Resource recovery from pig manure via an integrated approach : a technical and economic assessment for full-scale applications

Manuscript Bioresource Technology Version 2

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Running title: Resource recovery from manure

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Declarations of interest: none

Abstract

Intensive livestock farming cannot be uncoupled from the massive production of manure, requiring adequate management to avoid environmental damage. The high carbon, nitrogen and phosphorus content of pig manure enables targeted resource recovery. Here, fifteen integrated scenarios for recovery of water, nutrients and energy are compared in terms of technical feasibility and economic viability. The recovery of refined nutrients with a higher market value and quality, *i.e.*, $(NH_4)_2SO_4$ for N and struvite for P, coincided with higher net costs, compared to basic composting. The inclusion of anaerobic digestion promoted nutrient recovery efficiency, and enabled energy recovery through electricity production. Co-digestion of the manure with carbon-rich waste streams increased electricity production, but did not result in lower process costs. Overall, key drivers for the selection of the optimal manure treatment scenario will include the market demand for more refined (vs. separated or concentrated) products, and the need for renewable electricity production.

Keywords: anaerobic digestion, manure, nutrient recovery, renewable energy recovery, resource recovery.

1. Introduction

Intensive livestock production for human food purposes cannot be uncoupled from a massive production of manure. In the EU, about 1.3-1.8 billion tonnes (wet weight) of manure is produced each year (Foged et al., 2011; Meers, 2016), mainly in densely populated areas and nitrate vulnerable zones (Nitrates Directive 91/676/EEC). At the same time, agricultural land for the application of manure is limited. For example, in Belgium, over 1 million tons of pig meat for human consumption is processed on yearly basis (Eurostat, 2016), and the related high quantities of manure cannot simply be dispersed in the environment, due to the relatively small extension of land available (Coppens et al., 2016). To prevent environmental pollution, related to the uncontrolled application of pig manure for agricultural purposes, constant updating of the regulations on nutrient limitations are being enforced on the national and international level. Hence, adequate technologies to treat said manure are imperative to obtain a sustainable manure management (Bernet & Beline, 2009; Flotats et al., 2011).

The high carbon, nitrogen and phosphorus content of manure makes it highly suitable for targeted resource recovery. Resource recovery from manure can be applied in addition to an on-site conventional treatment system at the decentralised level, or in stand-alone centralised manure processing facilities. In recent years, the road to move towards advanced nutrient refinery and recovery systems is becoming more attractive, for several reasons (Verstraete et al., 2016). First, there has been a strong increase in technological developments for nutrient recovery from organic waste streams, such as (1) side stream ammonia stripping in anaerobic digestion (Pedizzi et al., 2017; Serna-Maza et al., 2014), (2) electrochemical nitrogen (N) and potassium (K) recovery from digestate (Desloover et al., 2015; Desloover et al., 2012) and urine (Luther et al., 2015), and (3) struvite crystallisation for a combined N and phosphorus (P) recovery (Cusick et al., 2014; Kataki et al., 2016). Second, these nutrient recovery technologies are becoming increasingly competitive with the conventional production techniques for reactive N (Haber-Bosch) and P (phosphate rock mining), with a current market value of around €1.0 kg^{-1} N and €1.9 kg^{-1} P (De Vrieze et al., 2016; Desmidt et al., 2015). Third, the ever stricter regulatory demands concerning manure treatment and its direct application as fertilizer on the land (FAO, 2015) are becoming a strong driver for nutrient recovery technologies. Nutrient recovery from manure is a promising approach, as it not only (1) reduces nutrient load into the environment, but it also (2) enables compliance with regulations, and (3) generates additional revenues from the recovered products. An important challenge for the implementation of resource recovery from manure is the selection of the most cost-effective combination of technologies.

The objective of this study was to determine to which extent different scenarios for resource recovery from pig manure are economically suitable to be applied as novel stand-alone entities or in combination with existing manure treatment facilities. Pig manure, rather than chicken or cattle manure, was chosen, because pigs are one of the most important animals for human consumption worldwide (McMichael et al., 2007). In contrast to cattle farms, pig breeding facilities usually have a lower self-sufficiency in terms of agricultural area for feed production. Consequently, insufficient area is available to spread the manure in an environmentally feasible way, and alternative strategies for manure processing are needed. Each scenario was derived from steady-state mass balances, and considered water recovery, nutrient recovery efficiency, energetic efficiency and economic viability. Different scenarios were designed for separate or combined N and P recovery at different grades of refined product quality. The different scenarios comprised simple technologies, commonly applied for manure treatment, such as solid-liquid separation and subsequent composting to obtain a compost product rich in P, to more complex and integrated processes that result in more high-quality products. Operational expenditure (OPEX), capital expenditure (CAPEX) and the revenues generated form the final products were determined for fifteen different sequences of treatments. The recovered nutrient quality and quantity per ton of manure processed were calculated, and will enable the selection of the most appropriate technology for target-specific full-scale pig manure treatment facilities.

2. Selection and optimisation of the different scenarios

2.1. Feedstock and unit technology selection

The composition of pig manure and the Ecofrit® co-feedstock, which is a carbon-rich mix of vegetable residues from supermarkets, was obtained from Pintucci et al. (2017). The different unit technologies thermophilic (54°C) anaerobic co-digestion with a sludge retention time of 30 days, ammonia stripping/absorption, centrifugation, ultrafiltration (UF), struvite crystallisation, and (chemical) acidification were implemented in the different scenarios based on pilot-scale manure treatment plant data (Pintucci et al., 2017). Acidification (chemical), followed by centrifugation was implemented to bring more P into solution, which makes it available for recovery from the liquid fraction. The centrifugation was assumed to generate a concentrated solid stream with a solids content of 20%, while the UF technology was assumed to produce a retentate with a solids content of 10%.

The design of the mesophilic (35°C) anaerobic digestion technology was considered similar as the thermophilic anaerobic digestion in the pilot-scale plant, but as no pilot-scale data were available for mesophilic digestion, literature data were used to determine its performance and required size (De Vrieze et al., 2016; Hashimoto, 1983). A higher sludge retention time of 50 days was considered for the mesophilic digester, compared to 30 days for the thermophilic digester. Nitrogen conversion/removal was realised with nitrification/denitrification (N/DN), nitritation/denitritation and partial nitritation/anammox (PN/A). The nitrogen conversion and removal efficiency values for these technologies were obtained from literature (Lackner et al., 2014; Maurer et al., 2003; Peng et al., 2017; Siegrist et al., 2008) and through personal communication with Ahidra, Agua y Energía S.L. and Colsen BV. The reverse osmosis (RO) technology was assumed to produce a concentrate with a solids content of 10%, and removal efficiencies were obtained through personal communication with Ahidra, Agua y Energía S.L. Composting data were obtained from literature (Bernal et al., 2009; Flotats et al., 2011) and in close consultation with Ahidra, Agua y Energía S.L.

2.2. Scenario performance parameters and costs

Each scenario contained different combinations of technologies, depending on the main target product of the scenario. The different scenarios were designed for the recovery of nutrients from a defined target flow of 100,000 tonnes of pig manure per year. Performance was evaluated through steady-state mass balancing of the different unit technologies. For those scenarios in which anaerobic digestion was included, the addition of the co-feedstock Ecofrit® was evaluated to increase biogas production. In the cases where Ecofrit® was applied, the yearly amount of manure processed remained at 100,000 tonnes. This implies that the addition of Ecofrit® resulted in an increase of the digester volume, and, consequently, higher CAPEX and OPEX, which are accounted for in the economic analysis (section 3.5).

The OPEX and CAPEX of the different scenarios were determined based on literature data, online market prices and the experience of the ManureEcoMine project partners (Table 1). The OPEX costs included electricity consumption, maintenance, chemicals consumption and labour, while CAPEX costs are average values based on the different sources. Labour cost was estimated at € 70,000 per person per year, based on an average cost of labour in this sector in Belgium. Based on experience from a technology provider (Colsen BV, the Netherlands), 20% of the employment cost of a full-time equivalent (FTE) was considered necessary for the operation of one unit technology. As an example, scenario NP4 required 1.6 FTEs, or € 112,000 per year in labour costs. The CAPEX costs were included in the calculations of the cost of each scenario, assuming a depreciation period of 10 years with an interest rate of 5%. The costs of added chemicals, electricity, and the estimated market value of the refined fertilizer products, compost and electricity sales (Table 2) were obtained from literature data and through the information provided by the industrial partners of the ManureEcoMine project (http://www.manureecomine.ugent.be). These data cover a wide range of European regions and literature sources (Table 2). To account for this variability, and to provide robust cost estimations that are applicable across multiple regions, the OPEX costs were determined in a probabilistic approach through 1000 Monte Carlo simulations for each scenario. Only OPEX costs were considered, because these comprise, averaged over the different scenarios, with a value of 73.7 ±

7.5 %, the majority of the total gross cost. The probability distributions used as an input for the Monte Carlo simulation were based on the variability of the data collected (Table 2). Uniform distributions were created using the *RAND()* function in excel.

