Evaluation of a new approach for swine wastewater valorisation and treatment: a combined system of ammonium recovery and aerated constructed wetland

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HIGHLIGHTS

- A combined ammonia stripping and aerated constructed wetlands system is evaluated at pilot scale.
- 32% of ammonium nitrogen is recovered from swine wastewater by ammonia stripping process.
- An alternative approach to biological nitrification-denitrification treatment is tested.
- Aerated constructed wetland nutrients and organic matter removals are higher than 80%.

ABSTRACT

Nitrate Vulnerable Zones (NVZs) are faced with a surplus of animal manure due to intensive livestock production, and the high use of mineral nitrogen (N) fertilisers in crop production. Recovery of N from animal manure to replace synthetic mineral fertilisers is considered a key strategy to close the N loop for more sustainable agriculture and to meet strict legal frameworks. In this study, N recovery from swine wastewater by an ammonia (NH_3) stripping process followed

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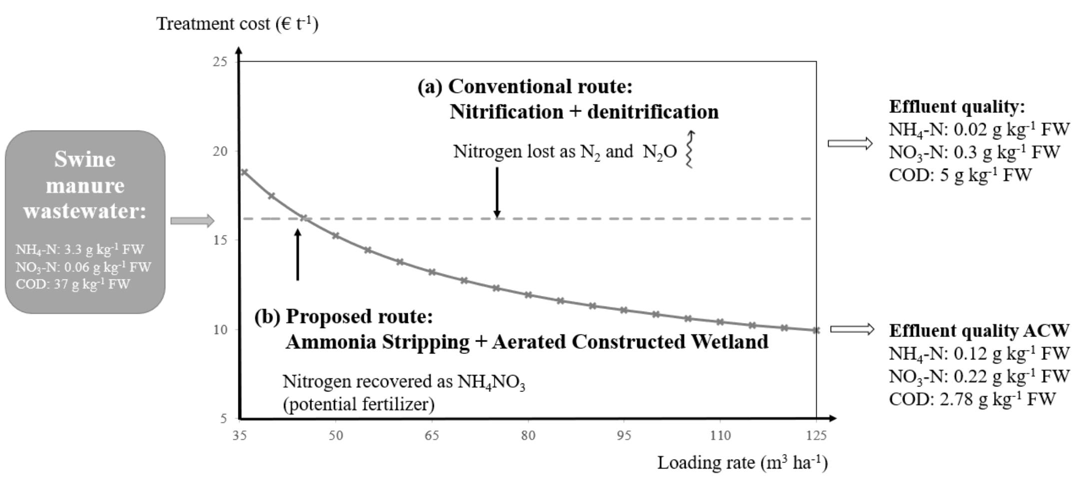
by purification via an aerated constructed wetland (ACW) was proposed as an alternative approach to conventional systems based on biological nitrification-denitrification (NDN) treatment. The performance of the NH_3 stripping pilot as well as the ACW was monitored in 2019-2020 over three periods, to evaluate the quality of recovered ammonium nitrate (AN) solution and the effluent of the ACW. Results showed that the NH_3 stripping unit recovered 21% of total-N (32% of mineral-N) in the form of AN solution. This could be used as a mineral fertiliser according to the criteria of the European Fertilising Products Regulation 2019/1009 and the technical proposal of manure-derived RENURE (REcovered Nitrogen from manURE) products by the European Joint Research Centre. As a RENURE product, AN solution would reach an end-of-manure status and could be used as a synthetic N fertiliser replacement. The tested ACW achieved a high removal efficiency with respect to suspended solids (96%), biological oxygen demand (96%), chemical oxygen demand (90%), total-N (80%), and total phosphorus (97%). The quality of ACW effluent was comparable to that of NDN treatment. Though the overall cost of the proposed pilot-scale process consisting of NH_3 stripping (5.1 € t^{-1}) and ACW (12 € t^{-1}) was calculated slightly higher than conventional NDN treatment (16 € t^{-1}), it is foreseen to outcompete at a higher loading rate (over 45 m^3 ha^{-1} d^{-1}). Furthermore, post-purification will be needed for the ACW effluent to meet the requirements for discharge to surface water.

KEYWORDS

Animal manure, Wastewater treatment, Ammonia stripping, Aerated constructed wetland, RENURE

GRAPHICAL ABSTRACT



1. INTRODUCTION

Due to intensive livestock production, the region of Flanders (Belgium) has a nutrient surplus available in the form of animal manure which cannot be applied directly on agricultural land as it must comply with the Nitrates Directive (EU) 676/1991 application limit of 170 kg total nitrogen (N) ha^{-1} y^{-1}. Therefore, each year 3,700 kt of excess manure, containing 34 kt N, is treated by different manure processing techniques such as biological treatment (i.e., nitrification/denitrification), anaerobic digestion (AD) and drying (VLM, 2020). Swine manure accounts for around 70% of the total input of the Flemish manure processing and is mainly treated by biological process, occasionally preceded by AD. During the biological wastewater treatment, reduced N compounds are oxidized to nitrate (NO_3^-) and subsequently removed as nitrogen gas (N_2) through the nitrification-denitrification (NDN) pathway. As 0.035% of the N load is converted into nitrous oxide (N_2O) during NDN (Kampschreur *et al*., 2009), wastewater treatment contributes almost 5% of the global N_2O emissions (Olivier *et al*., 2017). Recent studies reported up to 0.78% of N_2O losses during the treatment of NH_4-N-rich wastewaters (Wu *et al*., 2014). In 2019, about 16 kt of N were converted into N_2 by biological manure treatment in Flanders (VLM, 2020). As the nitrification of ammonium ions (NH_4^+) to form NO_3^- requires oxidising power of oxygen, i.e., 4.57 g O_2 per g of N oxidised (Magdum and Kalyanraman, 2017), oxygen-rich conditions must be created. Thus, aeration is required resulting in an energy-demanding process. Meers *et al*. (2005; 2008) were the first to propose and subsequently successfully implement constructed wetland (CW) systems for the post-treatment of biologically treated effluents towards dischargeable water. This further increased the sustainability of biological treatment systems as it removed the need to transport biologically treated effluents for spreading on land. Instead, the wetlands allowed in-situ complete treatment from manure towards dischargeable water.

Although intensive livestock is producing surplus N that needs extra treatment, arable farming and horticulture have an additional need for N in the form of mineral fertilisers, which are produced by the energy-intensive Haber-Bosh process. Recovering N as a high-end product and recycling it as mineral N fertiliser could help to overcome this paradoxical situation by reducing the N load in manure processing installations, while partially replacing the demand for mineral fertilisers. Alternative routes to conventional biological removal of N from swine wastewater can be classified as membrane filtration and physicochemical processes. The advantage of such technologies is the simultaneous removal of N, coupled with the production of biobased N fertilising products which are gaining attention as replacements for synthetic mineral fertilisers (Zarebska *et al*., 2015). Ammonia (NH_3) stripping is a robust technology that usually requires simple pre-treatment. It is a two-step process where in the first step NH_3 is transferred from the liquid effluent to the gas phase (NH_3 stripping). Usually, this step takes place in a packed tower with an inert material to enhance NH_3 removal. Subsequently, the gas phase enriched with NH_3 is washed with an acid solution to recover NH_3 in the form of ammonium (NH_4) salts (NH_3 absorption). Sulphuric acid (H_2SO_4), nitric acid (HNO_3), and gypsum ($CaSO_4.2H_2O$) have been recorded at full-scale NH_3 stripping installations as washing agents. The use of H_2SO_4 or $CaSO_4.2H_2O$ would result in ammonium sulphate ($(NH_4)_2SO_4$) solutions, whereas the addition of HNO_3 would form ammonium nitrate (NH_4NO_3, AN) solution (Brienza *et al*., 2020). Compared to $(NH_4)_2SO_4$, AN contains twice the amount of mineral N, thus representing a more interesting mineral N fertilising product (Sigurnjak *et al*., 2019). Recently, the Joint Research Centre (JRC) defined a set of criteria to define which manure-derived products (RENURE products) could be applicable as mineral fertilisers in Nitrate Vulnerable Zones (NVZs), adhering to the same regulations of synthetic fertilisers (Huygens *et al*., 2020). Furthermore, the fertilisers' regulatory framework ascribes to the recently approved Fertilising Products Regulation (FPR) (EU) 1009/2019, which includes manure-derived materials. On the other hand, the reuse of NH_4 salts derived from NH_3 stripping in agriculture is hindered by the Nitrates Directive, which restrains the application of N not only from animal manure but also from manure-derived products (i.e., AN solution). As a result of this limitation, manure-surplus regions (e.g., Flanders, Belgium) recourse to synthetic mineral fertilisers to meet crop N requirements despite the availability of N in manure excess.

NH_3 stripping itself is not effective in removing other components such as organic matter (OM) and phosphorus (P), thus its implementation for swine wastewater treatment should be accompanied by other technologies. The constructed wetlands (CW) for wastewater treatment, also known as treatment wetlands, are engineered systems designed and constructed to utilize natural processes and remove pollutants from contaminated water within a more controlled environment. CWs have been widely used for the treatment of various types of wastewater such as domestic sewage, metallurgical, agricultural, swine manure, mine drainage, landfill leachate, urban runoff, etc., worldwide (Donoso, 2018; Donoso *et al.*, 2019; Gupta *et al.*, 2020; Kadlec & Wallace, 2009;

Li et al., 2020; Maine et al., 2019; Maine et al., 2022; Nguyen et al., 2019; Wu et al., 2015, 2016; Zhang et al., 2014). Yet, some of the constraints could be the low N and recalcitrant OM removal, limited oxygen transfer, and the need for land availability. The effects of aeration and recirculation on constructed wetlands treating swine wastewater have been assessed by Wu *et al.* (2016b), Masi *et al.* (2017) and Lin *et al*. (2020) with the main goal to achieve higher OM and nutrients content removal rates in less time and area of land needed (He *et al.*, 2016; Ilyas and Masih, 2017a,b). It has been shown that in aerated horizontal flow (HF) and aerated vertical flow (VF), N removal is more effective than only horizontal flow or vertical flow designs. In fact, removal efficiencies on N increased by applying intermittent aeration with multiple on-off aeration cycles per day (Dotro *et al*., 2017). In addition, Borin *et al*. (2013) evaluated the performance of different hybrid-constructed wetlands treating swine effluents. Among the hybrid systems presented in their study the system dealing with the highest chemical oxygen demand (COD) and total nitrogen (TN) concentrations reached 4,413 mg COD l^{-1} and 709 mg TN l^{-1}. Comparing Borin's *et al*. (2013) work with the hybrid (vertical and horizontal subsurface flow) aerated constructed wetland (ACW) under evaluation in this study, this latter would treat six to four times higher concentrations.

Although the success of intermittent aeration and recirculation strategies have been established, most were carried out at a lab scale (Feng *et al*., 2020a; Jia *et al*., 2020). Furthermore, a system, which combines NH_3 stripping technology with an ACW at a pilot scale has not been tested before. Considering the facts mentioned above, this study aims to:

- Evaluate the technological potential of NH_3 stripping with HNO_3 as a washing agent and the quality of AN solution recovered
- Evaluate the replacement of biological NDN treatment of swine wastewater by NH_3 stripping together with ACW in terms of nutrient removal and treatment cost.

2. MATERIALS & METHODS

2.1 Conventional manure treatment at swine husbandry farm

The current study was conducted at the swine husbandry farm in Gistel-Zevekote, Belgium, with a capacity to raise 11,000 porkers and 5,400 piglets. The conventional manure processing system consists of an AD for biogas production, a decanter centrifuge for physical separation and a biological wastewater treatment plant for the removal of organic and inorganic residues. Manure is first separated into a solid (SF) and a liquid fraction (LF), and the SF is anaerobically treated. The anaerobic digester has the capacity to yearly process about 12,500 t of manure and co-substrates, producing around 1,400 MWh of electricity. The generated digestate is firstly separated by centrifugation into a LF and a SF. The SF is subsequently composted whereas the LF of digestate is mixed with the LF of manure for subsequent biological NDN treatment (about 29,565 t y^{-1}). The effluent from this biological step needs further purification via a CW to meet the Flemish discharge limits.

2.2 Alternative manure processing for mineral-N recovery and water purification

In the alternative process (Figure 1), the biological treatment was replaced by a two-step treatment consisting of NH_3 stripping and ACW. The NH_3 stripping is a pilot installation developed by Detricon BV (Belgium) with the capacity to process about one tonne of liquid stream per hour. The NH_3 stripping installation is a cylinder with a height of 8 m and a diameter of 3 m. The packaging material is made of steel and has the form of an open cylinder with a diameter of 10 cm. The stream treated in the NH_3 stripping pilot is a mixture of LF digestate and LF manure, usually in a ratio of 1:1. The stripping column is partially filled with packing material and has an air speed of 0.2 - 0.8 m s^{-1}. The air enriched with stripped NH_3 is sent to a scrubber column where 60% HNO_3 solution is added as a sorbent to generate AN solution.

The N-reduced effluent from the NH_3 stripper is then treated by a two-stage pilot process, a vertical subsurface flow (VSSF) and a horizontal subsurface flow (HSSF) ACW. The ACW is based on a concept called "Forced Bed Aeration (FBA)™ developed by Naturally Wallace Consulting (USA). The hybrid ACW (VSSF-HSSF) was divided into two equal parts which were linked with a pressure-driven tubing system, making the effluent of the first part the influent of the second part. The ACW was sealed from the underground by means of a flexible polypropylene liner, 1 mm thick and filled with 100 m^3 of round expanded clay aggregates (Argex®) as substrate. The ACW has a 20 m length, 5 m width, and 1.25 m depth of which 1.10 m are filled with the expanded clay aggregates. This was used because it has a higher specific surface (porous) and it is more economic than gravel in Belgium. The substrate was continuously water-saturated, containing 32 m^3 of water in the pore space. It was equipped with perforated tubes at the bottom of the ACW to provide the required airflow through the water column. For this, 41 pipes with a 12 cm separation between each were incorporated into the system. Dimensioning of the ACW was based on 100 gBOD/m².day on the VSSF part. Figure 2 shows a scheme of the ACW. Aeration was set for 50% of the cycle (240 minutes of aeration followed by 240 minutes off). When aeration was reduced due to clogging of the aeration tubing by iron deposits (seven months after installation), a longer aeration cycle (240 minutes on and 30 minutes non-aeration) was applied.

