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In the quest for sustainable management of liquid fraction of manure -Insights from a life cycle assessment

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ABSTRACT

To mitigate the risk of eutrophication and minimize adverse environmental impacts, surplus manure in nitratevulnerable zones is frequently divided into a liquid and solid fraction. Managing the liquid fraction (LF) typically presents a greater challenge due to its substantial volume. This study aimed to assess the environmental impacts by region and trade-offs of managing surplus LF with inventory data from pilot facilities using a life cycle assessment (LCA). The LF-treatment technologies assessed were (i) nitrification-denitrification (NDN) with field application of effluent (ii) NDN with ammonia stripping and nitric acid scrubbing as a pre-treatment step followed by polishing in constructed wetlands, and (iii) nutrient up-concentration using vacuum evaporation and/ or membrane filtration.

The LCA results suggested that 60 to 80 % of the environmental impacts occurred locally. Nutrient upconcentration from LF via membrane filtration (reverse osmosis) and vacuum evaporation indicated a better environmental performance, albeit with high uncertainty when compared to the other scenarios. Although ammonia stripping-scrubbing showed environmental benefits, these were offset by high environmental burdens from fugitive N₂O emissions and energy demand during NDN. Furthermore, the study identified that managing the effluent after NDN, a source of potassium (K), requires a nuanced approach from policymakers. Firstly, when K fertilization requirements are not met, direct land application of the effluent as a fertigation source can be a viable option. This minimizes the need for synthetic K fertilizer production and its ensuing freshwater ecotoxicity impacts. However, tertiary treatment of NDN effluent via constructed wetlands can be considered to prevent deterioration of soil from the influx of K. Policymakers are encouraged to engage with local stakeholders to tailor solutions based on these trade-offs. Furthermore, future research should focus on the implications of K on soil quality as well as the life span of nutrient up-concentration technologies for LF.

1. Introduction

Pigs constitute the largest category of livestock in the European Union (EU) with a population of 150 million (Augère-Granier and Marie-Laure, 2020), with Belgium experiencing manure surpluses due to intensive animal husbandry. For instance, in Flanders (BE), approximately 49 % of the total pig manure (PM) generated is classified as surplus (Vingerhoets et al., 2023). Under typical circumstances, PM is applied to arable land as an organic fertilizer source that is rich in Nitrogen (N) and Phosphorus (P). In addition, for bio-based substrates such

as raw PM and anaerobically digested PM, the N is predominantly present in its organic form and only gradually mineralizes with time. If N mineralization does not coincide with the plant growth period, there is a high probability of nitrate (NO_3^-) leaching (Cabrera et al., 2005). Thus, fertilization with organic fertilizers requires that it is managed correctly to optimize supply and demand of the crop.

Owing to a steady influx of organic N via PM and subsequent mineralization in the soil over the years, Flemish regions with a manure surplus have developed a susceptibility to NO_3^- leaching and some have been designated as 'nitrate vulnerable zones', following the Nitrate

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Directive (EU) 676/1991. Furthermore, Flemish soils are saturated with P, by far the highest in the EU. As a result, strict limits on PM application have been set to mitigate NO_3^- leaching and to contain soil P levels (170 kg N ha⁻¹, 65–95 kg P₂O₅ ha⁻¹, depending on crop requirements) (Amery and Schoumans, 2014). Farmers are required to manage excess PM for each incremental increase in PM production above the maximum application rate. This procedure involves solid-liquid separation of either raw or digested PM. Much of P binds to the solid fraction (SF) during separation, whereas N concentrates in the liquid fraction (LF). Before being transported to P-deficient regions, the SF is typically dried, hygienized, composted, or pelletized. Around 90 % of the SF produced in Flanders is exported to Germany and France.

Managing N in the LF of PM and digested PM, is more challenging. Transportation of LF to N-deficient regions is unsustainable owing to high volumes and transport costs (Melse and Verdoes, 2005). Therefore, the solution is to (i) remove and stabilize the N from the LF into inert nitrogen gas (N₂), (i.e. nitrification-denitrification) or (ii) process/upconcentrate the N in the LF to a plant available, mineral form i.e. Nutrient Reuse and Recovery technologies (NRR) (Sigurnjak et al., 2019; Vaneeckhaute et al., 2017). Selected NRRs include (a) ammonia (NH₃) stripping and scrubbing (SAS) to yield ammonium nitrate/ammonium sulfate, (b) membrane filtration (MF) to yield concentrate and (c) evaporator systems (ES) which may or may not be combined with MF to yield concentrates (Tampio et al., 2016). Furthermore, to vet the use of products derived through NRR, the EU's Joint Research Centre (JRC) recently developed technical proposals for the safe use of animal manure-derived products in nitrate vulnerable zones (Huygens et al., 2019). These products have been categorized as "RENURE" (Recovered Nitrogen from Manure).

Around 55 % of the surplus LF in Flanders is processed via nitrification-denitrification (NDN), in which ammonium nitrogen (NH⁴₄) in the liquid fraction is oxidized or 'nitrified' into nitrate (NO₃⁻) under aerobic conditions by autotrophic nitrifying bacteria. Subsequently, the facultative, heterotrophic bacteria reduce or 'denitrify' the NO₃⁻ to produce N₂ (Metcalf et al., 1991). As a result, the readily available N in the LF is made unreactive or is "lost" in the form of N₂. Drawbacks during NDN include (i) loss of reactive N as N₂, (ii) the possibility of nitrous oxide (N₂O) emissions due to incomplete/partial denitrification in the system, and (iii) high energy demand for aeration (Fabbricino and Pirozzi, 2004; Olivier et al., 2017). Despite these disadvantages, manure treatment installations in Flanders prefer NDN due to its ease of operation.

