immobilizing the unwanted compounds. This can be achieved by the smart use of organic feedstock (Wang et al., 2021), through diversification and optimization of the nutrient content in the solid and liquid fertilizer products.

Different technologies are developed to enable the handling and/or recovery of the digestate (Lu and Xu, 2021) according to their water content (Flotats et al., 2009; Vaneckhaute et al., 2017). Solid and low moisture digestate can be dried to facilitate its transport and its final use as a fertilizer, with or without pelletizing, or can be composted to stabilize its organic matter while performing its hygienization. For liquid digestate, a mechanical separation of phases (screening, screw press, or centrifugation) is often the first step, which allows concentrating the suspended solids and some associated elements in the solid fraction. The solid phase obtained after separation is treated as described above, while the liquid phase can undergo various post-treatment methods depending on the objective to either remove or concentrate nutrients to produce a suitable fertilizer. Membrane separation is a technology that through physical barriers for selective nutrient capture can concentrate the desired substances in smaller volumes, achieving reduced costs of storage and transportation, while minimizing the losses of nitrogen (N) and phosphorus (P) compounds (Masse et al., 2007; Thörneby et al., 1999).

Technical and scientific knowledge on the treatment technologies is highly variable. Among them, phase separation has been widely characterized in terms of separation efficiency and energy consumption, especially concerning manure (Béline et al., 2004; Burton and Turner, 2003; Daumer et al., 2003) and more recently about digestate from other types of feedstock (Guilayn et al., 2019). Areas with intensive livestock production often face excess of nutrients, especially N, sourced from animal manure compared to the nutrient and fertilization needs of the available adjacent arable land and grassland soil. Post-treatment technologies can be incorporated to reduce the unsustainable excess of nutrients by increasing mineralization and by partitioning the N content between the liquid and solid fertilizer products. This partitioning lowers the N/P ratio in the solid fertilizer and results in higher fertilizer value due to the more balanced fertilization effect. However, by 2015 only 3% of the European digestate was upgraded to high-value products (Dahlin et al., 2015).

The evaluation of the environmental performance of these post-treatment technologies is important to ensure future optimized circular nutrient management systems. One of the major environmental challenges of digestate utilization is the N losses through volatilization, which contributes to air emissions (especially ammonia (NH<sub>3</sub>) and nitrous oxide (N<sub>2</sub>O)) and result in losses of fertilizing value. Post-treatment and spreading of digestate products can generate different types of greenhouse gases, such as carbon dioxide (CO<sub>2</sub>), N<sub>2</sub>O, and volatilized NH<sub>3</sub> (Bertora et al., 2008) but certain post-treatment practices can also help reducing a share of these emissions compared to conventional practices without pre-treatment (Styles et al., 2018). For this reason, it is necessary to assess the sustainability of these post-treatment technologies regarding their efficiency in recycling of nutrients and associated environmental impacts.

In this study, a life cycle assessment (LCA) was performed to compare the environmental performance of two post-treatment technology designs and a baseline scenario corresponding to direct spreading of raw digestate.

The digestate post-treatment technology scenarios are simulated based on a French AD plant treating manure, which is collected from several farms and is co-digested with agro-food industry residues and sludge. The measurement of emissions related to digestate application on soil after post-treatment were performed with experimental setups. The LCA aims at exploring the environmental impacts of the potential conversion pathways processing digestate into high quality organic fertilizer products according to the European (European Commission, 2019) and national regulations (Danish Ministry of Environment, 2018; French Ministry of Agriculture and Food, 2013).

## 2. Materials and methods

The LCA follows four main steps: i) goal and scope definition, ii) life cycle inventory (LCI), iii) life cycle impact assessment (LCIA) and iv) interpretation, performed in accordance with the international standards ISO 14040-44 (International Organization for Standardization, 2006).

### 2.1. Goal and scope definition

The goal of the study is to compare three designs for the management of digestate from a centralized AD plant. The study takes as point of departure a French case study with the aim to increase the efficiency in the recovery and reuse of nutrients from the raw digestate. The organic fertilizer can be used locally and/or by exporting the excess nutrients to regional and global markets, thereby closing the biological nutrient loop and increasing the European fertilizer self-supply.

The functional unit (FU) is defined as the “utilization of 1 kg dry matter (DM) raw digestate produced by a centralized AD plant”.

### 2.2. Life cycle inventory

The study focuses on the utilization of digestate from a centralized mesophilic digester plant, with a retention time of 60 days and a post-digestion with a retention time of 14 days, treating mainly pig liquid manure from several farms mixed with sewage sludge and waste from agro-food industries. The assessment covers all the steps of the raw digestate management and utilization, excluding its production. The system boundary includes the conversion steps (e.g., separation, drying, and membrane filtration) and spreading of the products on agricultural soil.

Three different utilization pathways of raw digestate are assessed. The system boundary is shown with dashed lines in Fig. 1.

Scenario 1 (S1) describes a system where the raw digestate is directly spread on local agricultural soil surrounding the AD plant, without post-treatment. In scenario 2 (S2), the raw digestate is processed by phase separation (centrifugation). Two phases (liquid and solid digestate) are obtained without any further processing. Both the liquid and the solid digestate are applied on local agricultural soils. Scenario 3 (S3) describes a more advanced post-treatment system. The raw digestate is phase separated by centrifugation followed by drying of the solid digestate and further processing of the liquid digestate with a membrane filtration unit. The first step of filtration for the liquid digestate consists of an ultra-filtration (UF) that produces two liquid fractions, the UF retentate and the UF filtrate. The UF retentate is applied on local agricultural soil. The UF filtrate undergoes a second filtration through a reverse osmosis (RO) unit. The RO concentrate is locally valorized as liquid fertilizer, while the filtrate, i.e., demineralized water, substitutes tap water used in the digestate processing. The chemical characteristics of the raw digestate, the solid and liquid fertilizer in S1, and the dried digestate, UF retentate, RO concentrate and demineralized water in S3, were measured using triplicate samples collected from the centralized plant. The detailed analytical methods and the quality of the digestate products are presented in Appendix A1.