2.3. Scenario selection: focus on N and/or P recovery

Five categories of scenarios were considered, each with a different target, combination of unit technologies and, hence, varying degree of complexity (Figure 1).

2.3.1. Scenarios for exclusive P recovery (P1-4)

The first group contains scenarios in which the main focus was P-recovery (P1-4). Scenario P1 comprised a mere centrifugation of the manure mix after which the solid fraction (rich in carbon (C) & P) was composted, and the liquid fraction (rich in N) was subjected to a N/DN step for N removal. In scenario P2, the manure mix was treated by means of mesophilic anaerobic digestion for energy recovery, and the digestate was centrifuged. The solid fraction (rich in C and P) was subjected to composting, and a final liquid fraction (rich in N) was treated with nitritation/denitritation for N removal. In scenario P3, the manure mix was subjected to an acidification step to keep the P in solution, followed by centrifugation. The solid fraction (rich in C) was composted, and the liquid fraction treated with UF of which the retentate was sent back to the centrifugation step. The liquid fraction after the UF (permeate) was subjected to struvite crystallisation for P recovery, followed by nitritation/denitritation for N removal. Scenario P4 contained an additional step in comparison with scenario P3 in which the manure mix was subjected to mesophilic anaerobic digestion for energy recovery, followed by acidification of the digestate.

2.3.2. Scenarios for exclusive N recovery (N1)

The objective of the second group was N recovery (N1). As almost all scenarios included the recovery of P, only one relevant scenario for recovery of N alone was considered. In scenario N1, the manure mix was treated by means of mesophilic anaerobic digestion, followed directly by an ammonia stripping/absorption unit for N recovery in the form of $(NH_4)_2SO_4$. The digestate can be used as a suitable fertilizer rich in C and P.

2.3.3. Scenarios for combined N and P recovery (NP1-6)

The third group contains six scenarios (NP1-6) for combined N and P recovery. In scenario NP1, the manure mix was treated *via* mesophilic anaerobic digestion for energy recovery. The digestate was centrifuged, and the solid fraction (rich in C and P) sent for composting, while the liquid fraction was subjected to ammonia stripping/absorption for N recovery. In scenario NP2, the manure mix was first subjected to ammonia stripping/absorption for N recovery, followed by thermophilic anaerobic digestion for energy recovery. The digestate was separated *via* centrifugation into a solid fraction (rich in C and P) for composting and liquid fraction that was subjected to conventional N/DN for residual N removal. The final liquid product can be used to dilute the manure mix before ammonia stripping, but this was not considered here. Scenario NP3 was similar to NP2, but the ammonia stripping/absorption unit for N recovery was directly connected to the thermophilic digester in a side stream operation, which also eliminated the need to reuse the final liquid product to dilute the manure mix. In scenario NP4, mesophilic anaerobic digestion was used to treat the manure mix, followed by direct ammonia stripping/absorption of the digestate for N recovery and subsequent digestate acidification. Next, the acidified digestate was centrifuged, and the solid fraction (rich in C) composted, while the liquid fraction was treated *via* UF. The retentate of the UF was sent back to the centrifuge, and the permeate was subjected to struvite crystallisation for P recovery, followed by PN/A for residual N removal. In scenario NP5, the manure mix was treated with thermophilic anaerobic digestion with side stream ammonia stripping/absorption for N recovery. The digestate was acidified, centrifuged, and the solid fraction (rich in C) composted, while the liquid fraction was sent to the UF unit. The retentate of the UF unit was sent back to the centrifuge, while the permeate was treated in the struvite crystallisation unit for P recovery. A PN/A step for residual N removal was included after the struvite crystallisation unit. Scenario NP6 comprised direct ammonia stripping/absorption for N recovery from the manure mix, followed by acidification and centrifugation. The liquid fraction was sent to a UF unit, followed by a struvite crystallisation unit for P recovery after which the liquid fraction was mixed again with the solid fraction, and sent to a thermophilic digester for energy recovery. The digestate was centrifuged, resulting in a solid fraction (rich in C) and a liquid end product.

2.3.4. Scenarios for basic separation of C from N and P (C1-2)

A fourth group contains two scenarios (C1-2) whose main objective was to obtain a solid stream with high C-content and a liquid fraction containing N and P. In scenario C1, the manure was acidified, followed by centrifugation, which resulted in a solid fraction (rich in C) suitable for composting and a liquid fraction containing the N and P. Scenario C2 was similar to C1, but with an additional N/DN step for the liquid fraction after the centrifuge for N removal to obtain a final liquid fraction containing almost only P.

2.3.5. Scenarios for water recovery (R1-2)

The objective of the two scenarios in this group (R1-2) was to obtain a reusable effluent by incorporating RO as a final treatment step for the liquid fraction. In scenario R1, the manure mix was centrifuged, and the solid fraction (rich in C and P) was composted. The liquid fraction was treated *via* UF, followed by RO. The retentate of the UF was sent back to the centrifuge, while the concentrate of the RO was recovered as a N-rich end product. In scenario R2, in contrast to R1, the manure mix was subjected to acidification and centrifugation, which resulted in a solid fraction rich in C only, and a RO concentrate containing both N and P.

3. Results and discussion

The key issue to move towards nutrient recovery rather than nutrient removal from pig manure is the economic viability of the scenario. This is influenced by the costs for installation and operation of the technologies, as well as the market value of the final product. The market value will depend on the macroeconomic variables, such as the global demand for P and the natural gas price for N, but also the product quality, reflected in the degree of refining. Intensive livestock farming in densely populated areas implies that there is a need to transport the nutrients away from their site of production to avoid environmental issues, such as surface water bodies eutrophication (Conley et al., 2009; Smith et al., 1999). The recovery of nutrients may prevent (1) environmental issues, related to the energy-intensive Haber-Bosch process for reactive N production (Erisman et al., 2008), (2) geopolitical issues related to phosphate mining (Chowdhury et al., 2017), and (3) the dissipative and uncontrolled release of both N and P into the environment (Steffen et al., 2015). A direct environmental impact assessment of the different scenarios was not carried out, as this would require an in-depth life cycle assessment to estimate the impact on different environmental parameters (Lijó et al., 2018; Pedizzi et al., 2018). As transport entails an additional cost, the need to reduce the volume of the matrix (*e.g.*, water or sludge) in which the nutrients are contained is apparent. This also coincides with an increase in value by refining the nutrients to fertilizers, such as ammonium sulphate and struvite. Hence, the combination of volume reduction with increased purity of the nutrients, mainly N and P, makes it possible to develop several scenarios that will allow a cost-efficient recovery of nutrients from pig manure.

3.1. Phosphorus recovery

A simple centrifugation step to separate the solid from the liquid fraction in the pig manure (P1) resulted in 78.5 % of the total P and 82.4 % of the phosphate P contained within the solid fraction that goes to composting (Figure 2b). This is mainly due to the relatively high pH of fresh pig manure (6.5- 8.0) and related co-precipitation of P with cations, such as Ca, Mg and Fe, contained within the manure (De Vrieze et al., 2015; Hashimoto, 1983; Smith et al., 2001). On the contrary, 76.7 % of total Kjeldahl nitrogen (TKN) and 79.2 % of total ammonia nitrogen (TAN) remained in the liquid fraction prior to the N/DN stage (Figure 2a). Although the P content will show a certain degree of variation between different batches of pig manure, acidification of the pig manure is needed to achieve a transfer of the P from the solid to the liquid fraction. This was demonstrated in scenario P3 in which, due to acidification, the total P content in the solid fraction was reduced to 42.6 % (Figure 2b). The high residual TKN concentration in the liquid fraction of 7.3 g N L^{-1} (P1) and 6.5 g N L^{-1} (P3) after solid-liquid separation of the manure requires additional effluent polishing, before it can be discharged into the environment. Nitrification/denitrification is most commonly used for biological nitrogen removal from N-rich wastewaters, and has been applied successfully for treatment of pig manure in farms (Riaño & García-González, 2014). The incorporation of nitritation/denitritation for the treatment of the liquid fraction after centrifugation presents a novel approach that can be operated virtually without emissions of the greenhouse gas N_2O (Peng et al., 2017). This resulted for scenario P1 in a total net cost (OPEX and CAPEX) of € 10.95 per ton of manure processed, related to a gross cost of € 11.49 and a revenue of € 0.54 for the compost (Figure 3). Within the reported costs, the electricity consumption cost for scenario P1 was estimated at € 3.77 per ton of manure processed (Figure 4a).