To overcome the challenges associated with NDN, the use of NRR strategies is gaining precedence. It is estimated that only 2 % of the manure treatment installations in Flanders use NH_3 stripping and scrubbing (SAS) (Vingerhoets et al., 2023). The working principle of SAS involves shifting the NH_4^+ :NH₃ equilibrium in LF to gaseous NH_3 . This is done by increasing either the temperature, pH or both. The stripped NH_3 is then brought in contact, or, "scrubbed" with nitric (HNO₃) or sulfuric (H₂SO₄) acid and recovered as ammonium salt solution (ammonium nitrate or ammonium sulfate, respectively). Alternatively, also gypsum has been implemented as scrubbing agent (Brienza et al., 2021). SAS is mostly seen as a precursor to NDN and its benefits are two-fold. Firstly, the mineral N is recuperated in the form of ammonium nitrate (NH₄NO₃), which complies with RENURE criteria. Secondly, there is a reduced burden on the NDN system, owing to a reduced N load in the influent LF (Fabbricino and Pirozzi, 2004).

Evaporation systems (ES) are another long-standing and well-tested technology used to concentrate N and potassium (K) from LF (evaporator concentrate) and to distil water from LF (condensate) (Vondra et al., 2018), reducing the volume. Nevertheless, the condensates generated from the evaporation process contain volatile components (e. g., NH₃ and volatile fatty acids), thus requiring further treatment (such as ion exchangers) before being discharged to surface water. While vacuum evaporators are robust and reliable, they are energy-intensive, with the heat demand reaching hundreds of kWhth per m^3 of

condensate (Vondra et al., 2018). ES could further be integrated with pressure-driven techniques such as ultrafiltration and reverse osmosis to filter LF (Bolzonella et al., 2018; Van Puffelen et al., 2022).

A review paper on LF management/valorization by Vondra et al. (2019) identified a notable shortcoming in existing system analyses for LF treatment. Previous system analyses have either focused on mass and energy flows or techno-economic assessments. While there have been life cycle assessments (LCA) evaluating LF usage, the comparison has either been NDN or no treatment (Corbala-Robles et al., 2018). Tampio et al. (2016) examined the mass and energy flows of the NRRs (SAS, ES, MF) and more recently, Feiz et al. (2022), compared examined the impact of no treatment versus LF treatment via SAS from an environmental and an economic standpoint. Yet, no thorough LCA comparing all the existing NRRs has been conducted to our knowledge. An overview of existing LCAs regarding LF treatment have been highlighted in Table 1.

This study focuses on utilizing primary data from LF treatment installations, some of which have information available at an early stage of construction, as well as the propagation of uncertainty in the available data and its impact on environmental assessment outcomes. These findings are then utilized to determine the technology's potential in terms of environmental benefits as well as the regional impacts. Thus, the primary goal of this work is to discover the trade-offs between the different LF treatment techniques in nutrient surplus regions and compare their performance to the baseline, i.e., NDN. These insights are aimed at helping farmers, stakeholders, and policymakers to identify the relevant nutrient recovery technologies in promoting NPK recycling and reducing reliance on synthetic fertilizers, whose production has potential environmental and geopolitical impacts.

2. Methods

We consider 4 scenarios for managing LF of manure, and the geographical scope is set to a typical manure surplus region in Northwestern Europe, specifically Flanders, Belgium. The primary function of the system is to treat the liquid fraction in compliance with the following discharge norms: 250 mg L⁻¹ Chemical Oxygen Demand (COD), 25 mg L⁻¹ Biochemical Oxygen Demand (BOD), 35 mg L⁻¹ suspended solids,

Table 1

Overview of existing studies concerning LCA of anaerobic digestion and/or manure treatment.

Study	Focus of study	Key findings/Objectives
Finzi et al. (2020)	Techno-economic and environmental assessment of energy and nutrient recovery from manure treatment facilities	Profit of \pounds 1.61 t ⁻¹ treated manure and 70 % reduction in global warming potential (20.79 kg CO ₂ eq tonne ⁻¹).
Corbala- Robles et al. (2018)	AD vs. untreated swine manure	46 % reduction in climate change impact (9.80 kg CO_2 eq m ⁻³) with AD treatment.
Duan et al. (2020)	LCA of swine manure treatment	GW potential ranged from -11 to 64.7 kg CO ₂ eq tonne ⁻¹ for different treatment technologies.
Vondra et al. (2019)	Literature review on LF (livestock farming) management/valorization	Highlighted the lack of comprehensive LCA comparing various LF waste treatment approaches.
Vázquez- Rowe et al. (2015)	Comparison of NDN vs. nutrient recovery technologies	Examined environmental aspects of different LF waste management approaches.
Tampio et al. (2016)	Mass and energy flows analysis of various nutrient recovery technologies	Investigated the mass and energy flows of LF waste treatment options.
Feiz et al. (2022)	Comparison of LF waste treatment via various nutrient treatment technologies	Evaluated the environmental and economic trade-offs of LF waste treatment options.

15 mg L^{-1} N and 1 mg L^{-1} P.

The study's functional unit (FU) is the treatment of 1tonne of LF of raw and/or digested PM, and the characteristics are listed in the Supplementary information. The FU was analyzed in a number of scenarios described below. A graphic representation can be found in Fig. 1 (NDN: nitrification-denitrification, CW: constructed wetlands, RO: reverse osmosis; (T) denotes transport of the product and hashed boxes represent avoided product). The primary step, i.e. solid-liquid separation of



Fig. 1. System boundaries comparing liquid fraction (LF) manure management.

