Enhancing swine manure treatment : a full-scale techno-economic assessment of nitrogen recovery, pure oxygen aeration and effluent polishing

Reference:
Vingerhoets Ruben, Sigurnjak Ivona, Spiller Marc, Vlaeminck Siegfried, Meers Erik.- Enhancing swine manure treatment : a full-scale techno-economic assessment of nitrogen recovery, pure oxygen aeration and effluent polishing
Journal of environmental management - ISSN 1095-8630 - 356(2024), 120646
Full text (Publisher's DOI): https://doi.org/10.1016/J.JENVMAN.2024.120646
To cite this reference: https://hdl.handle.net/10067/2046640151162165141

Enhancing swine manure treatment: a full-scale techno-economic assessment of nitrogen recovery, pure oxygen aeration and effluent polishing

Ruben Vingerhoets[a,b], Ivona Sigurnjak[a], Marc Spiller[b,c], Siegfried E. Vlaeminck[b,c], Erik Meers[a]

[a]Ghent University, Department of Green Chemistry and Technology, RE-SOURCE - Laboratory for biobased resource recovery, 9000 Gent, Belgium.
[b]University of Antwerp, Department of Bioscience Engineering, Research Group of Sustainable Energy, Air and Water Technology, 2020 Antwerpen, Belgium
[c]Centre for Advanced Process Technology for Urban Resource Recovery (CAPTURE), Frieda Saeysstraat 1, 9052 Gent, Belgium

List of acronyms, abbreviations and definitions

AN - ammonium nitrate
AS - ammonium sulphate
BOD – Biological oxygen demand
CAPEX - capital expenditure
COD – Chemical oxygen demand
CW – Constructed wetland
EU – European Union
FA – Free ammonia
FPR - Fertilising Products Regulation
LF – Liquid fraction
MLSS - Mixed Liquor Suspended Solids
NDN – Nitrification – denitrification
NVZ - Nitrate Vulnerable Zones
OPEX - operational expenditure
PO – Pure oxygen aeration
Re - Recovery efficiency
RENURE - Recovered nitrogen from manure
Rm- Removal efficiency
Se - Separation efficiency
SF – Solid fraction
SS – Stripping-scrubbing

ABSTRACT

In regions with intensive livestock production, managing the environmental impact of manure is a critical challenge. This study, set in Flanders, Belgium, evaluates the effectiveness of integrating process intensification measures into the treatment of piggery manure to mitigate nitrogen surplus issues. The research investigates the techno-economic benefits of implementing three key interventions: pure oxygen (PO) aeration, ammonia (NH_3) stripping-scrubbing (SS) pretreatment, and tertiary treatment using constructed wetlands (CW), within the conventional nitrification-denitrification (NDN) process.

Conducted at a full-scale pig manure treatment facility, our analysis employs steady-state mass balances for nitrogen (N) and phosphorus (P) to assess the impact of these process intensification strategies. Findings indicate that the incorporation of advanced treatment steps significantly enhances the efficiency and cost-effectiveness of the manure management system. Specifically, the application of PO aeration is shown to reduce overall treatment costs by nearly 4%, while the addition of an NH_3 SS unit further decreases expenses by 1 to 2%, depending on the counter acid utilized. Moreover, the implementation of a CW contributes an additional 4% in cost savings.

Collectively, these measures offer substantial improvements in processing capacity, reduction of by-product disposal costs, and generation of additional revenue from high-quality fertilizing products. The study highlights the potential of advanced treatment technologies to provide economically viable and environmentally sustainable solutions for manure management in livestock-dense regions, emphasizing the cumulative economic benefit of a holistic approach to process intensification.

1. INTRODUCTION

Since global meat consumption and production has grown considerably over the last century, a large amount of manure is produced contributing to a significant part of anthropogenic N emissions and the exceedance of the 'planetary boundaries' capacity (Campbell et al., 2017). Annually about 1.3–1.8 billion tonnes of manure are produced in the European Union (EU) (De Vrieze et al. 2019), mainly concentrated in densely populated regions where the Nitrates Directive (EU) 676/1991 application limit of 170 kg N ha^{-1} y^{-1} is in effect, limiting application of animal manure and hence resulting in its excess. Strict manure management regulations in Flanders (Belgium), exemplifying a livestock-dense region with a manure surplus, enforce the processing of manure. Around half of the produced pig manure in Flanders is processed by the conventional treatment line consisting of a centrifuge for separation and an activated sludge tank to remove organic and inorganic contaminations (Coppens et al., 2016). The excess manure is first separated into a solid (SF) and a liquid fraction (LF) by centrifugation, whereafter the SF is dried and exported. The LF is subsequently processed by biological nitrification-denitrification (NDN) treatment followed by a clarifying step to produce an effluent with a reduced N content that can be applied on agricultural land (Brienza et al., 2023). The NDN process in Flanders removes 21 kilotonnes of N from manure by converting reactive N into N gas (N_2) (Vingerhoets et al., 2021). Nitrosomonas sp. bacteria primarily limit the rate of ammonia (NH_3) oxidation to nitrite (NO_2^-), consequently restricting the process capacity of the treatment line to the critical N load in the NDN tank (Yu et al., 2020). N loads above the rate-limiting nitrification reaction capacity induces accumulation of NH_4^+ which can inhibit nitrification and further reduce the whole processing rate of the NDN tank (Nehmtow et al., 2016). Increasing the capacity of the NDN tank by expanding the reactor’s dimensions is associated with high capital costs or infeasible because of space constraints (Derden and Dijkmans, 2020).

Elevating NDN processing capacity by process intensification can resolve this problem. The usage of pure oxygen (PO) instead of air during the aeration process increases the dissolved oxygen concentration in the wastewater, which improves the rate of the nitrification process and potentially increases the capacity of the NDN system (Rodriguez et al., 2012), but will also increase cots associated with the installation and maintenance of a PO tank. Skouteris et al. (2020) reviewed 25 studies evaluating PO aeration instead of air aeration for different wastewaters including municipal, industrial, leachate, mill effluent, food processing, petrochemical and synthetic. This review suggests a research gap on PO in swine manure treatment, indicating a broader lack of available studies or data on the use of PO aeration in swine manure treatment. Therefore, this study provides a first (to the authors knowledge) dataset to evaluate the economic feasibility of PO aeration for swine manure treatment at full scale.

To enhance the processing capacity of the NDN unit, another pathway involves reducing the N load entering the NDN tank. This is achieved by recovering N before NDN treatment through the addition of an extra recovery step, without compromising the amount of treated manure. This is particularly relevant for long-stored manure, where the elevated concentration of Ammonium N (NH_4-N), following separation, poses a challenge to the efficiency of subsequent biological treatment steps due to its inherent toxicity (Hollas et al., 2021). However, this issue can be effectively addressed through the application of physio-chemical recovery technologies, where NH_4-N is easily targeted. By reducing the N load introduced into the NDN system, a substantial decrease in nitrous oxide (N_2O) emissions associated with manure treatment can be achieved. Current estimates place these emissions in the range of 0.035-1.1% of the total input into the NDN (de Haas and Andrews, 2022; Kampschreur et al., 2009 and Ravi et al., 2023). N removal in wastewater treatment therefore contributes almost 5% of the global N_2O emissions (Olivier et al., 2017). As arable farming and horticulture in Flanders require 81 kilotonne additional N in the form of mineral fertilizers (Vingerhoets et al., 2023b), which are produced through the energy-intensive Haber-Bosch process converting atmospheric N_2 to NH_3, N recovery as a mineral N fertiliser equivalent also counters the additional need for N in the form of mineral fertilisers and reduce costs of purchasing N mineral fertilisers (Zarebska et al., 2015). Membrane filtration (e.g., nanofiltration and reverse osmosis) and physicochemical processes (e.g., NH_3 SS and chemical precipitation) are alternative treatment configuration with a high technology readiness level and adoption potential (Brienza et al., 2023). The implementation of membrane filtration involves high investment costs, intensive maintenance, and thorough pre-treatment whereas NH_3 SS is well suited for small-scale implementation and more concentrated wastewaters (van Puffelen et al., 2022). The rollout of full-scale NH_3 SS technology installations at manure treatment facilities located in livestock-dense regions to complement the current NDN treatment process could reduce the mineral fertilizer demand with 8% (Vingerhoets et al., 2023). NH_3 SS towers utilize a two-step process, where the initial step involves inducing NH_3 volatilization from the liquid phase into the gas phase by circulating ventilation air over the LF sprayed in a packed tower with inert material. NH_3 in the recirculation air is absorbed by sending the NH_3-rich gas over a counter acid solution in the scrubber unit, which recovers NH_3 in the form of ammonium (NH_4) salts. NH_3 SS installations have employed sulfuric acid (H_2SO_4) and nitric acid (HNO_3) for this purpose, as reported by Brienza et al. (2021), which results in the production of ammonium sulphate (($(NH_4)_2SO_4$, AS) and ammonium nitrate (NH_4NO_3, AN) solutions respectively. The recovery of N in the form of NH_4^+ salts presents a promising opportunity for the integration of manure-derived products into the mineral fertiliser market and thereby combines two key nutrient management strategies: circularity and system's efficiency (Spiller et al. 2024). In particular, these products are likely to conform to the recently established RENURE (REcovered Nitrogen from manURE) product criteria, which dictate the suitability of manure-derived products as mineral fertilisers in Nitrate Vulnerable Zones (NVZs) under the same regulations as synthetic fertilisers (Huygens et al., 2020). These criteria are in line with the existing Fertilising Products Regulation (FPR) (EU) 1009/2019 regulation. To this end, this study seeks to evaluate the economic viability of two scrubbing acids (AS and AN) and assess their compliance with the RENURE and FPR criteria.

As the effluent of NDN treatment tank does not comply with the Flemish discharge limits, transport and land spreading of the produced effluent also contributes to significant part of the manure processing costs (Willeghems et al., 2016). To comply with Flemish discharge limits, Donoso et al. (2015) and Meers et al. (2008) proposed the use of CW structed wetlands as an economically and ecologically beneficial solution for the tertiary treatment of NDN effluent. Therefore, in our study the hypothesis that the implementation of this in-situ post-treatment step can further increase economic profitability and sustainability of manure processing plants as it reduces costs associated to transport and gate fees was assessed.

The primary focus of this study is to conduct an economic analysis of intensive manure management strategies and assess the extent to which the integration of process intensification measures into the swine manure treatment configuration proves economically advantageous as an addition to existing manure treatment facilities. The economic assessment of each configuration (i.e. implementation of an additional measure) was based on technological assessment with steady-state mass balances, including mass, N and P, derived from a monitoring campaign executed for this study. By analysing the mass balances, this study determined operational expenditure (OPEX), capital expenditure (CAPEX), product quality and revenues from the final products for five configurations, which enables the determination of the most economical set of intensification measures for a full-scale pig manure treatment facility.

2. MATERIAL AND METHODS

2.1. Bio Sterco site description and employed nutrient recovery configurations

The Bio Sterco farm (Hooglede, Belgium) has a capacity to raise 454 sows, 5 boars and 5524 and exploits its own manure treatment installation (current maximum capacity 52000 t y^{-1}) which has been operational since 2011. The Bio Sterco farm implemented the following configurations from 2011 to 2022: (i) configuration 1: Centrifugation + NDN, (ii) configuration 2: Centrifugation + NDN with PO aeration, (iii) configuration 3: Centrifugation + NH_3 SS + NDN with PO aeration, and (iv) configuration 4: Centrifugation + NH_3 SS + NDN with PO aeration + CW (Figure 1).