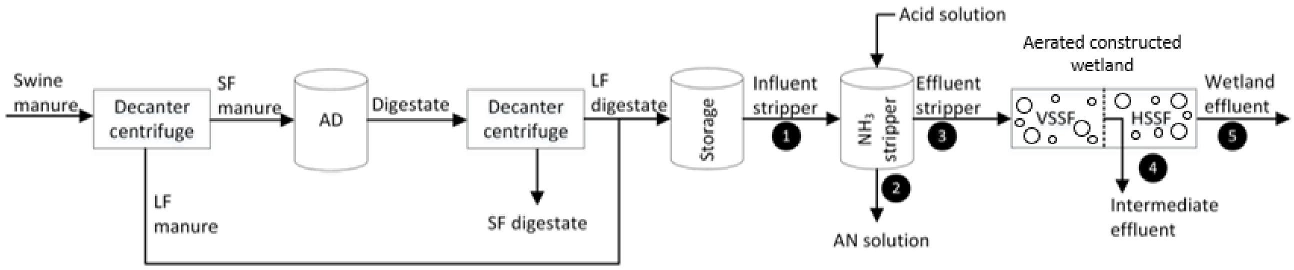


Figure 1. Process flow of the proposed swine wastewater processing steps by subsequent NH_3 stripping and aerated constructed wetland: vertical subsurface flow (VSSF) and horizontal subsurface flow (HSSF). Numbers in black show the five different sampling locations: influent stripper (1), ammonium nitrate (AN) solution (2), effluent stripper (3), intermediate wetland effluent (4) and wetland effluent (5).

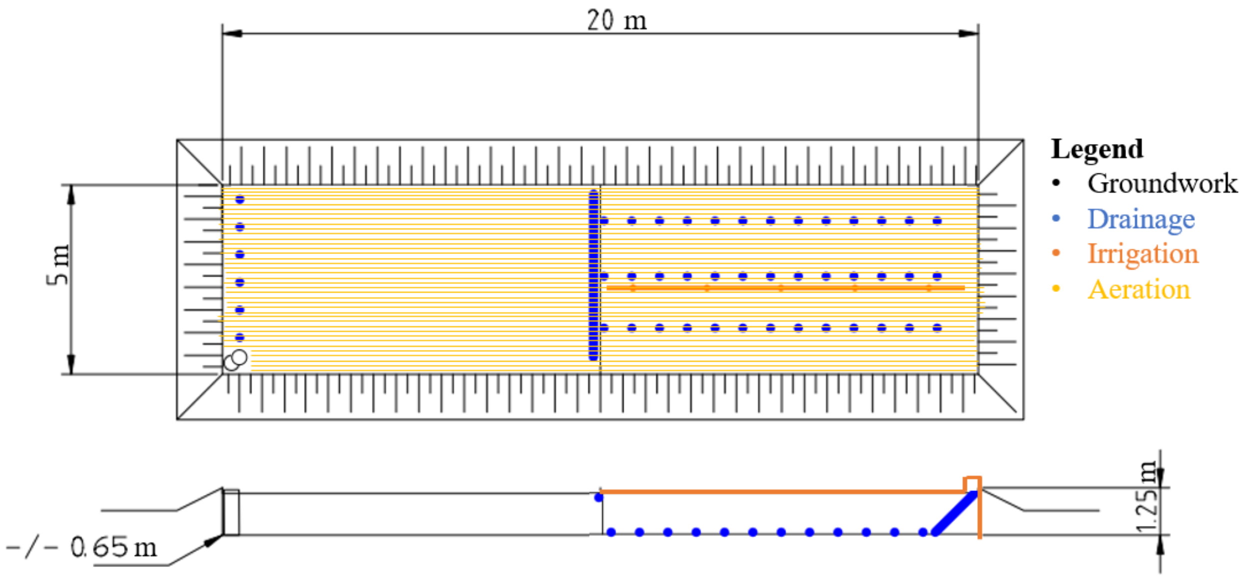


Figure 2: Scheme of the ACW. In black, the groundwork is shown indicating the dimensions of the ACW. At the bottom left the -/- 0,65: represents that the basis of the wetland is 65 cm deep under the rest of the terrain. The 1.25 m to the right indicates the total depth of the basin. The central pipe (in orange) represents the distribution pipe of influent coming from the stripper/scrubber. This pipe is connected to the pump in the pump well. The orange pipe on top of the wetland has holes of 8 mm Ø every 3 meters. There are 5 holes in total.

2.3 Monitoring and sampling overview

The monitoring of the proposed process was performed over three periods:

- Period 1: from May 9th, 2019, to August 18th, 2019
- Period 2: from August 18th, 2019, to December 10th, 2019
- Period 3: from September 10th to December 3rd, 2020.

During Periods 1 and 2 samples were collected on a weekly basis, whereas during Period 3 samples were collected once every two weeks. Five different sampling locations, numbered in Figure 1, were considered for monitoring purposes:

1. influent of the NH_3 stripping unit (IS)
2. AN solution
3. effluent of the NH_3 stripping unit (ES), which also corresponds to the influent of the ACW
4. intermediate effluent of the wetland (IW)
5. effluent of the wetland (EW).

The NH_3 stripping installation was operated a few hours a day to ensure enough feed for the ACW. The ES in excess was treated with the conventional biological NDN system in operation at the swine husbandry farm. The NH_3 stripping pilot was monitored over two sampling campaigns (periods 1 and 2), whereas, the ACW was monitored over all three sampling periods. Regarding the ACW, the initial proposed loading rate was 1 m^3 day^{-1}. However, due to the clogging of the aeration tubing during preliminary tests, this was lowered on average to 0.571 m^3 d^{-1} during period 1 and period 2. Period 1 is considered the acclimatisation phase. Between July 18th and August 16th 2019, no data was recorded due to reparations performed to the ACW, during which the ACW was partly cleared and refilled with fresh water coming from the effluent of an adjacent CW that was in operation for 14 years. Once the ACW was filled, ES was fed again during period 2 (0.571 m^3 d^{-1}). Between period 2 and period 3 the ACW was not fed due to nine months of COVID restrictions, after which the third monitoring campaign was carried out (period 3). In this period, the ACW came back into operation at a reduced loading rate (0.357 m^3 d^{-1}) to allow longer retention time, thus higher removal rate.

2.4 Laboratory analyses

After sampling, water samples were immediately transported to the laboratory and stored at 4 °C to be analysed respecting the holding time of each parameter. The executed physicochemical analyses per sample are shown in Table S1 (supplementary material). The values of pH and Electrical Conductivity (EC) were measured directly with a pH probe (Orion Star A211 USA) and a conductivity probe (Orion Star A212 USA). Dry matter (DM) was assessed as the residual weight after 24 h drying at 105 °C. Suspended solids (SS) were measured by filtering a known weight of a sample, drying the filter with the solids, and then weighing the filter to determine the difference between the weight of the clean filter and the filter with solids. Equation 1 shows the formula to calculate SS concentration:

$$SS\ (mg\ g^{-1}) = (Wfss - Wf) / Ws * 1000 \qquad (1)$$

where Wfss (g) is the weight of the filter with suspended solids, Wf (g) is the weight of the clean filter, and Ws (g) is the weight of the sample.

Biochemical oxygen demand (BOD) concentrations were determined through a respirometric method according to the Standard Methods for the Examination of Water and Wastewater (Eaton *et al*., 1998). BOD concentrations (mg kg^{-1}) were determined based on the amount of dissolved oxygen consumed by aerobic biological organisms at 20°C for 5 days of incubation. COD content was determined through the spectrophotometric method using NANOCOLOR® test kits (MACHEREY-NAGEL GmbH & Co. KG), range 15–160 mg l^{-1}. TN, ammonium-N (NH_4-N) and nitrate-N (NO_3-N) concentrations were measured by quick test kits (NANOCOLOR, MN985088, MN985005 & MN985064) respectively. Potassium (K), sulphur (S), calcium (Ca), magnesium (Mg) and sodium (Na) were determined using Inductively Coupled Plasma – Optical Emission Spectrophotometry (ICP-OES Varian MPX, USA). IS, ES and IW were analysed after microwave digestion (10 ml 65% HNO_3), whereas AN solution and EW were analysed after wet digestion (2 ml of 65% HNO_3 + 1 ml of H_2O_2). Cu and Zn were detected following the same procedure but only on AN solution. Total Organic Carbon (TOC) was calculated as the difference between total carbon (TC) and inorganic carbon (IC), previously determined via a C/N analyser (Skalar B.V., the Netherlands).

2.5 Calculations & statistical analyses

2.5.1 Material balance

To evaluate the performance of the NH_3 stripping pilot unit, a material balance was carried out per tonne of swine wastewater processed (IS). The mass of ES was calculated based on the difference in DM content between IS and ES. The mass flow of AN solution was calculated assuming that NH_4-N removed in the stripping step was entirely recovered, and thus no NH_3 losses occurred during the adsorption step. The amount of 60% HNO_3 solution was calculated based on the mass of NO_3-N in the AN solution.

2.5.2 Calculation of recovery and removal efficiencies

Recovery efficiencies (Rc) of the NH_3 stripping unit stands for the mass of TN, NH_4-N and COD in AN solution as a proportion of the total input from the stripper influent (Eq. 2) (Svarovsky, 1985)

$$\% Rc = ((X * Cx) / (Y * Cy)) * 100 \tag{2}$$

where X (kg) is the mass of AN solution; Cx (g kg^{-1} FW) is the concentration of NH_4-N or COD in AN solution; Y (kg) is the mass of IS; Cy (g kg^{-1} FW) the concentration of TN, NH_4-N or COD in the IS.

To determine the percentage of removal efficiencies (Rm) achieved by the ACW design, the difference between the effluent and influent concentrations of the above-mentioned physicochemical parameters was considered (Eq. 3)

$$\% Rm = ((Cw - Cz) / Cw)*100 \tag{3}$$

Cz (mg kg^{-1} FW) the concentration of NH_4-N, NO_3-N, P, SS BOD, or COD in EW; Cw (mg kg^{-1} FW) the concentration of NH_4-N, NO_3-N, P, SS, BOD, or COD in ES.

2.5.3 Statistical modelling and parameters estimate

The difference between the ACW effluent and influent concentrations (Diff EW-ES) of the above-mentioned parameters (pH, EC, SS, BOD, COD, TN, NO_3-N, NH_4-N and P) were considered as the response variables. Two models were contrasted to test if the design parameters (air temperature, rainfall, and flow) influence the response variables for the tested parameters. The ordinary least-squares (OLS) linear regression model and the robust linear model (RLM) were selected to check for inference robustness due to the small sample size availability. As an additional note, missing values in measured parameters were interpolated via the spline function.

These models check the influence of the design parameters for each of the sampled days and then predict the difference (Diff EW-ES) for each. Eq. (4) describes the resulting model, as follows

$$\text{Diff EW-ES} = \beta 0t + \beta 1t\ \text{Air\_temp} + \beta 2t\ \text{Rainfall} + \beta 3t\ \text{Flow} + \mu t \quad (4)$$

Where Diff EW-ES is the average of the difference between the effluent and influent concentration of the parameter under study; β the estimated coefficient of the design parameter; t equals time or sampled day, β0 the intercept unconditional value of the difference, and μ the stochastic measurement error.

Table 1 shows the minimum and maximum reported percentages of removal efficiencies for hybrid and VSSF CWs treating similar types of wastewater under similar environmental conditions when this study was carried out. The presented ranges were used in the OLS and the RLM models to test if the achieved removal efficiencies by the studied design were between the range of what has been reported in literature.

Table 1. Removal efficiencies of hybrid and VSSF constructed wetlands treating similar types of wastewater reported in literature.

Parameter	Minimum removal efficiency (%)	Maximum removal efficiency (%)	Reference
EC	76	86	(Vázquez et al., 2013)
SS	40	80	(Klomjek, 2016; Torrens et al., 2020)
BOD	75	94	(Torrens et al., 2020)
COD	52	79	(Borin et al., 2013; Maine et al., 2019)
TN	64	75	(Gonzalez et al., 2009; Borin et al., 2013)
NH_4	60	87	(Comino et al., 2013; Maine et al., 2019)
NO_3	53	86	(Borin et al., 2013; Comino et al., 2013)
P	61	87	(Borin et al., 2013; Comino et al., 2013)

2.5 Economic assessment

To address the financial viability of the proposed system in comparison with conventional biological NDN treatment, an economic assessment of the NH_3 stripping step and the ACW was carried out. The cost for upstream mechanical separation of manure and digestate via decanter centrifuge was excluded from the study, as this step is also necessary prior to conventional NDN. A Cost-Benefit Analysis (CBA) was performed to evaluate the economic viability of the NH_3 stripping pilot. The capital costs for the pilot amounted to 250,000 € and included housing, tubing and valves, electro-mechanical compounds and heat exchanger, electrical board and PLC, ventilator, external heating, storage for nitric acid (22 m^3), storage for AN solution (100 m^3) and, sensors (pH, conductivity, temperature, pressure). The investment was amortised following Anon (1998) (Eq 5).

$$Q = C * (r (1 + r)^n) / ((1 + r)^n - 1) \quad (5)$$

where Q represents the periodic amortisation period, C the total investment, r the interest rate (3%) and n the lifespan of the installation (10 years). It was considered that the pilot requires 0.2 full-time equivalent (FTE) to be operated (Detricon, personal communication). Insurance, maintenance, and personnel cost represented 0.24%, 1.4% and 2.5% of the capital cost. Overall, they amounted to 10,488 € y^{-1}. The pilot has an annual working capacity of 8,000 h, meaning processing about 8,000 t y^{-1} of swine wastewater. During the sampling campaign, the electricity requirement of the installation was recorded onsite for four batches: 11 ± 4 kWhel t^{-1} processed for the stripping batch and 1.9 ± 0.27 kWhel t^{-1} to empty the stripped effluent and refill the batch with a fresh mixture of stripper influent. Since the farm is provided with solar panels and generates the electricity necessary for the operation of the pilot plant, the energy costs were not included in the economic evaluation. An estimation of the potential cost was carried out considering 0.10 € kWh^{-1}. For the scrubbing step, the cost of 60% HNO_3 solution and tap water amounted respectively to 200 € t^{-1} and 0.15 € t^{-1}. To evaluate the effect of the increasing energy costs on fertilising commodities prices, a comparison between urea prices over the last five years against the calculated price for AN solution in this study (2019) and in March 2022. The cost of 60% HNO_3 for AN solution production was retrieved at 200 and 795 € per tonne of acid solution used in this study and in 2022, whereas urea prices were obtained from the Index Mundi data warehouse. As previously mentioned, the cost of electricity for the operation of the NH_3 stripping pilot was neglected.

As part of the CBA, the potential benefit from the trade of AN solution was calculated considering a price of 650-750 € t^{-1} N (NUTRIMAN project, 2019).

Regarding the ACW, the cost assessment was performed for a large-scale system, assuming that all wastewater generated at the swine husbandry farm (29,565 t y^{-1}) would be treated via subsequent NH_3 stripping and ACW. The cost for the initial investment was set at 150 € m^{-2}, of which 10% was for the aeration infrastructure and 90% for the construction of the wetland. Maintenance costs were estimated at 2,000 € y^{-1} (Rietland BV, personal communication). The purchase of the agricultural land necessary was defined at 70,000 € ha^{-1} (Notaris, 2021). Following Eq. (5), investment for the wetland and the aeration were amortised at 10% and 20% respectively. The land purchase was amortised at 20 years with an interest rate set at 3%. Electrical energy consumption was estimated considering that the pilot ACW was implemented with a 0.8 kW blower set to work 50% of the time. To study the effect of the ACW feeding rate on the total cost of the proposed process, a single variable sensitivity analysis was included. The range of feeding rates considered was between 0.357 and 1.5 m^3 d^{-1} (36-125 m^3 ha^{-1}). For the analysis, it was assumed all other costs were not to vary.

