Performance of a full-scale processing cascade that separates agricultural digestate and its nutrients for agronomic reuse

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ABSTRACT

The application of animal manure on agricultural land in the Netherlands is bound by legal limits to prevent the leaching of nitrogen (N) and phosphorus (P) to ground and surface waters. The surplus of animal manure is transported abroad at high costs. In this study, a full-scale cascaded membrane filtration system (GENIUS) comprising two decanter centrifuges, microfiltration (MF) reverse osmosis (RO) and an ion exchanger was monitored. The system processed agricultural digestate from anaerobically co-digested animal manure into two solid fractions (SFs), RO concentrate, MF concentrate and purified water. The goal was to separate P and ammoniacal nitrogen (NH_4-N) and remove water from the digestate. From the initial digestate, 66% of P was recovered in the first SF, which constituted 15% of the total mass, without the addition of iron or aluminium salts or polymer flocculants. Another 29% of P was recovered in the MF concentrate and used as a liquid organic fertiliser. Of the P in the initial digestate, 98% was removed before RO. For N, 34% ended up in the RO concentrate and this product can be regarded as an alternative for synthetic N fertiliser as it contains N solely in mineral form. Overall, around 18% of the total mass of initial digestate was discharged as purified water and 31% was locally applied in the form of RO concentrate. We found that aqua regia digestion before chemical analysis can decrease the measured S content of processed digestate. Compared to the transport of raw (unprocessed)

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digestate, the implementation of the GENIUS system led to a 53% reduction in the mass-weighted average transport distance.

KEYWORDS

Manure treatment, Anaerobic digestion, Nutrient recovery, Membrane filtration, Biobased fertilisers

1 INTRODUCTION

In the Netherlands and other parts of Europe, intensive animal husbandry has resulted in a surplus of animal manure in terms of nutrients compared to the legal nutrient application rates of soil-crop combinations. If not properly managed, this surplus of nutrients such as nitrogen (N) and phosphorus (P) can result in deterioration of groundwater and surface water quality and eutrophication of ecosystems. To avoid undesired emissions to soil, water and air, the Nitrates Directive (Council Directive 91/676/EEC) limits the application rate of N from animal manure on agricultural land in nitrate vulnerable zones (NVZs) to 170 kg ha^{-1} per year. Some countries, amongst others the Netherlands, also limit the application of P, which can further restrict the application of animal manure on agricultural land (Schoumans *et al*., 2017). Yearly animal manure production in the Netherlands, an NVZ, increased from 49 million tonnes (t) in 1950 to 74.3 million t in 2020 (CBS, 2021a). The surplus animal manure is transported to nutrient-deficient regions. In 2020, the Netherlands exported 3.07 million t of animal manure, accounting for 4% of its total animal manure production (RVO, 2021).

Paradoxically, Dutch farmers need to buy mineral fertilisers to meet the crop demand for N and P, in total respectively 220 and 4 million kg in 2020 (CBS, 2021b). Therefore, separating the nutrients in animal manure is important to facilitate their local use, thereby reducing environmentally and economically costly transport of animal manure.

Anaerobic digestion (AD) is a common way to process animal manure. It generates renewable energy in the form of biogas and reduces methane emissions from raw (unprocessed) animal manure (Burg *et al*., 2018). Furthermore, the produced agricultural digestate is a valuable organo-mineral fertiliser. In most cases, AD of sole animal manure is not economically viable because the revenue from biogas production does not outweigh the AD costs. Therefore, animal manure is often anaerobically co-digested with organic waste streams that have a higher biochemical methane potential than manure. However, this influences the digestate composition and increases the amount of digestate produced, a burden in regions with a manure surplus.

During AD, easily biodegradable organic matter (OM) is mineralised, thereby increasing the fraction of total-N (TN) that is present as ammoniacal N (NH_4-N), which is readily available for crop uptake (Möller and Müller, 2012). However, AD does not remove N or P and the field application of manure-derived digestate also falls under the application limit of 170 kg N ha^{-1} $year^{-1}$ in NVZs. Due to the digestate's high moisture content, usually more than 90%, its transport is environmentally and economically costly. Therefore, processing systems for (anaerobically digested) animal manure that concentrate nutrients (Sigurnjak *et al*., 2019; Brienza *et al*., 2021) and reduce manure or digestate volumes (Zarebska-Mølgaard *et al*., 2022) are key for environmentally friendly agriculture in regions with an animal manure surplus.

Mechanical solid-liquid separation, resulting in a solid fraction (SF) and a liquid fraction (LF) of anaerobically digested animal manure, is often performed as a first processing step. In practice, one or multiple solid-liquid separation steps are a prerequisite for membrane filtration. To date, hydraulic pressure-driven membrane filtration processes are the most commonly used membrane separation processes for the LF of anaerobically digested animal manure. To avoid membrane damage from abrasive solids (Masse *et al*., 2007) and remove microorganisms, macromolecules and suspended solids (Waeger *et al*., 2010), reverse osmosis (RO) is usually preceded by solid-liquid separation and subsequently microfiltration (MF) or ultrafiltration (UF) in processing cascades. An advantage of using MF as pre-treatment for RO is that MF membranes, having a pore size of 0.1-10 µm (Zhang *et al*., 2020), retain suspended (organic) solids and only require a low hydraulic pressure of a few bars. Also, ceramic MF membranes are well cleanable and can as such have a long operational lifetime.

The via RO produced concentrates from anaerobically digested animal manure (RO concentrates) are promising NK fertilisers. They can adhere to the recently, by the Joint Research Centre (JRC) of the European Commission proposed criteria for RENURE materials (Huygens *et al*., 2020). RENURE (REcovered Nitrogen from manURE) materials are manure-derived N fertilisers that could be applied in NVZs under the same regulations as those for synthetic N fertilisers.

Different membrane filtration cascades for the LF of anaerobically mono-digested or co-digested animal manure have been developed at pilot scale or full-scale (Vaneeckhaute *et al*., 2012; Ledda *et al*., 2013; Chiumenti *et al*., 2013; Adam *et al*., 2018; Bolzonella *et al*., 2018; Gienau *et al*., 2018; Bao *et al*., 2020). However, these studies mostly focus on N, P and potassium (K), omitting other plant macronutrients, such as sulphur (S). Moreover, the economic aspects of these cascades, including storage, transport and disposal of the end products are not, or only partially, evaluated.

This paper aims to evaluate the performance of a full-scale cascaded membrane filtration system (GENIUS) for the processing of agricultural digestate from anaerobically co-digested animal manure to separate nutrients for agronomic reuse and produce dischargeable water. As opposed to many conventional full-scale processing cascades for (anaerobically co-digested) animal manure in the Netherlands, no iron or aluminium salt is added. Also, an RO concentrate with relatively low S:TN and S:K ratios, respectively 0.18 and 0.19, and an SF free from polymers are produced. The former allows field application of more RO concentrate without exceeding the crop demand for S. The GENIUS system consists of two decanter centrifuges placed in series, an MF unit, a double-pass RO installation and, as a polishing step, an ion exchanger (IX). For the measured components, Mass balances over each process unit and the whole system, and recovery efficiencies were calculated. Additionally, each process unit's achieved separation efficiency (SE) for the measured components was calculated. The quality of the end products in terms of their nutrient content was investigated. Also, the capital and operational costs of the GENIUS system and the achieved transport reduction for the end products were assessed.

2 MATERIALS AND METHODS

2.1 The biogas plant

The biogas plant of Groot Zevert Vergisting (GZV) is located in Beltrum, the Netherlands, and has been operational since 2004. The hydraulic retention time (HRT) of the system of connected digesters and post-digesters is circa 50 days and AD occurs under mesophilic (38-42 ℃) conditions. The plant's co-digestion capacity is 130 kt feedstock per year. In 2020, about 60 kt of pig manure, 2 kt of dairy cattle manure and 9 kt of paunch (slaughterhouse) manure were fed to the digesters. Additionally, co-substrates were fed to the digesters, consisting of by-products from the dairy and feed industry (19 kt) and glycerine (3 kt). In 2020, AD of the animal manure and co-substrates resulted in 81 kt of digestate. The average TN and P content of all animal manure fed to the AD over 2020 and 2021 was 5.6 and 1.4 g kg^{-1}, respectively. Furthermore, the average TN and P content of all co-substrates fed to the AD over 2020 and 2021 was calculated at 12 and 2.2 g kg^{-1}, respectively. The added co-substrates influenced the digestate's composition, especially for TN, as the average TN and P content of all digestate produced over 2020 and 2021 was 7.0 and 1.6 g kg^{-1}, respectively.

Of the produced biogas 72% was sold to a nearby dairy processing factory via a 5.5-km long pipeline. The majority of the remaining biogas was fed to two combined heat and power (CHP) engines. The temperature of the last post-digester is permanently increased to 52 °C with heat produced by these engines to achieve hygienisation of the digestate it contains.

2.2 The GENIUS system

The produced digestate is processed in a full-scale processing cascade developed by Nijhuis Saur Industries called the GENIUS system (Figure 1). It has been in operation in various, gradually improved configurations since spring 2019. The GENIUS system was monitored from September 2020 up to and including February 2021. Of all digestate produced over this period, 98.8% was processed, and 1.2% was trucked off-site unprocessed.