PM, has been cut-off from the system since centrifugation and the enduse of the solid fraction (i.e. transportation to P-deficient regions) is common for all scenarios (Fig. 1). The LCA is modelled using a consequential approach and multifunctionality is handled through system expansion to account for the processes that are avoided by production of co-products (Niero et al., 2014). The background processes within the system boundary are based on the consequential system model of the ecoinvent database.

Scenario 1 (S1) represents the current baseline for managing surplus LF across manure treatment facilities in Flanders. Here, the LF is stabilized via NDN, with sludge as a by-product. However, the effluent from NDN does not typically meet the discharge norms, and its subsequent fate is region-specific. Ideally, the effluent from NDN should be directed to a constructed wetland (CW) for tertiary treatment. However, the reality is that this practice is not widely adopted among farms, and instead, most farmers opt to transport the effluent over short distances (around 10 km) and apply it to the land. The rationale is that the effluent has a low enough N to K ratio to not pose a threat of NO_3^- leaching, but at the same time, it can serve as a K fertilizer source. The residual sludge from NDN, owing to its high P content is transported to P deficit regions, in this case, France. Equivalent credits for avoided synthetic K and P fertilizer credits are ascribed for the NDN effluent and sludge, respectively.

In Scenario 2 (S2), the NH₃ in the liquid fraction is stripped and scrubbed with an acid, in this case, HNO₃, to form NH₄NO₃. The ensuing NH₄NO₃ solution serves as a substitute to conventional mineral N fertilizer (Brienza et al., 2023). The stripping residue is pumped to a NDN system, followed by tertiary treatment in a CW to meet the discharge norms. The treatment trail in S2 is selected to reflect the ideal case scenario for NDN systems. Similar to S1, the sludge from NDN is transported to regions with a P-deficit. However, it must be noted that the effluent from CW is not credited for avoided K fertilizer since it is discharge to water bodies.

Scenario 3 (S3) uses a combination of membrane filtration and an evaporation system to up-concentrate N and K from LF. This particular configuration first uses micro-filtration to filter the particulates from LF (i.e. filtrate), and the permeate is fed to the reverse osmosis (RO) unit. The RO unit concentrates N and K (RO concentrate), and the output is fed to an ion exchanger for tertiary treatment to meet discharge limits. The RO concentrate is fed to a vacuum evaporator to reduce the volume, thus yielding an NK-rich concentrate. This product is seen as a synthetic fertilizer substitute, whereas the permeate is discharged. The filtrate from the micro-filtration unit is transported to non- nutrient surplus regions, for which fertilizer credits are ascribed.

Scenario 4 (S4) focuses on evaporation without a membrane filtration set-up for the LF. Prior to evaporation, the LF undergoes aerobic treatment to reduce the BOD. Subsequently, in the evaporator, NH₃ and water evaporate which after condensation of the vapours forms condensed ammonia water. The condensed ammonia water has an NH₄-N content of around 10 %, and can be considered as a DeNOxing agent at incineration facilities. The evaporator also produces a condensate with a low N content (<0.1 %), called process water which is recirculated back to the aerobic treatment step. The main output from the system is a concentrate, which is a NK fertilizer substitute.

Impacts from field application of the products from all scenarios are included and the emission factors are provided in Table 2.

2.1. Life cycle inventory

The mass and energy flows for scenarios S1, S2 and S4 were based on primary data derived through measurement campaigns. Data quality for S3 was unreliable, and its uncertainty was quantified using a pedigree matrix approach. For NDN, the mass and energy flows were computed using the Activated Sludge Model (ASM1) and modelled in the opensource wastewater simulation tool STOAT (Stokes et al., 1997). The data related to auxiliary material usage and the infrastructure processes for all scenarios were collected from the ecoinvent database (Wernet et al., 2016) as well as peer-reviewed literature. The life cycle inventory information including the probability distribution of the exchanges necessary for the uncertainty analysis is presented in Table 3.

2.2. Life cycle impact assessment

The impacts were quantified using the Environmental Footprint (EF 3.0) life cycle impact assessment (LCIA) methodology since the geographical focus of the study was Europe and the LCA was modelled using Brightway2 and Activity Browser. Midpoint indicator results were normalized and weighted according to the EF guidelines to represent both best- and worst-case scenarios through the single score resulting from the weighting method from the EF methodology. Impact categories that collectively accounted for at least 80 % of the overall scores were identified as the most relevant (Zampori and Pant, 2019). Finally, for each scenario, the contribution analysis and uncertainty for the relevant impact categories are shown. The uncertainty analysis in the foreground system (Table 3) as well as in the background (ecoinvent processes) was performed using a dependent sampling approach, wherein all scenarios under comparison were sampled using the same technology and biosphere matrices for a given functional unit (Cucurachi et al., 2018). Furthermore, the regional impacts were identified using the Activity Browser's 'aggregate by region' feature, and further geospatial analysis and visualization was performed using the geopandas and matplotlib python packages. For regionalization, the impacts were aggregated by location and matched with its associated geographies through shapefiles provided by ecoinvent. Next, the results for the foreground system were isolated to better understand the impacts between local and global processes. This is done because of the "cut-out" locations (such as Europe without Switzerland or Europe without Russia etc) in the ecoinvent database, where regions are created without specific states or countries to link different market processes. Also impacts from "Rest of the world" processes were not represented since they do not have a set

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Emissions from field application of products generated in each scenario.