Mesophilic anaerobic digestion can be incorporated to recover part of the energy contained in the manure as electricity through biogas burning in a combined heat and power (CHP) unit, which was the case in scenario P2. A COD (chemical oxygen demand) conversion efficiency to methane of 18.5 % was obtained for pig manure during a BMP (biochemical methane potential) test that was carried out by the ManureEcoMine project partners using a standardised approach

(Angelidaki et al., 2009; De Vrieze et al., 2015). The pig manure COD conversion efficiency, as obtained by the ManureEcoMine project partners, was similar to other studies (Angelidaki & Ahring, 2000; Carrere et al., 2009; De Vrieze et al., 2015; Moller et al., 2007). Assuming a methane recovery efficiency of 95 % in the CHP unit, as a minor part of the methane is lost due to the so-called methane slip (Meyer-Aurich et al., 2012; Pucker et al., 2013), a CHP electrical efficiency of 38 % (Deublein & Steinhauser, 2008; Szarka et al., 2013), and a lower heating value (LHV) of 9.95 kWh m^{-3} CH_4, this amounts up to 19.4 kWh of electricity that can be recovered per ton of manure processed through mesophilic anaerobic digestion. Hence, the mesophilic anaerobic digestion of pig manure as single feedstock increased the total revenue to € 2.56 per ton of manure processed for scenario P2 (Figure 3). However, the increase in both OPEX and CAPEX costs, due to the construction and operation of the digester, resulted in a gross cost of € 13.74 per ton of manure processed for scenario P2, which led to a similar net cost of € 11.18 per ton of manure processed as for scenario P1.

Biogas production can be increased by co-digestion of the pig manure with carbon-rich feedstocks, such as crude glycerol (Astals et al., 2012; Fountoulakis et al., 2010) or waste streams from food processing industry (De Vrieze et al., 2013; Liu et al., 2013; Ye et al., 2015). The main requirements for these feedstocks are their (1) year-long availability (2) low cost, and (3) local accessibility. In this study, Ecofrit®, which is a mixture of supermarket organic waste with a relatively stable composition that is available all year, was considered. As the amount of pig manure that needs to be treated on yearly basis (100,000 tonnes) must remain constant, the addition of Ecofrit® as co-feedstock requires a higher anaerobic digester volume, which resulted in increased OPEX and CAPEX costs. For example, when adding 20% of Ecofrit® as co-feedstock, a total of 125,000 tonnes of manure mix needs to be treated, and the digester volume will increase with 25 %. In scenario P2, co-digestion of 60 % pig manure and 40 % Ecofrit®

(w/w) could reduce the total net cost to € 6.35 per ton of manure processed (Figure 4b), resulting from a gross cost of € 29.79 and revenue of € 23.43 per ton of manure processed. Up to 97 % of the revenue was due to the electricity being produced in the CHP unit. A further increase in the amount of Ecofrit® added would no longer have a beneficial effect with respect to the total net cost, related to the too high additional OPEX and CAPEX costs.

In scenario P3, the P-content in the compost decreased to 42.6 %, compared to the P1 & P2 scenarios, due the acidification step during which a certain fraction of the P was solubilized. The soluble P-fraction was partially recovered (46.7 %) as struvite ($NH_4MgPO_4.6H_2O$) (Figure 2b). The UF step was included to remove additional solids from the liquid fraction following the centrifugation step to be able to harvest high-quality struvite pellets (Ping et al., 2016). This enabled the production of 7.0 kg of struvite per ton of manure processed.

The production of struvite as a concentrated fertilizer product opens the potential for efficient transport of nutrients to the desired locations for application. However, the inclusion of the struvite crystallisation technology in scenario P3 resulted in a net cost of € 16.96 per ton of manure processed (Figure 3), which is a strong increase compared with the scenarios P1 and P2. This was mainly due to the strong increase in the gross cost to € 19.23 per ton of manure processed, related to the presence of the UF and struvite crystallisation unit, in contrast to the scenarios P1 and P2, and the limited expected revenues (€ 2.27 per ton of manure processed) for the struvite and compost.

The introduction of a mesophilic anaerobic digestion unit could reduce the total net cost in scenario P3, and this was implemented in scenario P4. However, despite a higher revenue of € 4.39 per ton of manure processed, due to the recovery of energy *via* the CHP and the production of struvite, the increased gross costs of € 23.68 per ton of manure processed resulted in a net cost of € 19.29 per ton of manure processed (Figure 3) which was 13.7 % higher than for scenario P3 and 72.5 % higher than for scenario P2. The addition of 10 % Ecofrit® to the manure mix for mesophilic anaerobic co-digestion resulted in an increase (4.5 %) of the net cost to € 20.16 per ton of manure processed, while a 40 % addition of Ecofrit® increased the net cost by 53.3 % to € 29.57 per ton of manure processed (Figure 4b), indicating that co-digestion was not a suitable option in this scenario.

3.2. Nitrogen recovery

Nitrogen recovery was targeted by implementing an ammonia stripping/absorption unit following mesophilic anaerobic digestion, which resulted in the formation of ammonium sulphate ($(NH_4)_2SO_4$). This method of N recovery was estimated at 24 kWh of electrical energy at mesophilic conditions per ton of manure processed, based on the ManureEcoMine pilot plant data (Table 1). A recovery efficiency of 68.1 % of TKN in the manure and 95.1 % of TAN in the digestate could be obtained (Figure 2a), resulting in 4.7 kg N or 21.5 kg of $(NH_4)_2SO_4$ recovered per ton of manure. This corresponds with an electricity consumption of 5.11 kWh kg N^{-1}, which was similar to values reported for urine (Maurer et al., 2003) and digestate (De Vrieze et al., 2016; Siegrist, 1996). Assuming a market value of about € 1.0 kg^{-1} N (Desmidt et al., 2015) and an electricity cost of € 0.108 kWh^{-1}, this allows a total product revenue of € 4.70 per ton of manure processed. However, maintenance, chemicals and CAPEX costs nullify this margin.

The incorporation of an anaerobic digester for energy recovery was essential to decrease the process cost, as it allowed an additional revenue of € 2.09 per ton of manure up to a total revenue of € 6.79 per ton manure processed. The gross cost of € 23.30, mainly related to the OPEX and CAPEX of the ammonia stripping/absorption unit, resulted in a net cost of € 16.51 per ton of manure processed (Figure 3). Co-digestion with Ecofrit® resulted in an increase in electricity production (Figure 4a), yet, the net costs increased to € 17.81 and € 33.15 per ton of manure processed at 10 and 40 % Ecofrit®, respectively (Figure 4b). Hence, co-digestion does not ameliorate the cost balance in scenario N1, similar to P4. Nitrogen can also be recovered in other ways, but because these approaches also engage P recovery, they are described in scenarios for combined N and P recovery.

3.3. Combined nitrogen and phosphorus recovery

The NP1 scenario combines the different elements of the P2 and N1 scenarios, which resulted in a separate N and P recovery. The centrifugation step following mesophilic anaerobic digestion allowed 75.8 % P and 9.1 % N recovery in the compost, while an additional 53.9 % of N could be recovered as $(NH_4)_2SO_4$ following ammonia stripping/absorption (Figure 2). This corresponds with 17.0 kg of $(NH_4)_2SO_4$ and 66 kg of compost recovered per ton of pig manure processed. This NP1 scenario had a net process cost of € 14.77 per ton of manure processed, which is higher than scenario P2, but lower than scenario N1 (Figure 2). Despite the presence of a centrifugation and composting unit in scenario NP1, in contrast to N1, the lower volumetric load to the ammonia stripping/absorption unit in NP1 compared with N1, yielded a lower total OPEX and CAPEX cost (Figure 3 & 5). Similar to scenario P4 and N1, co-digestion of the pig manure with Ecofrit® enabled net electricity production (Figure 4a), but this did not lead to a lower total net cost (Figure 4b).