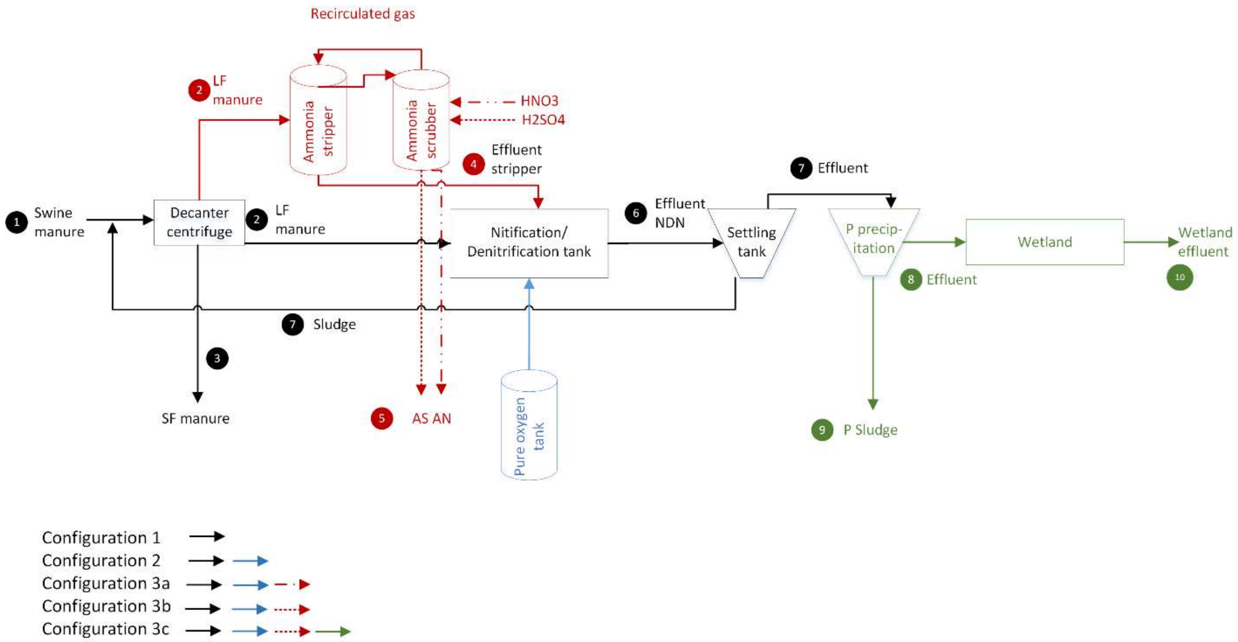


Figure 1 Different configurations of the treatment line of the Bio Sterco manure processing plant (1) Raw swine manure, (2) LF of manure, (3) SF of manure, (4) NDN tank, (5) Sludge, (6) Effluent settling, (7) Effluent SS, (8) NH4+ salt, (9) P-precipitate, (10) Influent of the wetland , (11) Effluent of the wetland

2.1.1 Configuration 1: Centrifugation + NDN

Configuration 1 follows the treatment path of a conventional manure processing system including a centrifuge for mechanical separation, an activated sludge tank focused on NDN and a settling tank to remove the activated sludge from the effluent. In this processing cascade, pig manure is first separated into a LF and SF fraction by centrifugation (Westfalia decanter AG) with a polymer dosing (cationic polyacrylamide) unit to improve separation efficiencies into SF and thereby increase the clarity of the LF. The SF is subsequently composted while the N-rich LF of manure is biologically treated through a NDN system. The NDN system consists of a tank of 3393m^3 with four submerged ejectors aerators developed by the Bio Armor. The oxygenation cycle consists of intermittent aeration (20−30 min oxygenation followed by 15−20 min anoxic period). The NDN effluent is sent to a settling tank of 320 m^3 to separate the produced sludge from the effluent, which are both transported and disposed on land. The NDN unit is the crucial factor determining processing capacity in the whole treatment line. By applying a negative feedback-loop in the NDN unit, the input quantity of LF of manure is regulated based on the prevailing NH_4^+ concentration in the NDN tank which in its turn manages the amount of raw manure processed by the centrifuge. This affects the manure level in the raw manure storage and subsequently the raw manure acceptance rate (Figure 2).

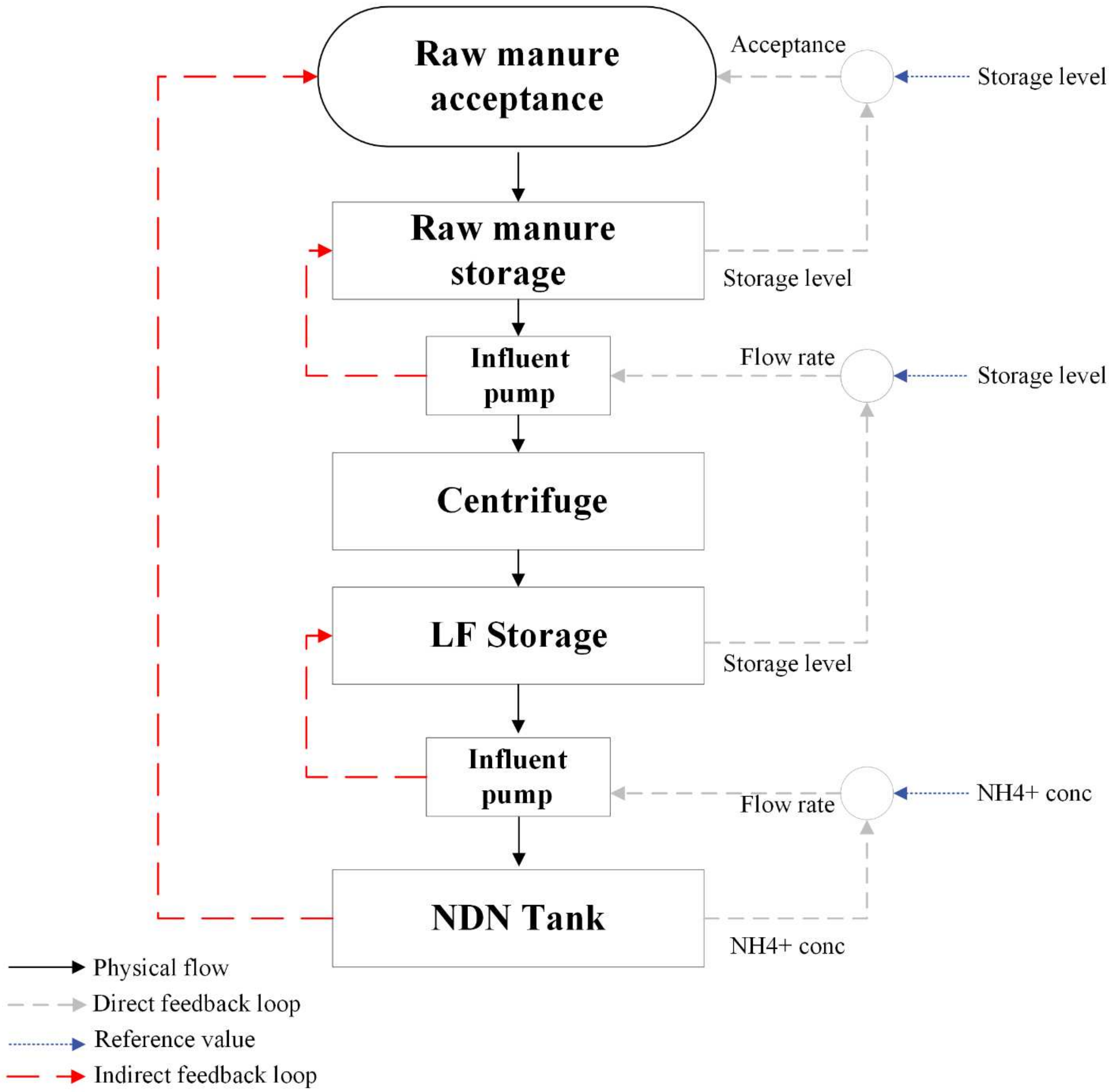


Figure 2 Control system and its feedback loops in a manure treatment plant

2.1.2 Configuration 2: Centrifugation + NDN with PO aeration

Like configuration 1, configuration 2 includes treatment steps such as a centrifuge for mechanical separation, an NDN tank for removing organic compounds and N, and a settling tank to eliminate activated sludge from the effluent. However, the NDN treatment was intensified by implementing PO aeration tank (Air products) with a volume of 22.7 m^3 and a maximal storage pressure of 22.2 bar. The aeration by PO can improve the dissolved oxygen concentration in the wastewater, which reduces the vulnerability to NH_4^+ toxicity and thereby potentially increases the capacity of the NDN system. This treatment set-up was active from 2015 till 2022.

2.1.3 Configuration 3: Centrifugation + NH_3 SS + NDN with PO aeration

In the processing cascade of configuration 3, pig manure is first separated into a LF and SF by centrifugation. A part of the N-rich LF of manure is treated in the NH_3 SS unit to reduce its NH_4-N content before being fed to the NDN system. The NH_3 SS unit, developed by Detricon bvba (Belgium) has a capacity to process 20,000 t y^{-1} and consists of two vertical acrylate stripping columns (2.5m x 3m x 2m) and scrubbing column (2.5m x 3m x 2.5m). The stripping columns are equipped with 9 spraying nozzles and are filled with pall rings as packing material. The ventilation flow of 1,440 m^3 per hour, with an air speed of 0.2 - 0.8 m/s, is divided equally between the two stripping columns. The NH_3-rich air is then directed to the scrubber column, where it encounters a diluted counter acid solution to produce a NH_{4+} salt solution as NH_3 is absorbed by this solution. Two sub-configurations (configuration 3a and 3b) were employed to evaluate the disparity in the usage of counter acids (HNO_3 and H_2SO_4), with NH_3 being recovered as AN or AS, respectively. The resulting stripped LF was then mixed with the non-stripped LF and biologically treated through a NDN system with PO aeration, whereafter the effluent was spread on land. From 2022 onwards, configuration 3 a/b and 4 were active simultaneously. whereby on average almost 79 t d^{-1} was following the treatment line of configuration 3 and 5 t d^{-1} was processed by configuration 4.

2.1.4 Configuration 4: Centrifugation + NH_3 SS + NDN with PO aeration + CW

Configuration 4 followed the same treatment line as in configuration 3b. However, a tertiary treatment line to polish the effluent of the NDN treatment to dischargeable water was added. To reduce the remaining P content of the NDN effluent before entering the wetland, P precipitation is induced by the addition of 4.5 l H_2SO_4 (55%) and 3.5 l of $FeCl_3$ (40%).in a settling tank. The acidification induced by H_2SO_4 establishes more optimal conditions for the formation of $FePO_3$ precipitates, thereby reducing the demand for $FeCl_3$, a crucial factor given the stringent discharge limits for chloride (Cl) in Flanders. Besides Fe precipitation in combination with pH reduction, the precipitation of P can also be achieved by elevating the pH and forming Ca complexes through the introduction of $Ca(OH)_2$. In this study only Fe precipitation in combination with acidification by H_2SO_4 addition was tested as the authors believed that this is the most economic option for the studied treatment line. After P removal, the effluent is polished in a CW of 1268 ha to comply with the surface water discharge norms. The CW system is divided into nine beds/lagoons including one underwater system, one U-turn wetland, four horizontal flow fields, two vertical percolation fields and one lagoon (Figure Supplementary Material (SM) A).

2.2. Sampling and physio-chemical analysis of process streams

We monitored the different configurations of the manure processing plant over the following periods for this study:

- Configuration 1: from February 12th, 2012, to April 21th, 2015
- Configuration 2: from April 16th, 2015, to February 1th, 2022
- Configuration 3a: from February 1th 2022 to March 15th 2022
- Configuration 3b: from March 16th 2022 to March 16th 2023
- Configuration 4: from March 16th 2022 to March 16th 2023

More detailed information on the monitoring periods of the different configurations can be found in SM B. The points sampled and the analysed parameters varied according to sampling period: Monitoring period 1 and 2 focused on Total N (TN) and P for all sample collection points, while also Chemical Oxygen Demand (COD), NH_4-N, NO_3-N, NO_2-N were monitored in the NDN tank. A wider range of parameters were analysed during period 3 for all sampling locations including pH, EC, Dry Matter (DM), Suspended solids (SS), COD, Biological Oxygen Demand (BOD), TN, NH_4-N, NO_3-N, P, K, Sulphur (S), Total Organic Carbon (TOC), Copper (Cu) and Zinc (Zn). The corresponding analysis methods are described in SM C.

2.3. Calculation of separation, recovery and removal efficiencies

Separation efficiency (Se) was determined for the centrifuge and indicates the amount of a component in the solid and liquid fraction respectively compared to the total input of this element (Eq. 1).

$$Se = \frac{X * C_x}{Y * C_y} \qquad \text{Eq. 1}$$

Where Se represents separation efficiency, X (kg) is the amount of outgoing fractions, Cx (g kg^{-1}) represents the concentration of a particular component (e.g. P) in the outgoing fractions, Y (kg) is the quantity of ingoing raw manure and Cy (g kg^{-1}) is the concentration of a a particular component in the ingoing substrate.