Emissions from field application of products generated in each scenario.							
Emission/substitution*	Unit	Biological effluent (S1)	Ammonium nitrate ^a (S2)		Mineral concentrate ^a (S3 and S4)		
			Grassland	Arable land	Grassland	Arable land	
NH ₃ -N ^a	%TAN	2.5	2.5		6	0.64	
N ₂ O-N ^a	%N	1	1.2	1	0.6	1.95	
NO-N ^a	%N	0	0		0.55	0	
NO ₃ -N ^b	%N	5	15.8		18.1		
P ^c	kg/kg P ₂ O ₅	0.00184	0		0		
N fertilizer replacement value ^a	%	0	100		60	70 %	

^{*} Note: S1 represents nitrification-denitrification (NDN) and field application of effluent, S2 represents stripping and scrubbing as pre-treatment with NDN followed by post-treatment in constructed wetlands, S3 represents Membrane filtration and vacuum evaporation, and S4 represents vacuum evaporation.

^a De Vries et al. (2012).

^b Roy et al. (2003).

^c Corbala-Robles et al. (2018).

Table 3

Life cycle inventory for all scenarios. Inputs are highlighted in bold and detailed exchanges for background processes are available in the Supplementary information.

Unit process*	Inputs/outputs per unit process	Unit	Probability	S1	S2	S3	S4
	Influent LF (functional unit)	tonne	distribution	1	1	1	1
Nitrification-denitrification (NDN)	Methanol	kg	Triangular	1.9-6.81	1.4-1.9		
	Electricity	kWh	Triangular	7.05-11.37	6.45–7		
	Ammonia	kg		0.02	0.02		
	Dinitrogen monoxide	kg		0.05	0.03		
	Sludge	kg		250	250		
	Biological effluent	kg		750	750		
Field application	Transport of product	t-km	Normal	4.7 ± 2	0.25 ± 0.02		
	Liquid manure spreading, by vacuum tanker	m ³		0.75	0.025		
	Inorganic potassium fertilizer, as K ₂ O	kg		-4.24			
	Inorganic nitrogen fertilizer, as N				-2.2		
	Nitrate	kg	Normal	0.12	0.35		
	Ammonia	kg	Normal	2.00E-03	0.02		
	Dinitrogen monoxide	kg	Normal	2.30E-03	0.02		
Sludge management	Transport of sludge	t-km		46	46		55
	Phosphate fertilizer, as P_2O_5	kg		-0.77	-0.82		-0.21
	Inorganic nitrogen fertilizer, as N						-0.72
	Inorganic potassium fertilizer, as K ₂ O	ha		220	16		-0.605
	Dinitrogon monovido	ĸg		230	40		220
	Nitrate						0.01
	Ammonia	kσ		0.01	0.01		0.10
	Methane	kg kg		0.33	0.33		0.03
Stripping and scrubbing	Market for nitric acid, without water, in 50 %	kø		0.00	4.54		0.00
surpping and serassing	solution state	0			110 1		
	Electricity	kWh			2.13		
	Tap water	kg			16		
	Stripped effluent	kg			1000		
	Ammonium nitrate	kg			25		
Constructed wetlands	Biological effluent	kg			750		
	Dinitrogen monoxide	kg			6.70E-04		
	Transformation, from arable land	m ² -			0.69		
		year					
Microfiltration	Electricity (trommel filter)	kWh				0.18	
	Trommel filter rejects	kg				50	
	Effluent from trommel filter	kg	· ·			950	
	Electricity	kwh	Lognormal			1.49 ±	
	Potontato	kα				0.45	
	Dermeate	kg kg				855	
Beverse osmosis	Electricity	kWh				3.56	
	Sulfuric acid	kg				2.09	
	Sodium hypchlorite	kg				9.50E-03	
	RO permeate	kg				213.75	
	Condensate from evaporator	kg				106.88	
	RO concentrate	kg				748.13	
Evaporator	Electricity	kWh	Triangular			20-25	22–23
	Antifoaming agent, adipic acid	kg				0.11	0.58
Field application_Evaporator	Transport of product	t-km				1.0685	0.94
concentrate	Inorganic potassium fertilizer, as K ₂ O	kg				-2.35	-4.34
	Inorganic nitrogen fertilizer, as N	kg				-1.42	-0.76
	Fertilizer spreading	m3				0.10	0.09
	Ammonia Dinitro con monovido	Kg Ive				0.01	0.00
	Nitrate	ka				0.02	0.01
Field application Retentate	Fertilizer spreading	m3				0.28	0.14
red application_recentate	Transport of product	t-km				23 75	
	Inorganic nitrogen fertilizer, as N	kg				-1.02	
	Inorganic potassium fertilizer, as K ₂ O	kg				-2.57	
	Phosphate fertilizer, as P_2O_5	kg				-1.72	
	Ammonia	kg				0.01	
	Dinitrogen monoxide	kg				0.03	
	Nitrate	kg				0.40	
Aeration tank	Electricity use	kWh					19.00
	Iron chloride	kg					0.07
	Recirculated process water	kg					276.63
	Dinitrogen monoxide	kg					0.01
	Ammonia	kg					0.00
	Sludge from aeration tank	kg					220
Credits for condensed ammonia	Ammonia	kg					-1.68
water Aerobic treatment of process water	Electricity	kwb					5.82
meropic meannent of process water	LICCULLY	IL VV II					5.65

(continued on next page)

Table 3 (continued)

Unit process*	Inputs/outputs per unit process	Unit	Probability	S1	S2	S3	S4
	Influent LF (functional unit)	tonne	distribution	1	1	1	1
	Ammonia	kg					3.00E-
	Dinitrogen monoxide	kg					04 1.90E- 02

* Note: S1 represents nitrification-denitrification (NDN) and field application of effluent, S2 represents stripping and scrubbing as pre-treatment with NDN followed by post-treatment in constructed wetlands, Scenario 3 represents Membrane filtration and vacuum evaporation, and Scenario 4 represents vacuum evaporation.