The application of thermophilic anaerobic digestion, as is the case for scenarios NP2 and NP3, could offer several advantages over mesophilic anaerobic digestion, such as a higher COD conversion efficiency and methane yield (Ge et al., 2011), and improved pathogens reduction (Kjerstadius et al., 2013). A full-scale trial in which waste activated sludge digestion was converted from a mesophilic to a thermophilic process confirmed that there was no net increase in energy consumption, as no additional heating was needed to sustain thermophilic conditions, yet, an 8.9 % increase in methane yield was observed (De Vrieze et al., 2016). An additional advantage lies in the higher potential for ammonia recovery, related to the (1) higher total ammonia concentration and (2) higher free ammonia fraction at thermophilic conditions (Anthonisen et al., 1976; De Vrieze et al., 2016). Together with the direct effect of an increased temperature and higher pH, thermophilic anaerobic digestion allowed a more cost-efficient N recovery from the pig manure (Gustin & Marinsek-Logar, 2011), which was reflected in a 12 kWh of electrical energy requirement for the ammonia stripping/absorption unit at thermophilic conditions per ton of manure processed, based on the ManureEcoMine pilot plant data (Table 1). The shift from mesophilic to thermophilic anaerobic digestion also coincides with a potential negative impact on the methanogenic community, due to free ammonia toxicity (Angelidaki & Ahring, 1993; Gallert et al., 1998), which is why the ammonia stripping/absorption units were incorporated as pre-treatment (NP2) or in side stream with the digester (NP3) to prevent ammonia toxicity. In scenario NP2, the application of ammonia stripping/absorption as pre-treatment resulted in 62.0 % N recovery (Figure 2a), similar to scenario NP3 with side-stream ammonia stripping/absorption in which 61.0 % of N in the pig manure could be recovered (Figure 2a). Similar values were obtained in other studies for fresh pig manure (Bonmatí & Flotats, 2003; Zhang et al., 2012), yet, higher values up to 90% could be obtained for digestate (Gustin & Marinsek-Logar, 2011; Lei et al., 2007; Pedizzi et al., 2017), which mainly relates to the high pH and release of ammonia due to degradation of organic N, which is an important factor that determines ammonia removal efficiency. The recovery of P was obtained through composting, with 75.8 % P recovery for both NP2 and NP3 (Figure 2b). A final N/DN step was needed to reduce the nitrogen in the effluent from 3.15 to 0.49 g L^{-1} in NP2 and from 3.22 to 0.49 g N L^{-1} in NP3.

The NP4 scenario is quite similar to the P4 scenario, yet, the inclusion of an ammonia stripping/absorption unit following the mesophilic digester allowed 84.5 % N recovery in the form of $(NH_4)_2SO_4$, and P was recovered as compost (41.1 %), and struvite (48.2 %) (Figure 2).

As PN/A can with deal with a certain load of biodegradable organics (Han et al., 2016; Scaglione et al., 2015), this process was included as a final treatment step to reduce the effluent N concentration from 0.56 to 0.13 g N L^{-1}. The N concentration in the final effluent was much lower than for scenarios NP3 and NP4, related to the inclusion of an ammonia stripping/absorption unit after the mesophilic anaerobic digester, as well as the inclusion of part of the N (5.7 %) in the struvite (Figure 2a). The recovery of N *via* ammonia stripping/absorption (€ 5.83) and P *via* struvite crystallisation (€ 1.72) yielded a total revenue of € 10.15 per ton of manure processed, which was the highest of all scenarios (Figure 3). The inclusion of both N and P recovery technologies, however, yielded high OPEX and CAPEX costs, which resulted in the highest total net cost for this scenario.

In scenario NP5, the combination of side stream ammonia stripping/absorption for N recovery during thermophilic anaerobic digestion (similar to scenario NP3) and struvite crystallisation for P recovery (similar to scenario NP4), allowed 39.0 % N recovery as $(NH_4)_2SO_4$, while the values for P recovery in compost and struvite were the same as for scenario NP4 (Figure 2). Given the lower fraction of N recovered *via* the side stream ammonia stripping/absorption unit, in comparison with the post-stripping/absorption unit in scenario NP4, a higher influent concentration of N to the PN/A had to be dealt with than in scenario NP5. The N concentration was reduced from 3.97 to 0.82 g N L^{-1} in the final effluent. Because a lower recovery of N was achieved compared with scenario NP4, a lower revenue for $(NH_4)_2SO_4$ (€ 2.69) was obtained, which also resulted in a lower total net revenue of € 7.06 per ton of manure processed (Figure 3). This led to a lower total net cost of this scenario compared with NP4, as OPEX and CAPEX costs were lower, mainly related to the ammonia stripping/absorption unit.

The NP6 scenario showed a certain similarity with scenario NP2, but the incorporation of an UF and struvite crystallisation step allowed a higher-value P recovery product, with 49.4 % of P recovered as struvite and 36.3 % P in the compost (Figure 2b). Direct ammonia stripping of the manure resulted in 57.7 % N recovery as $(NH_4)_2SO_4$ and an additional 6.7 % N in the struvite (Figure 2a). Due to the absence of a final polishing stage, a TKN concentration of 2.51 g N L^{-1} was obtained in the liquid effluent. Revenues could be obtained from the production of $(NH_4)_2SO_4$ (€ 3.98) and struvite (€ 1.76), resulting in a total revenue of € 8.11 per ton of manure processed. Similar OPEX and CAPEX costs as for scenario NP2 and NP5 were obtained, resulting in a similar total net cost (Figure 3 & 5).

3.4. Basic separation of C from N and P and clean effluent production

In the scenarios C1 and C2, the objective was to separate the N and P from the C and keep it in the liquid fraction, which resulted in a limited recovery of P (36.3 %) and N (8.4 %) in the compost, both for C1 and C2 (Figure 2). The incorporation of a N/DN step in scenario C2 resulted in a strong decrease in the effluent N concentration to 1.98 g N L^{-1}, compared to 7.20 g N L^{-1} for C1. This was, however, still far too high for discharge to surface water bodies. The high P concentration of 1.60 g P L^{-1} also prevented discharge of the liquid fraction, thus, imposing the need for alternative approaches. The liquid fraction of the pig manure, of either scenario C1 and C2, could serve as direct fertilizer, given its high N and P content (Chantigny et al., 2008). Gaseous NH_3 emissions (Chantigny et al., 2007) and the presence of pathogens might pose an issue (Vanotti et al., 2005), but the incorporation of a N/DN step, as is the case in scenario C2, could tackle both issues.

The alternative approach in the scenarios R1 and R2, related to the inclusion of a reverse osmosis step, resulted in much lower N (1.62 g N L^{-1} for R1 and 0.59 g N L^{-1} for R2) and P (1.08 g P L^{-1} for R1 and 0.05 g P L^{-1} for R2) concentrations in the final liquid effluents (Figure 2). The absence of an acidification step in R1, in contrast to R2, resulted in the majority of P recovered in the compost in R1 (90.5 %), while this was only 42.6 % in R2. As a result, most of the P (56.1 %)

was retained in the RO concentrate in R2, while this was only 0.5 % in R1. In both scenarios, most of the N was recovered in the RO concentrate, with 66.9 and 70.2 % N for R1 and R2, respectively. This RO concentrate can be considered a recovered mineral fertilizer, yet, it may lead to a higher overall environmental impact compared with the application of fresh (liquid) manure as fertilizer, due to the potential increase in methane and ammonia emissions (De Vries et al., 2012).

Total revenues did not exceed € 1.00 per ton of manure processed, related to the absence of anaerobic digestion and low market value of the recovered products (Figure 3). The C2 scenario showed a clearly higher total net cost than C1, related to the high OPEX and CAPEX cost of the N/DN unit. A similar observation could be made for R1 and R2, which was due to the OPEX and CAPEX costs of the acidification step in R2, compared with R1.

3.5. Unit process costs: OPEX vs. CAPEX

Although the results of the economic analysis of each scenario depend on the total revenues from the electricity and nutrient recovery products, the OPEX and CAPEX costs of the technologies included in the scenario have a stronger impact. Hence, even though the N, P and electricity revenues strongly depend on local market prices and willingness to pay, their temporal impact on the overall economic viability will be limited. A clear distinction can be made between OPEX and CAPEX costs.

The ammonia stripping/absorption unit, depending on its position in the scenario, has an OPEX costs between € 7.73 and 22.76 per ton of manure processed (Figure 5a). The value is mainly depending on to the nitrogen load and desired removal efficiency, both of which strongly increase the OPEX costs (De Vrieze et al., 2016; Siegrist, 1996). Compared with other unit processes, the N/DN (€ 4.26 – 6.13 per ton of manure processed) and nitritation/denitritation (€ 4.09 – 8.04 per ton of manure processed) technologies also have high OPEX costs, mainly due to the aeration energy consumption, up to 3.1 kWh kg^{-1} N (Maurer et al., 2003; Verstraete & Vlaeminck, 2011). In contrast, the OPEX cost for PN/A remains low at € 0.26-1.91 per ton of manure processed, due to the much lower O_2 requirements of this technology (Lackner et al., 2014; Maurer et al., 2003; Siegrist et al., 2008). Overall, the selection of a suitable N recovery/removal technology, depending on the scenario requirements, has a key impact on the OPEX costs.