The storage of digestate can contribute to the environmental impact with emissions, such as NH<sub>3</sub> and CH<sub>4</sub> (Styles et al., 2018), but could potentially reduce emissions during and after spreading. The emissions from storage were not directly measured by the experimental setup, but the

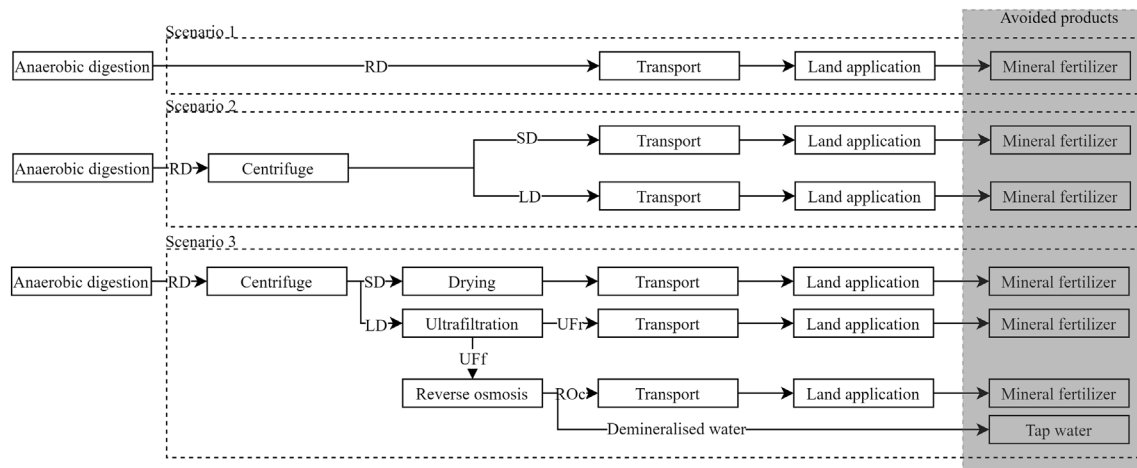


**Fig. 1.** Illustration of the assessed scenarios (RD: raw digestate, SD: solid digestate, LD: liquid digestate, UFR: Ultrafiltration retentate, UFF: ultrafiltration filtrate, ROc: reverse osmosis concentrate).

emissions after spreading were measured on non-stored digestate. The latter were used as a proxy in this study. However, long-term storage of digestate can have significant contributions to the environmental impacts. The digestate storage and its associated environmental impacts are further discussed in Section 3.

The digestate is considered both to be valorized locally and to be exported to other markets, as described below:

- In S1, the raw digestate is transported for 10 km prior to spreading on agricultural soils.
- In S2, both solid and liquid fractions are transported for 10 km prior to spreading on agricultural soils.
- In S3, the transport distance for the dried digestate is 200 km, while the UF retentate and RO concentrate are transported for 10 km prior to spreading on agricultural soils. The demineralized water is stored and used for dilution at the entry of centrifugation, therefore, substituting the consumption of local tap water.

The solid digestate in S2 and the dried solid digestate in S3 are solid fertilizer products, while the raw digestate in S1, the liquid digestate in S2, the UF retentate and the RO concentrate in S3 are liquid fertilizers products. The change in the nutrient content of the digestate products in the different scenarios does not affect the transportation distance of the digestate but is considered as a factor in the spreading activities and the potential area covered. The fuel consumption for transport is expressed in  $\text{tkm}\cdot\text{kg}^{-1}$  DM. A detailed inventory is presented in Appendix A2.

The French fertilization directive (JORF, 2011) limits the amount of N allowed to be spread on agricultural soil to  $170 \text{ kg N}\cdot\text{ha}^{-1}$ . Phosphorus is not regulated by law in France, but local prescriptions are given for equilibrated fertilization. Regulations exist in other countries, such as Germany, Netherlands, and Denmark (Danish Environmental Protection Agency, 2017). In the latter, limits for P spreading range between 30 and  $43 \text{ kg P}\cdot\text{ha}^{-1}$ . A limit of  $30 \text{ kg P}\cdot\text{ha}^{-1}$  has been chosen in this study, which results in an N/P ratio of 5.67. The spreading area covered by the digestate derived fertilizer products was calculated based on the N limit when the N/P ratio was above 5.67 and on the P limit when the N/P ratio was below 5.67 (see Eq. (1)).

$$SA = \frac{FC_{N \text{ or } P}}{N \text{ or } P \text{ limit}} \quad (1)$$

where SA is the spreading area expressed in  $\text{ha}\cdot\text{kg}^{-1}$  DM,  $FC_{N \text{ or } P}$  is the N or P content in the fertilizers in  $\text{kg P}$  or  $\text{N}\cdot\text{kg}^{-1}$  DM, and N or P limit is the N or P fertilizer regulation of, respectively,  $170 \text{ kg N}\cdot\text{ha}^{-1}$  and  $30 \text{ kg P}\cdot\text{ha}^{-1}$ .

The spreading of the digestate is modelled to be performed with an agricultural tractor and trailer. According to Lacour et al. (2012), the fuel

consumption (diesel) for spreading of solid products is  $11.25 \text{ L}\cdot\text{ha}^{-1}$ , while for the liquid products it is  $13 \text{ L}\cdot\text{ha}^{-1}$ . The impact of digestate spreading is calculated according to Eq. (2), in order to transform the fuel consumption into units of the agricultural trailer used (tkm), by considering the spreading area calculated in Eq. (1) and the type of digestate (solid and liquid). The impacts from digestate spreading include the machinery and the emissions associated with its operation. The results for the spreading of digestate are summarized in Appendix A2.

$$DS = \frac{FC \cdot SW_{\text{Diesel}}}{FC_{\text{database}}} \cdot SA \quad (2)$$

where, DS is the impact from digestate spreading with an agricultural tractor, expressed in units of  $\text{tkm}\cdot\text{kg}^{-1}$  DM, FC is the fuel consumption in  $\text{L}\cdot\text{ha}^{-1}$  according to Lacour et al. (2012),  $SW_{\text{Diesel}}$  is the specific weight of diesel,  $0.832 \text{ kg}\cdot\text{L}^{-1}$ , and  $FC_{\text{database}}$  is the fuel consumption provided in the Ecoinvent database,  $0.0436 \text{ kg diesel}\cdot\text{tkm}^{-1}$  (Wernet et al., 2016).

The fertilizer products spread on the soil replace mineral N and P fertilizer according to the fertilizing value of the output products from the post-treatment scenarios. The bioavailable P content of the output products that substitutes mineral fertilizer has been measured as the citrate soluble fraction. The latter substitutes phosphate ( $\text{P}_2\text{O}_5$ ) mineral fertilizer. The avoided processes included the production of mineral phosphate fertilizer (Wernet et al., 2016), and the direct emissions of heavy metals (Gambus and Wiczorek, 2012) and phosphorus leaching to soil. The substitution factor for the N content of the output products was 40% of the total N (Møller et al., 2009) and the mineral fertilizer substituted was ammonium nitrate ( $\text{NH}_4\text{NO}_3$ ). The avoided processes included the production of mineral ammonium nitrate fertilizer (Wernet et al., 2016),  $\text{N}_2\text{O}$  emissions to air,  $\text{NO}_3^-$  and  $\text{NH}_4^+$  emissions to water adopted from Yoshida et al. (2016), and emissions of heavy metals (Gambus and Wiczorek, 2012) and N leaching to soil. The effect of the N and P substitution methodology is further assessed with a sensitivity analysis.