Recovery efficiency (Re) was calculated for the NH_3 SS unit and P precipitation unit and stands for the mass of component in the recovered fertiliser (i.e. NH_4^+ salt solution or $FePO_4$ precipitate) respectively as a proportion of the total input from the unit process (Eq. 2).

$$Re = \frac{X * C_x}{Y * C_y} \qquad \text{Eq. 2}$$

The mass of the recovered fertiliser is represented by X (in kg), the concentration of the considered compound in the recovered fertiliser is Cx (in g kg^{-1}), the mass of the influent is Y (in kg), and the compound concentration is Cy (in g kg^{-1}).

Removal efficiencies (Rm) for NDN and CW were determined by the difference between the effluent and influent concentrations of the considered component (Eq. 3).

$$Rm = \frac{C_y - C_z}{C_y} \qquad \text{Eq. 3}$$

Where Cz (mg kg^{-1}) is the concentration of the compound under consideration in the effluent, while Cy (mg kg^{-1}) reports on the concentration of this compound in the influent.

2.4. Economic assessment

This study estimated the CAPEX and OPEX of the different configurations based on quotes from constructors received by the studied manure treatment facility and online market prices. The CAPEX costs are determined by the actual investment costs paid for each technology unit, while the OPEX costs included electricity use, resource consumption (i.e. polymer, H_2SO_4, HNO_3, Methanol and $FeCl_3$), insurance, maintenance and labour. The maintenance of the PO tank is managed by the PO supplier, with associated costs included in the PO's selling price. Consequently, we did not separately account for additional maintenance and labour costs for the treatment facility beyond these considerations.

Table 1 The total investment cost of the treatment plant and maintenance costs per technology unit

	Total investment cost (€)	**Maintenance (€ t^{-1} manure)[a]**	**Labour (FTE[b])**

Centrifuge	254 350	1	0.2
SS unit	225 000	0.1	0.1
NDN + clarifier	1 199 350	1.78	0.4
PO tank	30 000	0.0	0.0
P precipitation + CW	185 000	0.1	0.2

[a]De Vrieze et al. (2019) [b]full-time equivalent

The capital cost of all unit processes was amortised according to Eq. 4 assuming the process unit specific investment cost (C) (Table 1) with an interest rate (r) of 5% depreciation period (n) of 10 years (Møller, 2000).

$$Q = C * \frac{r((1+r)^n)}{(1+r)^n - 1} \qquad \text{Eq. 4}$$

The OPEX costs associated with the use of chemicals, labour, electricity, disposal of by-products and the estimated market value of the RENURE fertilizer products were gathered from online databases and through accounting data from the treatment facility under study (Table 2). As prices of these consumables are volatile, the cost of it was assessed by applying a probabilistic approach through Monte Carlo simulation with uniform distributions created by RAND() function in excel for the data provided in Table 2. The OPEX and CAPEX of each configuration was determined based on the total volume of manure treated and the associated electricity and resource consumption in a certain monitoring period.

Table 2 The considered cost price of the used chemicals, electricity, labour and by-products disposal and the estimated market value for the produced fertilisers.

Chemicals, products or energy	**Standard price (€ t^{-1})**	**Range in price (€ t^{-1})**
Polymer[a]	1,700	1,000 – 4,000
H_2SO_4[a]	120	100 - 300
HNO_3,[a]	200	150 - 400
$FeCl_3$[a]	100	80 - 250
O_2[a]	1.3	0.7 - 3.3
$NH_4(SO_4)_2$ solution[b]	79	0 - 260
NH_4NO_3 solution[b]	210	0 - 400
SF disposal[c]	20	15 - 25
NDN effluent disposal[c]	4	3 - 5

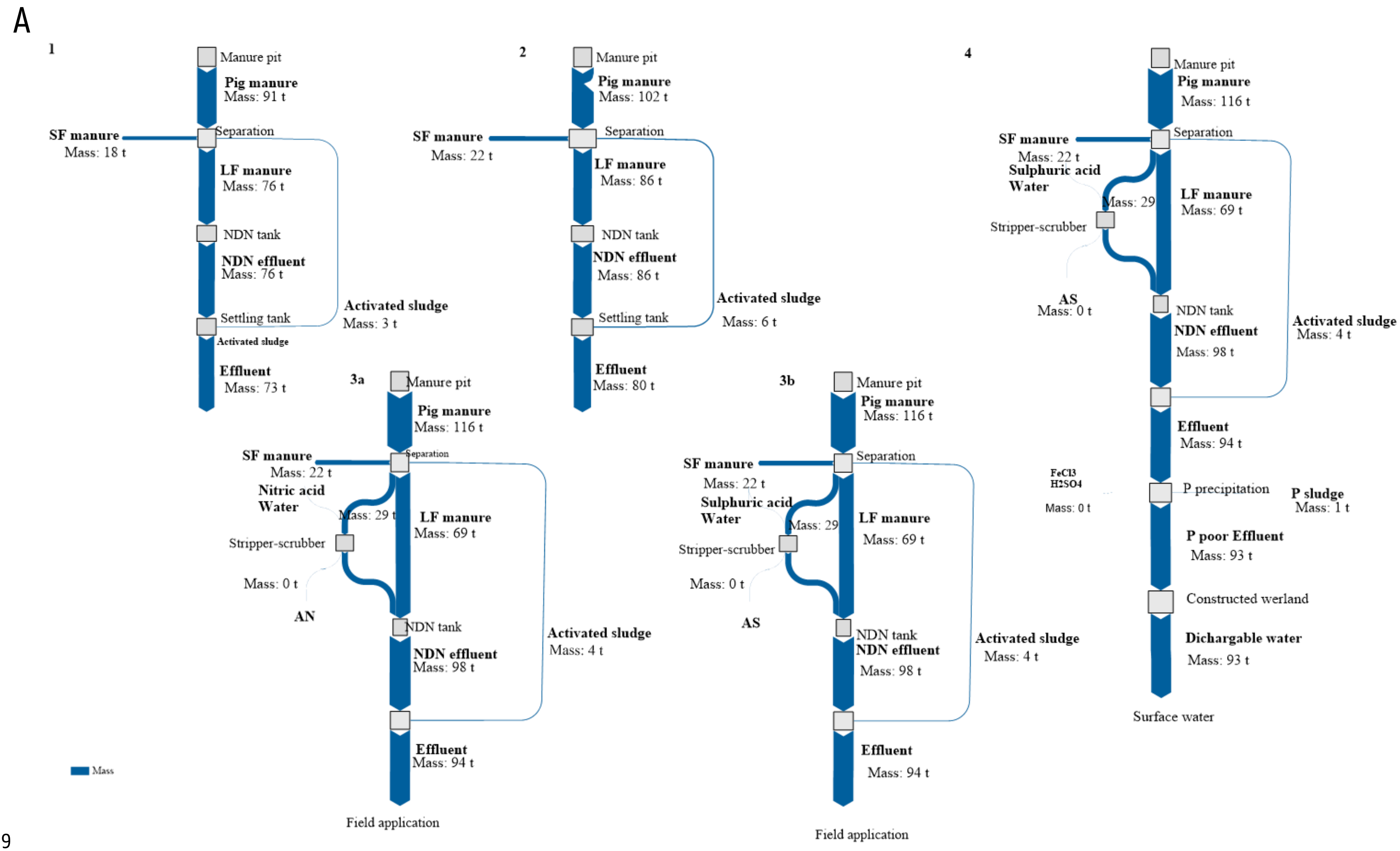
P sludge disposal[c]	20	15 - 25
Electricity[b]	0.15	0.05 - 0.4
Labour[d]	70,000	10,000 – 80,000

Sources: [a] Price quote of retailer [b]Vingerhoets et al. 2023a. , 2023 [c]Derden and Dijkmans, 2020 [d]De Vrieze et al. 2019

3. RESULTS AND DISCUSSION

3.1. Overall mass balance of the different treatment configurations

On average a higher processing capacity was achieved after the implementation of the changes to intensify the recovery process. The daily processing capacity for configuration 1, 2 and 3 was on average 91, 102 and 116 tonne raw manure, respectively (Figure 3A).


A
1
Manure pit
Pig manure
Mass: 91 t
SF manure
Mass: 18 t
Separation
LF manure
Mass: 76 t
NDN tank
NDN effluent
Mass: 76 t
Settling tank
Activated sludge
Activated sludge
Mass: 3 t
Effluent
Mass: 73 t
2
Manure pit
Pig manure
Mass: 102 t
SF manure
Mass: 22 t
Separation
LF manure
Mass: 86 t
NDN tank
NDN effluent
Mass: 86 t
Settling tank
Activated sludge
Mass: 6 t
Effluent
Mass: 80 t
3a
Manure pit
Pig manure
Mass: 116 t
SF manure
Mass: 22 t
Separation
Nitric acid
Water
Mass: 29 t
Stripper-scrubber
Mass: 0 t
AN
LF manure
Mass: 69 t
NDN tank
NDN effluent
Mass: 98 t
Activated sludge
Mass: 4 t
Effluent
Mass: 94 t
Field application
3b
Manure pit
Pig manure
Mass: 116 t
SF manure
Mass: 22 t
Separation
Sulphuric acid
Water
Mass: 29
Stripper-scrubber
Mass: 0 t
AS
LF manure
Mass: 69 t
NDN tank
NDN effluent
Mass: 98 t
Activated sludge
Mass: 4 t
Effluent
Mass: 94 t
Field application
4
Manure pit
Pig manure
Mass: 116 t
SF manure
Mass: 22 t
Separation
Sulphuric acid
Water
Mass: 29
Stripper-scrubber
AS
Mass: 0 t
LF manure
Mass: 69 t
NDN tank
NDN effluent
Mass: 98 t
Activated sludge
Mass: 4 t
Effluent
Mass: 94 t
FeCl3
H2SO4
Mass: 0 t
P precipitation
P sludge
Mass: 1 t
P poor Effluent
Mass: 93 t
Constructed werland
Dichargable water
Mass: 93 t
Surface water
Mass

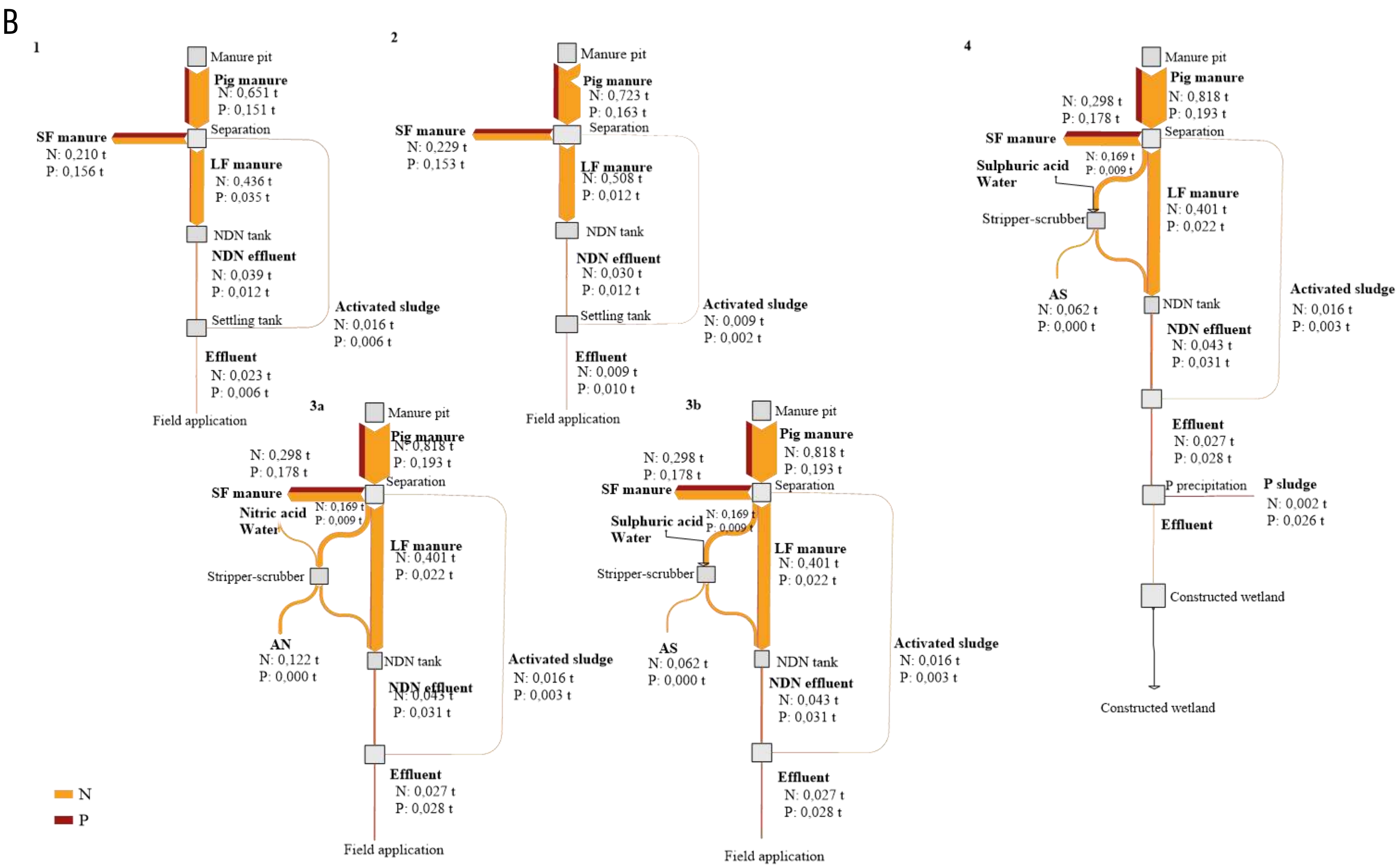


Figure 3 Mass balance for A) overall mass B) nitrogen and phosphorus expressed in t d^{-1} of solid and liquid products within the process flows of the manure treatment plan