The CAPEX costs per ton of manure processed were a factor 1.37 to 8.88 lower than the OPEX costs, depending on the scenario (Figure 5b). The inclusion of a mesophilic (€ 2.47 per ton manure processed) and thermophilic (€ 1.52 per ton manure processed) anaerobic digestion process had a distinct impact on the overall CAPEX cost. The CAPEX cost for thermophilic anaerobic digestion was slightly lower than for mesophilic anaerobic digestion, due to the lower sludge retention times that can be applied at thermophilic conditions (De Vrieze et al., 2016). The total CAPEX cost increased if a higher amount of waste needed to be treated on daily basis, simply because a higher volume was needed to sustain the same sludge retention time and volumetric loading rate. This explains why the addition of Ecofrit® as co-feedstock is only beneficial in 4 out of 9 cases (Figure 4b), because the revenues from biogas production do not outweigh the expanses for CAPEX and OPEX for the anaerobic digester and further downstream processing steps. The different N removal technologies also entailed the same CAPEX costs of € 2.25 per ton manure processed, while the CAPEX cost of the ammonia stripping/absorption technology for N recovery remained limited to 0.75 € per ton manure processed (Figure 5b). The majority of CAPEX costs, hence, could be contributed to the anaerobic digestion and N removal processes. The strong impact of OPEX and, to lesser extent CAPEX in relation to the gross costs limits the contribution of the product revenues to the net costs for each scenario. This results in the fact that none of the fifteen scenarios has a lower net costs than plain transport and local spreading of raw manure on the land, which corresponds with an estimated cost of € 4.9 per ton of manure (Nolan et al., 2012), in contrast to net costs ranging between € 7.28 and 31.61 per ton of manure processed. However, this economic estimation does not consider the externalized environmental costs associated with each scenario. Moreover, the local spreading of manure as reference scenario does not allow uncoupling of pig manure treatment and direct land application, which is an essential aspect to obtain more sustainable pig manure treatment.

3.6. Impact market prices and chemicals cost

The Monte Carlo simulations revealed a different impact for the various scenarios. For the scenarios targeting only P recovery, the value of compost and cost for electricity and labour did not have a strong impact, as demonstrated for scenario P1 and P2 (Figure 6a). An increase of only 4.7 and 0.7 %, respectively, could be observed for the total net cost at the 80 % probability level in relation to the standard value (Figure 3). A stronger impact was observed for the market price of H_2SO_4, necessary for acidification in the scenarios P3 and P4, which resulted in an increase of 23.2 % for P3 and 18.8 %for P4, for the total net cost (80 % probability level in relation to the standard value). The higher electricity consumption in the scenarios P3 and P4 (Figure 4b) also had a pronounced effect on the total net cost, but it was much lower than the impact of the H_2SO_4 market price. The impact of the struvite market price was also limited, because even if the struvite yielded no revenue, an increase in net cost of only 9.8 % for P3 and 8.9 % for P4 was observed.

In scenario N1, in decreasing order, the market price of H_2SO_4, the market value of $(NH_4)_2SO_4$ and the electricity cost (as this dominated over the electricity production) had a major impact on the total net cost. This was reflected in the fact that an increase of the total net cost with 49.3 % could be observed (80 % probability; Figure 6a). This strong impact was caused by the high consumption of H_2SO_4 and electricity and the production of $(NH_4)_2SO_4$ by the stripping/absorption unit at 470 tons of N per year.

The scenarios for combined N and P recovery yielded a similar overall impact of the variation in the different parameters (Figure 6b), except for scenario NP4, which was a consequence of the high electricity consumption (127 kWh per ton of manure), in contrast to lower values for NP1, NP2, NP3, NP5 and NP6 (ranging between 36 and 63 kWh per ton). Like the scenarios for P3, P4 and N1, the market price of H_2SO_4 and $(NH_4)_2SO_4$, in addition to the electricity cost, had the highest impact on the total net cost for combined N and P recovery.

The total net cost of scenarios C1 and C2 was also mainly impacted by the market price of H_2SO_4 for the acidification unit. A decrease in the net process cost, compared to the standard value, of 27.5 % for C1 and 11.4 % for C2 could be observed, when the price of H_2SO_4 was lowered from € 200 to € 100 per ton. The lower impact of the H_2SO_4 price for C2 compared to C1 was due to the 4 times higher electricity cost, related to the operation of the nitrification/denitrification unit. The strong impact of the H_2SO_4 market price was also the cause of the main difference between scenario R1 and R2, as only scenario R2 included an acidification unit (Figure 6c). This resulted in an increase of the total net cost with only 2.7 % (80 % probability) for R1, while this was 24.3 % for R2.

Overall, the market price of H_2SO_4 as a chemical and $(NH_4)_2SO_4$ as a product strongly impacted the total net cost of the different scenarios. The impact of the electricity market price logically increased with increasing electricity consumption, while labour cost did not have a determining impact on the total net cost of the different scenarios. The major cost, related to the consumption of H_2SO_4 in the acidification and/or stripping/absorption unit could be tackled by (1) biological acidification, *e.g.*, through pre-fermentation of the manure mix, or (2) the usage of a cheaper source of acid. Combined with the future expected decline in electricity price in the coming decades, related to the advances in *e.g.*, solar power technology (van Wijk et al., 2017), this could reduce the total net costs for nutrient recovery from manure.

4. Conclusions

The comparison of different scenarios that combine unit technologies for water, nutrient and energy recovery from pig manure revealed a strong variation in recovery product quality and value, as well as total net process costs. Nitrogen recovery through ammonia stripping/absorption, and phosphorus recovery through struvite crystallisation enabled the production of high-value products, and co-digestion allowed increased energy recovery. However, this coincided with higher OPEX and CAPEX costs in comparison with basic technologies, such as composting. Hence, the economic viability of the different scenarios for case-specific full-scale applications strongly depends on the required degree of resource recovery and effluent quality.

E-supplementary data of this work can be found in online version of the paper.

Acknowledgements

The presented work was carried out in the frame of the ECFP7 project 'ManureEcoMine – Green fertilizer upcycling from manure'. ManureEcoMine was co-funded by the European Community's Framework Program (FP7/2007–2013) under Grant Agreement no. 603744. Jo De Vrieze is supported as postdoctoral fellow from the Research Foundation Flanders (FWO-Vlaanderen). All authors declare no conflict of interest.

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Tables

Table 1 Overview of the main operational expenditures (OPEX) and capital expenditures (CAPEX) of each unit technology. N/DN = nitrification/denitrification, PN/A = partial nitritation/anammox. The OPEX and CAPEX cost are based on literature data, as cited for each technology, and were also obtained through personal communication with Ahidra, Agua y Energía S.L. and Colsen BV. The manure mix represents only raw manure or a mixture of raw manure and Ecofrit® in case a digestion step is included. NA = not applicable.

Unit technology	Treatment capacity	Electricity consumption (kWh ton^{-1} manure mix processed)	Maintenance (€ ton^{-1} manure mix processed)	Total CAPEX (€ x 1,000)	Reference
Centrifugation	240-1200 m^3 d^{-1}	4.00	1.00	100	(Derden et al., 2012; Flotats et al., 2011)
Ultrafiltration	240 m^3 d^{-1}	3.00	2.00	225	(Flotats et al., 2011; VITO, 2015)
Reverse osmosis	240 m^3 d^{-1}	8.00	5.00	185	(Flotats et al., 2011)
Mesophilic digestion (35°C)	280 m^3 d^{-1} (13,800 m^3)	1.61	0.10	0.12[a]	(Flotats et al., 2011)
Thermophilic digestion (55°C)	280 m^3 d^{-1} (8,500 m^3)	4.50	0.10	0.15[a]	(Flotats et al., 2011)
Ammonia stripping/absorption in side stream of thermophilic anaerobic digestion	240-360 m^3 d^{-1}	12.00[b]	0.10	500	(Flotats et al., 2011)
Ammonia stripping/absorption	240-360 m^3 d^{-1}	24.00[b]	0.10	500	(Flotats et al., 2011)
N/DN	275 m^3 d^{-1}	15.00	2.23	1,500	(Flotats et al., 2011)
Nitritation/denitritation	275 m^3 d^{-1}	12.00	1.78	1,500	(Flotats et al., 2011)
PN/A	275 m^3 d^{-1}	6.00	0.89	1,500	(Flotats et al., 2011)
Struvite crystallisation	160-200 m^3 d^{-1}	0.50	0.10	6[a]	(Flotats et al.,

					2011)
Composting	25-30 tonne d^{-1}	4.00	0.10	470	(Flotats et al., 2011)
Acidification	NA	0.10	0.10	20	Personal communication only

[a] CAPEX per ton of manure processed.
[b] Electricity consumption per unit of nitrogen recovered (kWh kg^{-1} N).

Table 2 Market value and prices of the chemicals that are used in the different unit technologies and final products obtained in the different scenarios. Market values are obtained from literature data and/or provided by the technology providing companies Ahidra, Agua y Energía S.L. and Colsen BV. The standard value or price was selected as the most typical value, while the range was used for the sensitivity analysis of the value/price assumptions. For the revenues of $(NH_4)_2SO_4$ and $NH_4MgPO_4{\cdot}6H_2O$, a minimum value of zero was considered, related to a local situation without suitable market, while the maximum value was assumed a potentially future doubling of the standard value. RO = reverse osmosis, n.a. = not applicable (as RO concentrate has no monetary value), FTE = full-time equivalent.