Laboratory measurements were performed to define the mineral and organic content of N in the output products. The  $\text{NH}_3$  emissions after spreading for each digestate product were measured through laboratory experiments with wind tunnels. The  $\text{NH}_3$  at the entry and exit points of the tunnel was trapped in acid solution and measured, and the  $\text{NH}_3$  emitted was calculated as the ratio of  $\text{NH}_3$  trapped per unit of digestate product applied for the experiment. The  $\text{N}_2\text{O}$  emissions were also measured using a laboratory set up for three consecutive months after the digestate application. The modelling of the heavy metal emissions follows a conservative approach with all the content to be considered as emissions to soil. Further information is provided in Appendix A3.

Details about the digestate-derived products and the quality of the digestate in each scenario are provided in Appendix A1. Data on energy consumption and transportation distances for the three scenarios are provided in Appendix A2. Data on the nutrient flows and associated air and soil emissions from the treatment of the digestate and the methods for the laboratory emission measurements are provided in Appendix A3, along with the methods and experimental setup used for the laboratory measurements. Data on N/P ratio, the agricultural area covered (spreading area) and the amount of mineral fertilizer products substituted are provided in Appendix A4. A detailed list of the French standards for soil improvers and fertilizers are given in Appendix A5. The mass flows of the post-treatment scenarios are given in Appendix A6, and last, selected product quality measures and Danish and European regulatory standards are given in Appendix A7.

### 2.3. Life cycle impact assessment

The LCIA was performed using the ReCiPe method (Huijbregts et al., 2017) at midpoint level. The impact assessment was performed in SimaPro software (7.3.3 version) (PRé Sustainability, 2021). The main emissions from the treatment are CO<sub>2</sub>, N<sub>2</sub>O, CH<sub>4</sub>, and NH<sub>3</sub> emissions to air after the spreading of the digestate, contributing to global warming and terrestrial acidification. As N leaching to soil was considered the mineral N content, thus, contributing to marine eutrophication potential through the characterization factors applied by the impact assessment methodology (1 kg N to agricultural soil = 0.13 kg N eq (Wernet et al., 2016)). A sensitivity analysis was performed to evaluate the effect on the environmental performance when the total content of N was considered as emission to soil. The impact assessment methodology implicitly models N leaching to soil as a potent emission to marine eutrophication and P leaching to soil as a potent emission to freshwater eutrophication. Emissions to air from the energy consumption during post-treatment and transport are associated with the impact categories climate change, fossil resource depletion, and acidification. Moreover, like mineral fertilizer products contain cadmium as an impurity, digestate can contain heavy metals and organic micro-pollutants transferred to the soil through spreading. The substitution of mineral fertilizer through the production and utilization of organic fertilizers can reduce mineral resource scarcity. Thus, eight impact categories quantifying the resource use, human health, and environmental impacts were selected to assess the performance of the post-treatment designs. The selected impact categories for this study are:

- Climate change; in kg CO<sub>2</sub> equivalents, [kg CO<sub>2</sub> eq]
- Fossil resource depletion; in kg oil equivalents, [kg oil eq]
- Mineral resource scarcity, in kg copper equivalents [kg Cu eq]
- Human toxicity, non-carcinogenic; in 1,4-dichlorobenzene equivalents, [kg 1,4-DCB eq]
- Terrestrial ecotoxicity; in kg 1,4-dichlorobenzene equivalents, [kg 1,4-DCB eq]
- Freshwater eutrophication; in kg P equivalents, [kg P eq]
- Marine eutrophication; in kg N equivalents, [kg N eq]
- Terrestrial Acidification; in kg sulphur dioxide equivalents, [kg SO<sub>2</sub> eq]

### 2.4. Sensitivity analysis

A sensitivity analysis was performed to evaluate the effect of the substitution methodology followed in the study. In the scenarios modelled, only a fraction of P, the citrate soluble, and N, 40% of the total N, substitute mineral fertilizers. This can have a strong effect on the calculated avoided impacts. In the sensitivity analysis, N and P are modelled to substitute mineral fertilizers according to their total content.

As N emission to soil was considered only the mineral N, in the main scenarios. This decision was taken to evaluate the N emissions on the same basis as the N substitution rates. In the sensitivity analysis, the total amount of N (mineral + organic) was considered as emission to soil, accounting for the total marine eutrophication potential caused by the

spreading of the different types of digestate. A similar approach was followed for P, and the freshwater eutrophication potential in the sensitivity analysis.

A fourth scenario (S4) was modelled to evaluate the influence of transport versus stationary energy on the environmental performance. A comparison between S3, in which the solid fraction is dried, and S4, in which the solid fraction is transported 200 km from the AD-plant (versus 10 km in S2) was performed.

## 3. Results and discussion

The results from performing the LCIA on the three scenarios, using the methodology ReCiPe at midpoint (H), are shown in Fig. 2. The bars visualizing positive values represent environmental burdens while negative values represent avoided impacts and potential environmental benefits. The impacts presented in Fig. 2 result from 18 individual processes and are aggregated in three emission categories, i.e., direct, indirect, and avoided emissions, which are presented in Table 1. The direct emissions are defined as process emissions to soil and air from the biomass management. These are the emissions from the application of the raw (S1), solid (S2), liquid (S2), and dried (S3) digestate, and the UF retentate (S3), RO concentrate (S3), and the demineralized water (S3). An additional direct emission is the emission during post-treatment, which refers to the loss of resources during the treatment processes, only measured for S3. The indirect emissions include emission associated to all inputs necessary to operate the system. These are emissions from 8 processes, i.e., electricity for centrifugation, for drying, and for ultrafiltration, the production of sulfuric acid, the production of polymer, dilution water, transport, and spreading activities. The avoided emissions are related to the production and use of the substituted mineral fertilizer and the avoided consumption of tap water.

The net results show that S1 performs better at the impact categories of human non-carcinogenic toxicity, terrestrial ecotoxicity, and fossil resource scarcity, S2 performs better at the impact categories of global warming, freshwater eutrophication, and mineral resource scarcity, and S3 performs better at the impact categories of terrestrial acidification and marine eutrophication.

Direct emissions, or process emissions, reflect the environmental impacts contained in the sphere of influence of the system while indirect emissions reflect the energy and material intensity of the system originating from the background system. The direct emissions are responsible for the greater part of total impacts for all impact categories, except for the impact categories of fossil resource depletion, mineral resource scarcity, and terrestrial ecotoxicity. The fossil and mineral resource depletion impact categories have zero contribution to the direct emissions since all the anthropogenic fossil and mineral products are included in the indirect emissions. Mitigation of environmental impacts is due to the avoided emissions from mining activities performed by the mineral fertilizer industry, since there is a decreased market demand corresponding to the amount of mineral fertilizers substituted by the organic fertilizers.