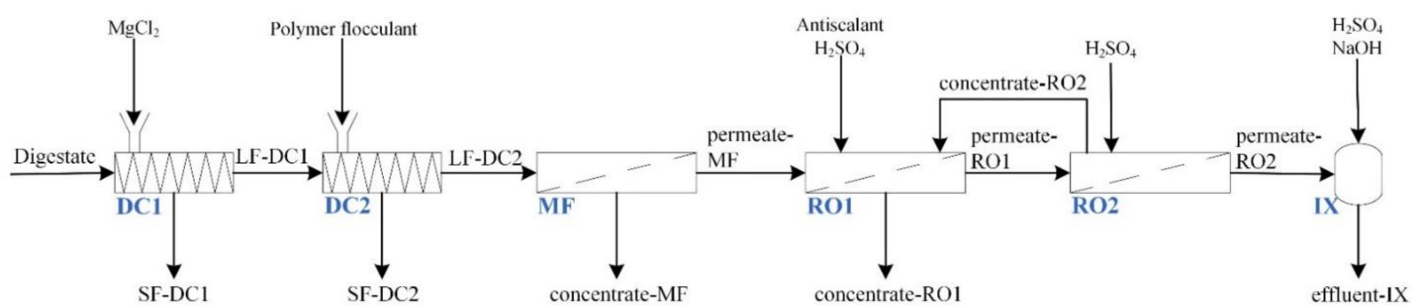

Figure 1. Process scheme of the GENIUS system including the major process streams and dosed additives. Additives: magnesium chloride ($MgCl_2$) solution; polymer flocculant solution, sulphuric acid (H_2SO_4), antiscalant and sodium hydroxide (NaOH). Process streams: solid and liquid fraction of the first decanter centrifuge (SF-DC1 and LF-DC1); solid and liquid fraction of the second decanter centrifuge (SF-DC2 and LF-DC2); concentrate and permeate of the microfiltration unit (concentrate-MF and permeate-MF); concentrate and permeate of the first reverse osmosis unit (concentrate-RO1 and permeate-RO1); concentrate and permeate of the second reverse osmosis unit (concentrate-RO2 and permeate-RO2); effluent of the ion exchanger (effluent-IX).

Digestate is mechanically separated by the first decanter centrifuge (DC1) after the addition of 32% magnesium chloride solution (1.75 L t^{-1} digestate) to precipitate P. The solid fraction of DC1 (SF-DC1) is applied on agricultural land in regions with a demand for P fertiliser at about 250-300 km from the plant. Dosage of polymer flocculant on DC1 is avoided to produce a polymer-free SF that can be further processed by the RePeat system (Regelink *et al.*, 2019) into raw material for potting soil production. The LF-DC1 is, after the addition of polymer flocculant solution to improve the separation of fine solids, separated by the second decanter centrifuge (DC2) in an SF (SF-DC2) and an LF (LF-DC2). The majority of LF-DC2 (about 94%) is subsequently processed by an MF unit and the remaining circa 6% is fed back to the influent of DC2.

From December 2019 until January 2021, part of the concentrate-MF was fed back to the post-digesters. This was not prolonged as the operators observed unwanted accumulation of fine particles in the GENIUS system that impacted its performance. Instead, all concentrate-MF was disposed of as a liquid organic fertiliser. Over the six-month monitoring period, between 0% and 71% (on average 37%) of concentrate-MF was fed back to the post-digesters. The permeate of the MF unit (permeate-MF) is cooled in cooling towers to increase NH_4-N retention in the subsequent RO installation. Both RO units, RO1 and RO2, of the double-pass RO installation consist of three stages. Permeate of RO1 (permeate-RO1) is fed to RO2 and the concentrate of RO2 is fed back to the influent of RO1. The majority of sulphuric acid is dosed on RO2 with smaller amounts dosed on RO1. The concentrate of RO1 (concentrate-RO1) is an NK-rich liquid fertiliser.

To meet the discharge limits, permeate from RO2 (permeate-RO2) is polished by first a degassing tower and second an IX. The purpose of the degassing tower is stripping of carbon dioxide to decrease the treatment load on the IX. The IX consists of a cation exchanger and a subsequent anion exchanger. The IX effluent (effluent-IX) is reused amongst others for the preparation of the magnesium chloride solution and the polymer flocculant solution, the remainder is discharged. The technical specifications and process conditions of each process unit are summarised in Table 1.

	Decanter centrifuges	Microfiltration unit	First reverse osmosis unit	Second reverse osmosis unit
Abbreviation	DC1 and DC2	MF unit	RO1	RO2
Brand (model)	GEA (UCF 465-00-35)	Metawater	Dow Filmtec (SW30 HRLE 400)	Hydranautics (CPA5 MAX)
Membrane type		Ceramic	Polyamide	Polyamide

Filtration surface area	25 m^2	37 m^2	41 m^2
Pore size	0.1 µm		
Operating temperature	35-40 °C	≈ 25 °C	≈ 25 °C
Operating pressure	2-3 bar	30-70 bar	15-23 bar

Table 1: Technical specifications and process conditions of the process units of the GENIUS system.

Maintenance on DC1 and DC2 was performed by an external party every 4,000 operational hours. Additionally, they were cleaned once per week. Cleaning of the MF and RO1 membranes was performed respectively once per month and once or twice per week. The membranes of RO2 were not cleaned. For cleaning the MF membranes 50% citric acid and 10% hydrochloric acid, respectively circa 0.014 and 0.11 kg per t of processed digestate, were used in 2020. Additionally, hydrogen peroxide was used for this. Regeneration of the IX required 1 L sulphuric acid (96%) and 2 L caustic soda (50%) and was performed once per circa 100 m^3 of influent of the IX. This equals circa 0.007 kg sulphuric acid (96%) and 0.01 kg caustic soda (50%) per t of processed digestate. Since June 2019 the membranes of the MF did not need to be replaced. Also, since then the membranes of RO1 have been replaced once or twice per year and the membranes of RO2 were replaced on average twice per year.

In total, 25 continuously logging flow meters, placed on the process streams between the process units and the recirculated process streams and end products, were logged. Additive dosages were logged via the pumping speed.

2.3 Sampling and physicochemical analysis

2.3.1 Sampling

On five days (17 September, 26 October and 8 December 2020, and 12 January and 10 February 2021), samples were taken from all ingoing and outgoing process streams of the previously mentioned process units, including major recirculated process streams. On these days, samples were taken within a period of maximum 3.5 hours. Sampling taps were flushed for at least five seconds before sampling to remove any stagnant liquid present. Sampling bottles were closed immediately after sampling to minimise losses of volatile compounds.

Permeate-RO1, permeate-RO2 and effluent-IX were sampled in 100 ml borosilicate glass bottles with a gas-tight PTFE coated silicone seal in the cap made of TpCh260. These bottles were filled to the brim, such that the headspace was negligibly small. These samples were initially stored at 4

°C and before physicochemical analysis stored maximally for eight weeks at 20 °C. Changes in the matrix of the permeate-RO1 and permeate-RO2 samples during storage are unlikely to have occurred due to the anaerobic conditions in the GENIUS system, resulting in the reduction of any oxidants present. Samples of permeate-RO1, permeate-RO2 and effluent-IX were analysed on physicochemical parameters by the Soil Chemistry Laboratory (CBLB) of Wageningen University & Research.

All other liquid process streams were sampled in 1 L polyethylene bottles. These bottles were filled for about two-thirds of their volume to minimise pressure increase due to biogas formation during storage. The loss of volatile compounds to the headspace of these bottles during storage was minimised by refrigeration. The pH of these samples ranged between 8.2 and 8.5. Therefore at least 85% of NH_4-N would have been present as the non-volatile NH_4^+, minimising NH_4-N losses. The volatilization of relevant amounts of H_2S is not likely due to these pH values. Digestate was sampled downstream of the addition of magnesium chloride solution. Solid process streams were sampled in plastic bags or 1 L polyethylene bottles. These liquid and solid samples were analysed on physicochemical parameters by the laboratory of the 'Landwirtschaftliche Untersuchungs- und Forschungsanstalt Nordrhein-Westfalen' (LUFA NRW) in Münster, Germany. They were stored refrigerated during the three weeks between sampling and analysis.

Analysis of the total organic carbon (TOC) content of the concentrate-RO1 was not performed for the samples mentioned above. Additional concentrate-RO1 samples were taken for this purpose on 27 October 2020, 4 March 2021 and 17 May 2021 and analysed by the laboratory of the 'Landwirtschaftliche Untersuchungs- und Forschungsanstalt Nord-West' (LUFA Nord-West) in Hamelin, Germany. These samples were stored refrigerated for most of the time between sampling and analysis. Also, additional samples of several process streams around RO1 and RO2 were taken, for analysis on S content, on 4 March 2021 and 9 July 2021.

2.3.2 Physicochemical analysis

For the samples analysed by LUFA NRW, the dry matter (DM) content was determined as the weight loss due to drying at 105 ºC for 24 hours. The OM content was determined as loss on ignition at 550 ºC for four hours. Total contents of P, K, S, calcium (Ca), magnesium (Mg), sodium (Na), copper (Cu) and zinc (Zn) were determined after digestion (i.e. boiling for two hours) with aqua regia followed by filtration over filter paper. For liquid samples, this aqua regia digestion was performed on the fresh sample. For solid samples, this digestion was performed on the dried sample. The filtrate was analysed via inductively coupled plasma optical emission spectrometry (ICP-OES).