The cost for the treatment of the (digested) LF of manure with the conventional biological NDN system at the swine husbandry farm was determined as follows. The investment was derived knowing that the daily treatment of 50,000 t ranges between 14 and 24 € t^{-1} (Santonja *et al*., 2017) and is amortised following Eq. (5) with an equal interest rate amortisation period. This resulted in an investment cost ranging between 1.6 and 2.8 € t^{-1}. Operational costs included the treatment process (9 € t^{-1}), as well as the disposal of final effluent and sludge (4.4 € t^{-1}) following Derden (2020). Overall total cost was defined at 16 € t^{-1}. It must be considered that treatment costs can increase up to 14 € t^{-1}, depending on the use of chemical additives. Thus, the overall cost can be as high as 21 € t^{-1}.

3. RESULTS & DISCUSSION

3.1 NH_3 stripping unit

3.1.1 Characterisation of ingoing and outgoing streams

Table 2 summarises the physicochemical composition of the investigated NH_3 stripping streams. The stripping phase increased the pH from 8.0 ± 0.35 to 8.5 ± 0.36. Given the fact that CO_2 is about 1,000-fold more volatile than NH_3, an increment in pH is usually ascribed to CO_2 stripping (Crittenden *et al*., 2012). Removal of NH_4-N resulted in a lower EC, TN, and NH_4-N of ES. Even though the NH_3 stripping system did not influence the SS composition of the treated wastewater, it contributed to the reduction of total COD content. The IS treated was characterised by high COD content, probably due to a poor separation step or a low OM degradation during the AD step. On average, the COD content decreased by 13% from 37 ± 4.9 to 32 ± 8.8 g kg^{-1} FW after approximately 1 hour of HRT at ambient temperature. Finally, no effect was found on NO_3-N and all other macronutrient content, which stayed stable before and after air stripping. Based on laboratory scale experiments, Bonmatı and Flotats (2003) observed a reduction of COD between 20 and 30% when air stripping was applied to raw and digested swine manure.

AN solution (21%) was characterised by high EC (246 ± 9.4 mS cm^{-1}) due to the presence of N ionic compound (NH_4^+ stripped from IS and NO_3^- added via HNO_3) and a neutral pH. The NH_4-N: TN ratio decreased from 0.66 in IS to 0.58 in ES and reached the highest value of 1 in AN solution (81 ± 14 g TN kg^{-1} FW). The presence of COD and other nutrients in the AN solution was negligible.

Table 2. Recorded composition (mean ± standard deviation) on fresh weight (FW) of influent NH_3 stripper (IS), effluent NH_3 stripper (ES), and ammonium nitrate (AN) solution in Periods 1 and 2.

	Unit	IS	ES	AN solution
pH		8.0 ± 0.35	8.5 ± 0.36	6.2 ± 0.25
EC	mS cm^{-1}	35 ± 3.5	28 ± 5.8	246 ± 9.4
DM	g kg^{-1} FW	35 ± 3.3	34 ± 5.2	210 ± 9.0
SS	mg g^{-1} FW	17 ± 3.5	16 ± 7.4	-
COD	g l^{-1} FW	37 ± 4.9	32 ± 8.8	0.52 ± 0.19
TN	g kg^{-1} FW	5.1 ± 0.75	4.0 ± 1.0	81 ± 14
NH_4-N	g kg^{-1} FW	3.3 ± 0.49	2.2 ± 0.62	39 ± 2.6
NO_3-N	g kg^{-1} FW	0.063 ± 0.012	0.072 ± 0.041	39 ± 4.6
P	g kg^{-1} FW	0.33 ± 0.041	0.33 ± 0.080	0.074 ± 0.0093
K	g kg^{-1} FW	4.1 ± 0.34	4.2 ± 0.65	1.4 ± 0.15
S	g kg^{-1} FW	0.54 ± 0.072	0.58 ± 0.15	0.37 ± 0.061
Ca	g kg^{-1} FW	0.73 ± 0.12	0.77 ± 0.22	0.51 ± 0.061
Mg	g kg^{-1} FW	0.15 ± 0.044	0.15 ± 0.058	0.080 ± 0.0052
Na	g kg^{-1} FW	1.4 ± 0.069	1.5 ± 0.043	0.58 ± 0.085

3.1.2 Material balance and mineral nitrogen recovery

A thorough material balance of macronutrients was assessed for the NH_3 stripping unit relying on 60% HNO_3 solution as an absorption agent. The treatment of 1 t of swine wastewater resulted in the production of 27 kg of 21% AN solution, amounting to 1.1 kg of NH_4-N recovered per tonne processed in the NH_3 stripping unit. The recovered NH_4-N makes up 50% of the TN content of the produced fertilising solution because the added HNO_3 solution (absorption agent) provides the remaining 50% in the form of NO_3-N. The use of HNO_3 instead of H_2SO_4 results in higher N concentrations in the recovered NH_4 salts which translates into important agronomic advantages. Overall, 21% of TN (32% of NH_4-N) contained in IS was recovered in the form of AN solution

(81 ± 14 g kg^{-1} of TN). Since only mineral-N is removed during the process, the amount of organic-N was not affected (Figure 3). This NH_4-N removal efficiency was achieved neither by heating the ingoing mixture of LF manure and LF digestate nor by adding any base to increment pH conditions. Therefore, these results represent the removal efficiencies with the lowest energy input and chemical use. It can be expected that the removal efficiencies can be significantly increased at higher temperatures and pH conditions (Zarebska *et al*., 2015).

As a result of the material balance performed in this study, the consumption of 60% HNO_3 amounted to 7.7 kg kg^{-1} N recovered. Similarly, Brienza *et al*. (2021) reported the use of 7.3 kg of 50% H_2SO_4 and 8.4 kg of $CaSO_4.2H_2O$ (75% DM) to recuperate 1 kg of N in different full-scale NH_3 stripping units.

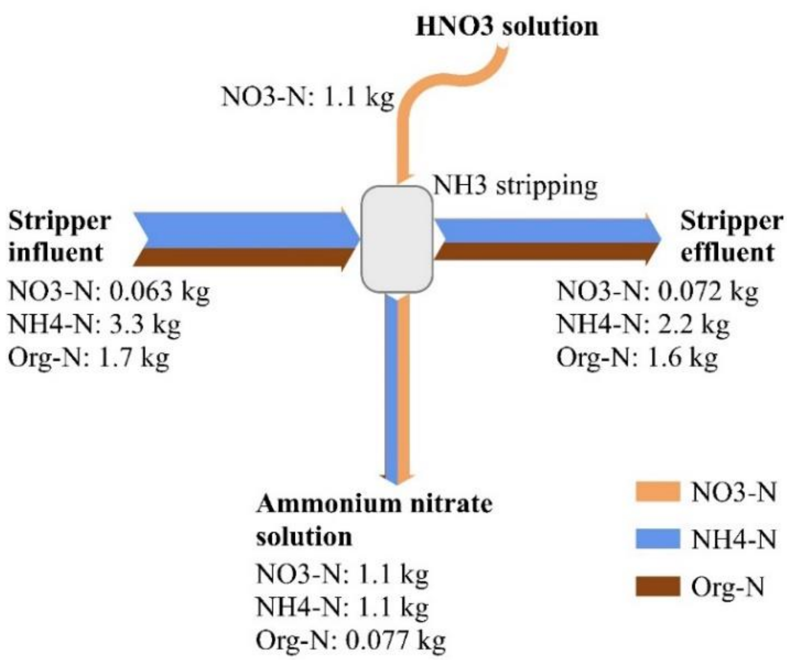


Figure 3. Material balance of organic nitrogen (Org-N), nitrate nitrogen (NO_3-N) and ammonium nitrogen (NH_4-N) for 1 tonne of animal wastewater processed.

Regarding all other macronutrients (P, K, S, Ca, Mg and Na), their contents were similar before and after NH_3 stripping and thus their mass flow resulted in equilibrium (Table 3). Differently from vacuum NH_3 stripping (Brienza *et al*., 2021), the ambient conditions operated by Detricon did not result in water evaporation and up-concentration of the IS components. On the other hand, although air stripping contributed to removing around 13% of the COD from IS, less than 1% was found in the recovered AN solution. Bonmatı and Flotats (2003) recorded COD losses higher than 5% when H_2SO_4 was used to recover stripped N. These results suggest that neither HNO_3 nor H_2SO_4 successfully fixed volatile organics, and therefore further air treatment is required to prevent detrimental effects on the environment. Yet, further research should aim to investigate possible COD degradation pathways during NH_3 stripping.

Table 3. Material balance of the NH_3 stripping unit in the Periods 1 and 2.

Parameter	IS (kg)	ES (kg)	60% HNO_3 solution (kg)	AN solution (kg)
Mass	1000	998	8.1	27
Water	965	965	3.2	22
DM	35	33	4.8	5.7
COD	37	32	-	0.014
P	0.33	0.33	-	0.0020
K	4.1	4.2	-	0.038
S	0.54	0.58	-	0.010
Ca	0.73	0.77	-	0.014
Mg	0.15	0.15	-	0.0022
Na	1.4	1.5	-	0.016

The implementation of NH_3 stripping technology to the AD of animal manure has been investigated at both pilot and full-scale. On a pilot scale, Pintucci *et al*. (2017) recovered between 35 and 39% of NH_4-N, depending on the air recirculation rate. Bolzonella *et al*. (2018) monitored a system where the LF of digested swine and cow manure entered in an NH_3 stripping unit and 22% of TN was recovered from the stripper influent, resulting in ammonium sulphate solution (26 g kg^{-1} TN). In 2018, Baldi and co-authors conducted a series of trials at different operative conditions and recorded NH_4-N removals ranging between 22% and 66%. Brienza *et al*. (2021) monitored a full-scale vacuum side stream NH_3 stripping installation relying on flue gas desulphurisation with $CaSO_4.2H_2O$ as an absorption agent. The authors recorded 31% of TN (57% of NH_4-N) recovery from raw digestate in the form of ammonium sulphate solution (46 ± 3.6 g kg^{-1} of TN). Differently from the two previous cases, Ledda *et al*. (2013) described a digestate processing cascade where LF digestate was processed in a membrane filtration system and subsequently its retentate flowed in an NH_3 stripping system. The N recovery rates differed when digestate originated from cattle or swine manure. The former resulted in 74% TN recovery (78% NH_4-N), while the latter led to 71% recovery of TN (73% of NH_4-N) from the ingoing reverse osmosis retentate. In both cases, H_2SO_4 was used as an absorption agent, generating ammonium sulphate solution (51-61 g kg^{-1} TN). The efficiency of the NH_3 stripping pilot in our study achieved 21% of TN (32% of NH_4-N), which is overall equal to or lower than the literature results. However, this was achieved at ambient pH and temperature. Also, the high COD content could have jeopardised the rate of NH_3 volatilisation, due to the binding of NH_4^+ by OM (Kinniburgh *et al*., 1996; Hafner *et al*., 2006), limiting the efficiency of the stripping process.

3.1.3 Agricultural value of ammonium nitrate solution

The biobased AN solution generated by Detricon pilot plant fulfils all quality criteria needed to be recognised as both RENURE product (Huygens *et al*., 2020) and as straight liquid inorganic macronutrient fertiliser (PFC 1(C)(I)(b)(i)), according to the European FPR (EU) 1009/2019 (Table 4). TN content is 1.6 times of the minimum content required and TOC is 40-fold lower than the maximum allowed by the FPR. In the proposal drafted after the public consultation, the amendments add a new component material category (CMC 15, "Recovered high purity materials"), which would include NH_4 salts if these are 95% pure, (with no more than 0.5% of OC) on DM basis. If so, AN solution from Detricon may be used as mineral fertiliser under the same prescriptions of synthetic fertilisers.

Regarding RENURE criteria, AN solution complies with both the maximal TOC:TN and the mineral N:TN ratio, despite it being sufficient to meet just one of the two. Regarding Cu and Zn, their content in the fertilising solution is largely below RENURE and FPR limits. According to its compositional characteristics, the AN solution generated by Detricon represents an interesting option to replace synthetic N fertilisers and to recycle mineral-N of manure origin. Currently, ammonium sulphate generated from NH_3 stripping plants and retentate from membrane filtration installations have demonstrated to meet RENURE quality standards proposed by the JRC (Brienza *et al*., 2021; van Puffelen *et al*., 2022).

Table 4. Characteristics requirements for the denomination of different fertilisers defined by the Fertilising Product Regulation (EU) 1009/2019 and Joint Research Centre (JRC) RENURE products (Huygens *et al*., 2020), in comparison with biobased ammonium nitrate generated by Detricon (FW: fresh weight; DW: dry weight).

Fertiliser type	TN (g kg^{-1} FW)	TOC (g kg^{-1} FW)	TOC:TN	mineral-N_l:TN (%)	Cu (mg kg^{-1} DW)	Zn (mg kg^{-1} DW)
PFC 1(C)(I)(b)(i) (Fertilising Product Regulation)	≥ 50	≤ 10			≤ 600	≤ 1500
RENURE product (JRC)			≤ 3*	≥ 90*	≤ 300	≤ 800
Ammonium nitrate (Detricon)	81 ± 14	0.24 ± 0.042	0.0029	100	62 ± 16	118 ± 42

*For RENURE products either the threshold for TOC:TN ratio or NH_4-N:TN ratio should be met.

To evaluate the potential of AN solution as a replacement for broadcast synthetic fertilisers, pot and field trials were set up by Sigurnjak *et al* (2019). The authors also investigated the environmental impact of AN application in terms of postharvest NO_3^- residue. The agronomic performance of biobased AN generated by Detricon was assessed in comparison with calcium ammonium nitrate (CAN) of synthetic origin. Pot experiments were performed on lettuce where the application of AN solution resulted in slightly higher crop yields and consequently N uptake compared to the commercial mineral fertilisation regime. AN performance on a field scale was assessed in maize cultivation, in comparison with a reference treatment of animal manure, and CAN: crop yields and N uptake were similar in both cases. Moreover, the postharvest NO_3-N residue after AN fertilisation was below the amount allowed by the Flemish legislation (90 kg NO_3-N ha^{-1} in 0-90 cm soil) and comparable to the reference treatments (Sigurnjak *et al*., 2019). The neutral pH of this biobased fertiliser reduces the risk of machinery erosion, yet, its application can lead to NH_3 volatilisation and loss in the atmosphere. As such, correct agronomical practices (e.g. injection into soil or fast incorporation after surface application) are required to alleviate adverse environmental effects (Huygens *et al*., 2020).