### 3.1. Global warming

The results for global warming show that S1 (−0.36 kg CO<sub>2</sub> eq) and S2 (−0.47 kg CO<sub>2</sub> eq) have greater avoided emissions compared to S3 (−0.14 kg CO<sub>2</sub> eq). This is mainly due to the increased electricity consumption in S3. The emissions of digestate spreading in S2 and S3 are comparable, calculated to 0.24 kg CO<sub>2</sub> eq. However, indirect emissions from the post-treatment processes, e.g., electricity, in S2 and S3 counterbalance the higher direct emissions from spreading in S1 (0.41 kg CO<sub>2</sub> eq). This results in lower environmental performance of S3 compared to S2. It was only possible to measure emissions during the drying phase of the solid fraction of the digestate in S3. The emissions from other post-treatment steps in S2 and S3 have not been measured directly. Uncertainties on mass flows between entry and exit of the phase separation process did not allow for a conclusion on the emission flows during post-treatment. For this reason, the emissions of post-treatment in S2 and other post-treatment steps, except

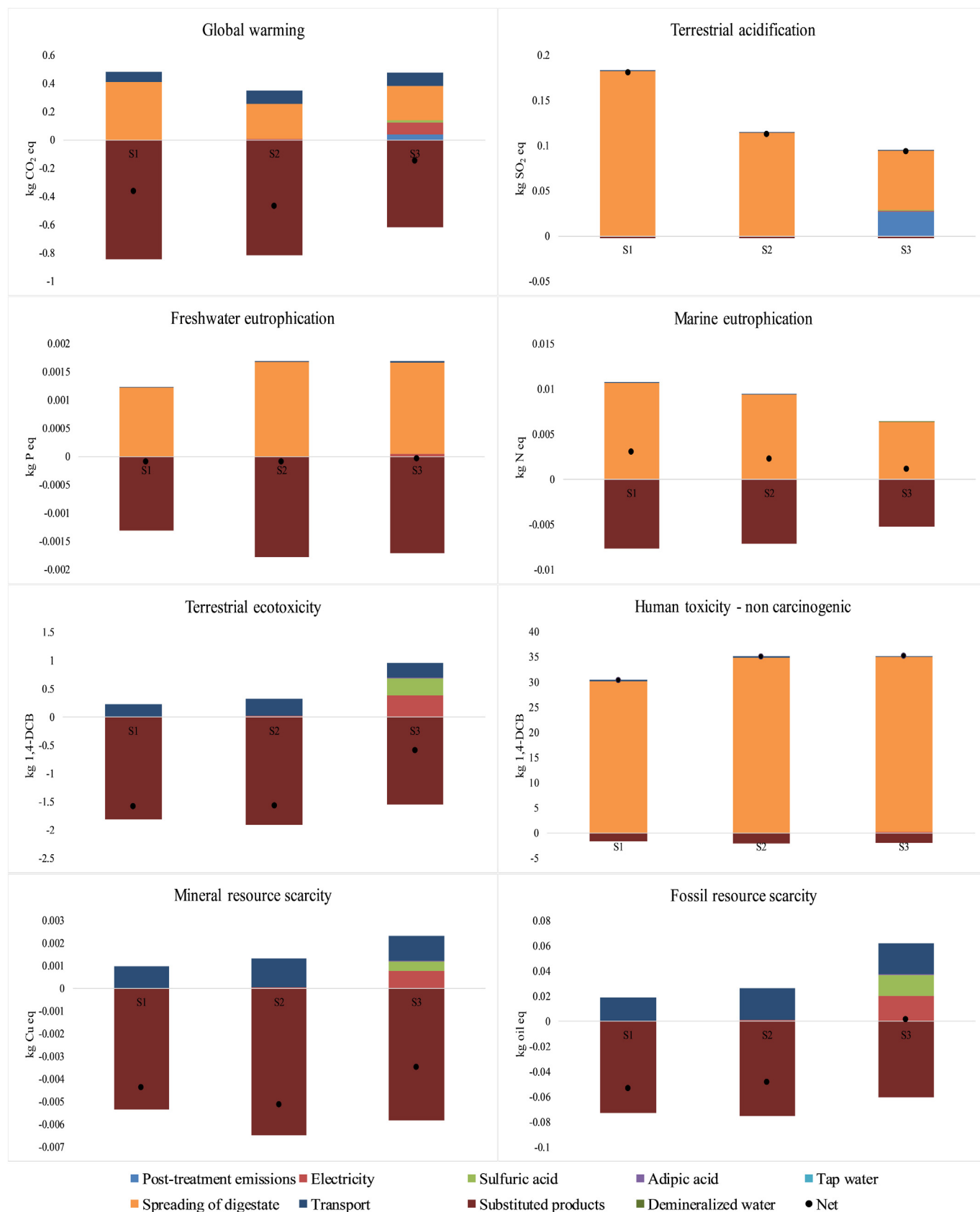


Fig. 2. Environmental impact of the three scenarios.

drying in S3, have been assumed negligible. The fuel consumption required for transport and spreading of the digestate has also a negligible impact on global warming compared to the other processes.

Regarding the substitution of mineral fertilizer according to the citrate soluble P content in the digestate, the post-treatment technologies applied in S2 and S3 enhances the concentration of citrate soluble P. This results

**Table 1**

Emissions related to each scenario (IC: impact category, S: scenario, GW: global warming, FRS: fossil resource scarcity, MRS: mineral resource scarcity, HT: human toxicity, TE: terrestrial ecotoxicity, FE: freshwater eutrophication, TA: terrestrial acidification, ME: marine eutrophication).