For liquid samples analysed by LUFA NRW, TN was determined via dry combustion in an elemental analyser. A few drops of diluted acetic acid were added to the fresh sample to prevent the volatilization of NH_3. For solid samples, TN was measured with the Kjeldahl method.

NH_4-N was determined by adding a phosphate buffer solution to the fresh sample and distilling off the NH_3. The NH_3 was collected in sulphuric acid followed by titration of the excess sulphuric acid.

LUFA Nord-West determined the TOC content of the concentrate-RO1 after dry combustion in a CN analyser. pH was determined by mixing 50 ml of sample with 100 ml of 0.01 M $CaCl_2$ solution. After letting the mixture stand for a few hours its pH value was measured by a pH meter. Electrical conductivity (EC) was determined by adding 300 grams of demineralised water to 30 grams of the fresh sample, followed by shaking and filtration. The EC of the filtrate was measured and multiplied 11 times to correct for the dilution. Reported EC values are for a temperature of 25 °C.

The samples analysed by the CBLB (permeate-RO1, permeate-RO2 and effluent-IX) were not acidified. NH_4-N content was determined spectrophotometrically via the Berthelot reaction with a segmented flow analyzer (San++ continuous flow analyzer, Skalar Analytical, the Netherlands). Total contents of P, K, S, Ca, Mg, Na, Cu and Zn were determined via ICP-OES (iCAP 6500 Duo, Thermo Fisher Scientific, USA) or inductively coupled plasma mass spectrometry (ICP-MS) (Element 2, Thermo Fisher Scientific, USA). To prevent volatilization of H_2S before these analyses, no aqua regia digestion was performed. pH was analysed by a pH meter (PHI 34, Beckman Coulter, USA). EC was measured with a conductivity meter (K810, Consort, Belgium) and automatically converted to the reported value for 25 °C.

EC, DM, OM, TN, NH_4-N, P, K, S, Ca, Mg, Na, Cu and Zn contents were measured for all five sampling days and their averages were calculated. If the measured concentration of a certain component was for some samples of a process stream above and other samples below the limit of quantification (LOQ), this was dealt with as follows. For calculation of the average over the samples, the value $LOQ/\sqrt{2}$ was used for concentrations below the LOQ (Croghan and Egeghy, 2003).

DM content of permeate-RO1, permeate-RO2 and effluent-IX was not measured. Instead, it was calculated as the sum of all its measured components, which likely resulted in a slight underestimation. DM content of the dosed additives was calculated as the mass sum of the non-water components they contain. The spent regeneration water was not sampled and analysed, it was assumed to have a negligible DM and salt content. This is plausible because the IX was regenerated often, approximately once per 100 m^3 of permeate-RO2 treated, which had a negligible calculated DM content as well.

Aqua regia digestion is commonly performed in the chemical analysis of animal manure or digestate. Chemical analysis via ICP-OES was tested with and without prior aqua regia digestion to investigate the influence of this digestion on the measured S content. For this, samples of process streams around RO1 and RO2 were taken in duplicate, all in polyethylene bottles, on the 4th of March 2021. Of each duplicate, one sample was analysed by LUFA NRW with prior aqua regia digestion and one sample was analysed by the CBLB without prior digestion. Additionally, three samples of the concentrate-RO1 were taken on the 9th of July 2021. Two in polyethylene bottles and one in a gas-tight borosilicate glass bottle. One of the polyethylene bottles was analysed by LUFA NRW with prior aqua regia digestion, the CBLB analysed the other bottles without prior digestion.

2.4 Mass balances and separation and recovery efficiencies

For the mass of the components DM, OM, TN, NH_4-N, P, K, S, Ca, Mg and Na in the ingoing and outgoing process streams of a process unit the following holds:

$$m_F * w_{Fi} = m_L * w_{Li} + m_S * w_{Si} \qquad \text{Equation (1)}$$

m_F = total mass of the feed of the process unit (kg)

m_L = total mass of the liquid fraction (kg)

m_S = total mass of the solid fraction (kg)

w_{Fi} = mass fraction of component i in the feed of the process unit (g kg^{-1})

w_{Li} = mass fraction of component i in the liquid fraction (g kg^{-1})

w_{Si} = mass fraction of component i in the solid fraction (g kg^{-1})

m_F, m_L and m_S were known for most process streams as day totals of volumetric flow rates measured by flow meters. These were all included in the performed calculations. This was possible as the measured density of the liquid and slurry process streams, having DM contents less than 15%, was very close to 1,000 kg m^{-3}. Densities of the additives were obtained from literature and dosages of all additives, except for the sulphuric acid added on RO1 and RO2, were logged.

For each of the five sampling days, the average volumetric flow rates over two weeks were used, one week before and one week after the sampling day. This was done to reduce the influence of volume changes in cellars and buffer tanks on the measured volumetric flow rates. Also, some process units operated in cycles with small breaks instead of continuously. Therefore, the used two-week daily averaged volumetric flow rates likely differ slightly from the actual volumetric flow rates at the moment of sampling.

For the SF-DC1 and SF-DC2, no volumetric flow rates could be measured because of their high DM content (> 15%). For these process streams, m_S was calculated via Equations (1) and (2).

$$a = (w_{Fi} - w_{Li}) / (w_{Si} - w_{Li}) \qquad \text{Equation (2)}$$

a = fraction of the total mass of the feed of the process unit that ends up in the solid fraction (-)

In Equations (1) and (2) DM content was used as i for DC1 and DC2.

With Equations 1 and 2 average mass balances were calculated over the five sampling days. No data reconciliation was performed, the reported mass balances are therefore not 100% fitting. A process unit's achieved SE for a component to the SF was calculated as described by Svarovsky (1985). SE was defined as the total mass or the mass of an individual component that ends up in the SF of a process unit as a portion of the total mass or the mass of that individual component in the feed (Equation 3).

$$SE = (m_S * w_{Si}) / (m_F * w_{Fi}) \qquad \text{Equation (3)}$$

The recovery efficiency for a component was calculated by dividing the mass of this component that ends up in a specific end product by the mass of this component in the digestate from the main digester (Equation 4).

$$\text{Recovery efficiency} = (m_{eP} * w_{EPi}) / (m_D * w_{Di}) \qquad \text{Equation (4)}$$

EP stands for end product and D for digestate from the main digester.

2.5 Energy consumption and economic evaluation

From 22 until 27 April 2021 and from 1 May until 5 May 2021, electricity consumption measurements of the individual power groups of the GENIUS system were performed for periods of 24 hours each. Over these periods the system functioned normally.

For the system's economic evaluation, a yearly digestate production of exactly 100,000 t and total investment costs of 2 M€ for the process units were used (excluding housing, air treatment installations and logistic facilities). Yearly maintenance costs were assumed to be 12.5% of the initial investment costs. GZV estimated required personnel at 1 full-time equivalent (on top of personnel already employed at the biogas plant). For electricity consumption, a cost of 0.07 € kWh^{-1} was used (price level 2020/2021). Only the following additives, necessary for the system's daily operation, were taken into account: 96% sulphuric acid at 10 € kg^{-1}, 32% magnesium chloride solution at 15 € kg^{-1}, polymer flocculant powder at 3.2 € kg^{-1}, antiscalant at 6 € kg^{-1} and antifoaming agent at 5.5 € kg^{-1}. Chemicals used for cleaning the membranes were not included. Depreciation of the system's investment costs was calculated with Equation (5) (Anon, 1998):

$$Q = C * (r(1+r)^n) / ((1+r)^n-1) \qquad \text{Equation (5)}$$

Where Q is the periodic payment, C is the system's investment cost (2 M€), r is the interest rate (3%) and n is the assumed depreciation period (10 years).

To quantify the differences in mass and transport distance of transported end products before and after the implementation of the GENIUS system, a mass-weighted average transport distance (MWATD) was calculated for both situations following Equation (6):

$$\mathrm{MWATD} = m * d \qquad \text{Equation (6)}$$

Where m is the transported total mass of digestate before implementation of the GENIUS system or the total mass of each end product (SF-DC1, SF-DC2, concentrate-MF, concentrate-RO1 and effluent-IX) after implementation of the system. d is the average distance over which the digestate or each end product is transported.

3 RESULTS AND DISCUSSION

3.1 Composition of process streams and achieved separation efficiencies

Table 2 shows the composition of the major solid and liquid process streams of the GENIUS system. The digestate had a high TN (7.3 ± 0.7 g kg^{-1} FW) and high NH_4-N (5.0 ± 0.3 g kg^{-1} FW) content compared to the average TN content of raw pig manure (4.7 g kg^{-1}) reported by Hoeksma *et al.* (2021) for 17 full-scale pig manure processing systems in the Netherlands. This is partially the result of the co-substrates fed to the AD process. The SF-DC1 had a 1.6-fold higher TN content and a 1.3-fold higher NH_4-N content than the ingoing raw digestate. These results suggest that centrifugation separates organically bound N (organic-N) more efficiently than NH_4-N. The concentrations of P, Ca and Mg in the SF-DC1 were between 4.3 and 6.4 times higher than those in the raw digestate (after the addition of magnesium chloride solution).