3.2 Aerated constructed wetland

3.2.1 Removal efficiencies

The percentages of removal efficiencies (Rm) achieved by the ACW design were calculated and are presented in Table 5. Considering that Period 1 represented the acclimatisation stage, average removal efficiencies were determined based on data collected during Periods 2 and 3. The comparison of Rm achieved between the ACW compartments proves the efficiency of the designed system. It is seen that Rm increase from the first to the second compartment and from the beginning to the end of the ACW system. It is important to note that readily biodegradable COD is promptly removed in the first compartment, different from the other parameters, which removal efficiency increases as wastewater passes through the system. Overall, the Rm for all parameters of the whole ACW ranged between 80% and 97%, except for NO_3-N (Table 5).

Table 5. Average concentrations of influent wetland (effluent stripper, ES), intermediate aerated constructed wetland effluent (IW), effluent aerated constructed wetland (EW) and removal efficiencies achieved by the aerated constructed wetland (ACW) by the end of Period 3, compared to effluent composition of typical biological nitrification-denitrification (NDN) treatment plant reported by Lemmens *et al*., (2007) (FW: fresh weight).

Parameter	ES (mg kg^{-1} FW)	IW (mg kg^{-1} FW)	EW (mg kg^{-1} FW)	Rm in the first part of the ACW (%)	Rm in the second part of the ACW (%)	Rm of the whole ACW (%)	Effluent NDN (mg kg^{-1} FW)
SS	14	5.0	0.55	62%	87%	96%	
BOD	2,370	554	80	76%	86%	96%	10 - 100
COD	26,875	13,843	2,787	90%	76%	90%	1,000 - 5,000
TN	2,861	2,186	509	27%	74%	80%	500
NH_4-N	1,788	1,346	119	18%	93%	95%	0 - 20
NO_3-N	67	228	224	-204%	-181%	-632%	250 - 300
P	344	131	8.7	57%	91%	97%	130 - 220

Figures 4 (a), 5 (a) and 6 (a) show higher variability in recorded data given the slow recovery of the ACW system after the performed repairs between periods 1 and 2. Conversely, Figures 4 (b), 5 (b) and 6 (b) show major stability in the system after nine months of continuous work under lower flow, which results in a longer retention time and higher removal rates.

The NO_3-N concentrations increased over time due to limited denitrification combined with effective nitrification of the NH_4-N by the aerated system. OC and NO_3-N concentrations, wetland vegetation, pH, water depth and temperature are parameters that have been assessed to determine their influence on denitrification rates. Among these, the available carbon, NO_3-N concentration, and water depth were the most influential factors (Hunt *et al*., 2003; Songliu *et al*., 2009). In this study, available carbon and high NO_3-N concentration could partially explain the limited denitrification. Readily available carbon sources could be scarce, due to the lack of plant litter as no plants grew in the system, this could happen due to high TN concentrations that resulted in no plants' survival or mainly due to the type of wastewater treated by the system.

Constructed wetlands treating the liquid fraction of piggery manure have to deal with fractions of recalcitrant or non-biodegradable OM which shows high COD concentrations and relatively low BOD concentrations (Donoso *et al*., 2019). Thus, a relatively high COD could imply that there was not sufficient biodegradable OC thus incomplete denitrification prevailed as was observed by Donoso *et al.* (2019) for this type of effluent to be treated. Additionally, Fan *et al*. (2013), Wu *et al*. (2016a), Hou *et al*. (2017) and Donoso *et al*. (2019) concluded that intermittent aeration could favour TN removal when there are longer non-aerated periods than aerated ones. In this study, however, non-aerated periods were reduced to minimise the continuous clogging of aeration pipes with large particles contained in the ES. Another reason that can explain the limited nitrate removal, lies in the prompt availability of ammonia-oxidizing bacteria to promote the oxidation of NH_4^+ to nitrite (NO_2^-), and then to NO_3^- at the beginning of Period 2. However, during mid Period 2 and 3, Figure 4 shows that aeration could have altered the microbial community properties and composition. Shirdashtzadeh *et al*. (2022) after studying factors that influence microbial communities and their behaviour on N removal, reported that among the regulating factors, dissolved oxygen and N concentration significantly influence microbial diversity and composition.

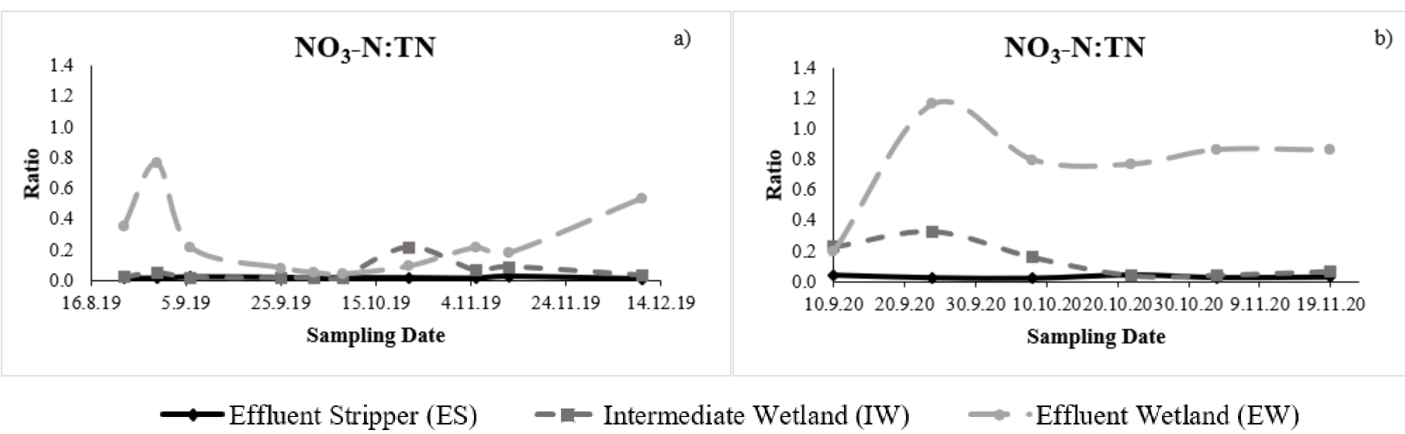


Figure 4. NO_3-N:TN ratio of removal concentrations achieved in the ACW during the second period in (a) and third period in (b) considering the sampling date.

Contrary to the accumulated NO_3-N effect, the ideal conditions encountered in the system for nitrification, (such as oxic conditions and pH values above 6.8) resulted in high NH_4-N removal rates (Figure 5).

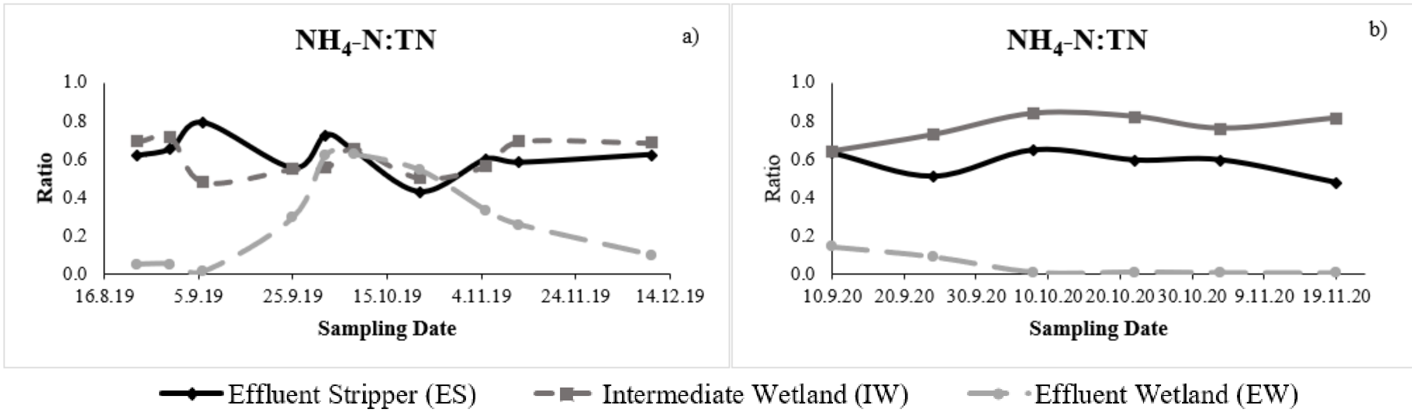


Figure 5. NH_4-N:TN ratio of removal concentrations achieved in the ACW during the second period in a) and third period in b) considering the sampling date.

According to literature (Samudro and Mangkoedihardjo, 2010; Abdalla and Hammam, 2014; Lakhlifi *et al*., 2017), BOD:COD ratios below 0.3 indicate non-biodegradable wastewater. Thus, the smallest recorded ratios were the ones of the wetland effluent where the COD values represent recalcitrant OM. The low BOD:COD ratios in ES (<0.3, Figure 6) but high COD removal rates (90%, Table 5) indicate that this ACW can treat wastewater containing not easily biodegradable OM as the residue after AD and stripping.

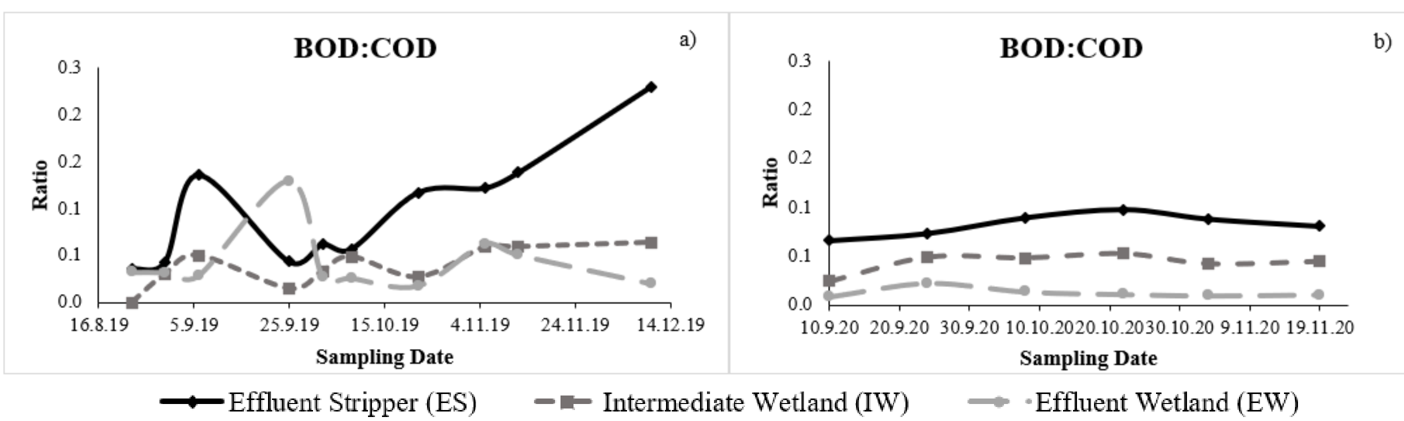


Figure 6. BOD:COD ratio of removal concentrations achieved in the ACW during the second period in a) and the third period in b) considering the sampling date.

Furthermore, the P removal was enhanced due to artificial aeration given the redox, managing with aeration strategies that facilitate processes, such as chemical precipitation and/or binding to iron in the substrate. Ilyas and Masih (2018) reported that major processes participating in P removal are soil sorption and chemical precipitation, whereas plant and microbial uptake play a moderate to low role.

On the other hand, 96% of SS removal is similar to the 97% achieved by Masi *et al*. (2017) in a system that coupled an anaerobic sludge blanket with an intensified constructed wetland (aerated CWs) to treat swine wastewater at pilot scale. At the intensified aerated vertical subsurface flow CWs, the maximum removal was achieved. The processes through which SS are removed are the absorption and retention of inert SS inside the substrate, the biodegradation of OM that converts into biosolids and the transformation of biomass residues into inert solids through microbial endogenous respiration (Hua *et al*., 2013).

Comparing the effluent composition achieved by the NH_3 stripping and ACW system, with the effluent quality of a typical biological NDN treatment plant (Table 5), the EW concentrations are similar for BOD, COD and TN. The P content in EW (224 mg kg^{-1}) is 15-25 times lower compared to the average NDN effluent. Yet, further research would be necessary to establish if this is a long-term removal process or if the ACW will reach a quick saturation. On the other hand, the achieved effluent NH_4-N concentrations of 119 mg.kg^{-1} at the EW compared to the 0–20 mg.kg^{-1} of the effluent NDN indicates that the proposed system did not reach as high removal concentrations of NH_4-N as the NDN and, in consequence, of NO_3-N. Thus, the tested system combing NH_3 stripping and ACW could replace a conventional biological treatment, provided that higher NH_4-N and NO_3-N removal rates are achieved. Increased NH_4-N removals could be achieved for instance by increasing the temperature during the NH_3 stripping step, with the excess heat generated by the AD plant. CW can also contribute to further polishing of the effluent prior to discharge on surface water.

3.2.2 Statistical modelling and parameter estimates

This section presents an interpretation of the results for the two contrasted models, the OLS and RLM. Statistical analyses were conducted for all the response variables of the parameters under study. Appendix A. Supplementary material shows the results and graphs for the run models (Nyieku *et al*., 2021). For illustration purposes and to explain how results were interpreted Table 6 and Figure 7 show OLS results, while Table 7 and Figure 8 present RLM results. The median value (-0.0017) less or close to zero indicates the model can be interpreted, and that there is no indication of specification problems. The statistically significant intercept indicates that BOD concentrations' removal is on average greater than 0. Regarding the control parameters among air temperature, rainfall, and flow, only flow affects statistically the BOD removal according to OLS model. The adjusted R-squared, indicates that for the BOD the design parameters together explain 13% of its variability. Predictions in blue, in Figure 7 indicate that in this study the average removal of the BOD is higher than the removal reported in the literature (75-94%; Table 1), for CWs treating the same type of wastewater working at similar environmental conditions.