| IC       |    | GW                    | TA                    | FE                    | ME                    | HT         | TE                   | FRS                   | MRS                   |
|----------|----|-----------------------|-----------------------|-----------------------|-----------------------|------------|----------------------|-----------------------|-----------------------|
|          | S  | kg CO <sub>2</sub> eq | kg SO <sub>2</sub> eq | kg P eq               | kg N eq               | kg 1,4-DCB | kg 1,4-DCB           | kg oil eq             | kg Cu eq              |
| Direct   | S1 | 0.41                  | 0.18                  | $1.21 \cdot 10^{-3}$  | $1.07 \cdot 10^{-2}$  | 30.26      | $8.10 \cdot 10^{-7}$ | –                     | –                     |
|          | S2 | 0.25                  | 0.11                  | $1.67 \cdot 10^{-3}$  | $9.35 \cdot 10^{-3}$  | 34.79      | $1.43 \cdot 10^{-6}$ | –                     | –                     |
|          | S3 | 0.28                  | $9.24 \cdot 10^{-2}$  | $1.61 \cdot 10^{-3}$  | $6.32 \cdot 10^{-3}$  | 34.73      | $6.83 \cdot 10^{-8}$ | –                     | –                     |
| Indirect | S1 | 0.07                  | $3.13 \cdot 10^{-4}$  | $1.55 \cdot 10^{-5}$  | $1.44 \cdot 10^{-6}$  | 0.20       | 0.23                 | $1.94 \cdot 10^{-2}$  | $9.70 \cdot 10^{-4}$  |
|          | S2 | 0.10                  | $4.31 \cdot 10^{-4}$  | $2.32 \cdot 10^{-5}$  | $2.16 \cdot 10^{-6}$  | 0.28       | 0.33                 | $2.64 \cdot 10^{-2}$  | $1.34 \cdot 10^{-3}$  |
|          | S3 | 0.19                  | $2.30 \cdot 10^{-3}$  | $6.98 \cdot 10^{-5}$  | $6.70 \cdot 10^{-6}$  | 0.48       | 0.97                 | $6.19 \cdot 10^{-2}$  | $2.34 \cdot 10^{-3}$  |
| Avoided  | S1 | –0.84                 | $-2.19 \cdot 10^{-3}$ | $-1.31 \cdot 10^{-3}$ | $-7.60 \cdot 10^{-3}$ | –1.64      | –1.81                | $-7.24 \cdot 10^{-2}$ | $-5.34 \cdot 10^{-3}$ |
|          | S2 | –0.82                 | $-2.31 \cdot 10^{-3}$ | $-1.78 \cdot 10^{-3}$ | $-7.10 \cdot 10^{-3}$ | –2.04      | –1.90                | $-7.49 \cdot 10^{-2}$ | $-6.47 \cdot 10^{-3}$ |
|          | S3 | –0.62                 | $-1.89 \cdot 10^{-3}$ | $-1.72 \cdot 10^{-3}$ | $-5.23 \cdot 10^{-3}$ | –1.86      | –1.55                | $-6.04 \cdot 10^{-2}$ | $-5.82 \cdot 10^{-3}$ |
| Total    | S1 | –0.36                 | 0.18                  | $-7.92 \cdot 10^{-5}$ | $3.07 \cdot 10^{-3}$  | 28.82      | –1.58                | $-5.30 \cdot 10^{-2}$ | $-4.37 \cdot 10^{-3}$ |
|          | S2 | –0.47                 | 0.11                  | $-9.04 \cdot 10^{-5}$ | $2.26 \cdot 10^{-3}$  | 33.03      | –1.57                | $-4.85 \cdot 10^{-2}$ | $-5.13 \cdot 10^{-3}$ |
|          | S3 | –0.14                 | $9.28 \cdot 10^{-2}$  | $-3.17 \cdot 10^{-5}$ | $1.09 \cdot 10^{-3}$  | 33.34      | –0.58                | $1.57 \cdot 10^{-2}$  | $-3.48 \cdot 10^{-3}$ |

in an increased amount of mineral phosphate fertilizer that can be substituted in S2 (–0.09 kg CO<sub>2</sub> eq) and S3 (–0.08 kg CO<sub>2</sub> eq) compared to S1 (–0.06 kg CO<sub>2</sub> eq). Regarding the substitution of mineral N fertilizer, the direct emissions of NH<sub>3</sub> during post treatment in S3, result in less ammonium nitrate substituted in S3 (–0.54 CO<sub>2</sub> eq), compared to S1 (–0.78 kg CO<sub>2</sub> eq) and S2 (–0.73 kg CO<sub>2</sub> eq). The resource losses through the NH<sub>3</sub> emissions are the determining factor for the lower environmental performance of S3, compared to the other two scenarios. Substitution of other types of mineral fertilizer could potentially result in other environmental footprints.

The experimental setup for the digestate spreading provides insights on the emissions of different digestate products. The post-treatment scenarios S2 and S3 perform better than S1 due to reduced loss of resources in terms of air emission after spreading. More precisely, the digestate spreading activities in S1, S2, and S3, emit 1.37, 0.84, and 0.81 g N<sub>2</sub>O per FU, allowing for higher nutrient cycling in the digestate products. A similar pattern between the scenarios is observed for NH<sub>3</sub> emissions during spreading. Rehl and Müller (2011) calculated a global warming potential of 0.06 kg CO<sub>2</sub> eq for solar drying and 0.1 kg CO<sub>2</sub> eq for belt drying. Styles et al. (2018) calculated the environmental performance of biofertilizer production and the results show that from the best to worse practices, the global warming potential ranges from 0.22 to 0.41 kg CO<sub>2</sub> eq. Vázquez-Rowe et al. (2015) calculated a global warming potential of 0.06–0.14 kg CO<sub>2</sub> eq per FU. Previous studies present a higher global warming potential mostly related to emissions during the storage phase of the digestate. In the current study and the experimental set-up, the different types of digestate were applied directly on soil, thus, not accounting for direct emissions during storage. Emissions during storage are CH<sub>4</sub> emissions from the storage unit and spreading due to anaerobe conditions and N<sub>2</sub>O emissions from the storage and digestate application (Rehl and Müller, 2011). Higher storage emissions are measured in the conventional management of digestate, when it is stored for a long period of time, while they are radically reduced when the digestate is dried and phase separated. Emissions from digestate storage, in this study, could compromise the calculated avoided emissions, especially for S1, where the management of raw digestate is considered. The storage emissions during conventional digestate management, according to Rehl and Müller (2011), are approximately 0.05 kg CO<sub>2</sub> eq per FU, which would reduce the avoided emissions in S1 to –0.31 kg CO<sub>2</sub> eq.

It worth noting that the direct emissions in S3 are lower than S1. The higher indirect emissions in S3, which define the final performance of the scenario, are generated from the production and consumption of chemicals and energy that are based on fossil resources. In a future scenario where fossil fuels are replaced by renewable energy, S3 could potentially perform better. A similar trend is noticed for the impact categories of terrestrial acidification, marine eutrophication, and terrestrial ecotoxicity.

Emission reduction may be obtained through optimization of the method applied for spreading, as it influences the amount of NH<sub>3</sub> emissions to air significantly. As assessed by Vázquez-Rowe et al. (2015), injection of digestate into the soil can lead to lower emissions to air, while lower odor

emissions can also be achieved with the same method (Orzi et al., 2018; Zilio et al., 2021). Such spreading method would require increased fuel consumption and, thus, needs further assessment.

In terms of treatment, a comparison of a scenario where the drying step (included in S3) is replaced by composting would also be interesting (Grigatti et al., 2020). Composting would increase the NH<sub>3</sub> losses through air emissions but would avoid the energy consumption for the drying process.

Increasing the substitution of mineral fertilizer products with the use of organic fertilizer has a great potential for increased environmental impact mitigation but it is crucial to monitor increased recycling through avoided air emission losses. The LCIA only considered the digestate as providing fertilizing value, while it also brings back organic matter to soil, providing carbon sequestration services. The environmental benefit of carbon stock increase in the soil was not included in the present study. An additional mitigation of global warming may be calculated assuming that 10% of the carbon remains in the soil at a time horizon of 100 year (Seghetta et al., 2016a), resulting in a reduction in the net CO<sub>2</sub> emission. This reduction corresponds to 0.157, 0.136, and 0.145 kg CO<sub>2</sub> eq for S1, S2 and S3, respectively. Such ecosystem services should be addressed in the future methodological development of LCIA and policy, as also suggested by Seghetta et al. (2016b).