On a mass basis, the majority of P and most of Mg in the raw digestate (after the addition of magnesium chloride solution) ended up in the SF, respectively 63 and 84%. In the whole GENIUS system, P and Mg concentrations were highest in the SF-DC1 (8.9 ± 0.8 and 6.4 ± 0.5 g kg^{-1} FW respectively). On the other hand, DM, OM and Ca were, on a mass basis, equally distributed over the two process streams (Figure 2a). For all components, the calculated total mass in the ingoing streams of DC1 was similar to that in the outgoing streams of DC1, except for a 7% higher outgoing total mass for Mg. During the first two sampling rounds and the fifth sampling round, magnesium chloride solution was continuously dosed on DC1, yielding respectively the following SEs for P: 58%, 60% and 64%. In between, during the third and fourth sampling round, no magnesium chloride solution was dosed. No clear difference in the achieved SEs for P between the sampling rounds with and without dosage of magnesium chloride solution was found.

Møller *et al.* (2002) separated anaerobically co-digested pig manure with a decanter centrifuge and observed similar SEs as in the current study. They found, for pig slurry co-digested with, on a mass basis, either 2% fatty waste or 25% other organic waste, SEs to the SF of respectively 60 and 65% for DM, respectively 25 and 31% for TN and respectively 83 and 64% for P. This was achieved without the addition of any salts or polymer flocculants. HRTs of the performed co-digestion were 12 and 16 days and thus considerably shorter than in the current study.

Process streams	pH	EC (mS cm^{-1})	DM (g kg^{-1} FW)	OM (g kg^{-1} FW)	TN (g kg^{-1} FW)	NH_4-N (g kg^{-1} FW)
Digestate[1]	8.2 ± 0.1	47 ± 3	81 ± 4	59 ± 3	7.3 ± 0.7	5.0 ± 0.3
SF-DC1	8.8 ± 0.1	-	313 ± 3	242 ± 5	12 ± 0.4	6.6 ± 0.3
LF-DC1	8.3 ± 0.1	49 ± 3	49 ± 2	32 ± 2	6.8 ± 0.6	4.7 ± 0.3
SF-DC2	8.5 ± 0.3	-	196 ± 10	146 ± 10	15 ± 0.4	5.1 ± 0.3
LF-DC2	8.3 ± 0.1	45 ± 3	31 ± 1	18 ± 1	5.2 ± 0.6	4.2 ± 0.3
Concentrate-MF	8.4 ± 0.1	43 ± 3	49 ± 3	35 ± 2	7.1 ± 0.5	4.2 ± 0.3
Permeate-MF	8.5 ± 0.1	45 ± 3	17 ± 1	6.7 ± 0.7	4.2 ± 0.4	4.0 ± 0.3
Concentrate-RO1	8.4 ± 0.2	89 ± 6	37 ± 5	14 ± 4	8.1 ± 0.8	8.0 ± 0.8
Permeate-RO1	8.7 ± 0.2	7.0 ± 1.1	-	-	0.94 ± 0.17	0.88 ± 0.2
Permeate-RO2	6.9 ± 1.0	0.92 ± 0.50	-	-	0.11 ± 0.06	0.10 ± 0.07
Effluent-IX	5.3 ± 1.1	0.045 ± 0.090	-	-	0.00028 ± 0.00008	0.00020 ± 0.00010

Process streams	P (g kg^{-1} FW)	K (g kg^{-1} FW)	S (g kg^{-1} FW)	Ca (g kg^{-1} FW)	Mg (g kg^{-1} FW)	Na (g kg^{-1} FW)
Digestate[1]	1.7 ± 0.1	4.5 ± 0.2	0.67 ± 0.04	1.8 ± 0.1	1.0 ± 0.1	1.6 ± 0.2
SF-DC1	8.9 ± 0.8	4.6 ± 0.3	1.9 ± 0.1	7.7 ± 0.6	6.4 ± 0.5	1.5 ± 0.2
LF-DC1	0.62 ± 0.07	4.7 ± 0.2	0.51 ± 0.03	0.94 ± 0.05	0.24 ± 0.07	1.7 ± 0.2
SF-DC2	4.6 ± 0.6	4.5 ± 0.3	2.5 ± 0.3	8.5 ± 0.9	2.0 ± 0.7	1.6 ± 0.1
LF-DC2	0.21 ± 0.07	4.1 ± 0.2	0.39 ± 0.24	0.17 ± 0.03	0.061 ± 0.025	1.5 ± 0.2
Concentrate-MF	0.42 ± 0.05	4.1 ± 0.2	0.63 ± 0.28	0.38 ± 0.06	0.11 ± 0.04	1.6 ± 0.2
Permeate-MF	0.077 ± 0.056	4.0 ± 0.2	0.35 ± 0.21	0.027 ± 0.012	0.016 ± 0.015	1.5 ± 0.2
Concentrate-RO1	0.15 ± 0.13	7.9 ± 0.4	1.5 ± 0.5	0.059 ± 0.013	0.040 ± 0.036	3.1 ± 0.4
Permeate-RO1	0.0034 ± 0.0035	0.38 ± 0.06	1.2 ± 1.0	< 0.0012	0.00061 ± 0.00041	0.12 ± 0.03
Permeate-RO2	< 0.00010	0.015 ± 0.006	1.3 ± 1.0	< 0.0012	< 0.00015	0.0034 ± 0.0024
Effluent-IX	< 0.00010	< 0.0004	0.0029 ± 0.0033	< 0.0012	< 0.00015	< 0.0003

[1] sampled after addition of magnesium chloride solution.

Table 2. Physicochemical composition in fresh weight (FW) of the major process streams of the GENIUS system (end products shown in bold). Parameters: pH, electrical conductivity (EC), dry matter (DM), organic matter (OM), total nitrogen (TN), ammoniacal nitrogen (NH_4-N), phosphorus (P), potassium (K), sulphur (S), calcium (Ca), magnesium (Mg) and sodium (Na).

Of the DM mass in the influent of DC1, 47% ended up in the SF. Of the whole GENIUS system, the SF-DC1 was the process stream with the highest DM content (313 ± 3 g kg^{-1} FW). DC2 separated 34% of the ingoing DM mass to the SF (Figure 2b). The SE to the SF for TN was similar for DC1 and DC2, respectively 81% and 79%. For DC2, the SEs to the SF for total mass and the soluble components (NH_4-N and K) were similar. For both decanter centrifuges, the separation of P to the SF was similar to the separation of Ca and Mg to the SF (Figure 2).
During the third sampling round, iron sulphate solution, to improve P separation, and a different polymer flocculant were dosed on the influent of DC2. However, this did not alter the SE for P.

In their review of solid-liquid separation techniques for animal manure, Hjorth *et al.* (2010) found that the DM content of the influent of a decanter centrifuge correlates with the achieved SE for DM. This is in line with our results given that the SE for DM of DC1 was higher than that of DC2 and that the DM content of the DC1 influent (raw digestate) was 1.7 times higher than that of DC2 (LF-DC1). Gravitational separation of colloids by a decanter centrifuge is more challenging than that of larger particles since colloids do not settle by themselves in a standing liquid. The DC2 influent (LF-DC1) therefore likely still contained relatively many colloids compared to larger particles, which will have impacted the achieved SE for DM of DC2.