The centrifuge unit first separated raw manure into LF and SF, recovering the majority of P in the SF with a separation efficiency above 85% for all configurations. A review of 11 studies evaluating mechanical separation technologies used in manure treatment by Lyons et al. (2021) found that the P separation efficiencies for a decanter centrifuge without chemical addition varied between 30–91%, while screw presses recorded a P separation efficiency of only 4–34%. Polymer addition (cationic polyacrylamide), inducing coagulation between solids, along with the most efficient swine slurry separation technology (i.e. centrifugation), resulted in elevated separation efficiencies compared to the literature findings in our study (Hjorth et al., 2010).The majority of N was found in the LF as separation efficiencies for N did not exceed 35%.

NDN treatment achieved similar removal efficiencies for LF in configurations 1 and 2, but the utilization of PO as the aeration agent resulted in an increased capacity for LF treatment (76 m^3 LF d^{-1} m^{-3} vs 86 m^3 LF d^{-1} $m^{-}3$) with a higher daily N load (0.135 kg N $m^3 d^{-1}$ vs 0.154 kg N $m^3 d^{-1}$) as compared to configuration 1. The suspension was further processed using a settling tank to obtain 3-6 t d^{-1} of activated sludge and 73-80 t d^{-1} of effluent depending on the configuration. The effluent of the settler was sold to neighbouring farmers to bring it to the crop land.

Configuration 3 involved a more complex LF treatment, processing an average of 29 t d^{-1} of LF in the NH_3 SS unit, where a counter-airflow captures the NH_3 of the liquid phase. The recirculation gas - rich in NH_3 - was sent over a NH_3 absorber containing 0.4 t HNO_3 diluted in water to produce 0.7 t AN solution (15% of N) in configuration 3a or 0.2 t H_2SO_4 diluted in water to produce 0.8 t d^{-1} of AS solution (7% of N) in configuration 3b/4. Between 32 and 36% of TN contained in the SS influent was recovered as fertilizer suspension depending on the counter acid used, corresponding to an NH_4–N recovery efficiency of 56 and 57 %. The N-poor stripped LF is then mixed with 69 t d^{-1} non-stripped LF and biologically treated through an NDN system. The NDN unit was fed with 508 kg N d^{-1} of TN, of which 59 % was present in the form of NH_4–N and 41 % in the form of Org-N, and achieved a N removal efficiency of 92% resulting in an effluent containing 43 kg N d^{-1} and 31 kg P d^{-1}. The NDN effluent proceeded to the settling tank where we recovered 4 m^{-1} d^{-1} of activated sludge from the effluent containing 16 kg N d^{-1} and 3 kg P d^{-1}. In configuration 3a/b, the effluent was applied on land, whereas the effluent was further polished in configuration 4. In the polishing step, P precipitation is induced in the effluent by adding 4.5 H_2SO_4 (55%) and 3.5 l of $FeCl_3$ (40%) per tonne effluent before being fed to the CW. P was mostly recovered in the P sludge (26 kg P d^{-1}), while TN, mainly in the form of NH_4–N and NO_3-N, was associated with effluent proceeding to the CW (25 kg N d^{-1}). The CW further removed N, P, BOD and COD by plant uptake, microbiological degradation, and sedimentation, resulting in the effluent meeting surface water discharge criteria (15 mg l^{-1} for N, 1 mg l^{-1} for P, for 250 mg l^{-1} COD, 25 mg l^{-1} for BOD and 35 mg l^{-1} for SS) (VCM, 2021).

3.1.1. NH_3 SS unit

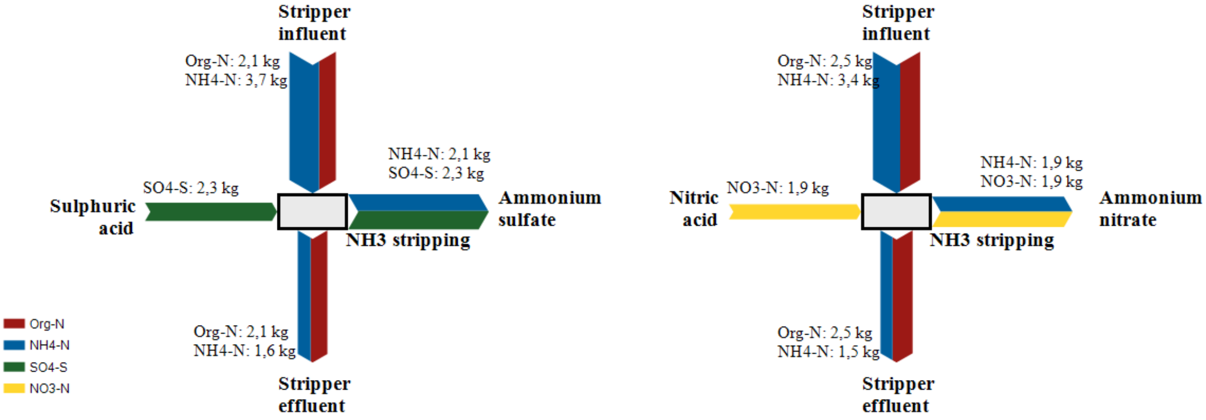


Figure 4 Mass balance for the SS unit using different counter acids; a) sulphuric acid b) nitric acid

The physicochemical characteristics of the influent, effluent, AS and AN can be found in Table 3. Caustic soda or lime were not added during the SS process of LF manure, however, there was an observed increase in pH from 8.1-8.2 to 8.5-8.6 through partially venting the recirculation air which induces CO_2 stripping (Palakodeti et al., 2022). This increase can be attributed to the transfer of the carbonate buffer from LF into the ventilation flow, which occurs at a higher rate than NH_3 due to the volatile nature of CO_2. Vingerhoets et al. (2023a) found that performing CO_2 stripping to increase the pH and thus the NH_3/NH_4 ratio in the LF manure during the SS process is economically more desirable than adding caustic acid. When H_2SO_4 was used as scrubbing agent, slightly higher NH_4^+ removal efficiencies could be achieved as compared to HNO_3. Therefore, the decrease in NH_4^+ content and NH_4-N:TN ratio was less significant for the configuration with HNO_3 as scrubbing agent. Because of the lower density of HNO_3 compared to H_2SO_4, the rate of ventilation flow had to be reduced to prevent HNO_3 from being carried with the ventilation air from the scrubbing unit to the stripping unit which would acidify the LF of manure and reduce stripping efficiency. However, the reduced air flow rate hampers the stripping efficiency because it has a strong impact on mass transfer coefficient, mixing, and gas-liquid interfacial area, which are important parameters determining gas-liquid transfer rate of NH_3. For example, Liu et al. (2015) showed that increasing the air flow rate from 60 m^3 h^{-1} m^{-3} to 840 m^3 h^{-1} m^{-3} enhances the hourly stripping efficiency from 8.6 to 86.4% when performing a stripping experiment on pig urine at a temperature of 50°C and an increased pH of 10. However, further increasing the air flow rate above 600 m^3 h^{-1} m^3 showed reduced benefits on NH_3 removal rates.

Table 3 Composition (mean ± standard deviation) of influent NH_3 SS (IS), effluent NH_3 SS (ES), ammonium nitrate (AN) solution, and ammonium sulphate (AS) solution for configuration 3a and 3b

	Unit	Configuration 3a			Configuration 3b		
		IS	ES	AN	IS	ES	AS
pH		8.2 ± 0.1	8.6 ± 0.1	6.0 ± 0.6	8.1 ± 0.2	8.5 ± 0.1	5.7 ± 1.1
EC	mS cm^{-1}	33.2 ± 1.5	31.1 ± 1.3	178 ± 13	31.3 ± 2.3	30.1 ± 2.3	242 ± 27
TOC	g kg^{-1}	n.a.	n.a.	0.12 ± 0.03	n.a.	n.a.	0.82 ± 0.11
COD	g kg^{-1}	48.6 ± 6.1	44.8 ± 4.7	0.21 ± 0.04	55.4 ± 2.8	52.1 ± 2.2	0.32 ± 0.09
TN	g kg^{-1}	5.91 ± 0.26	3.97 ± 0.45	153.1 ± 26.4	5.82 ± 0.4	3.73 ± 0.51	74.4 ± 8.4
NH_4-N	g kg^{-1}	3.34 ± 0.35	1.46 ± 0.21	76.2 ± 18.1	3.73 ± 0.3	1.61 ± 0.38	74.3 ± 8.1
NO_3-N	g kg^{-1}	n.a.	n.a.	77.4 ± 15.0	n.a.	n.a.	n.a.
P	g kg^{-1}	0.54 ± 0.09	0.52 ± 0.06	0.06 ± 0.01	0.31 ± 0.11	0.32 ± 0.09	0.05 ± 0.01
Cu	mg kg^{-1}	n.a.	n.a.	1.2 ± 0.7	n.a.	n.a.	2.2 ± 0.9
Zn	mg kg^{-1}	n.a.	n.a.	3.4 ± 1.3	n.a.	n.a.	5.5 ± 2.4

Figure 4 shows the mass balance of N in the NH_3 SS unit using 60% HNO_3 solution and 98% H_2SO_4 as an absorption agent. When treating 1 t of LF, 24.5 kg of AN (15.3 % N) is produced wherein 1.9 kg of NH_4-N is recovered which only amount for half of the TN content in the produced AN solution as the addition of 14.3 kg of 60% HNO_3 to scrubbing solution contributes the remaining 50% in the form of NO_3-N. The treatment of 1 t LF with scrubbing solution containing 7.1 kg of H_2SO_4 resulted in the production 28.4 kg of 7.4 % N AS solution, which contains 2.1 kg of recovered NH_4^+-N. The use of HNO_3 instead of H_2SO_4 results in a slightly lower recovery ratio of TN from the influent in the form of AS and AN solution. However, the organic N content remained the same as only mineral N is recovered during the SS process. It was found that higher N concentrations could be achieved in the recovered NH_4NO_3 solution as compared to the $(NH_4)_2SO_4$

solution because of the low solubility of $(NH_4)_2SO_4$ resulting in crystallisation. A higher N concentration is more favourable as it has agronomical benefits and reduces costs of transportation and land spreading (Sigurnjak et al., 2019).