### 3.2. Fossil and mineral resource scarcity

The substitution of mineral fertilizer has a positive effect on the reduction of fossil-based material consumption. For S1 (–0.053 kg oil eq) and S2 (–0.049 kg oil eq), this results in a net negative fossil resource scarcity value, which means that the fossil resource depletion is reduced. On the other hand, S3 (0.002 kg oil eq) represents a more energy intensive post-treatment technology, and this results in a net positive value and depletion of fossil resources. The latter is the result of the fossil-based materials consumed for the post-treatment process, such as electricity and sulfuric acid production. For the impact category of mineral resource scarcity, S1 (– $4.4 \cdot 10^{-3}$  kg Cu eq) and S2 (– $5.1 \cdot 10^{-3}$  kg Cu eq), perform better than S3 (– $3.5 \cdot 10^{-3}$  kg Cu eq) due to the higher electricity consumption, the use of sulfuric acid, and the lower substitution of ammonium nitrate caused by the N losses in the post-treatment activities. Similar numeric results are calculated for aggregated abiotic resource depletion (fossil and mineral) from Styles et al. (2018) that range from –0.001 to 0.003 kg Sb eq. Vázquez-Rowe et al. (2015) calculated a fossil resource depletion equal to 0.01–0.02 kg oil eq.

Only fossil depletion impacts should be temporarily allowed to become worse upon advancement of the post-treatment processes because of an increased energy need within the post-treatment. However, this increase is dependent on the energy mix of the country where the post-treatment occurs. The impacts from fossil energy consumption will be reduced upon a gradual transformation into renewable energy supply. The current study did not consider the use of the energy from the AD plant as an input for

the post-treatment steps, which would have improved the environmental results, as shown by Rehl and Müller (2011) who compared different sources of energy inputs.

### 3.3. Human toxicity and terrestrial ecotoxicity

The impact categories human toxicity and terrestrial ecotoxicity show a similar pattern for all three scenarios. There is a trend for less avoided impacts in S3 ( $-0.58$  kg 1,4-DCB eq) that is associated with the higher electricity demand ( $0.38$  kg 1,4-DCB eq) and the production of sulfuric acid ( $0.31$  kg 1,4-DCB) in the terrestrial ecotoxicity category. For the human non-carcinogenic toxicity, the environmental impacts derive almost exclusively (98–99%) from the emissions of the digestate spreading. The environmental performance for the human non-carcinogenic toxicity for S1, S2, and S3 is 28.8, 33, and 33.3 kg 1,4-DCB eq while for the impact category of terrestrial ecotoxicity, S1, S2, and S3, have an impact of  $-1.58$ ,  $-1.57$ ,  $-0.58$  kg 1,4-DCB eq, respectively. S3 performs the lowest in both impact categories. Vázquez-Rowe et al. (2015) found a lower environmental impact of  $2.46 \cdot 10^{-6}$  kg 1,4-DCB eq for terrestrial ecotoxicity and  $0.003$  kg 1,4-DCB eq for human toxicity.

The partitioning of the hazardous substances prefers the solid fraction of the digestate (Appendix A4). An in-depth analysis of the processes contributing to the negative environmental impact on human toxicity showed that zinc and lead emissions to soil are the most relevant, while for terrestrial ecotoxicity, the higher contributors to the environmental impact are silver and mercury to air from the electricity consumption.

For the impact category of human toxicity, the direct emissions are responsible for the negative environmental impact, while for the terrestrial ecotoxicity, the direct emissions have a negligible contribution on the total impact. Thus, S1 performs the best for the human toxicity due to less direct emissions that are reflected, however, in worse performance in the global warming impact category. Similarly, S1 and S2 perform better in the terrestrial ecotoxicity category due to the higher substituted mineral fertilizer amount and the lower energy and material consumption compared to S3. Environmental and human health impacts should not increase upon closing the biological nutrient cycle.

According to the present regulatory standards (Appendix A5 and A7), the organic fertilizer presents a good product quality as its content in heavy metals is lower than the thresholds for organic soil improvers in France, except for zinc and cadmium in the liquid digestate in the S2. Excluding the liquid digestate, the concentration of cadmium in the post-treated digestate in this study is comparable to the Danish untreated manure and lower or comparable to the content in Danish sludge.

The concentration level of heavy metals in the organic fertilizer is below the limits adopted in the European fertilizer Regulation (European Commission, 2019) as well as below the quality criteria of the Danish Sludge Directive (Danish Ministry of Environment, 2018). Still, according to a draft proposal on the European regulation of cadmium in fertilizers (Chemicals Unit of DG Enterprise, 2003) a concentration level of  $136$  mg Cd·kg<sup>-1</sup> P ( $60$  mg Cd·kg<sup>-1</sup> P<sub>2</sub>O<sub>5</sub>) will lead to cadmium accumulation in agricultural soils. A concentration level of  $45$  mg Cd·kg<sup>-1</sup> P is considered an acceptable value, even though such product standard would still lead to accumulation in soil (Dam Jensen et al., 2015; Pizzol et al., 2014).

As presented in the study of Pizzol et al. (2014), it is possible to monetize human health cost of cadmium emission to soil as a consequence of the presence of micro-pollutants in the individual fertilizer products. Both the efficiency in recycling of the nutrient content in digestate, but also the fertilizing value, and the concentration of micro-pollutants influence the environmental performance of the technologies. This implies the importance of developing science-based policy regulations for the micro-pollutant content per mass unit of plant available P. Like N, the application of organic P fertilizer on agricultural soil needs to be included in the national regulations. This will allow the moderation of the mineral fertilizers' use, especially when it can be adjusted to the degree of self-supply and demand.

### 3.4. Freshwater and marine eutrophication

The net freshwater eutrophication potential is calculated close to zero due to the balanced impacts of the substitution of mineral by organic fertilizers and the eutrophication effect caused by the spreading of digestate. The substitution includes the production and use of mineral fertilizer, which explains the capacity to counter-balance the negative impact of the emissions after spreading. Negative values are obtained for all scenarios, with the total impact score for S1, S2, and S3 being  $-7.9 \cdot 10^{-5}$ ,  $-9.0 \cdot 10^{-5}$ , and  $-3.1 \cdot 10^{-5}$  kg P eq, respectively. Direct emissions from the digestate application and avoided mineral fertilizer are lower in S1, due to the higher availability of citrate soluble P in the post-treated digestate. The highest contributing process is the spreading of digestate on agricultural land, which for S1, S2, and S3 was calculated to  $1.2 \cdot 10^{-3}$ ,  $1.7 \cdot 10^{-3}$ , and  $1.6 \cdot 10^{-3}$  kg P eq, respectively.

The net marine eutrophication potential for S1, S2, and S3 is  $3.1 \cdot 10^{-3}$ ,  $2.7 \cdot 10^{-3}$ , and  $1.1 \cdot 10^{-3}$  kg N eq per FU, with a significant share of the contribution (<99%) to be from the digestate spreading activities. The eutrophication effect in the marine environment is due to the runoff of N and N compounds to soil and their transport to the marine system, with N being the limiting nutrient. The contribution of other processes to the marine eutrophication is negligible.