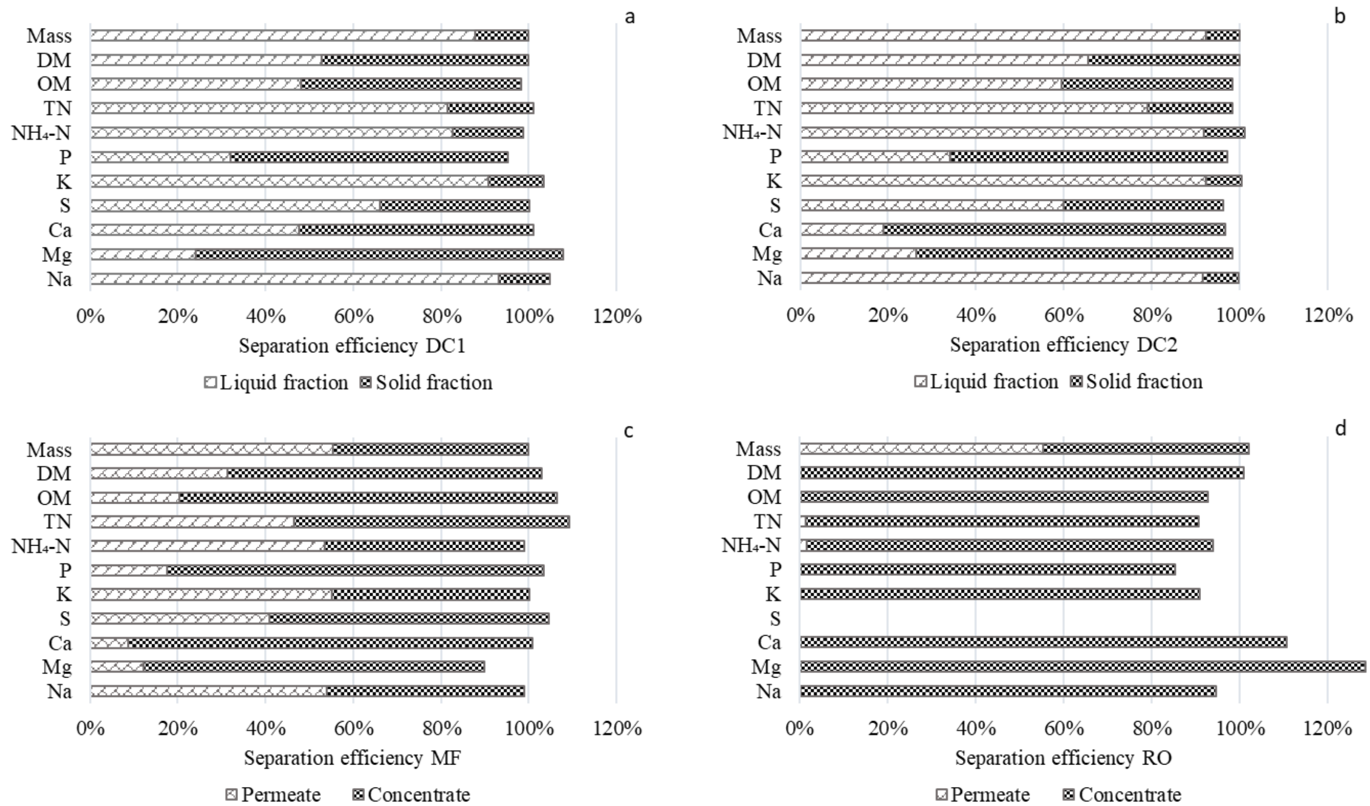

Figure 2. Separation efficiencies of the GENIUS process units: first decanter centrifuge (DC1) (a), second decanter centrifuge (DC2) (b), microfiltration (MF) unit (c) and reverse osmosis (RO) installation (d). Parameters: total mass (Mass), dry matter (DM), organic matter (OM), total nitrogen (TN), ammoniacal nitrogen (NH_4-N), phosphorus (P), potassium (K), sulphur (S), calcium (Ca), magnesium (Mg) and sodium (Na).

The MF membrane, with a pore size of 0.1 µm, retained most OM (86%), P (85%), Ca (92%) and Mg (78%) present in the MF influent. Mazzini *et al*. (2020) found that, as a percentage of the total feedstock, co-digesting more manure from cows and pigs (40.5-84.3% *w/w*), increased the digestate's fraction of total P associated with organic matter and amorphous Fe/Al (extracted with NaOH-EDTA). Therefore, the large share of cow and pig manure, 76% (*w/w*), in the AD feedstock of GZV possibly benefitted the retention of P by the MF unit.

SEs to the concentrate-MF were similar for NH_4-N (46%), K (45%) and total mass (45%), which indicates that all NH_4-N and K in the LF-DC2 is present in a form that can pass the MF membrane (Figure 2c).

Achieved SEs with microfilters strongly depend on the influent type. Mantovi *et al*. (2020) processed, with a screw press, agricultural digestate from livestock manure co-digested with energy crops. The resulting LF was processed with a microfilter (50 µm pore size). Of the total mass, total Kjeldahl N and NH_4-N in the microfilter influent respectively circa 19, 20 and 20% ended up in the MF concentrate. Finzi *et al*. (2021) co-digested 50% (*w/w*) pig and cattle slurry, 10% (*w/w*) poultry manure and 40% (*w/w*) agro-food byproducts. The digestate was processed by a screw press, and the resulting LF was processed with a microfilter (50 µm pore size). In contrast, approximately 70% of total Kjeldahl N and NH_4-N in the influent of the microfilter ended up in the MF concentrate. These high SEs to the MF concentrate are due to 68% of the total mass of microfilter influent ending up in the MF concentrate.

The permeate-MF was processed by a double-pass RO system into concentrate-RO1, containing 8.1 ± 0.8 g TN kg^{-1} FW, 8.0 ± 0.9 g NH_4-N kg^{-1} FW and 7.9 ± 0.4 g K kg^{-1} FW, and permeate-RO2. On average, 99% of this TN was NH_4-N and 1% was organic-N. Due to the high pH (8.4 ± 0.2) of concentrate-RO1, 12% of NH_4-N will be present as NH_3. Concentrate-RO1 has a high Na content (3.1 ± 0.4 g kg^{-1} FW) considering that Na is only a micronutrient, essential to many, though not all, C_4 plants (Kronzucker *et al*., 2013).
SEs were calculated for the RO installation as a whole (Figure 2d). Of the total mass of ingoing permeate-MF, 55% was collected as concentrate-RO1 and 46% as permeate-RO2, whereas all nutrients (NH_4-N, P, K, S, Ca, Mg, Na) primarily (> 85%) ended up in the concentrate-RO1. Contrary to DC1, DC2 and the MF unit, outgoing streams of the RO installation contained in total -15% P and +28% Mg compared to the ingoing stream(s). These calculated differences might be caused by the high variation in P (0.15 ± 0.13 g kg^{-1} FW) and Mg (0.040 ± 0.036 g kg^{-1} FW) content of concentrate-RO1. An earlier, on 24 April 2019, taken sample of concentrate-RO1 showed that at least 89% of, and possibly all, P was present in the form of orthophosphate.

The four times higher S content of concentrate-RO1 compared to permeate-MF is partly caused by volume reduction and partly by the addition of sulphuric acid. The total sulphuric acid dosage on the RO installation, calculated based on this difference in S content and the volumetric flow rates of permeate-MF and concentrate-RO1, amounts to 0.67 kg t^{-1} digestate (0.21 kg of S t^{-1} digestate). S in sulphuric acid is present as the anion SO_4^{2-} that, due to its negative charge, is retained by RO membranes and therefore ends up in the concentrate-RO1. Therefore, it is remarkable that permeate-RO1 and permeate-RO2 had unexpectedly high S contents, respectively 1.2 and 1.3 g S kg^{-1}. These contents are more than three times as high as the measured S content of the permeate-MF. This impossibility hampered the calculation of the SE for S of the RO installation (Table 2d). Concentrate-RO1 analysed via ICP-OES with and without prior aqua regia digestion resulted in three-sample average S contents of respectively 1.6 ± 0.1 and 1.9 ± 0.2 g kg^{-1} FW. This difference might be caused by volatilization, and thereby loss, of H_2S during the digestion due to the addition of acid and heating of the sample. The measured S content of two samples of concentrate-RO1, sampled as a duplicate, of which one was stored in a gas-tight borosilicate glass bottle and the other in a polyethylene bottle, was similar. This observation, in combination with the concentrate-RO1 having a high pH of 8.1, makes it unlikely that volatile losses of H_2S from the polyethylene bottle occurred.

Without aqua regia digestion prior to chemical analysis, measured S contents of permeate-RO1, sampled both before and after the feed tank of RO2, and permeate-RO2 (sampled before the degassing tower) were respectively 0.66, 0.99 and 7.1 g kg^{-1} FW. With aqua regia digestion prior to analysis, the measured S contents of these samples were all < 0.10 g kg^{-1} FW (LOQ). This difference in measured S contents with and without aqua regia digestion is also thought to be caused by the volatilization of H_2S. The S in permeate-RO2 is for circa 99% removed by the degassing tower (data not shown) which, after polishing by the IX, results in effluent-IX containing only 0.0029 ± 0.0033 g S kg^{-1} FW.

For the five sampling rounds, the S contents of permeate-MF and concentrate-RO1 were determined with prior aqua regia digestion, whilst the S content of permeate-RO2 was determined without prior digestion. In the calculated mass balance, in total more S leaves the RO installation via the permeate-RO2 and the concentrate-RO1 than can be explained by the S in the permeate-MF and the calculated sulphuric acid dosage. An explanation for this difference is that in reality the permeate-MF and all preceding process streams, including the digestate, have a higher S content, which was not measured here due to the performed aqua regia digestion.

Most studies on processing the LF of anaerobically digested animal manure with RO have been conducted using UF or nanofiltration (NF) as pre-treatment instead of MF. The separations of TN, NH_4-N and K by the RO installation in the current study are comparable with those for RO (preceded by UF) reported by Chiumenti *et al.* (2013) and Ledda *et al*. (2013). Adam *et al*. (2018) operated a pilot processing cascade for anaerobically co-digested cow manure, consisting of a screw press followed by NF of the resulting LF after which a double-pass RO system processed the NF permeate. The double pass RO system achieved SEs for TN, NH_4-N and K of 94, 93 and 95% respectively, similar values as in the current study.

3.2 Overall mass balances and recovery efficiencies

A mass balance over the GENIUS system and its process units for water and DM was calculated for 1,000 kg of processed digestate (Figure 3). Digestate from the main digesters flowed into the post-digesters, where it was mixed with 7.7 kg of cleaning water and 203 kg of concentrate-MF. Therefore, 1,219 kg of digestate from the post-digesters was, after the addition of 1.8 kg of 32% magnesium chloride solution, processed in the two decanter centrifuges placed in series. DC1 produced 148 kg of SF-DC1 and DC2 produced 96 kg of SF-DC2. Together, DC1 and DC2 removed about 73% of DM and 77% of OM present in the digestate from the main digester. On average, 99 kg of polymer flocculant solution (containing 0.29 kg of polymer flocculant powder) and 7.4 kg of rinse water were added to the influent of DC2. To the 1096 kg of LF-DC2 51 kg of IX regeneration water was added after which the mixture was separated into 673 kg of MF-permeate and 548 kg of MF-concentrate. The latter was partly recirculated to the post-digesters (203 kg).