Chemicals, products or energy	Standard value or price (€ $tonne^{-1}$ dry chemical/product)	Range in value or price (€ $tonne^{-1}$ dry chemical/product)	Reference or assumption
$Mg(OH)_2$	200	200-400	(De Vrieze et al., 2016)
H_2SO_4	200	100-500	(De Vrieze et al., 2016)
$(NH_4)_2SO_4$	200[a]	0-400	(De Vrieze et al., 2016; Desmidt et al., 2015)
$NH_4MgPO_4{\cdot}6H_2O$	250[b]	0-500	(Desmidt et al., 2015; The World Bank Group, 2015)
Compost	5	0-10	Ahidra, Agua y Energia; Colsen BV
RO concentrate	0	n.a.	Ahidra, Agua y Energia; Colsen BV
Electricity	0.108 € kWh^{-1}	0.03-0.15 € kWh^{-1}	(De Vrieze et al., 2016; Verstraete & Vlaeminck, 2011)
Labour	€ 70,000 FTE^{-1} $year^{-1}$	10,000-80,000 € FTE^{-1} $year^{-1}$	(Eurostat, 2017)

[a] The market value of $(NH_4)_2SO_4$ is based on the assumed market price of € 1.0 kg^{-1} N.
[b] The market value of struvite is based on the assumed market price of € 1.9 kg^{-1} P.

Figures

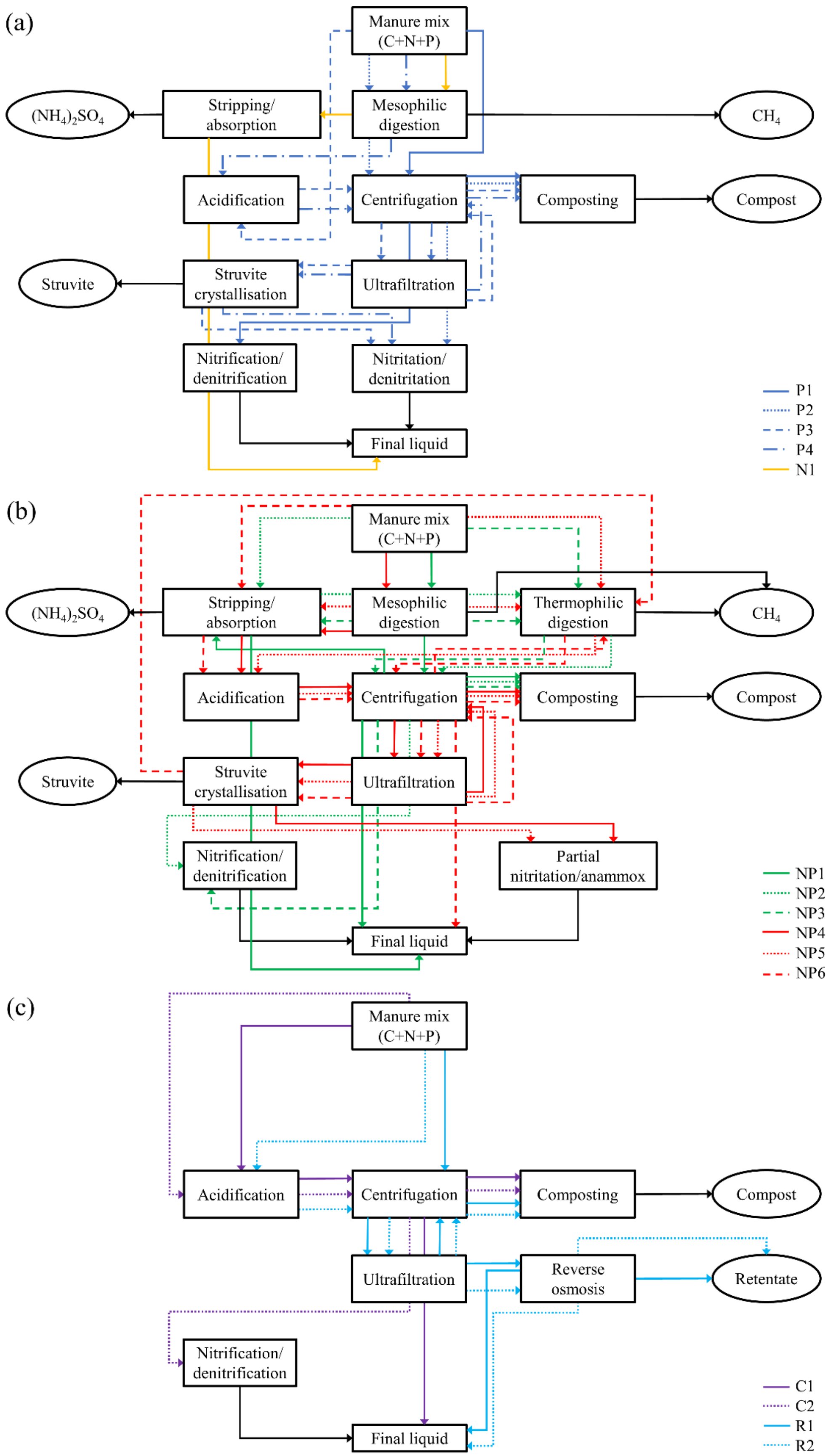
(a)
Manure mix (C+N+P)
$(NH_4)_2SO_4$
Stripping/ absorption
Mesophilic digestion
CH_4
Acidification
Centrifugation
Composting
Compost
Struvite
Struvite crystallisation
Ultrafiltration
Nitrification/ denitrification
Nitritation/ denitritation
Final liquid
P1
P2
P3
P4
N1
(b)
Manure mix (C+N+P)
$(NH_4)_2SO_4$
Stripping/ absorption
Mesophilic digestion
Thermophilic digestion
CH_4
Acidification
Centrifugation
Composting
Compost
Struvite
Struvite crystallisation
Ultrafiltration
Nitrification/ denitrification
Partial nitritation/anammox
Final liquid
NP1
NP2
NP3
NP4
NP5
NP6
(c)
Manure mix (C+N+P)
Acidification
Centrifugation
Composting
Compost
Ultrafiltration
Reverse osmosis
Retentate
Nitrification/ denitrification
Final liquid
C1
C2
R1
R2

Figure 1 Schematic overview of the combinations of the unit processes, and corresponding products, for each of the 15 treatment scenarios of the manure mix, *i.e.*, pig manure for schemes without anaerobic digestion, and pig manure supplemented with a co-substrate (Ecofrit) if digestion is included. The different scenarios include (a) exclusive P recovery (NP1-4) and N recovery (N1), (b) combined N and P recovery (NP1-6), and (c) basic separation of C from N and P (C1-2) and water recovery (R1-2). The coloured arrows represent the flows between the different unit technologies, while the black arrows link the recovered product or stream to the unit technology. Double arrows between the thermophilic digestion and stripping/absorption unit reflects a stripping unit in side stream operation to the digester.