### 3.5. Terrestrial acidification

For terrestrial acidification, a better environmental performance is observed for S2 ( $0.11$  kg SO<sub>2</sub> eq) and S3 ( $0.09$  kg SO<sub>2</sub> eq) compared to S1 ( $0.18$  kg SO<sub>2</sub> eq). Among the substances listed in the LCI, only the NH<sub>3</sub> emission has a characterization factor that describes its effect on terrestrial acidification. Direct emissions are the most relevant contributors to the total impact in this category. Lower NH<sub>3</sub> emissions during spreading of the digestate in S2 and S3 explain the better performance of the post-treatment scenarios in this impact category. S3 also considers NH<sub>3</sub> emissions during the drying post-treatment. High NH<sub>3</sub> emissions are reported in literature during the digestate storage, which could contribute to the acidification potential by  $0.007$  kg SO<sub>2</sub> eq (Rehl and Müller, 2011), further increasing the environmental burden in this impact category. In a more advanced scenario, NH<sub>3</sub> volatilized during drying would be trapped in acid and valorized which would still decrease the terrestrial acidification potential of S3. Styles et al. (2018) resulted in a similar terrestrial acidification potential of  $0.008$  kg SO<sub>2</sub> eq.

### 3.6. Sensitivity analysis

A sensitivity analysis was performed to evaluate the effect of the fertilizer substitution methodology and substitution factors used in the study. In the main scenarios only the citrate soluble P and 40% of the total N content were considered for substitution. In the sensitivity analysis the total content of N and P were considered to substitute ammonium nitrate and phosphate fertilizers, respectively. The results are presented in Fig. 3.

The order of the three scenarios in respect to their environmental performance is the same for all the impact categories, except freshwater eutrophication and mineral resource scarcity. The higher amount of substituted mineral fertilizer and its avoided direct emissions results in numerically higher avoided impacts for all impact categories. In the mineral resource scarcity impact category, for the sensitivity analysis, S1 ( $-0.009$  kg Cu eq) performs better than S2 ( $-0.0088$  kg Cu eq) and S3 ( $-0.006$  kg Cu eq). Similar for the freshwater eutrophication S1, S2, and S3 has a total impact of  $-1.63 \cdot 10^{-4}$ ,  $-1.59 \cdot 10^{-4}$ , and  $-8.1 \cdot 10^{-5}$  kg P eq, respectively.

The quantified environmental impacts associated with the energy and material consumption remain numerically the same in the main scenarios and the -respective to each scenario- sensitivity analysis, since no changes in the amount of energy and materials were considered. The impacts associated with the process emissions and the substituted products influence the results of the sensitivity analysis. In the freshwater eutrophication, the spreading of digestate and the substituted products have different numerical

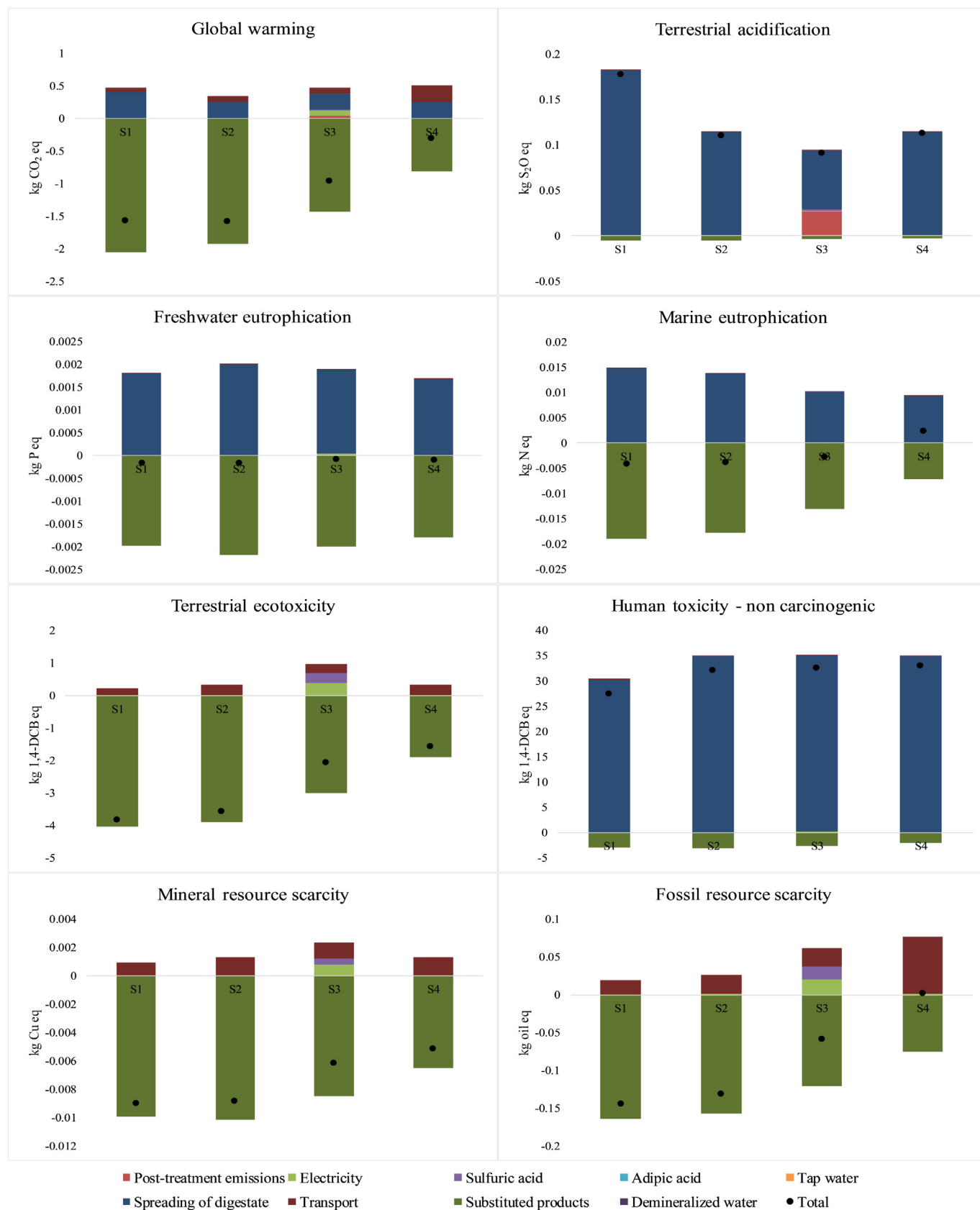


Fig. 3. Environmental impact of the four scenarios in the sensitivity analysis.

scores, resulting in the change of order for the scenarios' environmental performance. The digestate post-treatment increases the concentration of citrate soluble P. Thus, the fraction not substituted in the main scenarios is

higher for S1 than S2 and S3. For S1, in the freshwater eutrophication impact category, the substituted products in main scenario and the sensitivity analysis is  $-0.0013$  and  $-0.002$  kg P eq, while for S2 it is  $-0.0018$