KML description (ecoinvent 2022). The supporting code is included in the Supplementary information as jupyter notebooks.

3. Results

3.1. Overall impacts and contribution analysis

Fig. 2 compares the assessed scenarios based on their normalized and weighted scores of all impact categories under the EF method. From the overall impacts, it appeared that Scenario S3, i.e., membrane filtration



Weighted impact results

Fig. 2. Overall impact score after normalization and weighting per functional unit (managing 1 t of LF). S1 represents nitrification-denitrification (NDN) and field application of effluent, S2 represents stripping and scrubbing as pre-treatment with NDN followed by post-treatment in constructed wetlands, S3 represents Membrane filtration and vacuum evaporation, and S4 represents vacuum evaporation.

and vacuum evaporation (overall weighted score of -0.006), outranks the other scenarios (S4: -0.005; S1: -0,004, S2: 0.001) as the most environmentally friendly alternative. The highest burdens from S3 (membrane filtration and vacuum evaporation) are due to climate change potential (4 % of the overall score) as well as marine eutrophication potential (2 %). However, these burdens are to a large extent offset by benefits due to freshwater ecotoxicity potential and mineral and metals resource use, which contributed to 53 % and 32 % of the overall score respectively. The overall impact contributions for S4 (vacuum evaporation) and S1 (NDN + field application of effluent) followed a similar trend as S3. For S2 (Stripping-Scrubbing + NDN + Constructed wetlands), the net results showed an environmental burden, primarily because the benefits from freshwater ecotoxicity potential were much lower (10 %) compared to the other scenarios.

From Fig. 2, the impact categories for further analysis are relevant if they contribute to at least 80 % of the overall score. Based on this criterion, the impact categories of interest are climate change potential, freshwater ecotoxicity potential, acidification, and eutrophication potential. The impacts of mineral and metal resource utilization for contribution analysis are excluded due to high uncertainty.

Fig. 3 details the overall impacts for impact categories selected after normalization and weighting. Here, sub-scenarios (suffix _no K) have been included where fertilizer credits from avoided K fertilizer were not ascribed. It can be seen that across all impact categories for scenarios S1, S3 and S4, the environmental impacts increase when K fertilizer credits are not ascribed. For S2, however, there is no difference since the K is lost anyway due to treatment and subsequent discharge from constructed wetlands.

3.1.1. Climate change potential

The potential climate change impacts for S3 (median: 5.71 kg CO_2 -eq) appeared to be the least relative to the other scenarios (S2: 31 kg CO₂-eq; S1: 24 kg CO₂-eq; S4: 16 kg CO₂-eq) (Fig. 4).

The contribution analysis for the baseline, i.e. S1 showed that the majority of the burdens from climate change potential are due to fugitive N_2O emissions from NDN (14 kg CO₂-eq) as well as the energy demand



Fig. 3. Overall results for select impact categories per functional unit (managing 1 t of LF). S1 represents nitrification-denitrification (NDN) and field application of effluent, S2 represents stripping and scrubbing as pre-treatment with NDN followed by post-treatment in constructed wetlands, Scenario 3 represents membrane filtration and vacuum evaporation and Scenario 4 represents vacuum evaporation. The suffix "_no K" describes scenarios that do not include fertilizer credits for potassium.



Fig. 4. Overall impacts for selected impact categories per functional unit (managing 1 t of LF) after 1000 Monte Carlo runs. Violins represent probability distributions using kernel density estimation on either side. S1 represents nitrification-denitrification (NDN) and field application of effluent, S2 represents stripping and scrubbing as pre-treatment with NDN followed by post-treatment in constructed wetlands, S3 represents membrane filtration and vacuum evaporation and S4 represents vacuum evaporation.

for aeration (2 kg CO₂-eq) and methanol use for denitrification (4 kg CO₂-eq) (Fig. 5). Other significant contributors include the transportation and storage of sludge to P deficient regions (16 kg CO₂-eq). These burdens are offset by avoided synthetic K fertilizer use (-15 kg CO₂-eq) as a consequence of field application of the effluent from NDN. The results also showed that the environmental performance of S1 was highly dependent on the K fertilizer credits.

S2 (i.e. SAS + NDN + CW configuration) showed a 29 % increase in potential climate change impacts relative to S1 (Fig. 3). This increase is primarily due to the direct discharge of the effluent from the constructed wetlands and as a consequence, its constituent K leaves the system without fertilizer credits. Leaving the K fertilizer caveat aside, the inclusion of ammonia SAS in S2 reflected a benefit on the NDN step in S2, which showed a 38 % reduction compared to the NDN in S1. This is primarily due to reduced N loading during NDN as a consequence of SAS, which harvests mineral N in the form of NH₄NO₃. The burdens from the SAS system (4.57 kg CO₂) were mostly a consequence of nitric acid production. These emissions are partially offset by avoided production of synthetic N (-9 kg CO₂-eq) due to field application of NH₄NO₃. S3, MF + ES (5.71 kg CO2-eq) showed the least potential climate change impacts in relation to the other scenarios, although with high uncertainty. This can primarily be attributed to the up-concentration of N and K in the form of mineral concentrate (from RO and vacuum evaporator) and filtrate (residual fraction from microfiltration) and their subsequent field application (Retentate: -9 kg CO_2 -eq and Mineral Concentrate: -9 kg CO_2 -eq) create a net positive impact on climate change potential. The major burdens from S3 are due to the infrastructure for the evaporator (8 kg CO₂-eq) as well as its energy usage (7 kg CO₂-eq), which is higher compared to S1 and S2. The infrastructure burdens can be attributed to the stainless steel and brass needed for the evaporator, the RO, and ceramic membranes for microfiltration as well as the infrastructure modules for the ion exchanger.