A considerable number of experiments to assess the efficacy of the implementation of NH_3 SS technology to recover N from (digested) LF of pig manure has been performed at laboratory, pilot and full-scale over the last decades. Brienza et al. (2023) recovered on average 22% of N in the form of AN (81 g kg^{-1} TN) when stripping LF of digestate at ambient temperature and low pH (8.0) in a pilot-scale installation for 2 hours. A range of temperatures (55-65°C) were tested in series of trials by Pintucci et al. (2017) for a low pH (7.8) and found NH_3 removal rates ranging from 28 to 46%. Baldi et al. (2018) achieved an high removal efficiency of 62% NH_4^+ in a stripping experiment on digestate by subjecting it to a temperature of 48°C and pH of 9.5 over a period of 2 hours, while Bolzonella et al. (2018) recovered 22% of TN in the form of $(NH_4)_2SO_4$ solution (26 g kg^{-1} TN) when stripping a mixture of digested swine and cow manure at pilot scale. As compared to other studies, the efficiency found in our study (32 - 36% of TN and 56 – 67 % of NH_4^+-N) was at the higher end of the spectrum as it was performed at both high pH (8.5 – 8.6) and temperature (50 °C). Also, high N concentrations in the NH_4^+ solutions were achieved in our study compared to literature results.

Assuming the quality criteria for manure derived RENURE products and liquid inorganic macronutrient fertilisers set by European FPR are met, the net costs of the proposed N recovery pathway were calculated by considering the sale of the produced AS and AN solution at current N fertilisation prices. We found that the concentration of TOC was below the prescribed 1% while complying with the minimum required TN content, as shown in Table 4. Moreover, the AS and AN solutions satisfied the maximal TOC:TN and the mineral N:TN ratio requirements. The concentration of hazardous elements Cu and Zn in the NH_4^+ solutions also remained well below the RENURE and FPR requirements. Therefore, it can be concluded that the AS and AN solution obtained during the SS process can be regarded as a substitute for mineral N, which confirms the pricing assumption.

Table 4. Composition requirements for the different fertilisers products defined by the Fertilising Product Regulation (EU) 1009/2019 and Joint Research Centre (JRC) RENURE products (Huygens et al., 2020).

Fertiliser type	TN (g kg^{-1} FW)	TOC (g kg^{-1} FW)	TOC:TN	mineral-N_l:TN (%)	Cu (mg kg^{-1} DW)	Zn (mg kg^{-1} DW)
PFC 1(C)(I)(b)(i) (Fertilising Product Regulation)	≥ 50	≤ 10			≤ 600	≤ 1500
RENURE product (JRC)			≤ 3*	≥ 90*	≤ 300	≤ 800

Despite the NH_4^+ solutions recovered less than 1% of the COD embedded in the influent LF, a reduction in of 8% in the COD content of LF was found during the SS process. This trend was also described by Brienza et al. (2023) who found a loss in COD content of 13% when HNO_3 was used as absorbent, and Bonmatı and Flotats (2003) who recorded COD losses above 5% when using H_2SO_4 as scrubber acid. Therefore, it can be assumed that HNO_3 nor H_2SO_4 are not able to absorb volatile organics which stress the need for additional air treatment to reduce impact on the environment.

3.1.2. NDN system and settling tank

Figure SM D shows the long-term measurement data of the NDN tank, while Figure 5 presents the average physicochemical characteristics of the effluent for the different configurations. The N removal efficiency in the NDN tank was on average 91% of N for configuration 1, 94% of N for configuration 2 and 92% of N for configuration 3. Despite the similar N removal efficiencies observed for all configurations, the N effluent concentration varied depending on the configuration. The NH_4^+ concentration in the NDN tank during configurations 2 and 3 were considerably lower as compared to configuration 1. For all configurations, NH_4^+ concentrations in the effluent increased during the months August-February as the land spreading limitation increased the loading rate during these months. The high loading rates are causing elevated NH_4^+ concentration which eventually led to the free ammonia (FA) toxicity effects inhibiting NH_3 and NO_2^- oxidation (Nehmtow et al., 2016; Tian et al., 2013). The inhibition of this process causes the accumulation of NO_2^- during these months. When NH_4^+ concentration exceeds 160 mg/l, the negative feedback loop was triggered reducing the LF input into the NDN tank which enables the process to restore its concentration of NH_4^+ into healthy ranges. This negative feedback loop limits the process capacity of configuration 1 to 76 m^3 LF d^{-1} m^{-3} (0.135 kg N m^{3}d^{-1}), while a capacity of 86 m^3 LF d^{-1} m^{-3} (0.156 kg N m^{3}d^{-1}) could be reached after the implementation of PO aeration. Despite increasing the N loading rates in configuration 2, the number of occasions where NO_2^- accumulation occurred diminished compared to configuration 1 (9 vs 20 per year) indicating on an elevated nitrification rate in the NDN tank through the usage of PO. A similar N load was treated in configuration 3 (0.157 kg N m^3d^{-1}) compared to configuration 2 (0.156 kg N m^3d^{-1}), but an increased quantity (96 m^3 LF d^{-1} m^{-3}) could be treated because of the reduced N content in the stripped LF.

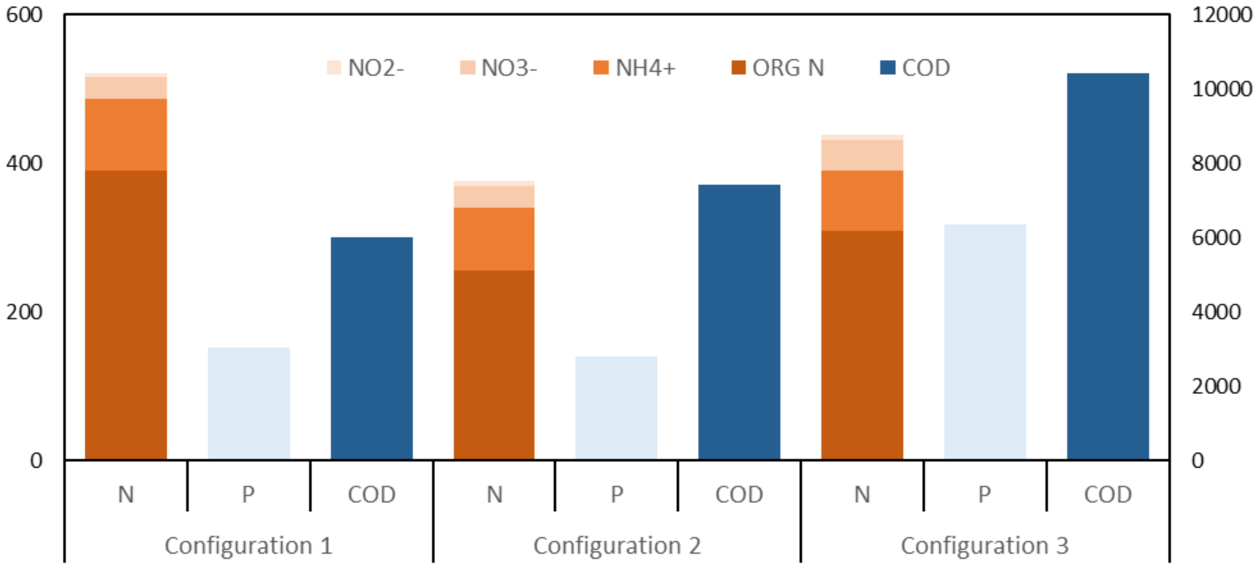


Figure 5 Average concentration of N, P and COD compounds in the NDN tank during the monitoring configurations of the different configurations.

Corbala Robles et al. (2018) reported on a typical wastewater treatment plant treating the LF of pig manure after centrifugation in Flanders with a tank volume of 2846 m^3 which could process on average 58 m^3 LF manure per day with an associated N loading rate of 0.09 kg N m^3 d^{-1}. Smet et al. (2003) evaluated 14 different biological manure treatment installations according to the Trevi concept with a total treatment capacity of 300,000 m^3 y^{-1} and found that the operation of the biological treatment installations allows a loading rate of 0.13 kg N m^3 d^{-1}. The average loading rate sustained by the NDN tank during configuration 1 in our study equalled 0.14 kg N m^3d^{-1} and is thus similar as the values found in literature. A review of 25 studies by Skouteris et al. (2020) showed that the replacement of ambient air by PO as aeration agent could enhance the treatment capacity by increasing the oxygen transfer rate through an elevated partial pressure of oxygen, especially for high strength wastewaters with high Mixed Liquor Suspended Solids (MLSS). Similar conclusions were yielded by Rodríguez et al. (2012) who compared the use of PO and air on the nitrification rate in a pilot-scale MBR system used for wastewater treatment and found that aeration by PO could enhance the nitrification rate with 8-13%. As the nitrification rate increases, it effectively diminishes the NH_4^+ concentration within the NDN tank, preventing the accumulation of NH_4^+ and thereby mitigating the inhibitory effects of FA. This inhibition generally impedes the entirety of the nitrification process when FA concentrations exceed 150 mg N l^{-1} (Elawwad, A. 2018). Conversely, the inhibition of NO_2^- oxidation commences at FA levels surpassing 2.8 mg N l^{-1} (Jubany et al. 2008). Hawkins et al. (2010) conducted a comprehensive review of 15 studies, revealing that the FA concentration threshold at which NO_2^- oxidation begins to be inhibited varies significantly across studies, yet predominantly initiates at FA concentrations of 2-3 mg N l^{-1}. This finding is consistent with observations of approximately 160 mg $l^{-1}$of total NH_4-N present in the NDN tank under standard operational conditions.

These findings support the increased processing capacity found for configuration 2. When the LF was partially pretreated by NH_3 SS, an increased volume of LF could be processed by the NDN tank. However, only a slight difference was found in processed N load between configuration 2 and 3, which could be due to the more favorable COD:N ratio (Phanwilai et al., 2020). Lower COD concentrations were found for configurations 1 and 2 as compared to configuration 3 due to the higher COD loading rates of configuration 3. In addition to augmenting the treatment capacity of the NDN system, N recovery by SS in configuration 3 has the potential to decrease N_2O emissions per m^3 of LF treated. Specifically, the N_2O emissions are estimated to be 0.057 kg N_2O-N in configuration 3, compared to 0.063 kg N_2O-N per m^3 of LF treated in configuration 1, assuming N_2O emissions account for 1.1% of the N load in the NDN (de Haas and Andrews, 2022), The conventional N_2O emission factors are applicable on concentrated piggery wastewaters (Ravi et al., 2023). However, the reduction in NO_2^- accumulation in configuration 3 is expected to contribute to an even more pronounced decrease in N_2O emissions, given the strong association between NO_2^- accumulation and N_2O emission rates (Van Hulle et al., 2011). The NDN effluent was further processed by a clarifier, which resulted in the production of 3-6 t d^{-1} activated sludge, containing 9-16 kg N d^{-1} and 2-6 kg N d^{-1}, and 73-94 t d^{-1} clarified effluent, containing 23-26 kg N $d^{-1}$and 6-28 kg P d^{-1}. As the settled activated sludge contains a considerable amount of water, it is looped to the centrifuge for further processing.

3.1.3. P precipitation and wetland

Table 5 presents the average influent and effluent concentrations of the P precipitation unit and effluent of the wetland. The results illustrate that the P precipitation unit achieves a high P recovery efficiency of 91.5 % by inducing P precipitation as $FePO_4$ though the addition of 4.5 H_2SO_4 (55%) and 3.5 l of $FeCl_3$ (40%). In the sedimentation tank, also the DM content was partly recovered, 19 and 7 % respectively. On average 0.01 t of sludge, recovering almost 10% of N and 92% of P, and 0.99 t P poor effluent, containing 0.26 kg of N, 0.03 kg of P and 10.9 kg of COD, were produced during the treatment of 1 tonne effluent. Meers et al. (2006) achieved a removal efficiency of 39, 88 and 95% of P when applying 1, 3 and 5 l of $FeCl_3$ to LF of pig manure treated by NDN with a P content of 332 mg P l^{-1}. Decreasing pH below 8 by the addition of H_2SO_4 increases the P removal rates for similar $FeCl_3$ dosage. Through the acidification of the NDN effluent in our study, high P removal efficiencies could be achieved for low $FeCl_3$ dosages which is crucial to meet the stringent local Cl discharge limits of 1000 mg Cl^- l^{-1} (VCM, 2021).