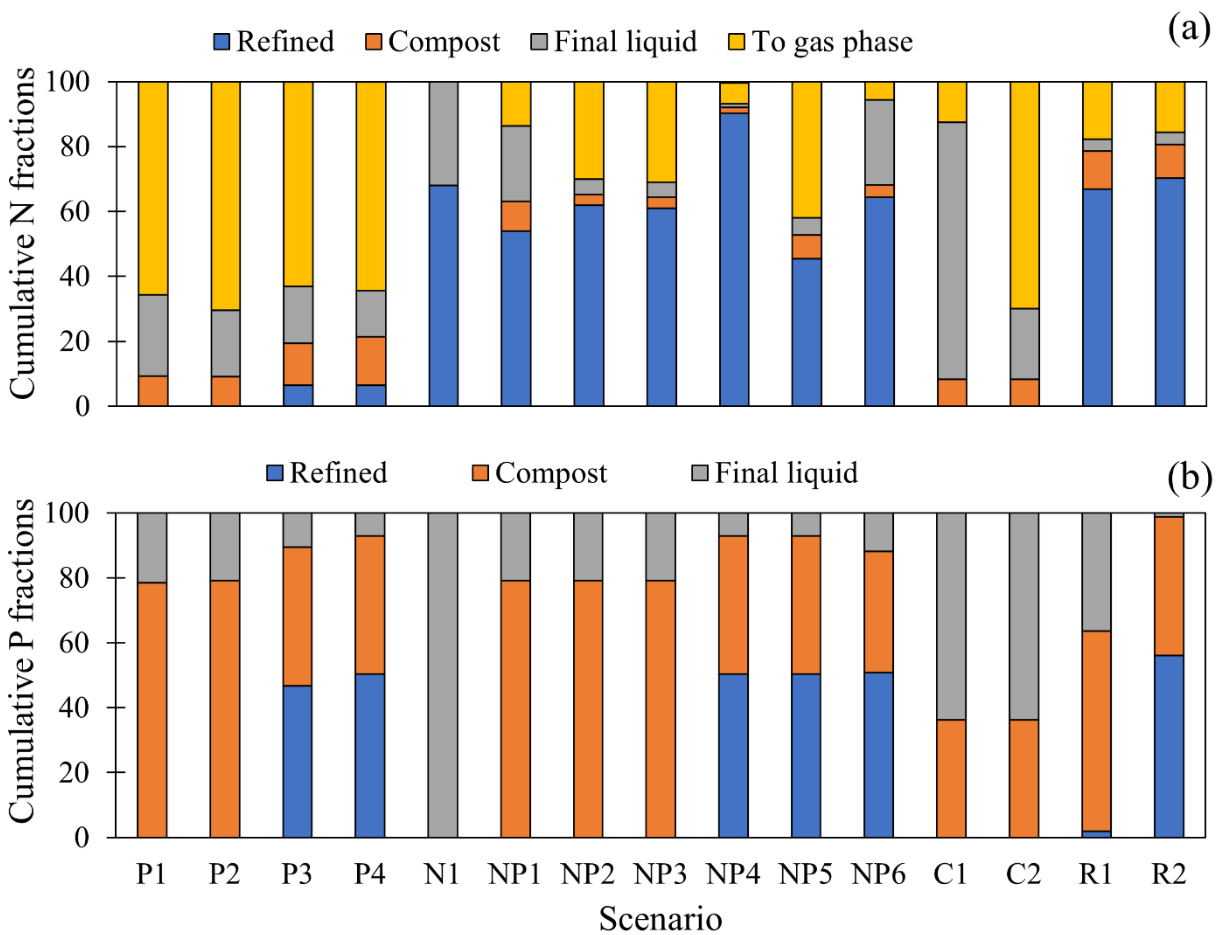

Figure 2 Percentage of (a) N and P (b) in the pig manure that ends up in the refined products (ammonium sulphate, struvite and concentrate for R1 & R2), compost, final liquid or gas phase. Due to the involatile character of P, no release to the gasphase was assumed. The refined products for N included $(NH_4)_2SO_4$, struvite, and concentrate from RO, while the refined products for P included struvite and RO concentrate.

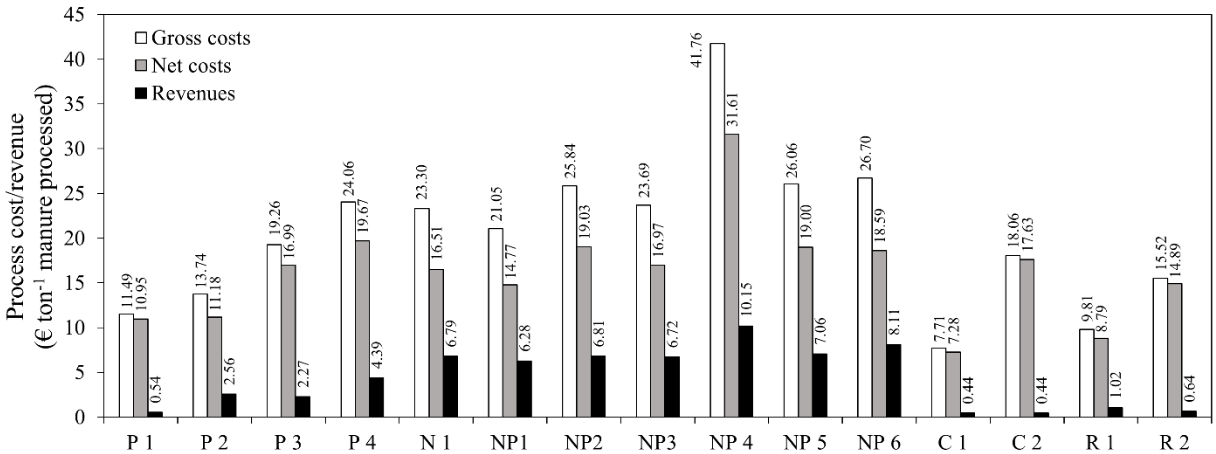

Figure 3 Overview of the gross costs, total net costs and revenues for each of the different scenarios. The net cost is determined as the difference between the gross cost and the revenues. Revenues are calculated based on the market value of the final products (ammonium sulphate, struvite, compost & electricity, Table 2), while gross costs represent both OPEX and CAPEX costs.

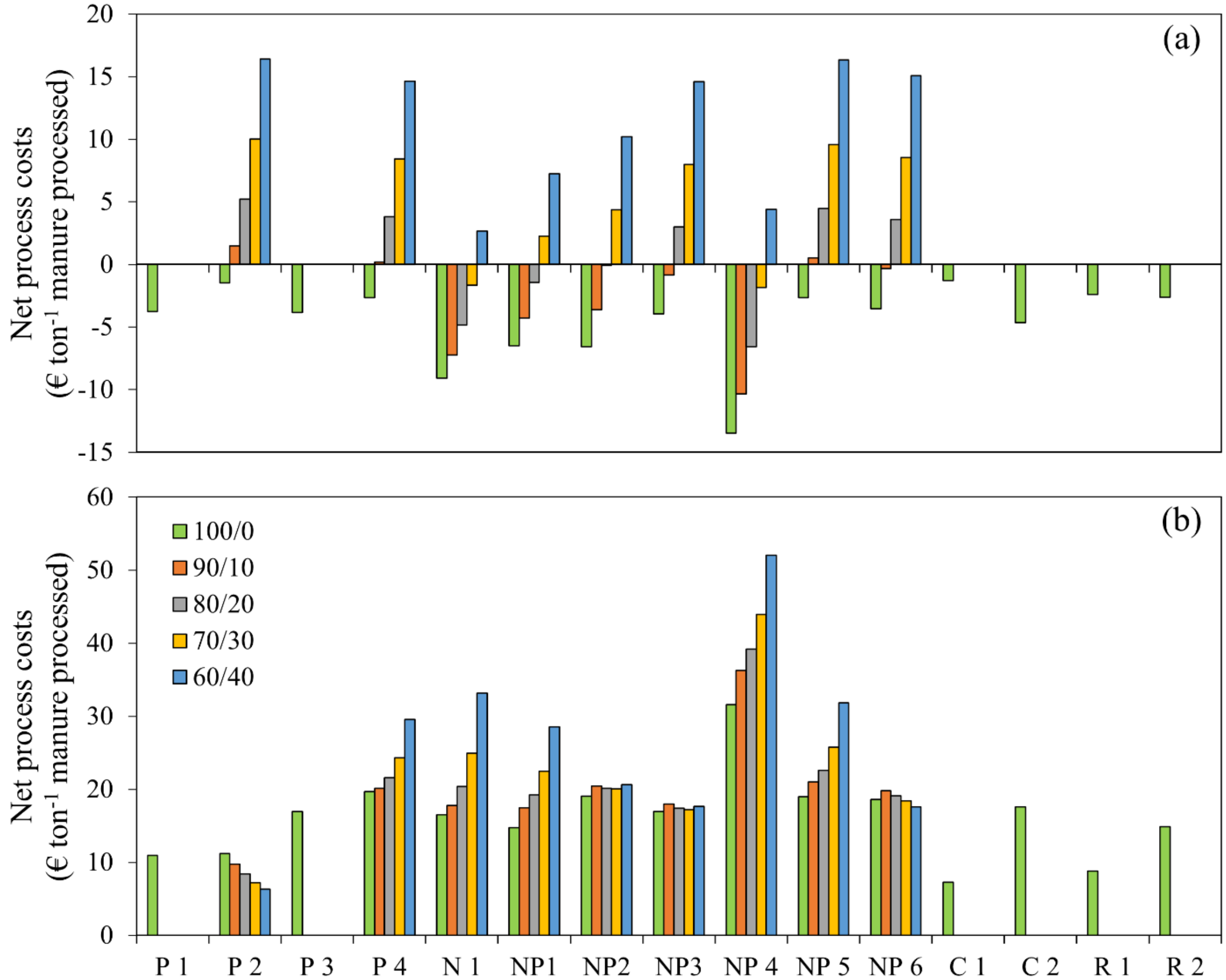

Figure 4 Net electricity production (a) and total net costs (b) for each of the different scenarios. A negative value indicates a net electricity consumption, while a positive value indicates a net electricity production. Different combinations of pig manure and the Ecofrit® co-feedstock were compared, ranging from 100 to 60 % pig manure. In case no anaerobic digestion stage was included in the scenario, no Ecofrit® addition was simulated. In each scenario, the total amount of pig manure to be treated was kept at 100,000 tonnes per year.