Table 6. Ordinary least-squares model output for biological oxygen demand & contrast

lm (formula = BODdiff.interp ~ Air_temp + Rainfall + Flow, data = wetland.data)					
Residuals:					
	Min	**1Q**	**Median**	**3Q**	**Max**
	-0.11689	-0.01248	-0.0017	0.02278	0.06412
Coefficients:					
	Estimate	**Std. Error**	**t value**	**Pr (>|t|)**	
(Intercept)	1.01	3.07e-02	32.99	<2e-16***	
Air_temp	4.05e-05	1.84e-03	0.02	0.98	
Rainfall	-7.42e-03	5.26e-03	-1.41	0.18	
Flow	-6.14e-05	2.86e-05	-2.15	0.05*	
Significant codes	0	0.001 ***	0.01 **	0.05 *	0.1
Multiple R-squared: 0.2567			Adjusted R-squared: 0.1255		
F-statistic: 1.957 on 3 and 17 DF			p-value: 0.1589		

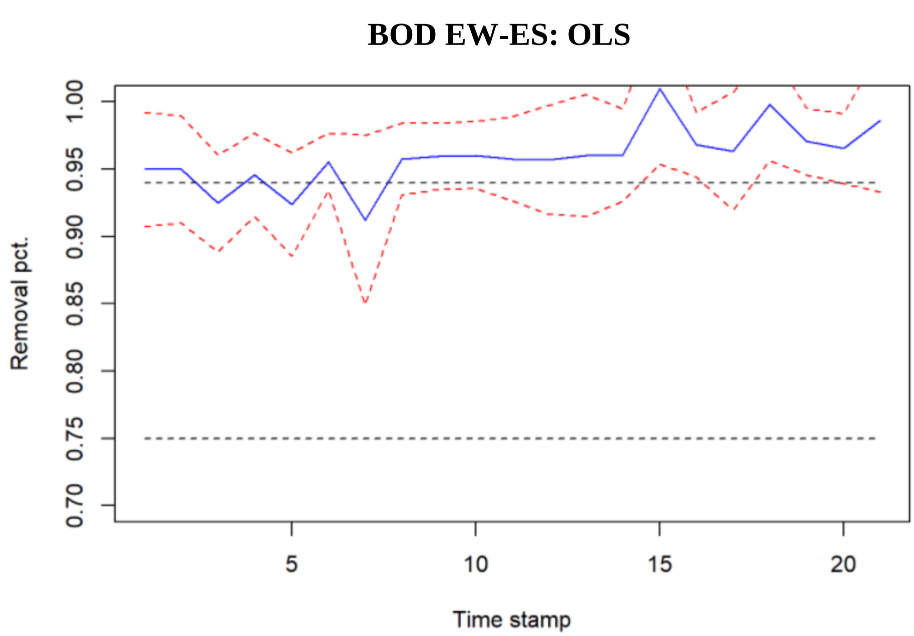


Figure 7. Ordinary least-squares model graph for biological oxygen demand indicating the difference between the ACW effluent and influent concentrations. The blue line shows the predictions. Confidence intervals in red are plotted vs. timestamps. Time stamps should be understood as the sampling times following the reported dates. The dotted lines in black show the maximum and minimum values reported in literature for BOD removal by VSSF or hybrid CWs treating swine wastewater. Removal pct represents removal percentage.

Following with the statistical analysis, the RLM model was estimated, due to the small sample size. Both models need to be contrasted to express results with more certainty. Differences between both models could imply, conclusions cannot be estimated or not with full certainty. For example, the RLM model results show that the effect of flow in the mean difference of BOD is not sufficiently high to be explained by this model, or the sample size is too small to conclude with certitude. This is seen by the absolute t value (-2.008), which is lower than the critical value for a two-tailed t-distribution, 2.11, with 17 degrees of freedom. Nonetheless, Figure 8 shows that the graph of this model is very similar to the prior behaviour, and the t value of the intercept in Table 7 proves that the mean difference of the dependent variable is not zero.

Table 7. Regression linear model output for biological oxygen demand & contrast

rlm (formula = BODdiff.interp ~ Air_temp + Rainfall + Flow, data = wetland.data)					
Residuals:					
	Min	**1Q**	**Median**	**3Q**	**Max**
	-0.1336	-0.0176	-0.0037	0.0227	0.0487
Coefficients:					
	Estimate	**Std. Error**	**t value**		
(Intercept)	0.997	0.024	41.66		
Air_temp	0.0005	0.0014	0.345		
Rainfall	-0.005	0.0041	-1.225		
Flow	0.000	0.000	-2.008		
Residual standard error: 0.02973 on 17 degrees of freedom					

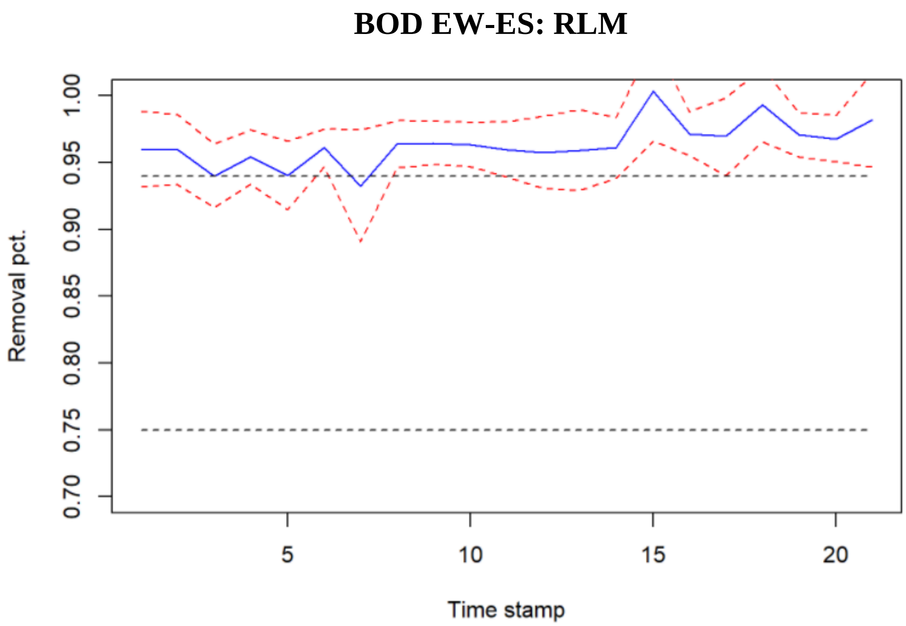


Figure 8. Regression linear model graph for biological oxygen demand indicating the difference between the ACW effluent and influent concentrations. The blue line shows the predictions. Confidence intervals in red are plotted vs. timestamps. Time stamps should be understood as the sampling times following the reported dates. Dotted lines in black show the maximum and minimum values reported in literature for BOD removal by VSSF or hybrid CWs treating swine wastewater. Removal pct represents, removal percentage.

Results of all the other studied parameters for both models indicate that the mean difference between the ACW effluent and influent concentrations of EC (Tables S4 and S5), SS (Tables S6 and S7), and TN (Tables S10 and S11) are influenced by the flow. For the specific case of P, only the RLM model indicates that its mean difference is also influenced by flow (Table S17). The adjusted R-squared indicates that for each of them, the design parameters together explain 77% of EC, 36% of SS, and 45% of TN of the observed variability. Differently, the mean difference between the ACW effluent and influent concentrations of pH and COD are influenced by temperature. For these parameters, the adjusted R-squared indicates that this design parameter explains 28% of the observed variability of pH, and 15% of COD. For the case of NH_4-N, none of the studied design parameters showed influence.

In conclusion, it can be presumed that for all cases there are other parameters that can be considered in the model to explain better the variability and decrease of the studied physicochemical concentrations. Otherwise, a larger sample size could define better the results of the tested models. It is important to mention that reported studies, among few (Gonzalez *et al*., 2009; Borin *et al*., 2013; Comino *et al*., 2013; Vázquez *et al*., 2013; Klomjek, 2016; Maine *et al*., 2019; Torrens *et al*., 2020) do not consider meteorological influence, as air temperature, rainfall, or design parameters as flow, to calculate or report their results.

Looking at the OLS and RLM graphs in Appendix A, Figures S3-S14 for EC, SS, COD, TN, NH_4-N and P indicate that in general, the average removal is higher than that reported in literature (Table 1). This was achieved thanks to the intermittent aeration in the wetland which helped to decompose OM, and in principle triggered TN removal. Microorganisms will break down carbon sources (BOD, COD) and use oxygen for that. When the aeration is off, they have to use the dissolved oxygen in the wastewater left from the aeration phase, which could be insufficient. This will lead to anoxic and possibly anaerobic conditions needed for denitrification and NO_3^- removal (Feng *et al*., 2020b; Parde *et al*., 2021). Therefore, for this study, further research and development are needed, regarding engineering design and automation in the constructed wetland, mainly aeration rates.

3.3 Process economics

3.3.1 Cost-Benefit Analysis (CBA) of the NH_3 stripping installation

The electricity needed for the pilot installation amounted to 13 kWh t^{-1} of IS processed (12 kWh kg^{-1} N recovered), whereas no thermal energy was required since the process was carried out at ambient conditions. Assuming the same energy consumption for a full-scale system, the amount of electrical energy necessary to process all wastewater generated by the pig farm (29,565 t y^{-1}) would represent about 27% of the total electricity generated at the AD plant (about 1,400 MWh).

Our results corroborate the findings of previous work in this field. Vaneeckhaute *et al*. (2017) revealed electrical consumptions between 1.5 and 12 kWh m^3; requirements in the range of 0.8-28 kWh kg^{-1} N recovered were identified by Tampio *et al*. (2016).

The results of the CBA analysis are summarised in Table 8. The cost of processing 1 t of mixed LF manure and LF digestate and to generate 27 kg of 21% AN solution amounted to 6.6 € t^{-1} processed, of which more than 50% consists of the initial amortised investment. This is in line with literature values (2.0-8.1 € m^3) indicated by Vaneeckhaute *et al*. (2017); nevertheless, it is reasonable to think that both investment and operational costs can be reduced when scaling up the installation to full-scale. Moreover, the valorisation of excess heat from CHP engines may increase the profitability of NH_3 stripping at full-scale thanks to advantageous industrial incentives. It must be pointed out, that energy costs were not included since the farm generates all electricity necessary for the operation of the pilot by means of solar panels. In case electricity was purchased, the overall cost would increase by roughly 20%, to 7.9 € t^{-1} processed. The potential benefit from the trade of biobased AN solution was calculated at 1.4-1.7 € t^{-1} processed, corresponding on average to 57 € t^{-1} AN solution produced (8.1% N), in accordance with the market value of N (650-750 € t^{-1} N, NITROMAN project). The calculated market value for AN solution generated by Detricon (about 57 € t^{-1}) is in line with prices estimated for ammonium sulphate solutions from NH_3 stripping installations. Market values ranging from 21 to 35 € t^{-1} (4.6-8% N) were reported by Laureni *et al*. (2013), Bolzonella *et al*. (2018) and Brienza *et al*. (2021), respectively in Spain, Italy and Germany. The variation of NH_4 salts value can be ascribed to the specificity of each regional market; nonetheless, the higher value AN solution is justified by the higher N content, almost double, in comparison to ammonium sulphate solutions with similar DM content.

Table 8. Cost Benefit Analysis of ammonium nitrate solution production via NH_3 stripping.

	Cost (€ t^{-1} processed)	Benefit (€ t^{-1} processed)
Amortised capital cost	3.7	
Electrical energy	0	
60% HNO_3 solution	1.6	
Insurance, maintenance, labour	1.3	
Ammonium nitrate value		1.5
Total	6.6	1.5

The cost of AN solution (8.1% N) amounting to 242 € t^{-1}, translates into a cost of 3.0 € kg^{-1} N. According to IndexMundi, in the same years of our study (2019 and 2020), the price of broadcast synthetic N fertiliser (urea 46% N) was on average 0.46 € kg^{-1} N (Figure S15). However, the increased energy prices over the last year and a half contributed to the increase in the price of urea by four times, up to 1.8 € kg^{-1} N. Similarly to urea´s cost, also the price for 60% HNO_3 quadruplicated from 200 in 2019 to 795 € t^{-1} in March 2022. Nevertheless, as AN solution produced by NH_3 stripping relies on the renewable electricity generated onsite, the overall production cost calculated in March 2022 increased to 5.2 € kg^{-1} N, only by 1.7 times, against an increment of 3.9-fold of urea. Although the purchase of synthetic urea is still economically more favourable compared to the cost of producing biobased AN solution, it is worth of notice that in 2019-2020 the cost per kg N in AN solution was 6.6 times higher than urea, whereas, in March 2022, it was 2.9.

3.3.2. Overall cost of the proposed system: NH_3 stripping + ACW

During the last monitoring period, the ACW was fed with 36 m^3 ha^{-1} d^{-1}. With such loading rate, a surface area of 2.3 ha (226 times larger than the actual ACW pilot), would be necessary to replace the biological NDN system of the pig farm. This translates into high investment costs, about 8.7 € t^{-1} of processed and operational costs of around 3.8 € t^{-1}. The overall cost of the proposed process, consisting of NH_3 stripping (5.1 € t^{-1}) and ACW (12 € t^{-1}) resulted to be slightly more expensive than the conventional biological NDN system (16 € t^{-1}).

To investigate the effect of increasing loading rates on the overall process cost, a sensitivity analysis was carried out. Results indicated that the process becomes competitive with NDN only for loading rates higher than 40 m^3 ha^{-1} d^{-1}, which is 12% more of the amount fed during the last period of the monitoring (Figure 9). At the envisaged loading rate in the initial experimental design (1 m^3 d^{-1}, corresponding to 100 m^3 ha^{-1}), the proposed process would cost 10 € t^{-1}, against 16 € t^{-1} of the current NDN system, thus cheaper than conventional treatment. Yet, to achieve such high treatment capacity, it is of utmost importance to implement baffles in the system and control clogging of pipes by improving solids removal from the treated wastewater.

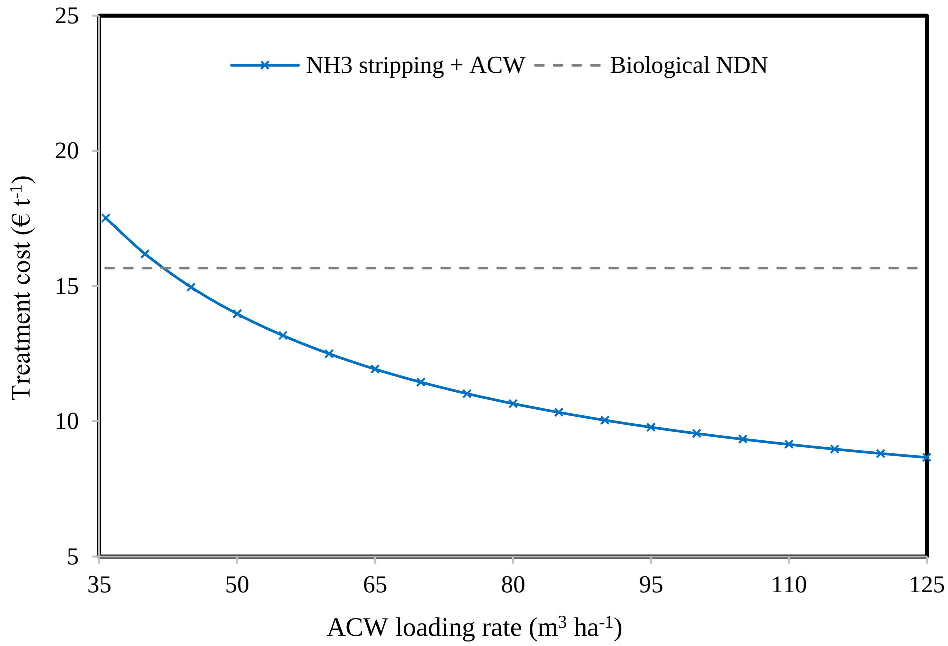


Figure 9. Effect of aerated constructed wetland (ACW) increasing loading rate on the overall cost of the proposed process (NH_3 stripping and ACW), in comparison with conventional biological nitrification-denitrification (NDN) treatment.