Table 5 Composition (mean ± standard deviation) of influent P precipitation unit (I-PU), effluent P precipitation unit (E-PU), and effluent constructed wetland (E-CW)

		I-PU	E-PU	E-CW
pH		8.1 ± 0.1	7.9 ± 0.2	7.9 ± 0.1
EC	mS cm^{-1}	33 ± 1.5	31 ± 1.3	9.8 ± 1.8
DM	g kg^{-1}	14 ± 1.7	11 ± 0.8	0.12 ± 0.03
SS	g kg^{-1}	0.56 ± 0.14	0.52 ± 0.21	0.01 ± 0.00
COD	g kg^{-1}	12 ± 2.2	8.7 ± 1.8	0.12 ± 0.07
BOD	g kg^{-1}	5.9 ± 1.7	5.8 ± 2.1	0.01 ± 0.00
TN	mg kg^{-1}	283 ± 153	258 ± 45	13 ± 5.9
NH_4-N	mg kg^{-1}	81 ± 100	75 ± 33	1.5 ± 0.2
NO_3-N	mg kg^{-1}	401 ± 70	37 ± 10	5.7 ± 1.4
P	mg kg^{-1}	304 ± 45	28 ± 8.9	0.32 ± 0.09

After precipitation, the effluent flows into the CW. The average influent and effluent concentrations of the CW can be found in Table 5. As on average 4.6 t d^{-1} of effluent were treated, the N and P loading rates equalled 0.94 g N m^{-2} d^{-1} and 0.11 g N m^{-2} d^{-1}, wherefore a removal efficiency of 95% of N, 99% of P and 99% of COD is obtained. Meers et al. (2006) conducted bench experiment with a corresponding loading rate of 0.23 g N m^{-2} d^{-1}, 0.27 g P m^{-2} d^{-1} and 3.2 g COD m^{-2} d^{-1} and achieved removal efficiencies between 73%–83% for N, 71% - 98% for P and 64 – 75% for COD, while Meers et al. (2008) reported on a CW of 4500 ha that could sustain a nutrient load of 0.75-1.22 g N m^{-2} d^{-1} and 0.04 g P m^{-2} d^{-1} with a removal efficiency of 96% and 99%, respectively. Lee et al. (2014) loaded 0.84 g N m^{-2} d^{-1} into a CW (4492 ha) in the form of piggery effluent and achieved a removal efficiency of 55% N. The plant uptake was estimated by the crop cut method (Sapkota et al. 2016) to account for 17 % of N and 26% of P removal, which is considerably lower than the values found by Meers et al. (2006), Meers et al. (2008) and Lee et al. (2014) as the biomass yield remained under expectations.

However, COD, BOD and N were mainly removed by microbiological degradation and denitrification processes resulting in C and N losses to the atmosphere as CO_2 and N_2, while the remainder of the P removal is attributed to sorption by substrate and sedimentation. Due to the finite capacity of these P removal mechanisms, sorption and sedimentation will not sustain long-term P removal (Meers et al., 2008). Therefore, it is crucial to establish a correlation between the loading rates of P and its uptake by the biomass that can be harvested to maintain the long-term effectiveness of a system in removing P. By reducing the P load through the introduction of a P precipitation before the effluent enters the CW, an elevated amount of effluent can be treated without compromising the sustainable operation of the CW.

The regional discharge criteria limit the effluent concentration to 15 mg l^{-1} for N, 1 mg l^{-1} for P, for 250 mg l^{-1} COD, 25 mg l^{-1} for BOD and 35 mg l^{-1} for SS (VCM, 2021). Figure SM E illustrates the effluent concentration of the tertiary treatment system over time. The CW effectively removed P, COD, BOD and SS during the entire year as the effluent concentration were continuously below discharge limits, while N was above the discharge limits during the winter period. Therefore, the results of this study indicated that N is the limiting component determining the CW operational capacity while being in correspondence with the legal discharge limits.

3.2. Energy balance

The energy balance was computed for each configuration based on the total consumed energy and amount treated during the period the respective configuration was tested. Table 6 shows the average consumed energy per tonne of raw manure treated. The electricity required by the manure processing ranged between 14.8-16.4 kWh t^{-1} raw manure depending on the used configuration. For conventional processing (configuration 1), NDN treatment was the most energy-intensive step, followed by the centrifugation. Replacing air by PO in the oxygenation step resulted in a significant reduction of energy requirement for the NDN process. When installing a SS unit before the NDN treatment, the costs by oxidation requirement even decreased further until 11.4 kWh t^{-1} raw manure.

However, N recovery by the NH_3 SS unit has an electrical energy requirements of 12.4 kWh t^{-1} LF manure stripped, which equals 3.0 kWh t^{-1} raw manure treated. P precipitation and CW consumed about 0.3 kWh t^{-1} raw manure treated. Similar energy requirements for centrifugal separation were reported by Willeghems et al. (2016), which are considerably higher than separation by a screw press. Tampio et al. (2016) provided a review study reporting an energy requirement between 0.8 and 28 kWh kg^{-1} N for N recovery through NH_3 SS of a wide range of substrates including manure, digestate and urine. As circa 2 kg N t^{-1} LF manure is recovered by the SS unit in our study, the energy requirement for AS and AN production are in accordance with data provided by literature. According to Brienza et al. (2023), the energy consumption of the SS unit ranged between 6.4-13.6 kWh kg^{-1} N recovered from digestate, while Bolzonella et al. (2018) and Brienza et al. (2021) reported 12.0 and 3.8-5.0 kWh kg^{-1} N recovered, respectively. Different values for energy consumption of NDN treatment are stated in literature. Corbala Robles et al. (2018) reported 17 kWh t^{-1} LF of swine manure for aeration and mixing, whereas Willeghems et al. (2016) mentioned two different energy consumption for the two systems used in Flanders, namely 16 kWh t^{-1} LF of manure (Bio Armor system) and 17 kWh t^{-1} of LF manure (Trevi). The reduced energy consumption when using PO aeration can be explained by the increased oxygen transfer efficiency for PO aeration which increases the aeration efficiency (kg O_2 kWh^{-1}). As nitrification demands an oxidizing power of 4.57 g O_2 per g of N oxidized, the reduction in N content in configuration 3 through the SS unit further reduces the energy demand per tonne LF treated.

Table 6 Energy consumption (kWh t^{-1} raw manure) per process technology unit

	Configuration				
	1	2	3		4
			A	B	
Separation	1.9	1.9	1.9	1.9	1.9
NH_3 SS			3.0	3.1	3.1
Thermal			1.7	1.7	1.7
Non-thermal			1.3	1.4	1.4
NDN tank and settler	12.9	12.4	11.4	11.4	11.4
P precipitation and CW					0.3
Total	14.8	14.3	16.3	16.4	16.7

3.3. Economic assessment of intensified manure processing

The different configurations were economically evaluated to find the effect of intensification of the processing configuration on the total costs associated with raw manure treatment. The cost benefit analysis for the different configurations is illustrated in Figure 6. CAPEX compromised capital costs, including technology cost and land usage, for centrifuge, NDN tank and settling tank in all configurations, while amortisation costs associated with the installation of a pure liquid oxygen oxygenation tank, NH_3 SS unit and CW were considered depending on the configuration. The amortisation costs associated with the centrifuge unit varied between 0.7 and 1.0 €, while the amortisation costs of the NDN treatment including the settling tank, depending on the treatment capacity of the configuration, amounted between 3.6 - 4.7 € t^{-1} with an additional cost of 0.09 - 0.1 € t^{-1} for the PO tank. Implementing the NH_3 SS unit involved a cost of € 0.23 M which resulted in a cost of 1.8 € t^{-1} LF manure or 0.7 € t^{-1} raw manure treated. Considering the found treatment capacity of the CW, the tertiary treatment via P precipitation and CW amounted for 2.2 € t^{-1}LF or 1.8 € t^{-1} raw manure.

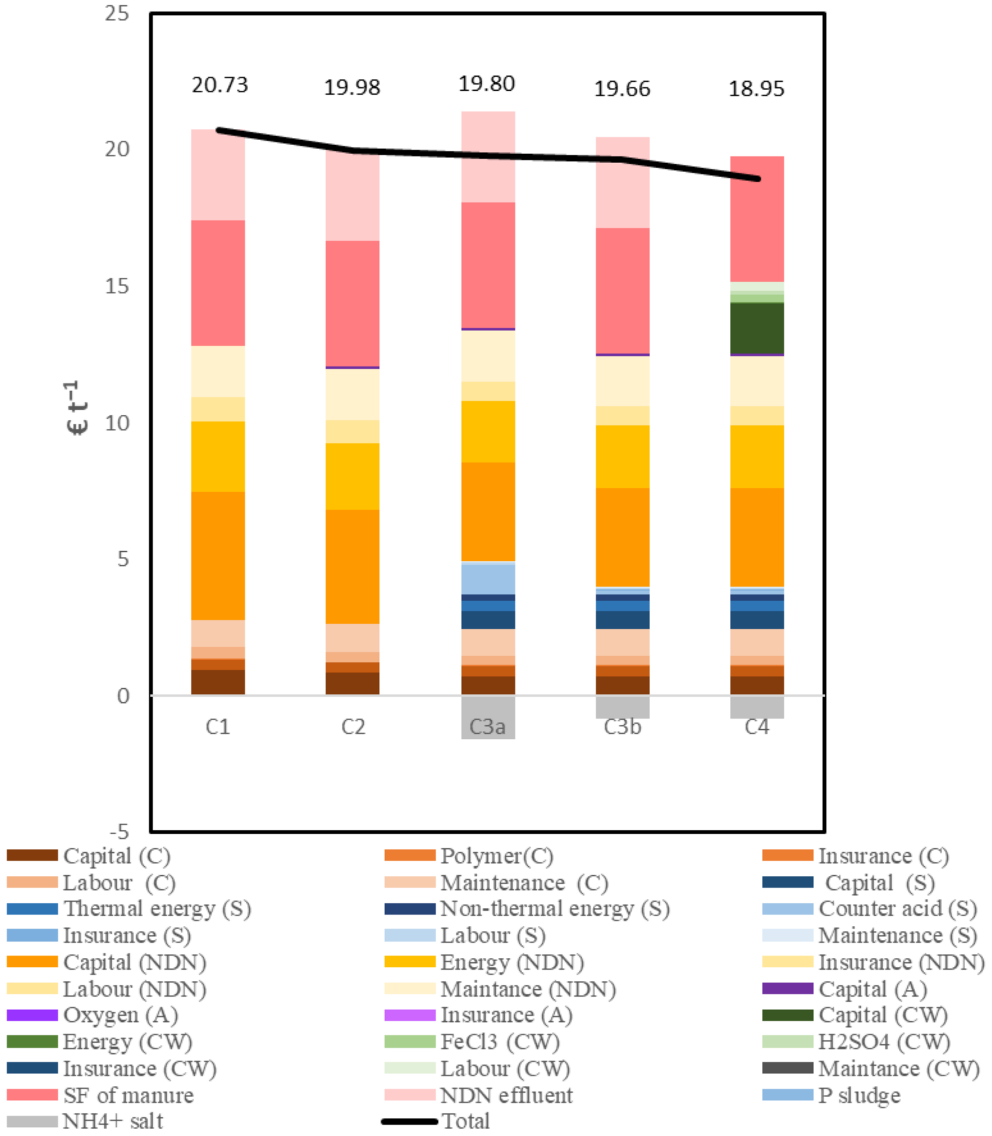


Figure 6 Economic results for the different configurations, calculated for the treatment of 1 tonne of raw manure. (C= Centrifuge, S= SS unit, NDN= Nitrification denitrification unit, A= Aeration tank and CW= Constructed wetland) (C1=Configuration 1, C2 =Configuration 2, C3a=Configuration 3a, C3b=Configuration 3b, C4=Configuration 4)

The OPEX comprised electrical energy requirements, chemicals, disposal, insurance, maintenance, and labour costs. The insurance costs for the separation, NDN and settling unit decreased per tonne raw manure as the capacity of these units increased while these costs remained the same. However, the alternative configuration implicated additional costs associated with insurance, maintenance, and labour for the PO tank, NH_3 SS unit, P precipitation unit and CW depending on the configuration. Because of the lower aeration requirements of configuration 2 compared to configuration 1, a lower cost for energy consumption was obtained. The incorporation of the NH_3

SS step in configuration 3 resulted in a further decrease in energy consumption during NDN processing, however, this was not enough to compensate additional energy requirements of the NH_3 SS unit itself resulting in an increased cost for energy in configuration 3. To recover N in configuration 3a and 3b, 6 kg of HNO_3 and 2.3 kg of H_2SO_4 was used amounting in a cost of 1.1 and 0.2 € t^{-1}, respectively. The N recovery via NH_3 SS unit provided an economic benefit of 0.8 – 1.6 € t^{-1} depending on the choice of counter acid as it can be sold as replacement for synthetic mineral N fertilisers, while the implementation of a CW saves farmers the costs associated with effluent disposal.