S4 ranked as the second-best alternative in terms of climate change potential. Despite up-concentration of N and K, the energy use during aeration increases the net climate change potential. Furthermore, the burden from infrastructure is like S3, albeit lower since an ion exchanger is not present in this scenario. These burdens are however offset by benefits due to avoided N and K fertilizer from the mineral concentrate and process water. The use of ammonia water as a denoxing agent avoids the use of conventional ammonia thereby benefitting the system $(-4 \text{ kg CO}_2\text{-eq})$.

3.1.2. Acidification potential

For acidification potential, S3 (median: 0.02 mol H^+ -eq) performed the best relative to the other scenarios (S2: 0.18 mol H^+ -eq; S1: 0.03 molH⁺-eq; S4: 0.09 mol H^+ -eq) (Fig. 4). The high acidification impacts in S2 can primarily be attributed to NH₃ emissions from field application of NH₄NO₃, since it has a higher emission factor (2.5 % of TAN for arable land) when compared to mineral concentrates in S3 and S4 (0.64 % of TAN).

3.1.3. Freshwater ecotoxicity potential

Freshwater ecotoxicity impacts are represented by the toxic effect on aquatic species in the water column and measured in comparative toxic unit equivalent (CTU-eq). As seen in Fig. 5, the impacts due to freshwater ecotoxicity potential are a function of whether K fertilizer credits is awarded to the system. For scenarios S1, S3, and S4, K was supplemented through field application of products, reflecting an increased environmental benefit, whereas in S2, where the K is lost through effluent discharge, the scores showed a comparatively lower ecotoxicity benefit. The major influence on freshwater ecotoxicity potential is due to sulphur and chloride emissions during potassium chloride and potassium sulfate production respectively.

3.1.4. Terrestrial eutrophication potential

With respect to terrestrial eutrophication, N is the limiting factor and the impacts resulted from NH_3 , and NO_3^- emissions due to field application of the ensuing products. Similar to acidification potential, S2 performed poorly relative to the other scenarios (Fig. 5).

3.2. Impacts by region

Because environmental variables vary widely across space (for example, water availability, land type, population density, and background pollution concentrations), regionalization in LCAs is extremely relevant (Verones et al., 2020), and can be performed at the inventory as well as the impact assessment level with regionalized characterization factors. This study regionalized the impacts on inventory level, but did not incorporate a spatially explicit Life cycle impact assessment (LCIA) method, (for instance AWARE, ImpactWorld+, LC-Impact). However, impact categories for regionalization were chosen such that either they used country-specific characterization factors or where there was no need to provide location-specific emission factors (for instance climate change potential, which is modelled as a global increase in radiative forcing). Here, we illustrated the impacts by region for climate change potential (Fig. 6) for Scenario 3. It can be seen that around 60-80 % of the impacts occurred in the foreground, mostly in Belgium and France. The rest of the impacts occurred worldwide and this can be attributed to auxiliary use for managing LF as well as avoided primary products.

4. Discussion

Comparison of the results with previous peer-reviewed works was possible although the system perspective and functional units were study-specific. In Table 4, we present a comparative analysis of the outcomes of our work with scenarios and technologies from similar LCAs as presented in Table 1. Perhaps the closest comparison could be made with the results of Corbala-Robles et al. (2018), although they compared $1m^3$ of direct landspreading of pig manure versus treatment via NDN in Flanders. The overall outcome from their study was inconclusive, with some impact categories favoring direct landspreading over NDN and vice-versa. However, they identified that NDN is an environmental hotspot for fugitive N₂O emissions, and also illustrated the impact of the high energy demand, corroborating our results. Furthermore, we identified that the inclusion of a stripping and scrubbing process prior to NDN, reduced its associated environmental burden owing to reduced N loads. The burdens from stripping and scrubbing itself pertains to acid and energy use validating the observation of Vázquez-Rowe et al. (2015), although they considered direct field application of the effluent from stripping and scrubbing. Furthermore, we identified that the use of HNO3 partially offset the benefits from producing NH4NO3 and its associated fertilizer credits since the production of HNO₃ follows the Ostwald process, which itself has a high environmental footprint. The use of a scrubbing substitute with a lower environmental impact, possibly organic acids could be tested (Brienza et al., 2020). Also, this study considered a pilot facility which implemented ammonia stripping and scrubbing with no additional heat as well as pH control, and as a consequence the NH3 recovery from the LF was on the conservative end i.e. 29 % N. Based on expert estimates on-site, the NH3 recovery from LF could further be increased to around 50-60 %, but this requires additional energy and auxiliary use.

Notwithstanding the environmental benefits from stripping and scrubbing, S2, which reflected the ideal treatment trail through stripping-scrubbing and post treatment through NDN and constructed wetlands performed worse relative to S1 (with NDN and field application of effluent from NDN). This appears counter-intuitive, despite the implementation of N recovery in S2. This is because the fertilizer credits given to K as a consequence of direct field application of the effluent from NDN results in a net environmental benefit especially with regard to freshwater ecotoxicity, thus favoring S2 over S1. This may not exactly match reality since K crop requirements might already be met with the rest of the fertilization plan (optimized on raw manure usage), and thus the spreading of NDN effluent may not save primary resources.