**Table 2**

Total value for the impact categories in scenarios S3 and S4.

| Impact category           | Unit                  | S3                     | S4                     |
|---------------------------|-----------------------|------------------------|------------------------|
| Global warming            | kg CO <sub>2</sub> eq | −0.14                  | −0.3                   |
| Terrestrial acidification | kg SO <sub>2</sub> eq | 0.09                   | 0.11                   |
| Freshwater eutrophication | kg P eq               | −3.17·10 <sup>−5</sup> | −9.03·10 <sup>−5</sup> |
| Marine eutrophication     | kg N eq               | 1.09·10 <sup>−3</sup>  | 2.26·10 <sup>−3</sup>  |
| Human toxicity            | kg 1,4-DB eq          | 35.2                   | 33.03                  |
| Terrestrial ecotoxicity   | kg 1,4-DB eq          | −0.58                  | −1.55                  |
| Mineral resource scarcity | kg Cu eq              | −3.48·10 <sup>−3</sup> | −5.13·10 <sup>−3</sup> |
| Fossil resource scarcity  | kg oil eq             | 1.57·10 <sup>−3</sup>  | 1.94·10 <sup>−3</sup>  |

and −0.0022 kg P eq, respectively. An even smaller difference than S2 is noticed for S3. The direct impacts from the digestate spreading also increase in the sensitivity analysis; more for S1 (0.0018 kg P eq), followed by S2 (0.002 kg P eq) and S3 (0.0018 kg P eq). However, this increase does not compensate for the avoided impacts from fertilizer substitution.

The changes in the impact category of mineral resource scarcity follows the same trend, however, it is only reflected in the avoided environmental impact of the substituted products. The larger substituted fraction in S1 in the sensitivity analysis result in S1 having the greatest avoided impact potential (−0.009 kg Cu eq), followed by S2 (−0.0088 kg Cu eq) and S3 (−0.0061 kg Cu eq).

A fourth scenario was also assessed, reflecting the difference between stationary energy use for digestate drying, modelled in S3, and longer transportation distances in S4. The results of the sensitivity analysis are provided in Table 2.

Table 2 shows that for global warming, freshwater eutrophication, human toxicity, terrestrial ecotoxicity, and mineral resource scarcity S4 performs better than S3. This implies that the emissions from the drying phase are more than the increased emission associated to the transportation of wet digestate. Thus, in the case where spreading is not feasible locally (e.g. excess nutrients zone), a drying post-treatment demonstrates to be a relevant option for solid digestate valorization, however, there are environmental impacts associated with specific impact categories that need to be considered. According to the regulations, the transport distance of the liquid concentrated products as UF retentate and RO concentrate is restricted to less than 20 km and has, therefore, been omitted from the sensitivity analysis.

### 3.7. Input data and model uncertainties

Measurements of the post-treatment process-based emission are performed at lab-scale and may differ from full-scale measurements. Data and model uncertainty should be considered when interpreting the LCIA results.

The gaseous emissions from spreading of the fertilizer products are measured at laboratory scale. Such emissions during spreading are the main contributor to global warming and terrestrial acidification and should be verified at real scale to lower the uncertainty in LCIA results. Furthermore, as for the carbon sequestration in soil, there seem to be a need for regulatory instruments acknowledging the implementation of emission reduction technologies within the agricultural and circular bioeconomy sector in general.

In this case study, some steps of the digestate post-treatment were omitted, such as the storage step before spreading. These storage steps could lead to air emissions, such as NH<sub>3</sub> and N<sub>2</sub>O. However, it could potentially decrease some emissions after spreading, which were measured for this study on non-stored products. Long digestate storage periods can increase the total emissions, especially when raw digestate is stored, without being post-treated (Rehl and Müller, 2011).

On a local scale, the spreading of the digestate may become restricted due to regulations or simply because the local supply exceeds the demand from local farmers (and/or the nutrient demand of the local cropland) (Damgaard Poulsen et al., 2019). One of the interests in performing advanced post-treatment of digestate is the ability to export organic soil improvers or fertilizers to markets outside the local areas. Further post-treatment of the dried solid digestate (S3), such as pelletizing, could produce other final products.

In a future low-carbon/renewable energy scenario, the total CO<sub>2</sub> footprint would equal the process CO<sub>2</sub> footprint implying no energy-related emissions from the post-treatment. Considering pelletizing of dried digestate, it would have increased the energy consumption through the post-treatment scenario, but it might have decreased emissions after spreading (Alemi et al., 2010), as for composted products (Laville et al., 2014). It must be yet noted that S3 is based on a drying up to 80% DM. Pelletizing could be performed on less dried digestate with lower energy consumption.

Considering the diverse situations that may exist, it would be interesting to assess scenarios where a fraction of the raw digestate is directly applied on soil (up to the spreading area limitations) and another fraction is upgraded through highly performant technologies for export and market valorization.

## 4. Conclusion

The study assessed three digestate utilization scenarios. Scenario 1 describes a system where the raw digestate is directly applied on soil. In scenario 2, the raw digestate is processed by phase separation (centrifugation) with two output phases (liquid and solid digestate) that are spread on agricultural soil. Scenario 3 describes a more advanced post-treatment system, where the raw digestate is phase separated by centrifugation followed by drying of the solid digestate and further processing of the liquid digestate with a membrane filtration unit and a reverse osmosis unit. Among the studied scenarios, in scenario 3 it was possible to obtain three different types of products that have fertilizing value. These and other high-value products might answer diverse agronomic needs and sufficient concentration flexibility for substituting synthetic fertilizers products. The digestate post-treatment is a sustainable solution able to tackle the problem of excess nutrients and their management in agricultural areas. It could replace conventional nitrogen removal processes (aerobic biological treatment) keeping more nutrients in the digestate products but minimizing the losses through air and soil emissions.

The life cycle assessment showed that for the impact categories of global warming, freshwater eutrophication, and mineral resource scarcity scenario 2 was evaluated to have the lowest environmental impact. For human non-carcinogenic toxicity, terrestrial ecotoxicity, and fossil resource scarcity scenario 1 has the lowest emissions and for terrestrial acidification and marine eutrophication scenario 3 was calculated as the most efficient. The environmental performance of the system could be further improved by including the contribution to climate change mitigation potential from an increased soil carbon stock. Methodological development to include such ecosystem services is ongoing and represents a central service delivered from circular management of organic residues.

### CRedit authorship contribution statement

**Elisavet Angouria-Tsorochidou:** Methodology, Investigation, Software modelling, Validation, Visualization, Writing – original draft, Writing – review & editing. **Michele Seghetta:** Conceptualization, Software modelling, Writing – review & editing. **Anne Trémier:** Conceptualization, Field experiments & data generation, Writing – review & editing, Funding acquisition. **Marianne Thomsen:** Conceptualization, Methodology, Validation, Supervision, Writing – review & editing, Funding acquisition.

### 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.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2021.152764>.

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