The total costs for pig manure treatment ranged between 18.9 - 20.7 € t^{-1}. The gradual implementation of additional treatment units could reduce the cost; PO oxygenation led to a cost reduction of almost 4%, the implementation of a NH_3 SS could further reduce the costs with 1 to 2% depending on counter acid used, while the expansion of the treatment line with a CW accounted for the lowest total cost (-9% compared to reference situation of using configuration 1).

Spiller et al. (2022) reported in review study on conflicting results regarding the economic feasibility of a direct N recovery via technologies such as NH_3 SS and membrane filtration. This is because the traditional "indirect" reuse loop, which involves reactive N removal by NDN in waste streams as N_2 and refixation via the Haber-Bosch process, appears to be the more financially viable in many cases. Derden and Dijkmans (2020) estimated the overall treatment costs involving separation and NDN for raw manure at 19 € t^{-1}, while the implementation of N recovery via NH_3 SS is expected to result in a cost increment of 10 € t^{-1}. A similar conclusion was made by De Vrieze et al. (2019) who found a cost increase of 51% when involving NH_3 SS as compared to the conventional treatment configuration. However, both studies did not consider the synergetic effects between NH_3 SS and NDN processing such as capacity increase and reduced oxygen requirement.

The Monte Carlo simulations demonstrated a different impact of uncertainty in resource pricing for the different configurations (Figure 7). For configuration 1 and 2, an increase of 42 and 43% in costs was observed at 80% probability as these configurations have a lower dependence on the external resources (including energy, counter acid, with varying prices and uncertain revenue from the produced NH_4^+ salts). Due to the strong dependency of configuration 3a-c on resource pricing, the increase in overall costs at 80% probability was more pronounced (45-46%). The uncertainty in the market price of counter acid and market value of NH_4^+ salt solution mainly impacted the total net cost of configurations 3. The impact of the NHO_3 price and market value of AN solution was stronger compared to the impact of H_2SO_4 and AS pricing, due to the higher acid consumption, revenue, and price range. The additional consumption of H_2SO_4 and $FeCl_3$ in configuration 4 compared to configuration 3b was reflected in the fact that the elevation of total net cost was higher at 80% probability (+ 0.7 %). Although the higher electricity consumption depicted in configuration 3 had a noticeable impact on the overall net cost, its effect was significantly less than that of the market price of chemicals. The sensitivity analysis of configuration 3a-c illustrates that process intensification can be highly economically beneficial compared to the conventional situation when high market values for the recovered NH_4^+ salts prevail, but can also turn out more expensive when resource costs for the use of chemical and energy are high and the market values of the produced products are not favourable.

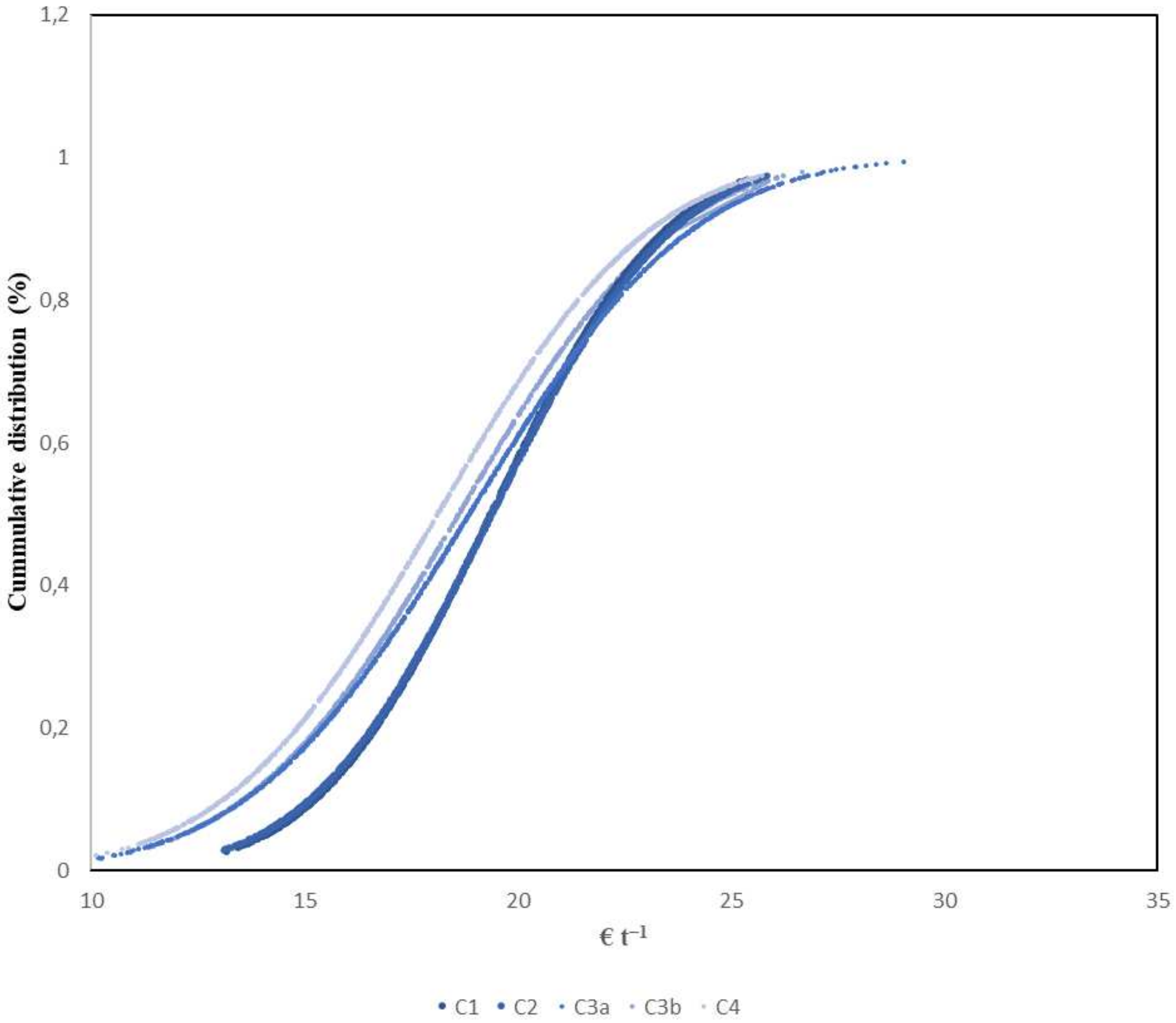


Figure 7 Monte Carlo probability distributions for the sensitivity analysis for the different manure processing configuration (C1=Configuration 1, C2 =Configuration 2, C3a=Configuration 3a, C3b=Configuration 3b, C4=Configuration 4)

4. CONCLUSION

This study presents a comparative techno-economic assessment of different configurations studying the effect of the implementation of process intensifying measures along the swine manure treatment line, including PO aeration, pretreatment by NH_3 SS and tertiary treatment by a CW. The techno-economic assessment was performed based steady-state mass balances derived from long-term monitoring campaigns at a full-scale pig manure treatment facility focusing on total mass, N and P. Inserting additional treatment steps to the treatment train was found to have a beneficial effect on the overall treatment costs as it enhances the processing capacity of the conventional configuration, reduces costs of by-product disposal and generates addition revenue by producing high quality fertilising products. The usage of PO oxygenation reduces the total with almost 4%, the addition of a SS unit further reduces the costs with 1 to 2% depending on counter acid used and the implementation of a CW even reduces the total costs further with 4%.

REFERENCES

Baldi, M., Collivignarelli, M., Abbà, A., & Benigna, I. 2018. The Valorization of Ammonia in Manure Digestate by Means of Alternative Stripping Reactors. Sustainability. 10(9), 3073–. https://doi.org/10.3390/su10093073

Bolzonella, D., Fatone, F., Gottardo, M., & Frison, N. 2018. Nutrients recovery from anaerobic digestate of agro-waste: Techno-economic assessment of full scale applications. J. Environ.l Manage. 216, 111–119. https://doi.org/10.1016/j.jenvman.2017.08.026

Bonmatí, A., & Flotats, X. 2003. Air stripping of ammonia from pig slurry: characterisation and feasibility as a pre- or post-treatment to mesophilic anaerobic digestion. Waste Manage. 23(3), 261–272. https://doi.org/10.1016/S0956-053X(02)00144-7

Brienza, C., Donoso, N., Luo, H., Vingerhoets, R., de Wilde, D., van Oirschot, D., Sigurnjak, I., Biswas, J. K., Michels, E., & Meers, E. 2023. Evaluation of a new approach for swine wastewater valorisation and treatment: A combined system of ammonium recovery and aerated constructed wetland. Ecol. Eng. 189, 106919- .https://doi.org/10.1016/j.ecoleng.2023.106919

Brienza, C., Sigurnjak, I., Meier, T., Michels, E., Adani, F., Schoumans, O., Vaneeckhaute, C., & Meers, E. 2021. Techno-economic assessment at full scale of a biogas refinery plant receiving nitrogen rich feedstock and producing renewable energy and biobased fertilisers. J. Clean. Prod. 308, 127408–127408. https://doi.org/10.1016/j.jclepro.2021.127408

Campbell, B. M., Beare, D. J., Bennett, E. M., Hall-Spencer, J. M., Ingram, J. S. I., Jaramillo, F., Ortiz, R., Ramankutty, N., Sayer, J. A., Shindell, D. 2017. Agriculture production as a major driver of the Earth system exceeding planetary boundaries. Ecol. Soc. 22(4), art8. https://doi.org/10.5751/ES-09595-220408.

Commission Regulation (EU), 2011. No 142/2011 of 25 February 2011 implementing Regulation (EC) No 1069/2009 of the European Parliament and of the Council laying down health rules as regards animal by-products and derived products not intended for human consumption and implementing Council Directive 97/78/EC as regards certain samples and items exempt from veterinary checks at the border under that Directive. Orkesterjournalen L, 54 (2011), pp. 1-254 26.2

Coppens, J., Meers, E., Boon, N., Buysse, J., Vlaeminck, S. E. 2016. Follow the N and P road: High-resolution nutrient flow analysis of the Flanders region as precursor for sustainable resource management. Resour. Conserv. Recycl. 115, 9–21. https://doi.org/10.1016/j.resconrec.2016.08.006.