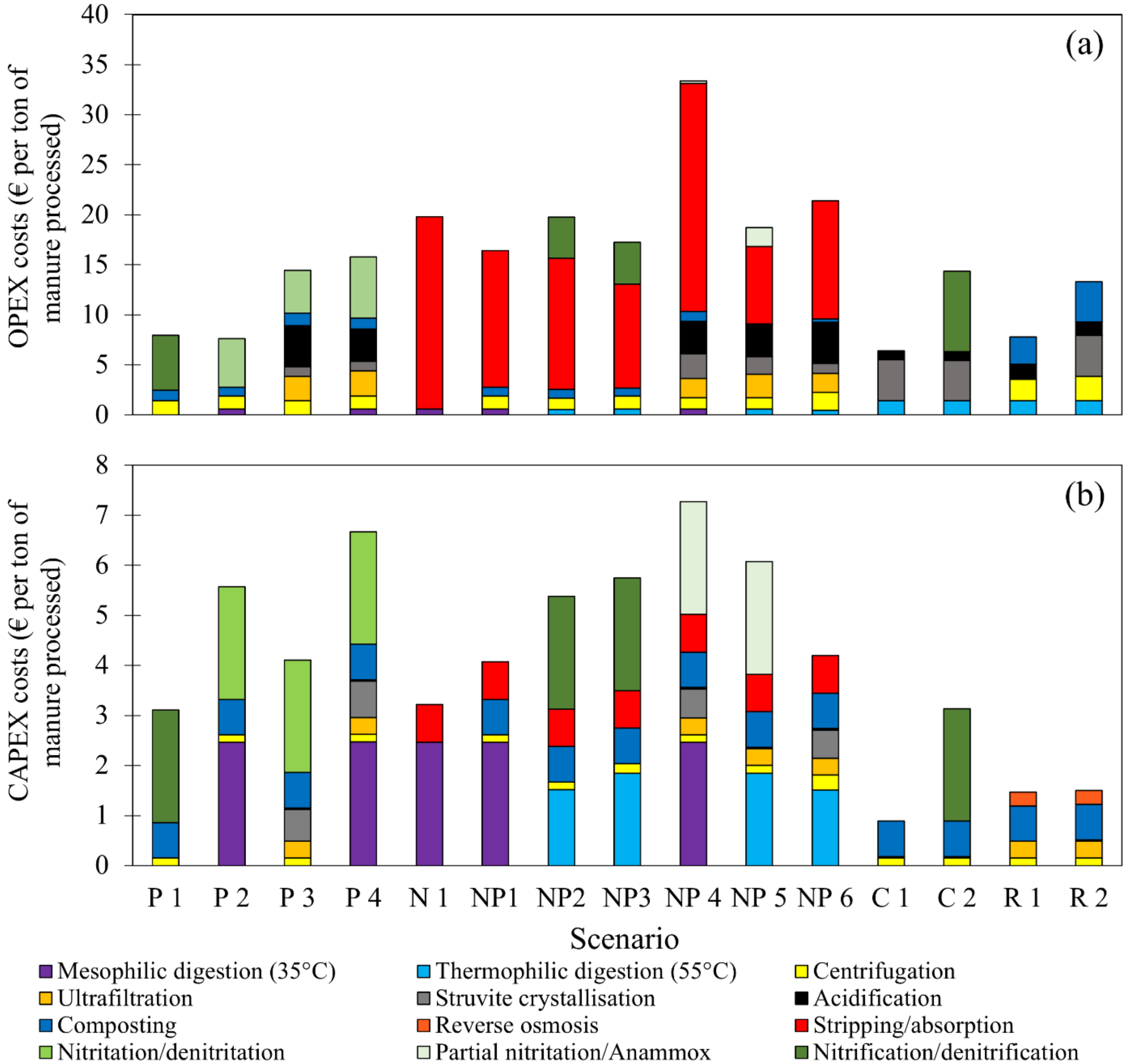

Figure 5 Overview of the (a) operational expenditure (OPEX), and (b) capital expenditure (CAPEX) for the different unit technologies that were used to build the different scenarios.

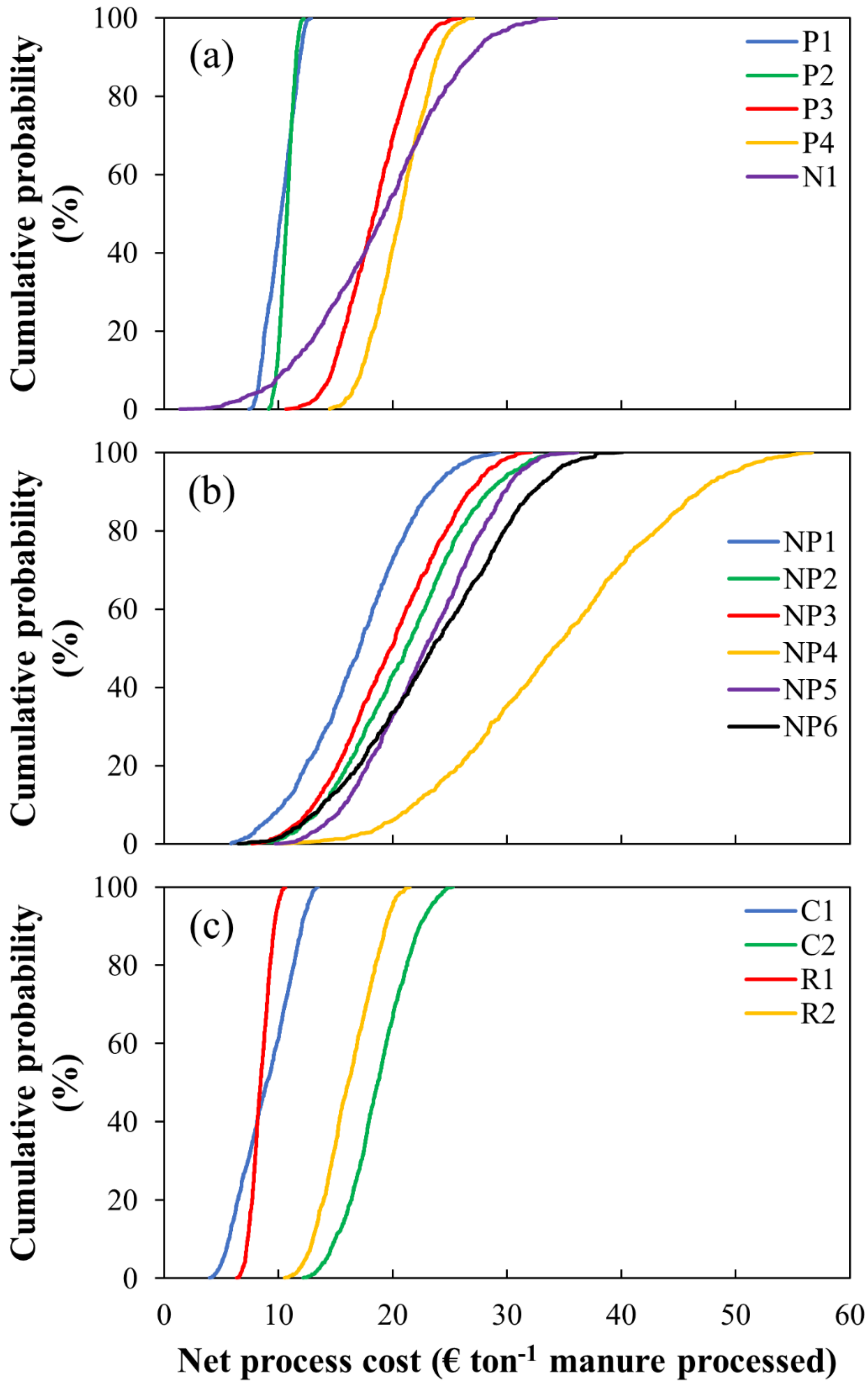

Figure 6 Monte Carlo probability distributions for the sensitivity analysis for the manure processing scenarios based on (a) exclusive P (P1-4) and N recovery (N1), (b) combined N and P recovery (NP1-6), and (c) basic separation of C from N and P (C1-2) and water recovery (R1-2).

Highlights manuscript "Resource recovery from pig manure via an integrated approach: a technical and economic assessment for full-scale applications"

- Technologies were combined to recover nutrients and energy from pig manure
- Anaerobic digestion lowered net energy consumption, but not OPEX costs
- High-value refined products (N and P) could be recovered from pig manure
- The OPEX costs per unit of manure were considerably higher than the CAPEX costs
- Economic viability strongly depended on the required nutrient and effluent quality