4. CONCLUSIONS

Considering the system design, overall efficiency and estimated costs, this study suggests two design options for alternative swine wastewater treatment. First, the effluent of NH_3 stripping plus ACW is brought to land in the right season, yet it must be buffered in winter. Alternatively, the effluent of the proposed process must be followed by hybrid CW so that the effluent could meet discharge standards limits for surface water. Despite not being yet economically competitive with conventional NDN systems, the proposed process has the potential to produce a biobased mineral fertiliser, AN solution, that meets both FPR and RENURE criteria.

In this study design, parameters such as rainfall, air temperature and flow, were considered in two models, the OLS, and the RLM. The reasoning behind this was to capture their possible incidence in each of the studied parameters (pH, EC, SS, COD, BOD, TN, NO_3, NH_4 and P) and compare it with removal ranges reported in the literature. Most of the removal efficiencies of the studied parameters (EC, SS, TN, partially P and BOD) were influenced by the flow. This proves that it was suitable to test different flow rates in each sampling period, as these influenced the most to the mean concentrations decrease. The overall analysis shows that at lower flow, higher removal efficiencies were achieved. The exception was for NO_3-N concentration whose decrease was limited by insufficient denitrification. Thus, the NH_3 stripping step could be optimised to remove more NH_4-N, improving the COD:N ratio in the influent to the ACW.

NH_3 stripping plus ACW has the potential to replace the conventional biological NDN system and improve N circularity in livestock-dominated food chains. However, further investigation should validate potential environmental benefits through comprehensive LCA analysis, especially to address concerns regarding intensive land use, and optimise process parameter settings to improve economic viability and environmental impact of these technologies combination.

ACKNOWLEDGEMENTS

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APPENDIX A. Supplementary data

Table S1. Overview of monitoring periods (Period 1-2-3) and physico-chemical characterisation for each sampling point: influent stripper (IS), ammonium nitrate (AN) solution, effluent stripper (ES), intermediate wetland effluent (IW) and wetland effluent (EW).

	IS	AN solution	ES	IW	EW
pH	Period 1-2	Period 1-2	Period 1-2-3	Period 2-3	Period 1-2-3
EC	Period 1-2	Period 1-2	Period 1-2-3	Period 2-3	Period 1-2-3
DM	Period 1-2	Period 1-2	Period 1-2-3	Period 2-3	
SS	Period 1-2	Period 1-2	Period 1-2-3	Period 2-3	Period 1-2-3
COD	Period 1-2	Period 1-2	Period 1-2-3	Period 2-3	Period 1-2-3
BOD			Period 1-2-3	Period 2-3	Period 1-2-3
TN	Period 1-2	Period 1-2	Period 1-2-3	Period 2-3	Period 1-2-3
NH_4-N	Period 1-2	Period 1-2	Period 1-2-3	Period 2-3	Period 1-2-3
NO_3-N	Period 1-2	Period 1-2	Period 1-2-3	Period 2-3	Period 1-2-3
P	Period 1-2	Period 1-2	Period 1-2-3	Period 2-3	Period 1-2-3
K	Period 1-2	Period 1-2	Period 1-2-3		
S	Period 1-2	Period 1-2	Period 1-2-3		
Ca	Period 1-2	Period 1-2	Period 1-2-3		
Mg	Period 1-2	Period 1-2	Period 1-2-3		
Na	Period 1-2	Period 1-2	Period 1-2-3		
TOC		Period 1-2			
Cu		Period 1-2			
Zn		Period 1-2			

Table S2: Ordinary least-squares model output pH & contrast

lm (formula = pHdiff.interp ~ Air_temp + Rainfall + Flow, data = wetland.data)					
Residuals:					
	Min	**1Q**	**Median**	**3Q**	**Max**
	-0.048920	-0.019464	-0.005367	0.016245	0.067451
Coefficients:					
	Estimate	**Std. Error**	**t value**	**Pr (>|t|)**	
(Intercept)	1.10e-01	2.31e-02	4.78	0.00018***	
Air_temp	-4.39e-03	1.39e-03	-3.18	0.0055**	
Rainfall	3.44e-03	3.97e-03	0.87	0.40	
Flow	1.44e-05	2.16e-05	0.67	0.51*	
Signif. codes	0 ***	0.001 **	0.01 *	0.05	0.1
Multiple R-squared: 0.3842			Adjusted R-squared: 0.2756		
F-statistic: 3.536 on 3 and 17 DF			p-value: 0.03723		

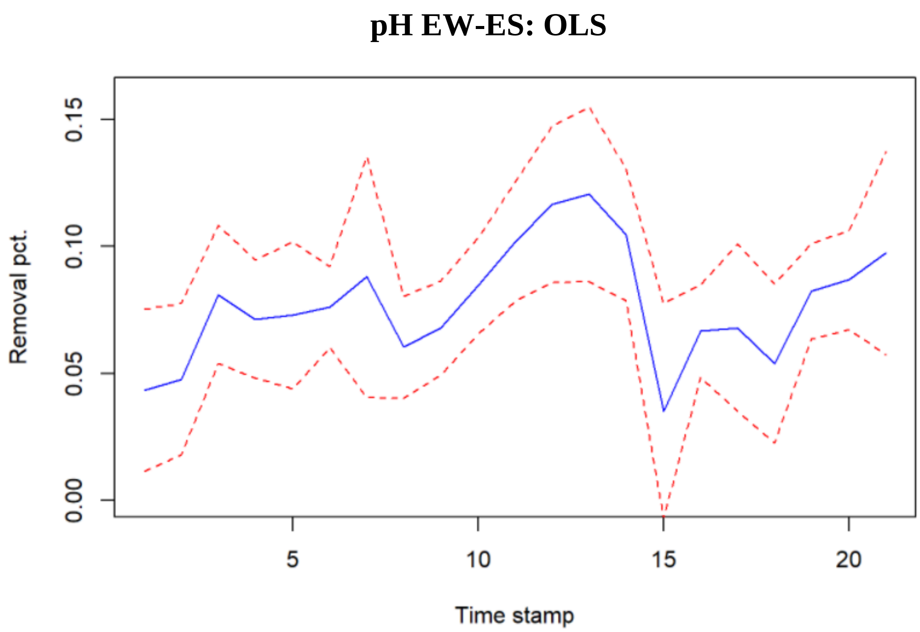

Figure S1: Ordinary least-squares model graph for pH indicating the modelled difference between the ACW effluent and influent concentrations

Table S3: Regression linear model output for pH & contrast

rlm (formula = pHdiff.interp ~ Air_temp + Rainfall + Flow, data = wetland.data)					
Residuals:					
	Min	**1Q**	**Median**	**3Q**	**Max**
	-0.04715	-0.01561	-0.005262	0.01655	0.06916
Coefficients:					
	Estimate	**Std. Error**	**t value**		
(Intercept)	0.106	0.0205	5.174		
Air_temp	-0.0046	0.0012	-3.722		
Rainfall	0.0031	0.0035	0.884		
Flow	0.000	0.000	1.05		
Residual standard error: 0.02454 on 17 degrees of freedom					

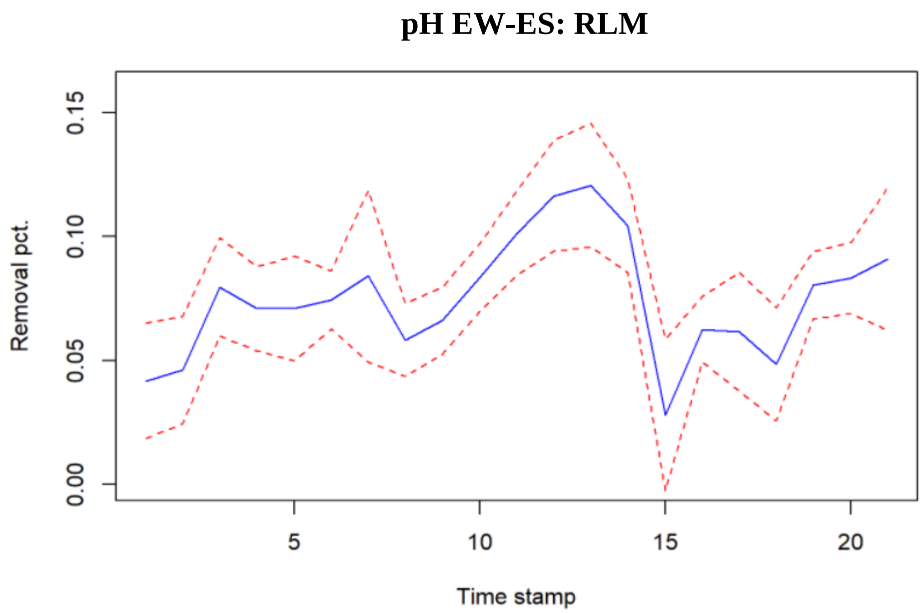

Figure S2: Robust linear model graph for pH indicating the modelled difference between the ACW effluent and influent concentrations

Table S4: Ordinary least-squares model output for electrical conductivity & contrast

lm (formula = ECdiff.interp ~ Air_temp + Rainfall + Flow, data = wetland.data)					
Residuals:					
	Min	**1Q**	**Median**	**3Q**	**Max**
	-0.15213	-0.07236	-0.01043	0.09291	0.22965
Coefficients:					
	Estimate	**Std. Error**	**t value**	**Pr (>|t|)**	
(Intercept)	5.10e-01	8.34e-02	6.12	1.14e-05***	
Air_temp	1.33e-02	4.99e-03	2.67	0.016 *	
Rainfall	2.03e-02	1.43e-02	1.42	0.17	
Flow	-5.76e-04	7.77e-05	-7.42	1.01e-06 ***	
Signif. codes	0 ***	0.001 **	0.01 *	0.05	0.1
Multiple R-squared: 0.8028			Adjusted R-squared: 0.768		
F-statistic: 23.07 on 3 and 17 DF			p-value: 3.158e-06		

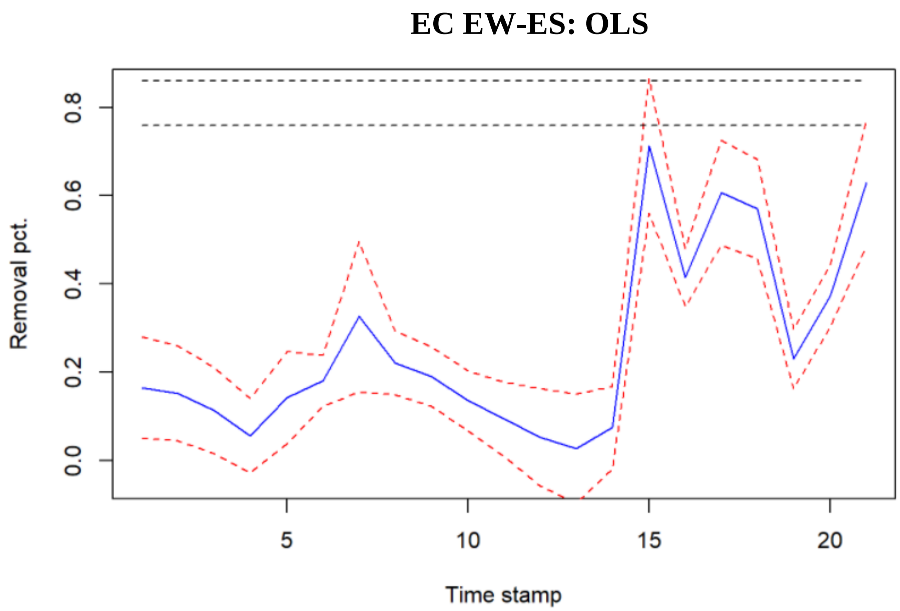

Figure S3: Ordinary least-squares model graph for electrical conductivity indicating the modelled difference between the ACW effluent and influent concentrations

Table S5: Robust linear model output for EC & contrast

rlm (formula = pHdiff.interp ~ Air_temp + Rainfall + Flow, data = wetland.data)					
Residuals:					
	Min	**1Q**	**Median**	**3Q**	**Max**
	-0.149548	-0.071860	-0.009106	0.095229	0.234199
Coefficients:					
	Estimate	**Std. Error**	**t value**		
(Intercept)	0.5056	0.0885	5.7129		
Air_temp	0.0131	0.0053	2.4720		
Rainfall	0.0202	0.0152	1.3312		
Flow	-0.0006	0.0001	-6.9215		
Residual standard error: 0.1315 on 17 degrees of freedom					

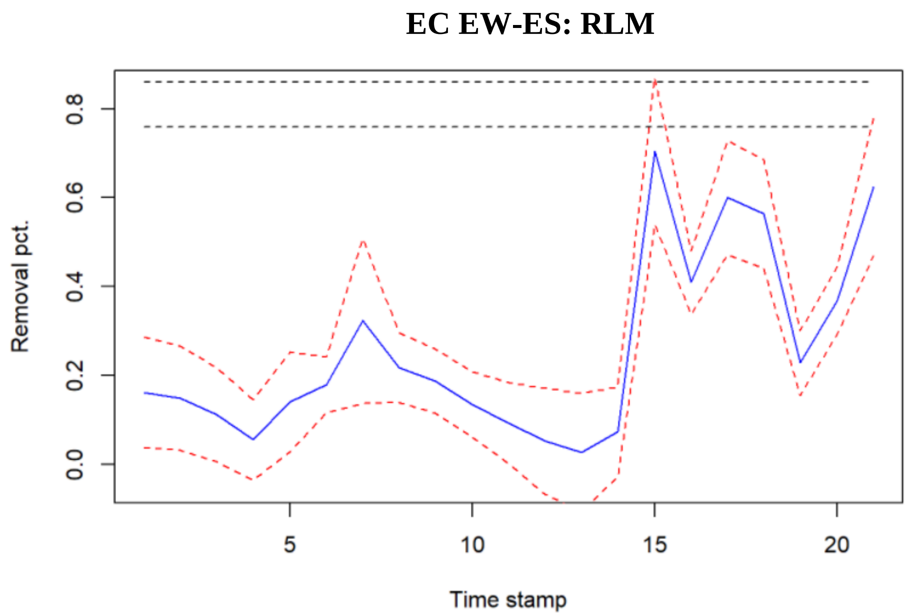

Figure S4: Robust linear model graph for electrical conductivity indicating the modelled difference between the ACW effluent and influent concentrations

Table S6: Ordinary least-squares model output for suspended solids & contrast

lm (formula = SSdiff.interp ~ Air_temp + Rainfall + Flow, data = wetland.data)					
Residuals:					
	Min	**1Q**	**Median**	**3Q**	**Max**
	-0.052045	-0.010550	-0.000823	0.015973	0.045562
Coefficients:					
	Estimate	**Std. Error**	**t value**	**Pr (>|t|)**	
(Intercept)	1.009	1.757e-02	57.417	<2e-16 ***	
Air_temp	-6.986e-04	1.052e-03	-0.664	0.5156	
Rainfall	-5.755e-03	3.014e-03	-1.909	0.0733	
Flow	-5.502e-05	1.637e-05	-3.362	0.0037 **	
Signif. codes	0 ***	0.001 **	0.01 *	0.05	0.1
Multiple R-squared: 0.4554			Adjusted R-squared: 0.3593		
F-statistic: 4.739 on 3 and 17 DF			p-value: 0.01402		