A treatment trail with Stripping and Scrubbing followed by NDN and field application of the NDN effluent appears to be the optimal choice if K crop requirements are not met. On the flipside farmers could process NDN effluent via constructed wetlands since the Na + K content is high in effluent compared to Ca + Mg. This implies an unfavorable SAR-ratio, which degrades alumosilicates (clay minerals) if consistently spread at high amounts. In other words, NDN effluent usage results in soil structure degradation by progressive substitution of divalents in the clay minerals by monovalents (and bridge collapse). Perhaps the use of constructed wetlands as a tertiary treatment step could be combined with the growth of floating wetland plants to valorize the nutrients from the NDN effluent and produce protein rich biomass (Beyers et al., 2023).

If the transport distances render it impossible for field application of the NDN effluent, then the use of constructed wetlands for tertiary treatment cannot be completely ruled out solely on the basis of the "lost K fertilizer" and the freshwater ecotoxicity impacts associate with the production of alternative sources of K fertilizer. As pointed out in this study, the sensitivities of K fertilizer credits has a significant bearing on the overall results (Fig. 3). Therefore future practitioners are recommended to consider this caveat for the sensitivity analysis. Furthermore, these results should be carefully interpreted due to two reasons. (i) The ecoinvent process of the 'market for inorganic potassium fertilizer, as K₂O, BE' is heavily associated with the emission of sulphur and chloride into freshwater bodies. It is noteworthy that sulphur constitutes the largest contributing factor to freshwater ecotoxicity in this analysis. (ii) the characterization factors for freshwater ecotoxicity used by the EF method relies on the USEtox®, a global scientific consensus model to measure ecotoxicity effects. While the model is primarily suited to measuring the effects of organic compounds, (Fazio et al., 2018), it includes inorganics and metals, albeit with an uncertainty factor of 0.1. Further scientific developments are required to cover more environmental compartments as well as reducing uncertainty for inorganics (Sala et al., 2022). What is perhaps giving even more cause for caution is that the high relative impact in the freshwater ecotoxicity is dependent on the weighting factors of the EF weighting methodology. These weighting factors are highly uncertain for the toxicity categories (Sala et al., 2022).



Fig. 5. Contribution analysis of select impact categories per FU (managing 1 t of LF). S1 represents nitrification-denitrification (NDN) and field application of effluent, S2 represents stripping and scrubbing as pretreatment with NDN followed by post-treatment in constructed wetlands, S3 represents membrane filtration and vacuum evaporation and S4 represents vacuum evaporation.



Fig. 6. Impacts by country and region for climate change potential. Scenario 3 represents membrane filtration and vacuum evaporation.

Despite the caveat of potential freshwater toxicity impacts, the use of K and its implications should not be understated even though it is not a limiting nutrient. This is because of Russia's sphere of influence in the synthetic fertilizer market; as of 2020, Russia's market share for N, P_2O_5 and K_2O totalled 14 %, 13 % and 18 % respectively. Also, the fertilizer prices haven risen by nearly 30 % since the advent of the Russia-Ukraine conflict (OECD, 2022). This puts the EU in a vulnerable position, especially since it relies on other countries for its P and K demand.

Considering these geopolitical limitations, the importance of N and K recovery via membrane filitration and/or vacuum evaporation becomes profound. From this study, nutrient up-concentration technology via reverse osmosis and vacuum evaporation (S3) pointed to an environmental benefit as opposed to stripping and scrubbing, where only N is recovered, and NDN, where the N is lost as N₂. The benefits can be attributed to the avoided production of conventional N and K fertilizer, which have a high environmental footprint. The work of Tampio et al. (2016) which assessed nutrient valorization from food waste digestate had a similar outcome, where reverse osmosis and vacuum evaporation was considered to be beneficial over stripping and scrubbing. In our

study, the hotspots from nutrient up-concentration were energy demand, auxiliary use, as well as the infrastructure needed for the treatment modules. Furthermore, in S4, in addition to mineral concentrate, condensed ammonia water is produced as a by-product from vaccum evaporation. Owing to its high pH (~11), the end-use of ammonia water in agriculture and its market demand need to be further researched. This study assumed that the ammonia water could possibly substitute urea in DeNOx (selective non-catalytic reduction) systems for treatment of the flue gasses of incineration plants, which may be too ambitious. Regarding infrastructure, the lifespan of the reverse osmosis membrane is highly dependent on the efficiency of manure separation as well as pre-treatment using microfiltration (Van Puffelen et al., 2022). Based on expert views, our study considered a rather conservative estimate of one year for the life span of reverse osmosis modules. Furthermore, we relied on secondary literature to build the inventory for the evaporator and microfiltration modules, which is a shortcoming. More emphasis is therefore needed to build a robust inventory for infrastructure processes related to nutrient recovery.

While the nutrient up-concentration scenarios benefit the

Table 4

Comparison of results from this study versus peer reviewed LCA studies in Table 1. Note that climate change potential has been considered since it is widely used impact category across the peer reviewed studies.