The 673 kg of MF-permeate was subsequently processed by the RO installation on which, as calculated, about 2.3 kg of 96% sulphuric acid was dosed. In addition, antiscalant was added before RO1, circa 0.015 kg per t of processed digestate from the main digester. The RO installation produced 313 kg of concentrate-RO1 and 374 kg of permeate-RO2. The latter was polished by an IX into purified water.

On average, 18% (183 kg) of the initial digestate mass was, as purified water, discharged to surface water or evaporated in the cooling tower. Of the initial digestate mass, 25% was recovered as SFs (15% as SF-DC1 and 9.6% as SF-DC2) and 65% as concentrated fertilising products (34% as concentrate-MF and 31% as concentrate-RO1). Calculated mass balances over the individual process units and the GENIUS system as a whole were not 100% fitting, as shown in Figure 3 and Table A.1 (Appendix A). This might be caused by small deviations in the measured volumetric flow rates compared to the actual volumetric flow rates and by temporal variation in the composition of process streams.

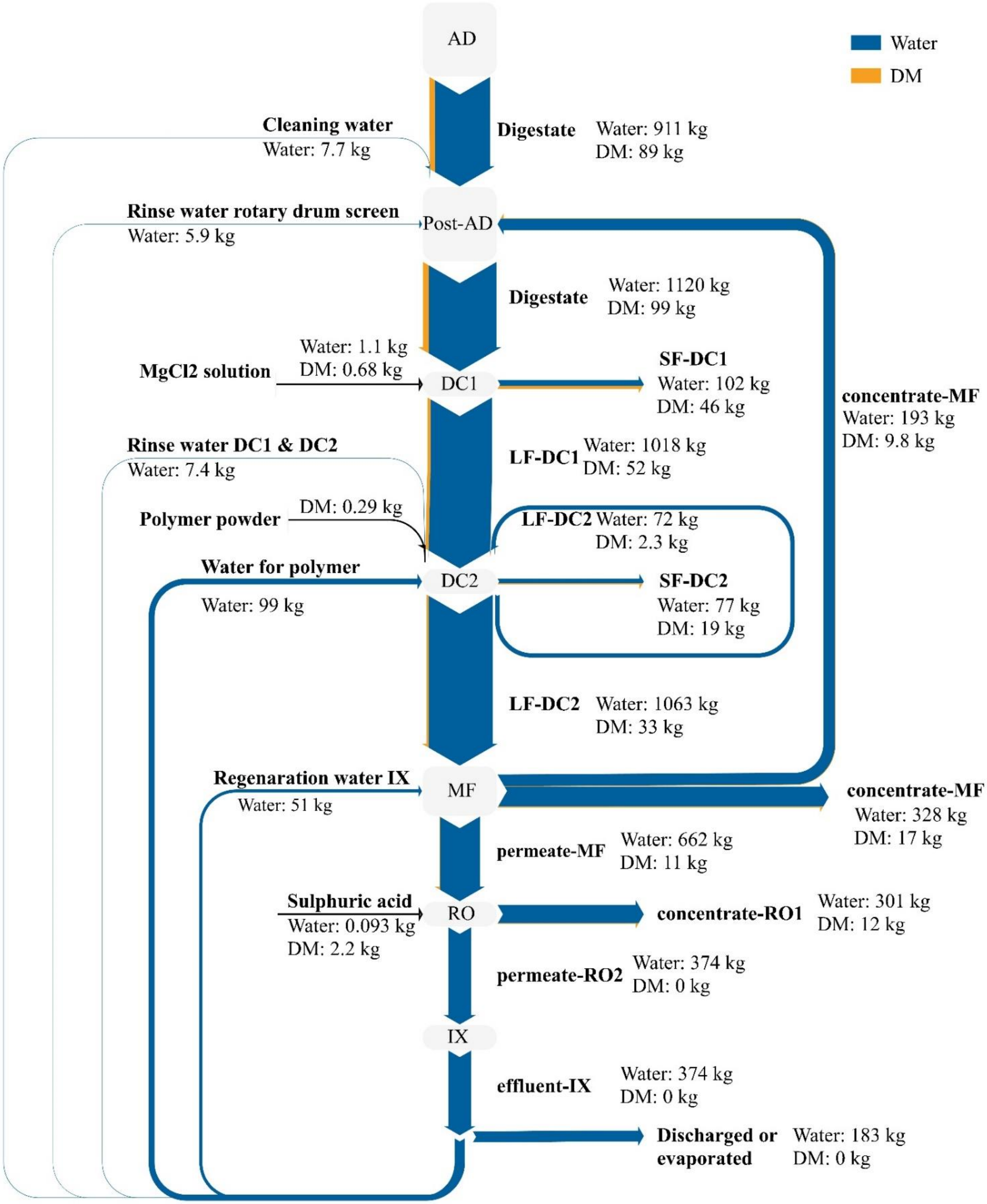

Figure 3. Sankey diagram for water and dry matter (DM) per 1,000 kg of digestate from the main digester processed by the GENIUS system consisting of two decanter centrifuges (DC1 and DC2); a microfiltration (MF) unit; a reverse osmosis (RO) installation and an ion exchanger (IX).

Figure 4 shows the calculated mass balance over the GENIUS system and its process units for organic-N, NH_4-N, P and K per 1,000 kg of processed digestate. NH_4-N was, with 70% of TN, the predominant form of N in the digestate. As no nitrates were found in the concentrate-RO1 and the RO permeates it was assumed that the rest of TN in the digestate was organic-N.

On a mass basis, NH_4-N from the digestate mainly went to the LFs of the decanter centrifuges with the majority ending up in the membrane filtration concentrates (28% in the concentrate-MF and 48% in the concentrate-RO1). Of the digestate's organic-N only 1.4% ended up in the concentrate-RO1.

The NH_4-N:TN ratio, which is important for the leaching of N from fertilisers to ground and surface water, increased from 0.70 in the initial digestate to 0.99 in the concentrate-RO1. Lowering the organic-N content of RO concentrates reduces N losses from their field application (Velthof *et al.*, 2012).

The majority of K in the initial digestate ended up in the membrane filtration concentrates (54% in the concentrate-RO1 and 31% in the concentrate-MF), with 15% ending up in the SF-DC1 and 9.3% in the SF-DC2. Overall, 2.5 kg of both NH_4-N and K were recovered as concentrate-RO1 per t of digestate processed. Most of the P in the initial digestate ended up in the SF-DC1 (66%), with an additional 22% ending up in the SF-DC2 and only 7.2% and 2.2% ending up in respectively the concentrate-MF and the concentrate-RO1.