Corbala-Robles, L., Sastafiana, W. N. D., Van linden, V., Volcke, E. I. P., & Schaubroeck, T. 2018. Life cycle assessment of biological pig manure treatment versus direct land application − a trade-off story. Resour., Conserv. Recycl. 131, 86–98. https://doi.org/10.1016/j.resconrec.2017.12.010

Council Directive 91/676/EEC of 12 December 1991 concerning the protection of waters against pollution caused by nitrates from agricultural sources. Official Journal L, 375 (31/12) (1991) 0001 – 0008

Derden, A., Dijkmans, R. 2020. Addendum Bij de Studie "Beste Beschikbare Technieken (BBT) Voor Mestverwerking-Derde Uitgave" Mestverwerkingstrajecten: BBT En "Technieken in Opkomst" Met Focus Op Nutriëntrecuperatie Eindrapport. https://emis.vito.be/sites/emis/files/study/Eindrapport_addendum_bij_BBT_mestverwerking_versie_sept_2020.pdf/ (accessed 15 March 2023).

de Haas, D., & Andrews, J. 2022. Nitrous oxide emissions from wastewater treatment - Revisiting the IPCC 2019 refinement guidelines. Environmental Challenges (Amsterdam, Netherlands), 8, 100557-. https://doi.org/10.1016/j.envc.2022.100557

De Vrieze, J. V., Colica, G., Pintucci, C., Sarli, J., Pedizzi, C., Willeghems, G., Bral, A., Varga, S., Prat, D., Peng, L., Spiller, M., Buysse, J., Colsen, J., Benito, O., Carballa, M., & Vlaeminck, S. E. 2019. Resource recovery from pig manure via an integrated approach: A technical and economic assessment for full-scale applications. Bioresour. Technol. 272, 582–593. https://doi.org/10.1016/j.biortech.2018.10.024

Donoso, N., Boets, P., Michels, E., Goethals, P. L. M., & Meers, E. 2015. Environmental Impact Assessment (EIA) of Effluents from Constructed Wetlands on Water Quality of Receiving Watercourses. Water Air Soil Pollut. 226(7), 1–18. https://doi.org/10.1007/s11270-015-2465-8

Elawwad, A. 2018. Optimized biological nitrogen removal of high-strength ammonium wastewater by activated sludge modeling. Journal of Water Reuse and Desalination, 8(3), 393–403. https://doi.org/10.2166/wrd.2017.200Hawkins, S., Robinson, K., Layton, A., & Sayler, G. 2010. Limited impact of free ammonia on Nitrobacter spp. inhibition assessed by chemical and molecular techniques. Bioresource Technology, 101(12), 4513–4519. https://doi.org/10.1016/j.biortech.2010.01.090

Hjorth, M., Christensen, K. V., Christensen, M. L., & Sommer, S. G. 2010. Solid—liquid separation of animal slurry in theory and practice. A review. Agron. Sustain. Dev. 30(1), 153–180. https://doi.org/10.1051/agro/2009010

Hollas, C. E., Bolsan, A. C., Chini, A., Venturin, B., Bonassa, G., Cândido, D., Antes, F. G., Steinmetz, R. L. R., Prado, N. V., & Kunz, A. 2021. Effects of swine manure storage time on solid-liquid separation and biogas production: A life-cycle assessment approach. Renewable & Sustainable Energy Reviews, 150, 111472-. https://doi.org/10.1016/j.rser.2021.111472

Huygens, D., Orveillon, G., Lugato, E., Tavazzi, S., Comero, S., Jones, A., Gawlik, B. and Saveyn, H., Technical proposals for the safe use of processed manure above the threshold established for Nitrate Vulnerable Zones by the Nitrates Directive (91/676/EEC), EUR 30363 EN, Publications Office of the European Union, Luxembourg, 2020, ISBN 978-92-76-21539-4, doi:10.2760/373351, JRC121636.

Jubany, I., Carrera, J., Lafuente, J., & Baeza, J. A. 2008F. Start-up of a nitrification system with automatic control to treat highly concentrated ammonium wastewater: Experimental results and modeling. Chemical Engineering Journal (Lausanne, Switzerland : 1996), 144(3), 407–419. https://doi.org/10.1016/j.cej.2008.02.010

Kampschreur, M. J., Temmink, H., Kleerebezem, R., Jetten, M. S. M., & van Loosdrecht, M. C. M. 2009. Nitrous oxide emission during wastewater treatment. Water Research (Oxford), 43(17), 4093–4103. https://doi.org/10.1016/j.watres.2009.03.001

Lee, S., Maniquiz-Redillas, M. C., Choi, J., & Kim, L.-H. 2014. Nitrogen mass balance in a constructed wetland treating piggery wastewater effluent. J. Environ. Sci. 26(6), 1260–1266. https://doi.org/10.1016/S1001-0742(13)60597-5

Liu, B., Giannis, A., Zhang, J., Chang, V. W.-C., & Wang, J.-Y. 2015. Air stripping process for ammonia recovery from source-separated urine: modeling and optimization. J. Chem. Technol. Biotechnol. 90(12), 2208–2217. https://doi.org/10.1002/jctb.4535

Lyons, G. A., Cathcart, A., Frost, J. P., Wills, M., Johnston, C., Ramsey, R., & Smyth, B. 2021. Review of Two Mechanical Separation Technologies for the Sustainable Management of Agricultural Phosphorus in Nutrient-Vulnerable Zones. Agron. 11(5), 836–. https://doi.org/10.3390/agronomy11050836

Meers, E., Rousseau, D. P., Lesage, E., Demeersseman, E., & Tack, F. M. 2006. Physico-Chemical P Removal from the Liquid Fraction of Pig Manure as an Intermediary Step in Manure Processing. Water Air Soil Pollut. 169(1-4), 317–330. https://doi.org/10.1007/s11270-006-3112-1

Meers, E., Tack, F. M. G., Tolpe, I., & Michels, E. 2008. Application of a Full-scale Constructed Wetland for Tertiary Treatment of Piggery Manure: Monitoring Results. Water Air Soil Pollut. 193(1-4), 15–24. https://doi.org/10.1007/s11270-008-9664-5

Møller, H. B., Lund, I., & Sommer, S. G. 2000. Solid–liquid separation of livestock slurry: efficiency and cost. Bioresource Technology, 74(3), 223–229. https://doi.org/10.1016/S0960-8524(00)00016-X

Nehmtow, J., Rabier, J., Giguel, R., Coulomb, B., Farnet, A. M., Perissol, C., Alary, A., & Laffont-Schwob, I. 2016. Evaluation of an integrated constructed wetland to manage pig manure under Mediterranean climate. Environ. Sci. Pollut. Res.Int. 23(16), 16383–16395. https://doi.org/10.1007/s11356-016-6808-9

Palakodeti, A. Rupani, P.F. Azman, S. Dewil, R. Appels, L. 2022. Novel approach to ammonia recovery from anaerobic digestion via side-stream stripping at multiple pH levels. Bioresour. Technol. 361, 127685-. 10.1016/j.biortech.2022.127685

Phanwilai, S., Noophan, P., Li, C.-W., & Choo, K.-H. 2020. Effect of COD:N ratio on biological nitrogen removal using full-scale step-feed in municipal wastewater treatment plants. Sustain. Environ. Res. 30(1), 1–9. https://doi.org/10.1186/s42834-020-00064-6

Pintucci, C., Carballa, M., Varga, S., Sarli, J., Peng, L., Bousek, J., Pedizzi, C., Ruscalleda, M., Tarragó, E., Prat, D., Colica, G., Picavet, M., Colsen, J., Benito, O., Balaguer, M., Puig, S., Lema, J. M., Colprim, J., Fuchs, W., & Vlaeminck, S. E. 2017. The ManureEcoMine pilot installation: advanced integration of technologies for the management of organics and nutrients in livestock waste. Water Sci. Technol. 75(5-6), 1281–1293. https://doi.org/10.2166/wst.2016.559

Ravi, R., Beyers, M., Vingerhoets, R., Brienza, C., Luo, H., Bruun, S., & Meers, E. 2023. In the quest for sustainable management of liquid fraction of manure - Insights from a life cycle assessment. Sustainable Production and Consumption. https://doi.org/10.1016/j.spc.2023.11.006

Rodríguez, F. A., Reboleiro-Rivas, P., González-López, J., Hontoria, E., & Poyatos, J. M. 2012. Comparative study of the use of pure oxygen and air in the nitrification of a MBR system used for wastewater treatment. Bioresour. Technol. 121, 205–211. https://doi.org/10.1016/j.biortech.2012.06.053

Sapkota, T.B., Jat, M.L., Jat, R.K., Kapoor, P., Stirling, C., 2016. Yield Estimation of Food and Non-food Crops in Smallholder Production Systems. Methods Meas. Greenh. Gas Balanc. Eval. Mitig. Options Smallhold. Agric. https://doi.org/10.1007/978-3-319-29794-1_8.

Sigurnjak, I., Brienza, C., Snauwaert, E., De Dobbelaere, A., De Mey, J., Vaneeckhaute, C., Michels, E., Schoumans, O., Adani, F., & Meers, E. 2019. Production and performance of bio-based mineral fertilizers from agricultural waste using ammonia (stripping-)scrubbing technology. Waste Manage. 89, 265–274. https://doi.org/10.1016/j.wasman.2019.03.043

Skouteris, G., Rodriguez-Garcia, G., Reinecke, S. F., & Hampel, U. 2020. The use of pure oxygen for aeration in aerobic wastewater treatment: A review of its potential and limitations. Bioresour. Technol. 312, 123595–123595. https://doi.org/10.1016/j.biortech.2020.123595

Smet, E., Debruyne, J., Deckx, J., Deboosere, S. 2003. Manure treatment according to the Trevi-concept. Commun Agric Appl Biol Sci. 2003;68(2 Pt A):125-31. PMID: 15296146.

Spiller, M., Vingerhoets, R., Vlaeminck, S.E., Wichern, F., Papangelou, A. 2024 Beyond circularity! Integration of circularity, efficiency, and sufficiency for nutrient management in agri-food systems. Nutr Cycl Agroecosyst. https://doi.org/10.1007/s10705-024-10339-8

Spiller, M., Moretti, M., de Paepe, J., Vlaeminck, S. E. 2022. Environmental and economic sustainability of the nitrogen recovery paradigm: Evidence from a structured literature review. Resourc. Conserv. Recycl. 184, 106406. https://doi.org/10.1016/j.resconrec.2022.106406.

Tampio, E., Marttinen, S., & Rintala, J. 2016. Liquid fertilizer products from anaerobic digestion of food waste: mass, nutrient and energy balance of four digestate liquid treatment systems. J. Clean. Prod. 125, 22–32. https://doi.org/10.1016/j.jclepro.2016.03.127

Tian, W.D., An K.J., Ma C., Han X.K. 2013. Partial nitritation for subsequent Anammox to treat high-ammonium leachate. Environ Technol. 34(8), 1063–1068. https://doi.org/10.1080/09593330.2012.733503

Van Hulle, S. W. H., Callens, J., Mampaey, K. E., van Loosdrecht, M. C. M., & Volcke, E. I. P. 2012. N2O and NO emissions during autotrophic nitrogen removal in a granular sludge reactor – a simulation study. Environmental Technology, 33(20), 2281–2290. https://doi.org/10.1080/09593330.2012.665492

van Puffelen, J. L., Brienza, C., Regelink, I. C., Sigurnjak, I., Adani, F., Meers, E., & Schoumans, O. F. 2022. Performance of a full-scale processing cascade that separates agricultural digestate and its nutrients for agronomic reuse. Sep. Purif. Technol. 297, 121501-. https://doi.org/10.1016/j.seppur.2022.121501

VCM, 2021. Nota VCM Waterrecuperatie uit mest. https://www.vcm-mestverwerking.be/nl/nieuws/153/vcm-nota-waterrecuperatie-uit-mest/ (accessed 20 May 2023).

Vingerhoets, R., de Backer, J., Adriaens, A., Verbesselt, S., de Corte, M., Vlaeminck, S., Spiller, M., Meers, E. 2021. Begroting van stikstof-, fosfor- en eiwitstromen in het agrovoedingssysteem in Vlaanderen: Indicatoren voor efficiëntie en circulariteit. https://omgeving.vlaanderen.be/nl/begroting-van-stikstof-fosfor-en-eiwitstromen-in-het-agrovoedingssysteem-in-vlaanderen-indicatoren/ (accessed 16 March 2023).

Vingerhoets, R., Brienza, C., Sigurnjak, I., Buysse, J., Vlaeminck, S. E., Spiller, M., & Meers, E. 2023a. Ammonia stripping and scrubbing followed by nitrification and denitrification saves costs for manure treatment based on a calibrated model approach. Chemical Engineering Journal (Lausanne, Switzerland : 1996), 477, 146984-. https://doi.org/10.1016/j.cej.2023.146984

Vingerhoets, R., Spiller, M., De Backer, J., Adriaens, A., Vlaeminck, S. E., & Meers, E. 2023b. Detailed nitrogen and phosphorus flow analysis, nutrient use efficiency and circularity in the agri-food system of a livestock-intensive region. J. Clean. Prod. 410, 137278–. https://doi.org/10.1016/j.jclepro.2023.137278

Willeghems, G., De Clercq, L., Michels, E., Meers, E., & Buysse, J. 2016. Can spatial reallocation of livestock reduce the impact of GHG emissions? Agric. Syst. 149, 11–19. https://doi.org/10.1016/j.agsy.2016.08.006

Yu, L., Wang, Y., Li, R., Zhang, R., Zhang, X., Hua, S., & Peng, D. 2020. The differential proliferation of AOB and NOB during natural nitrifier cultivation and acclimation with raw sewage as seed sludge. RSC Adv. 1(47), 28277–28286. https://doi.org/10.1039/d0ra05252c.

Zarebska, A., Romero Nieto, D., Christensen, K. V., Fjerbæk Søtoft, L., & Norddahl, B. 2015. Ammonium Fertilizers Production from Manure: A Critical Review. Crit. Rev. Environ. Sci. Technol. 45(14), 1469–1521. https://doi.org/10.1080/10643389.2014.955630