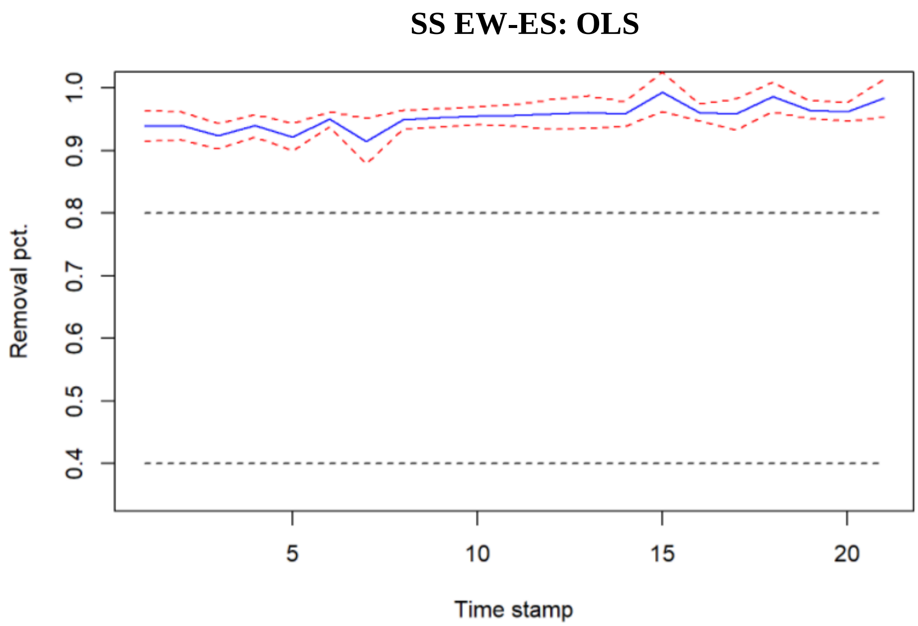

Figure S5: Ordinary least squares model graph for suspended solids indicating the modelled difference between the ACW effluent and influent concentrations

Table S7: Robust linear model output for suspended solids & contrast

rlm (formula = SSdiff.interp ~ Air_temp + Rainfall + Flow, data = wetland.data)					
Residuals:					
	Min	**1Q**	**Median**	**3Q**	**Max**
	-0.051946	-0.009336	-0.003394	0.014664	0.047536
Coefficients:					
	Estimate	**Std. Error**	**t value**		
(Intercept)	1.0131	0.0175	57.854		
Air_temp	-0.0009	0.0010	-0.8650		
Rainfall	-0.0057	0.0030	-1.8894		
Flow	-0.0001	0.000	-3.5321		
Residual standard error: 0.01935 on 17 degrees of freedom					

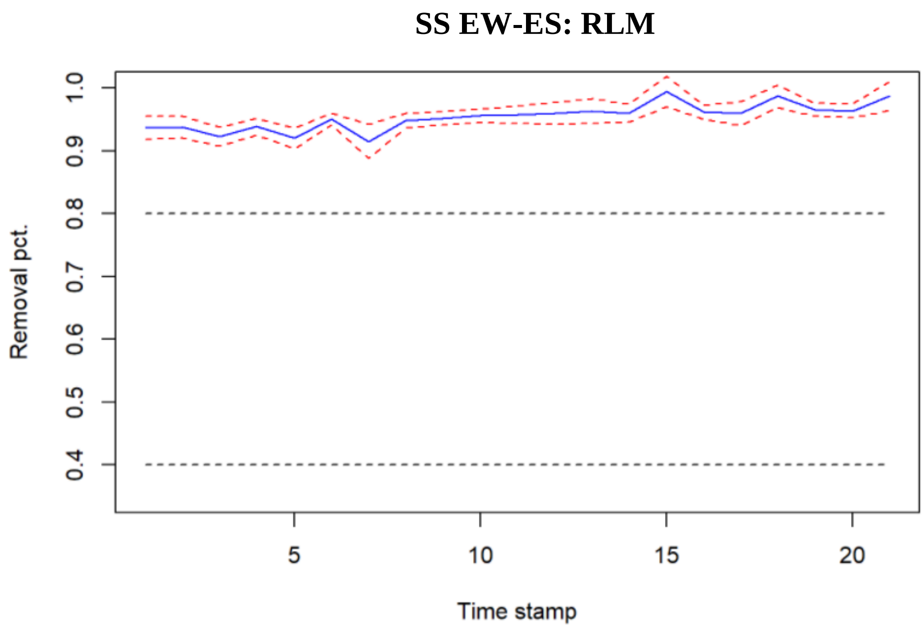

Figure S6: Robust linear model graph for suspended solids indicating the modelled difference between the ACW effluent and influent concentrations

Table S8: Ordinary least-squares model output for chemical oxygen demand & contrast

<table>
<tr><td colspan="6">lm (formula = CODdiff.interp ~ Air_temp + Rainfall + Flow, data = wetland.data)</td></tr>
<tr><td colspan="6">Residuals:</td></tr>
<tr><td></td><td>Min</td><td>1Q</td><td>Median</td><td>3Q</td><td>Max</td></tr>
<tr><td></td><td>0.077394</td><td>-0.044558</td><td>0.000001</td><td>0.000001</td><td>0.068759</td></tr>
<tr><td colspan="6">Coefficients:</td></tr>
<tr><td></td><td>Estimate</td><td>Std. Error</td><td>t value</td><td>Pr (>|t|)</td><td></td></tr>
<tr><td>(Intercept)</td><td>8.390e-01</td><td>3.651e-02</td><td>22.980</td><td>3.06e-14 ***</td><td></td></tr>
<tr><td>Air_temp</td><td>5.539e-03</td><td>2.186e-03</td><td>2.533</td><td>0.0214 *</td><td></td></tr>
<tr><td>Rainfall</td><td>-1.779e-03</td><td>6.264e-03</td><td>-0.284</td><td>0.7799</td><td></td></tr>
<tr><td>Flow</td><td>-6.138e-06</td><td>3.401e-05</td><td>-0.180</td><td>0.8589</td><td></td></tr>
<tr><td>Signif. codes</td><td>0 ***</td><td>0.001 **</td><td>0.01 *</td><td>0.05</td><td>0.1</td></tr>
<tr><td colspan="3">Multiple R-squared: 0.2746</td><td colspan="3">Adjusted R-squared: 0.1466</td></tr>
<tr><td colspan="3">F-statistic: 2.145 on 3 and 17 DF</td><td colspan="3">p-value: 0.1322</td></tr>
</table>

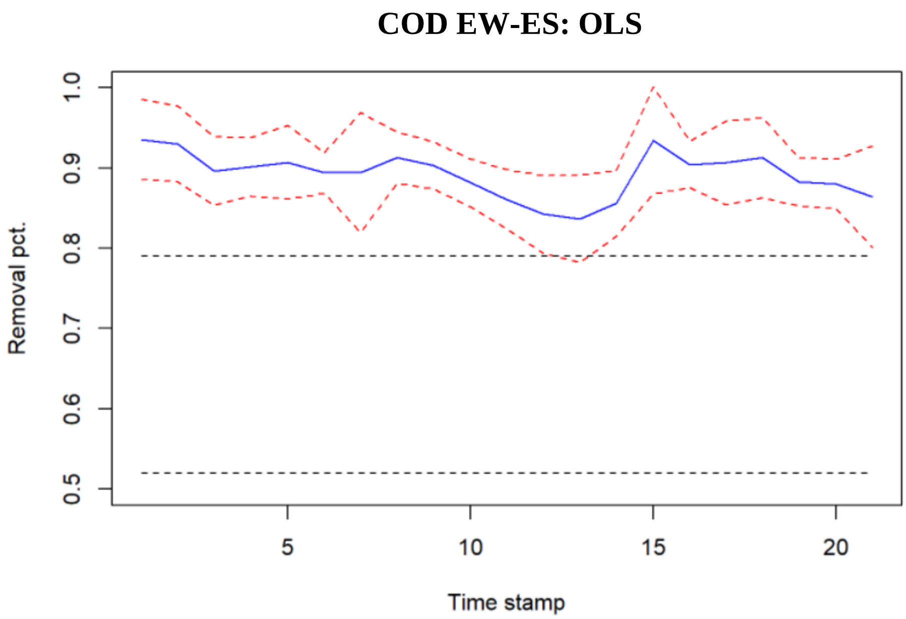

Figure S7: Ordinary least-squares model graph for chemical oxygen demand indicating the modelled difference between the ACW effluent and influent concentrations

Table S9: Robust linear model output for chemical oxygen demand & contrast

rlm (formula = CODdiff.interp ~ Air_temp + Rainfall + Flow, data = wetland.data)					
Residuals:					
	Min	**1Q**	**Median**	**3Q**	**Max**
	-7.739e-02	-4.456e-02	9.502e-07	4.662e-02	6.876e-02
Coefficients:					
	Estimate	**Std. Error**	**t value**		
(Intercept)	0.8390	0.0365	22.9800		
Air_temp	0.0055	0.0022	2.5334		
Rainfall	-0.0018	0.0063	-0.2839		
Flow	0.0000	0.000	-0.1805		
Residual standard error: 0.06628 on 17 degrees of freedom					

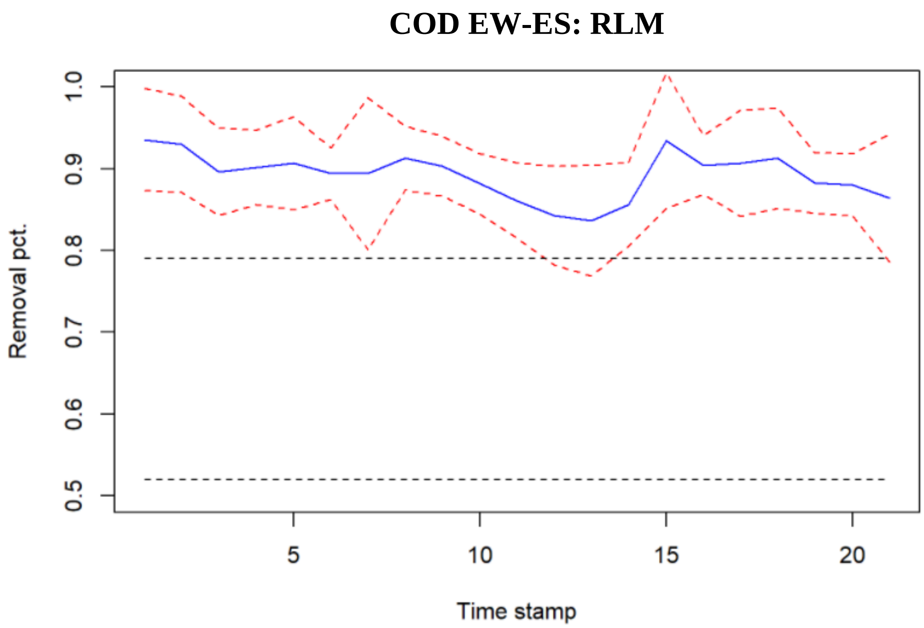

Figure S8: Regression linear model graph for chemical oxygen demand indicating the modelled difference between the ACW effluent and influent concentrations

Table S10: Ordinary least-squares model output for total nitrogen & contrast

lm (formula = TNdiff.interp ~ Air_temp + Rainfall + Flow, data = wetland.data)					
Residuals:					
	Min	**1Q**	**Median**	**3Q**	**Max**
	-0.24609	-0.02568	0.00356	0.03874	0.23256
Coefficients:					
	Estimate	**Std. Error**	**t value**	**Pr (>|t|)**	
(Intercept)	5.555e-01	7.617e-02	7.292	1.26e-06 ***	
Air_temp	3.590e-03	4.562e-03	0.787	0.44208	
Rainfall	-5.701e-03	1.307e-02	-0.436	0.66815	
Flow	2.991e-04	7.096e-05	4.215	0.000582 ***	
Signif. codes	0 ***	0.001 **	0.01 *	0.05	0.1
Multiple R-squared: 0.5339			Adjusted R-squared: 0.4516		
F-statistic: 6.49 on 3 and 17 DF			p-value: 0.003986		

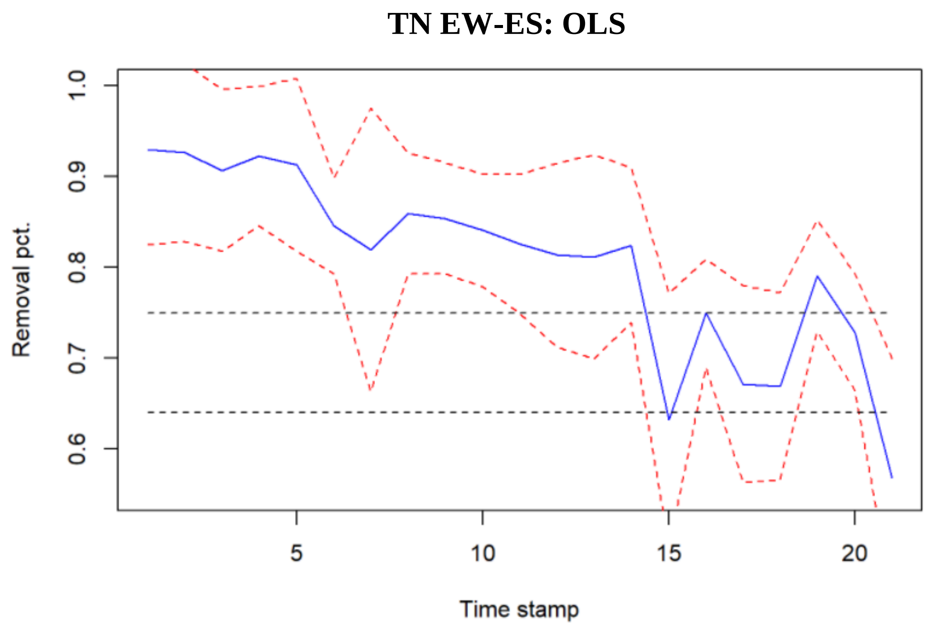

Figure S9: Ordinary least-squares model graph for total nitrogen indicating the modelled difference between the ACW effluent and influent concentrations

Table S11: Robust linear model output for total nitrogen

rlm (formula = TNdiff.interp ~ Air_temp + Rainfall + Flow, data = wetland.data)					
Residuals:					
	Min	**1Q**	**Median**	**3Q**	**Max**
	-0.230137	-0.036122	0.007232	0.024163	0.261321
Coefficients:					
	Estimate	**Std. Error**	**t value**		
(Intercept)	0.5682	0.0419	13.5681		
Air_temp	0.0011	0.0025	0.4486		
Rainfall	-0.0060	0.0072	-0.8414		
Flow	0.0003	0.0000	8.2713		
Residual standard error: 0.06628 on 17 degrees of freedom					