Literature and technological process considered		Results from literature	Scenarios from this study that can be possibly compared
Finzi et al. (2020)	Anaerobic digestion, solid-liquid separation, nitrogen removal and field application	20.79 kg CO_2 eq t ⁻¹ of treated manure	25.56 kg CO_2 eq t^{-1} of LF manure using nitrification- denitrification and field application (S1)
Corbala- Robles et al. (2018)	Solid-liquid separation, nitrification- denitrification and field application of solid and liquid fraction	9.80 kg CO ₂ eq/m ³	$25.56 \text{ kg CO}_2 \text{ eqt}^{-1} \text{ of}$ LF manure using nitrification- denitrification and field application (S1)
Duan et al. (2020)	Composting solid fraction and using treated liquid fraction for microalgae cultivation and composting solid fraction and producing powder biofertilizers via struvite precipitation with ammonia stripping	-11 to 64.7 kg CO ₂ eq t ⁻¹ of PM treated	32 kg CO ₂ eqt ⁻¹ of LF manure using Ammonia stripping-scrubbing, nitrification- denitrification and treatment of effluent via constructed wetlands (S2)
Vázquez- Rowe et al. (2015)	Solid, liquid separation, biological treatment, reverse osmosis and drying of digested PM	56.58 kg CO ₂ eq/m ³	25.56 kg $CO_2 eqt^{-1}$ of LF manure using nitrification- denitrification and field application (S1) and 10.36 kg kg $CO_2 eqt^{-1}$ for reverse osmosis, vacuum evaporation and field application (S3)
	Ammonia stripping and drying of digested PM	68.41 kg CO ₂ eq/m ³	32 kg CO ₂ eqt ⁻¹ of LF manure using Ammonia stripping-scrubbing, nitrification- denitrification and treatment of effluent via constructed wetlands (S2)
Feiz et al. (2022)	Solid liquid separation of digestate and ammonia stripping & scrubbing followed by field application	30 kg CO ₂ eqt ⁻¹	32 kg CO ₂ eqt ⁻¹ of LF manure using Ammonia stripping-scrubbing, nitrification- denitrification and treatment of effluent via constructed wetlands (S2)

environment, it is important to factor in their capital costs, operational costs and ease of operation. The capital costs for an evaporator are around 20 €/m³ LF processed and 7–18 €/m³ for a reverse osmosis unit (Derden, 2020) with the operational costs varying between 4 and 6 \notin/m^3 (Vaneeckhaute et al., 2017). In comparison, operating an NDN unit costs around 5 \notin /m³ for capital costs and around 7 \notin /m³ for operational costs. Adding a stripping and scrubbing unit would additionally cost 1.5–3.2 (m^3) and 5 (m^3) for capital and operational expenses respectively (Brienza et al., 2023). Thus, the cost difference between upconcentrating nutrients and using a stripping and scrubbing system plus an NDN module is nearly a factor of two. Furthermore, the SAS system might be easier to operate at a farm level compared to an evaporator system. Yet, it must be highlighted that the economical value of final products (e.i. concentrates form ES, as well as ammonium salts from SAS) is susceptible to variation and might have different impacts on the overall process cost from region to region. Data from a technoeconomic assessment, as well as inputs from a social life cycle assessment, might be used with the current LCA results to make an informed

choice on the suitable technology.

Measuring the intensity and estimating regional impacts of N emissions from manure management and agriculture is crucial to prevent the trespassing of regional and planetary N boundaries (Schulte-Uebbing et al., 2022). In that order, this study attempted to bridge the gap with regard to regionalization of manure processing. We analyzed the impacts by region at inventory level and for the LCIA, the EF method was used. Although the EF uses regionalized characterized factors for certain impact categories (eutrophication, for instance), producing reproducible regionalized LCAs remains a challenge (Frischknecht et al., 2019). An ongoing collaboration to harmonize regionalization is underway, with the LCIA method LC-IMPACT providing regionalized characterization factors for all relevant impact categories. These characterization factors are then translated to endpoint indicators in the form of human health, ecosystem damage and resource use. Future studies could aim at implementing LC-IMPACT and more information regarding the method is available in (Verones et al., 2020).

5. Conclusion

According to the LCA, concentrating nutrients via reverse osmosis and/or vacuum evaporation outperforms the treatment trails of stripping-scrubbing, nitrification-denitrification, and tertiary treatment in constructed wetlands as the most environmentally beneficial option for managing liquid fraction of manure.

We identified that fugitive N₂O emissions and energy demand during nitrification-denitrification of liquid fraction of manure are major environmental hotspots that can be reduced in part by introducing stripping and scrubbing as a pre-treatment step, but post-treatment of nitrification-denitrification effluent was the study's point of contention. The seemingly sub-optimal route of field application of nitrificationdenitrification effluent demonstrated a net environmental benefit due to avoided K fertilizer as opposed to tertiary treatment in a constructed wetlands system, where the K is lost to surface water. The environmental benefits were mostly due to freshwater ecotoxicity potential, whose characterization and weighting factors for inorganic compounds are highly uncertain. Therefore, these results must be cautiously considered. For liquid fraction management through reverse osmosis and vacuum evaporation, the production of mineral concentrate avoided the production of conventional N and K fertilizers causing a net environmental benefit. However, we recommend future studies to address the lifespan and data quality of the infrastructure concerning nutrient upconcentration.

Finally, because avoided synthetic fertilizer production affects the outcome of LCAs including nitrogen recovery from manure, future studies may include a socioeconomic variable connected to the geopolitical supply risk of crucial raw materials.

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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CRediT authorship contribution statement

Rahul Ravi: Writing and conceptualization, LCA modelling. Miriam Beyers: Conceptualization, LCA modelling, Review &

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Ruben Vingerhoets: Data collection, Review & Editing.

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Appendix A. Supplementary data

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