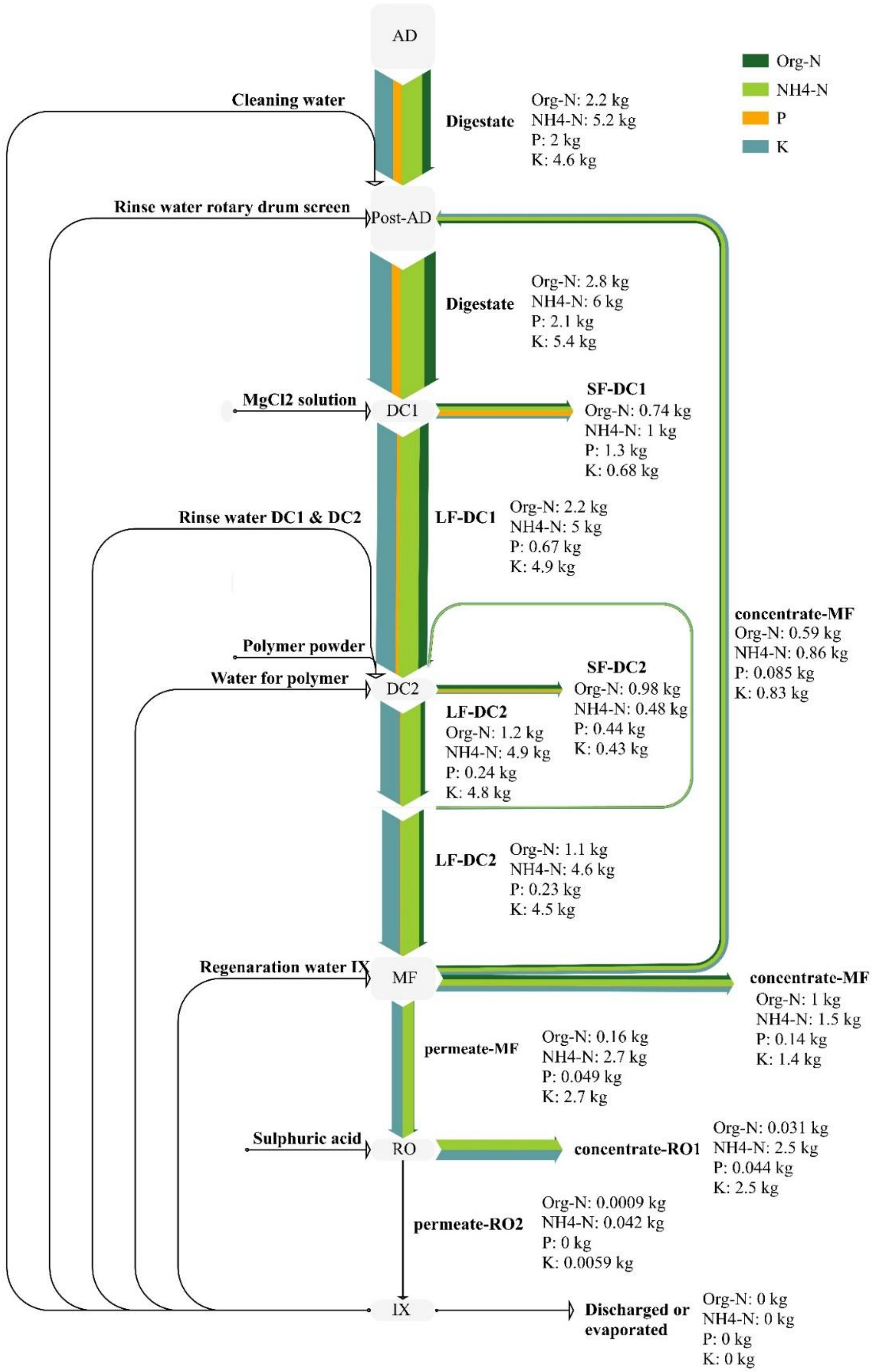

Figure 4. Sankey diagram for organic nitrogen (Org-N), ammoniacal nitrogen (NH_4-N), phosphorus (P) and potassium (K) per 1,000 kg of digestate from the main digester processed by the GENIUS system consisting of two decanter centrifuges (DC1 and DC2); a microfiltration (MF) unit; a reverse osmosis (RO) installation and an ion exchanger (IX).

3.3 Environmental discussion of the end products

For the effluent-IX, the average values of all measured parameters meet the discharge limits of GZV's environmental permit except for pH. The average pH value of 5.3 was 0.2 lower than the lower boundary of the discharge limit (5.5-8.5). However, the pH very easily deviates in such a purified matrix because of its very low buffer capacity. For Cu, the discharge limit (≤ 6 µg L^{-1}) is lower than the LOQ of the performed chemical analysis (10 µg L^{-1}). The other discharge limits, all met, are: total P (≤ 0.3 µg L^{-1}), TN (≤ 6 µg L^{-1}), NH_4-N (≤ 2.5 µg L^{-1}) and Zn (≤ 50 µg L^{-1}). The environmental permit specifically lists individual sample limits for total K (≤ 400 mg L^{-1}) and Na (≤ 50 mg L^{-1}) to which the samples of the effluent-IX easily adhere. In addition, the effluent-IX also meets the governmental water quality criteria set for the Berkel river for a score of 'very well', < 2.3 mg N L^{-1} and < 0.11 mg P L^{-1} (Altenburg *et al*., 2018).

The fertilising products produced by the GENIUS system are a liquid mineral NK fertiliser (concentrate-RO1), a solid organic fertiliser (SF-DC1) and a liquid organic NP fertiliser (mixture of SF-DC2 and concentrate-MF). The composition of the SF-DC1, SF-DC2, concentrate-MF and concentrate-RO1 was compared with the proposed criteria for RENURE materials (RENURE criteria).

Concentrate-RO1 meets all proposed RENURE criteria: its TOC:TN and NH_4-N:TN ratio are 0.78 and 0.98 respectively, its Cu content is < 100 mg kg^{-1} DW and its Zn content is < 550 mg kg^{-1} DW. Hence, concentrate-RO1 could be used as an alternative for synthetic N fertiliser if the European Commission will implement the proposed RENURE criteria.

However, the TN content of concentrate-RO1 is low (8.1 g kg^{-1} FW) compared to synthetic N fertiliser, which hampers its transport over long distances. Its high K content (7.9 ± 0.4 g kg^{-1} FW) makes concentrate-RO1 particularly suitable for crops with a high demand for K such as potato and maize. On the other hand, high application rates (> 50 t ha^{-1} y^{-1}) of K on grassland might cause grass tetany (hypomagnesemia) in cattle (Vaneeckhaute *et al*., 2012, originally from Hillel, 2007, Römheld and Kirkby, 2010 and Grunes and Welch, 1989). The high Na content (3.1 ± 0.4 g kg^{-1} FW) of concentrate-RO1 may be beneficial for the growing of sugar beet (Velthof, 2015). However, applying large amounts of Na via the LF of pig manure or any of its processing products can decrease crop growth due to Na toxicity or high soil salinity (Verlinden, 2005). The S content of concentrate-RO1 (1.5 ± 0.5 g kg^{-1} FW) is low compared to the average S content (3.6 ± 3.1 g kg-1 FW) of RO concentrates produced from raw pig manure in The Netherlands (Hoeksma *et al*., 2021). This is valuable as applying too much S can decrease crop uptake of micronutrients and can result in the leaching of S to ground and surface water. The low S content of concentrate-RO1 is achieved by the low sulphuric acid dosage of the GENIUS system and by avoiding the addition of iron sulphates.

In their agronomic assessment, Schils *et al.* (2015) concluded that the P content (0.2-0.3 g kg^{-1} FW) of the assessed RO concentrates produced from pig slurry, pig slurry digestate or cattle slurry digestate was the factor limiting their application on arable farms in the Netherlands. This is due to an oversupply of P, especially on sandy soils. Concentrate-RO1 could therefore, even though its P content of 0.15 ± 0.13 g P kg^{-1} FW is lower than 0.2-0.3 g kg-1 FW, possibly benefit from having an even lower P content. Regardless, the concentrate-RO1 seems a suitable NK-rich liquid fertiliser for areas where regulations limit P application.

Concerns regarding the field application of RO concentrate produced from (anaerobically digested) animal manure are the occurrence of NH_3 and NO_3^- emissions and a possible lower nitrogen use efficiency (NUE) compared to synthetic N fertilisers. The application of RO concentrate produced from pig slurry was compared with the application of calcium ammonium nitrate (CAN) in a greenhouse experiment with ryegrass (Klop *et al.*, 2012). When surface applied, the NUE for RO concentrate (22-38%) was lower than that for CAN (61%), when injected the NUEs were similar. Injection of this RO concentrate, having a pH of 7.8-7.9, effectively reduced volatile NH_3 emissions compared to its surface application. The concentrate-RO1 produced by the GENIUS system has an even higher pH (8.4 ± 0.2). Therefore, injecting it is important to minimise volatile NH_3 emissions from its field application.

Velthof and Rietra (2019) studied the application of RO concentrate from (anaerobically co-digested) pig slurry in a greenhouse pot experiment with ryegrass and in an incubation experiment. They found that, although soil incorporation of the RO concentrate reduced volatile NH_3 emissions compared to surface application, it did increase emissions of N_2O, a strong greenhouse gas. To minimise both NH_3 and N_2O emissions, they suggest soil incorporation of the RO concentrate with priorly added nitrification inhibitors or acidification of the RO concentrate followed by surface spreading. RO concentrates produced from pig manure have a high buffer capacity due to, amongst others, the carbonates they contain. For their acidification, attention should be paid to the type of acid used to avoid an oversupply of a certain component, for example S, to the soil.

In six field experiments, Schröder *et al.* (2014) found that injection of RO concentrate (containing 90-100% of TN as NH_4-N) yielded a nitrogen fertiliser replacement value (NFRV) of 72-84%, indicating that the applied NH_4-N was only partially available to the crops. The cause for this was unclear, although NH_3 volatilization possibly played a role.

In grassland experiments on sandy and clay soils, van Middelkoop and Holshof (2017) found that the risk of nitrate leaching for shallow injection of RO concentrate produced from pig slurry was similar as for surface-applying granulated CAN or shallow injection of liquid ammonium nitrate.

	TOC:TN	NH_4-N:TN	Cu mg kg^{-1} DW	Zn mg kg^{-1} DW
RENURE material (JRC) [1]	≤ 3	≥ 0.9	≤ 300	≤ 800
SF-DC1 (GZV)	11	0.55	242 ± 134	376 ± 30
SF-DC2 (GZV)	5.5	0.34	69 ± 10	221 ± 16
Concentrate-MF (GZV)	2.8	0.59	112 ± 53	<100
Concentrate-RO1 (GZV)	0.78	0.98	< 100	< 550

[1] For RENURE materials the threshold for either the TOC:TN ratio or the NH_4-N:TN ratio should be met.

Table 3. Proposed compositional requirements expressed as ratios and dry weight (DW) contents for classification as RENURE material compared with the end products of the GENIUS system. Parameters: total organic carbon (TOC), total nitrogen (TN), ammoniacal nitrogen (NH_4-N), copper (Cu) and zinc (Zn).

SF-DC1 and SF-DC2 do not meet the proposed RENURE criteria. However, P was recovered in the form of SF-DC1, SF-DC2 and concentrate-MF without dosing any iron or aluminium salts whose addition might reduce the end product's P availability to crops in the short term. For the SF of digestate from the sugar beet industry, Regelink and Rietra (2021) found a negative correlation between its bioavailable P fraction and its iron:phosphate ratio. The bioavailable P fraction was determined as orthophosphate (P-PO_4), extracted with 10 mM $CaCl_2$ at pH 5.5, as a percentage of total P. SF-DC1 does not contain any added polymer flocculants and is characterized by a high P (8.9 ± 0.8 g kg^{-1} FW) and DM (313 ± 3 g kg^{-1} FW). A study from Regelink *et al.* (2021) revealed that 87% of P in the SF-DC1 is present as easily available P to crops. This was determined using an adapted $CaCl_2$ extraction method. Moreover, SF-DC1 was found to have a slow N release in an incubation experiment with sandy-loam soil, making it useful for soil fertilisation in organic farming (Egene *et al.*, 2021).

SF-DC2 has a low NH_4-N:TN ratio (0.34) and its low DM content (196 ± 10 g kg^{-1} FW) makes transporting it over long distances undesirable. Concentrate-MF does comply with the proposed RENURE criteria thanks to its low TOC content. However, due to its low NH_4-N:TN ratio of 0.59 in combination with its P content of still 0.42 ± 0.05 g kg^{-1} FW, it does not find application in the region as an alternative for synthetic N fertilisers. Currently, the SF-DC2 and the concentrate-MF, together representing 44% of the initial digestate mass, are blended to form a liquid organic fertiliser with a high organic-N:TN ratio. The mixture is a by-product for which there is no market in the region. It is trucked to the northern part of the Netherlands where the demand for organic-N from animal manure is higher.

3.4 Energy consumption

The GENIUS system's measured electricity consumption amounts to 22 kWh per t of digestate processed, which is approximately 8.9 kWh kg^{-1} N recovered as concentrate-RO1. The system consumes no thermal energy. Chiumenti *et al*. (2013) reported energy consumption of 20-25 kWh m^{-3} digestate for a full-scale agricultural digestate processing cascade consisting of a screw press and subsequent decanter centrifuge followed by UF and RO. However, only 1.2-1.4 kg N per m^3 initial digestate was recovered as RO concentrate, whereas the GENIUS system recovered 2.5 kg N per m^3 initial digestate as concentrate-RO1. The energy required to recover a kg N as RO concentrate is nearly twice as low for the GENIUS system. Also, only 78% of TN in the RO concentrate reported by Chiumenti *et al*. (2013) was NH_4-N, compared to 99% for concentrate-RO1. Adam *et al*. (2018) reported a lower electricity consumption of 11.6 kWh t^{-1} digestate for a pilot processing cascade for agricultural digestate, consisting of a screw press followed by NF of the resulting LF after which the NF permeate was processed by a double-pass RO system.

3.5 Economic evaluation

The biogas plant is located in an area with a surplus of animal manure. Before GZV invested in digestate processing, raw digestate was exported to western Germany over distances ranging between 200 and 300 km (250 km on average). Net disposal costs for this export amount to about 20 € t^{-1} digestate (price level 2020/2021), including transport and sampling costs and revenues from the sale of digestate. GZV aimed to reduce net disposal costs by processing the digestate into, amongst others, concentrate-RO1 for the regional market and dischargeable purified water.

Overall costs for the GENIUS system are estimated at 21 € t^{-1} digestate processed, including CAPEX, OPEX and net disposal costs of all end products (price level 2020/2021). CAPEX consists of the amortised investment costs (2.3 € t^{-1} digestate, depreciation period of ten years), whereas OPEX consists of costs for maintenance and labour (2.2 € t^{-1} digestate), electricity consumption (1.5 € t^{-1} digestate) and dosed additives (2 € t^{-1} digestate). The CAPEX and OPEX together amount to 8 € t^{-1} digestate.

Disposal of the end products is still a net cost to GZV. The P-rich SF-DC1 is trucked to western Germany (on average over 300 km) at about 18 € t^{-1}, corresponding to 2.7 € t^{-1} of initial digestate. The mixture of SF-DC2 and concentrate-MF, a sludge, is trucked over roughly 150 km to the northern part of the Netherlands at about 19 € t^{-1}, corresponding to 8.5 € t^{-1} of initial digestate. Concentrate-RO1 is, after blending with a solution of urea and ammonium nitrate and a recovered ammonium sulphate solution, applied in a pilot project as an alternative for synthetic fertiliser within a range of 25 km from the biogas plant. Although the farmers pay to receive this blend, disposal of concentrate-RO1 is still a net cost to GZV as its revenues do not outweigh the costs for storage, chemical analysis, blending, transport and field application. Net disposal costs for the concentrate-RO1 are estimated to be 8 € t^{-1} (2 € t^{-1} initial digestate).

Table 4 shows the calculated MWATD of all end products produced by GZV with and without the implementation of the GENIUS system. Implementation of the system resulted in a calculated 53% reduction in the MWATD. Despite this reduction, the GENIUS system does not yet give an economic benefit over the handling of raw digestate, mostly due to the high net disposal costs of the end products. Reducing the produced volume of concentrate-MF and increasing the produced volume of purified water, which can be discharged for free, would improve the business case.

End product	Total mass (t)	Average distance (km)	MWATD (t * km)
Without GENIUS			
Digestate	1,000	250	250,000 (100%)
With GENIUS			
SF-DC1	148	300	51.800
SF-DC2	96	150	14,400
Concentrate-MF	345	150	51,750
Concentrate-RO1	313	25	7,825
Effluent-IX	183	0	0
Total			118,375 (47%)

Table 4. Mass-weighted average transport distance (MWATD) of the end products produced by Groot Zevert Vergisting with and without implementation of the GENIUS system.

The costs mentioned above refer to the price level of the monitoring years (2020 and 2021). OPEX has since then increased due to a strong increase in electricity and additive prices. Disposal costs for the end products also fluctuate. The performed economic evaluation is therefore only valid for a particular period in time.

Bolzonella *et al*. (2018) reported the costs for a full-scale processing cascade in Italy, for anaerobically co-digested cow or pig manure, consisting of a screw press and subsequent decanter centrifuge followed by UF and RO. They reported a comparable CAPEX (2.74 € t^{-1} digestate) and OPEX (4.23 € t^{-1} digestate) as in the current study. However, the costs for additives (flocculant and solutions for cleaning of the membranes) were six times lower with 0.33 € t^{-1} digestate. Vaneeckhaute *et al*. (2017) reported total costs (CAPEX and OPEX combined) of 12 € per t of animal manure processed for a pilot plant with a double pass RO system in France. In their survey study, de Hoop *et al*. (2011) reported for two full-scale Dutch processing cascades the total costs for processing anaerobically co-digested pig and cattle manure into RO concentrate, including the net disposal costs for all end products. The total costs were 13-14 € t^{-1} of raw animal manure, excluding costs for AD. Both processing cascades consisted of a decanter centrifuge followed by UF and RO.

4 CONCLUSIONS

Implementation of the GENIUS system led to a 53% decrease in the mass-weighted average transport distance (MWATD) for GZV's end products. The produced purified water meets the discharge limits set in the environmental permit of GZV. Of the P in the initial digestate, 98% was removed before RO without adding iron or aluminium salts, whereas 48% of the NH_4-N in the digestate was recovered in the NK-rich RO concentrate. This RO concentrate meets all proposed compositional requirements for RENURE materials and is, therefore, a suitable alternative for synthetic N fertilisers. However, 33% of TN in the initial digestate was recovered as concentrate-MF, which presses the business case. The GENIUS system consumes about 22 kWh_{el} per t digestate processed, and its overall processing costs are 21 € per t digestate (price level 2020/2021) including disposal of all end products.

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APPENDIX A

	Digestate	Additives	Total in (digestate + additives)	SF-DC1	SF-DC2	Concentrate-MF	Concentrate-RO1	Purified water	Total out (end products)	Difference between total out and total in
Mass	1000	5.6	1006	148	96	345	313	183	1084	78 (+7.8%)
DM	89		89	46	19	17	11	0.000	93	4.4 (+4.9%)
OM	64		64	36	14	12	4.1	0.000	66	1.7 (+2.7%)
TN	7.4		7.4	1.7	1.5	2.5	2.5	0.000	8.1	0.77 (+10%)
NH_4-N	5.2		5.2	0.97	0.48	1.5	2.5	0.000	5.4	0.22 (+4.2%)
P	2.0		2.0	1.3	0.44	0.14	0.044	0.000	1.9	-0.057 (-2.8%)
K	4.6		4.6	0.68	0.43	1.4	2.5	0.000	5.0	0.40 (+8.7%)
S	0.69	0.85	1.5	0.27	0.24	0.20	0.45	0.001	1.2	-0.38 (-24%)
Ca	2.1		2.1	1.1	0.81	0.13	0.018	0.000	2.1	0.025 (+1.2%)
Mg	0.86	0.25	1.1	0.94	0.19	0.038	0.013	0.000	1.2	0.080 (+7.2%)
Na	1.7		1.7	0.23	0.15	0.54	0.99	0.000	1.9	0.23 +13%)

Table A.1. Mass balance over the ingoing and outgoing process streams of the GENIUS system in kg per 1000 kg of digestate from the main digester. Parameters: total mass (Mass), dry matter (DM), organic matter (OM), total nitrogen (TN), ammoniacal nitrogen (NH_4-N), phosphorus (P), potassium (K), sulphur (S), calcium (Ca), magnesium (Mg) and sodium (Na).

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