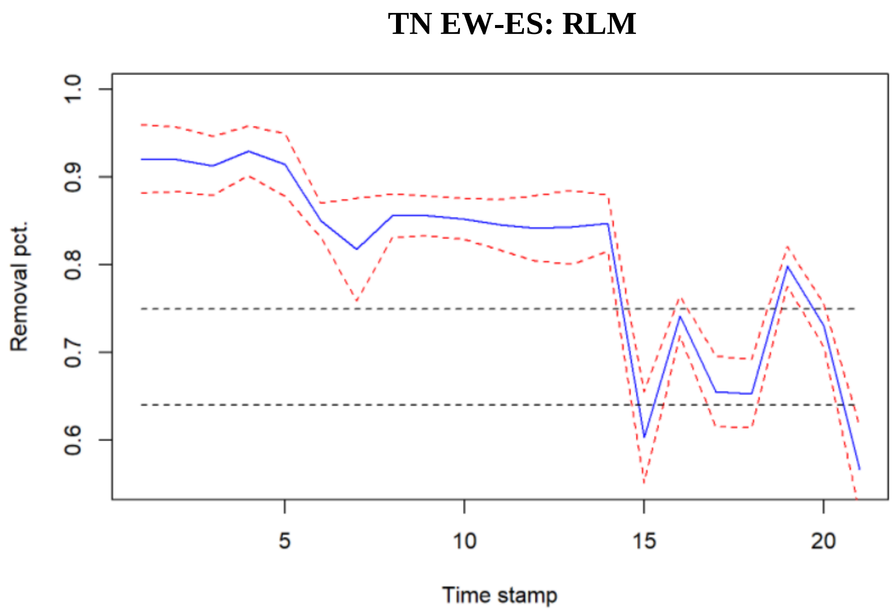

Figure S10: Robust linear model graph for total nitrogen indicating the modelled difference between the ACW effluent and influent concentrations

Table S12: Ordinary least-squares model output for ammonium & contrast

lm (formula = NH_4diff.interp ~ Air_temp + Rainfall + Flow, data = wetland.data)					
Residuals:					
	Min	**1Q**	**Median**	**3Q**	**Max**
	-0.183401	-0.008957	0.019756	0.043528	0.087156
Coefficients:					
	Estimate	**Std. Error**	**t value**	**Pr (>|t|)**	
(Intercept)	1.007e+00	6.105e-02	16.488	6.84e-12 ***	
Air_temp	-1.589e-03	3.656e-03	-0.435	0.669	
Rainfall	-4.669e-03	1.047e-02	-0.446	0.661	
Flow	-5.647e-05	5.687e-05	-0.993	0.335	
Signif. codes	0 ***	0.001 **	0.01 *	0.05	0.1
Multiple R-squared: 0.0724			Adjusted R-squared: -0.09129		
F-statistic: 0.4423 on 3 and 17 DF			p-value: 0.7258		

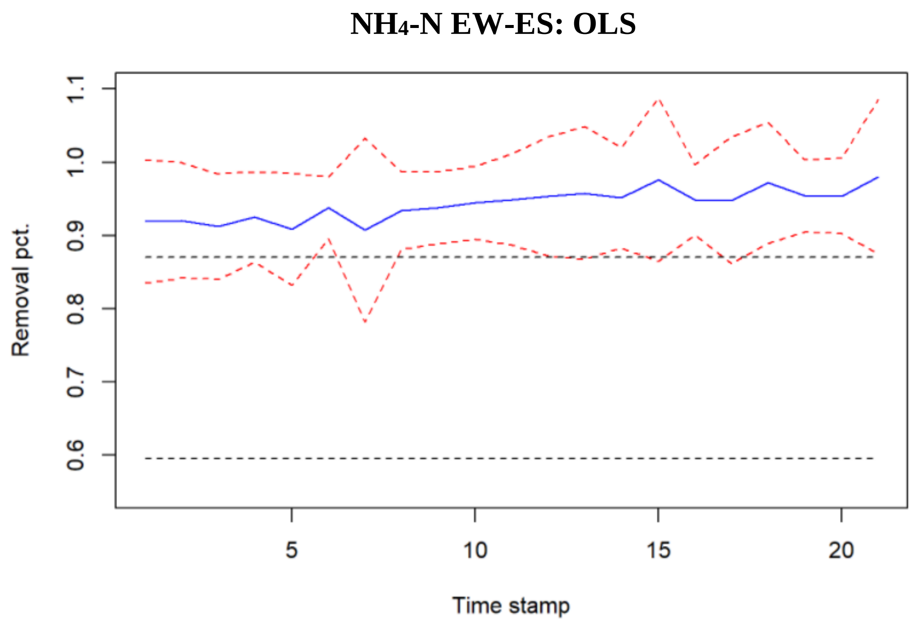

Figure S11: Ordinary least-squares model graph for ammonium indicating the modelled difference between the ACW effluent and influent concentrations

Table S13: Robust linear model output for ammonium & contrast

rlm (formula = NH_4diff.interp ~ Air_temp + Rainfall + Flow, data = wetland.data)					
Residuals:					
	Min	**1Q**	**Median**	**3Q**	**Max**
	-0.20739	-0.02225	0.01148	0.03639	0.06543
Coefficients:					
	Estimate	**Std. Error**	**t value**		
(Intercept)	0.9939	0.0470	21.1358		
Air_temp	0.0000	0. 0028	-0.0092		
Rainfall	-0.0059	0.0081	-0.7314		
Flow	0.0000	0.0000	-0.9002		
Residual standard error: 0.05396 on 17 degrees of freedom					

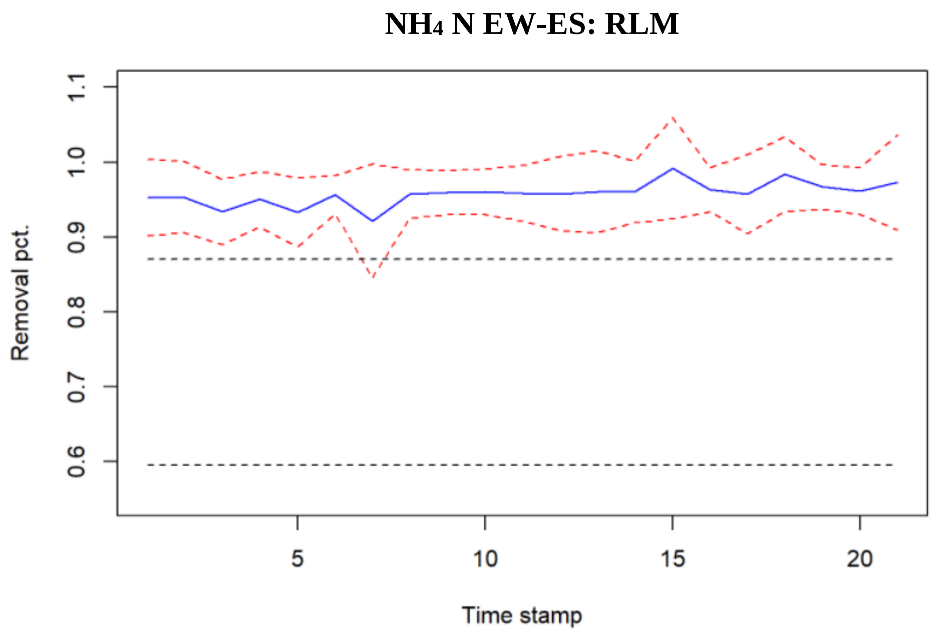

Figure S12: Robust linear model graph for ammonium indicating the modelled difference between the ACW effluent and influent concentrations

Table S14: Ordinary least-squares model output for nitrate & contrast

lm (formula = NO_3diff.interp ~ Air_temp + Rainfall + Flow, data = wetland.data)					
Residuals:					
	Min	**1Q**	**Median**	**3Q**	**Max**
	-37.113	-2.476	0.397	7.000	16.698
Coefficients:					
	Estimate	**Std. Error**	**t value**	**Pr (>|t|)**	
(Intercept)	-30.7593	8.822629	-3.486	0.00283 **	
Air_temp	0.763030	0.528332	1.444	0.16685	
Rainfall	-1.864858	1.513726	-1.232	0.23473	
Flow	0.026431	0.008219	3.216	0.00507 **	
Signif. codes	0 ***	0.001 **	0.01 *	0.05	0.1
Multiple R-squared: 0.4694			Adjusted R-squared: 0.3758		
F-statistic: 5.014 on 3 and 17 DF			p-value: 0.01137		

Table S15: Robust linear model output for nitrate & contrast

rlm (formula = NO_3diff.interp ~ Air_temp + Rainfall + Flow, data = wetland.data)					
Residuals:					
	Min	**1Q**	**Median**	**3Q**	**Max**
	-54.73776	-1.70858	-0.01938	1.70612	12.65026
Coefficients:					
	Estimate	**Std. Error**	**t value**		
(Intercept)	-13.6298	2.7330	-4.9872		
Air_temp	0.0397	0.1637	0.2424		
Rainfall	-0.3184	0.4689	-0.6791		
Flow	0.0141	0.0025	5.5567		
Residual standard error: 2.533 on 17 degrees of freedom					

Table S16: Ordinary least-squares model output for total phosphorus & contrast

lm (formula = Pdiff.interp ~ Air_temp + Rainfall + Flow, data = wetland.data)					
Residuals:					
	Min	**1Q**	**Median**	**3Q**	**Max**
	-0.046745	-0.007525	0.001187	0.013159	0.022936
Coefficients:					
	Estimate	**Std. Error**	**t value**	**Pr (>|t|)**	
(Intercept)	9.629e-01	1.405e-02	68.519	<2e-16 ***	
Air_temp	-3.307e-04	8.415e-04	-0.393	0.699	
Rainfall	-4.439e-04	2.411e-03	-0.184	0.856	
Flow	1.655e-05	1.309e-05	1.264	0.223	
Signif. codes	0 ***	0.001 **	0.01 *	0.05	0.1
Multiple R-squared: 0.1007			Adjusted R-squared: -0.05802		
F-statistic: 0.6344 on 3 and 17 DF			p-value: 0.603		

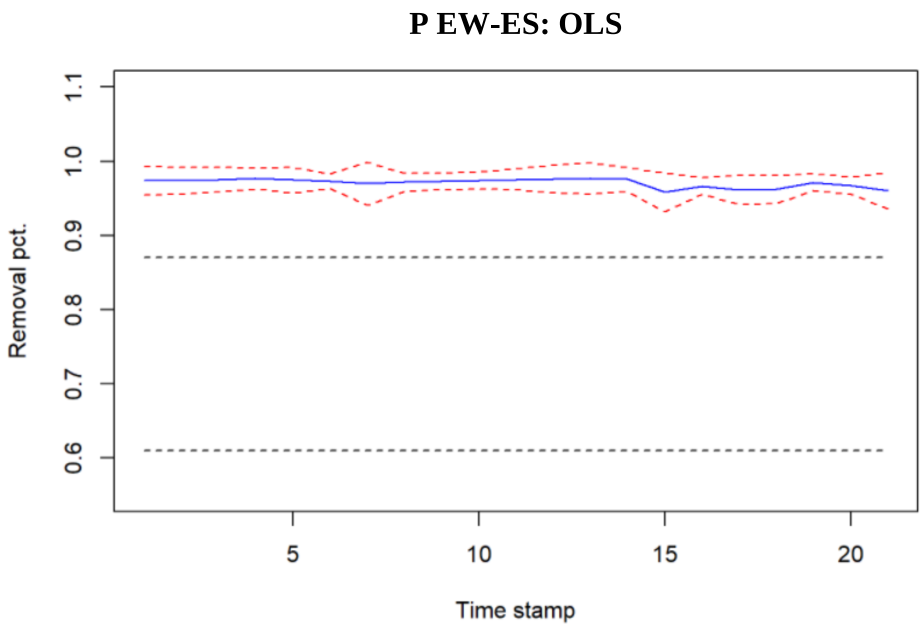

Figure S13: Ordinary least-squares model graph for total phosphorus indicating the modelled difference between the ACW effluent and influent concentrations

Table S17: RLM model output for total phosphorus

rlm (formula = Pdiff.interp ~ Air_temp + Rainfall + Flow, data = wetland.data)					
Residuals:					
	Min	**1Q**	**Median**	**3Q**	**Max**
	-0.0544326	-0.0060075	0.0004999	0.0098416	0.0190727
Coefficients:					
	Estimate	**Std. Error**	**t value**		
(Intercept)	0.9550	0.0115	82.7683		
Air_temp	-0.0001	0.0007	-0.1019		
Rainfall	0.0002	0.0020	0.1155		
Flow	0.0000	0.0000	2.4338		
Residual standard error: 0.01459 on 17 degrees of freedom					

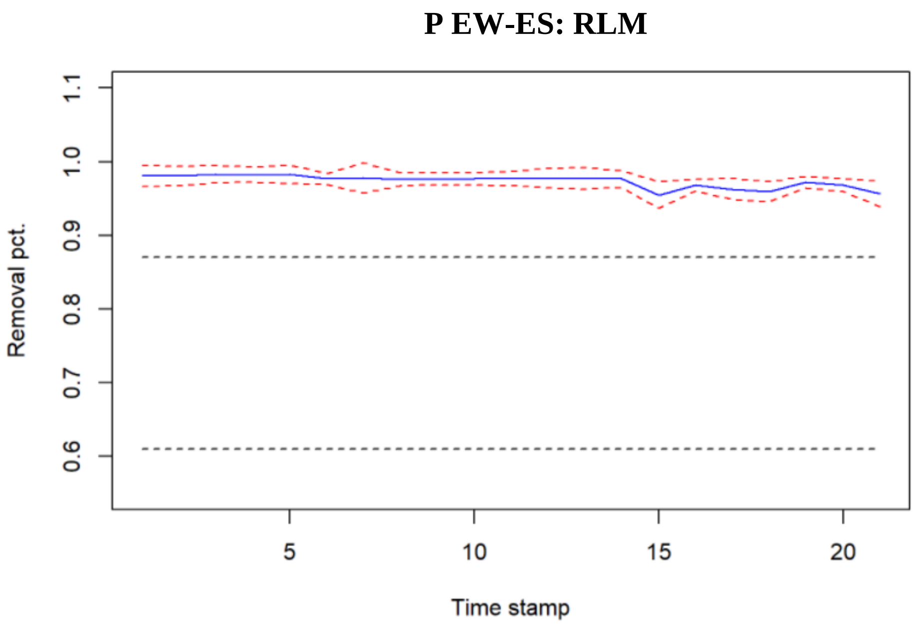

Figure S14: RLM model graph for total phosphorus indicating the modelled difference between the ACW effluent and influent concentrations

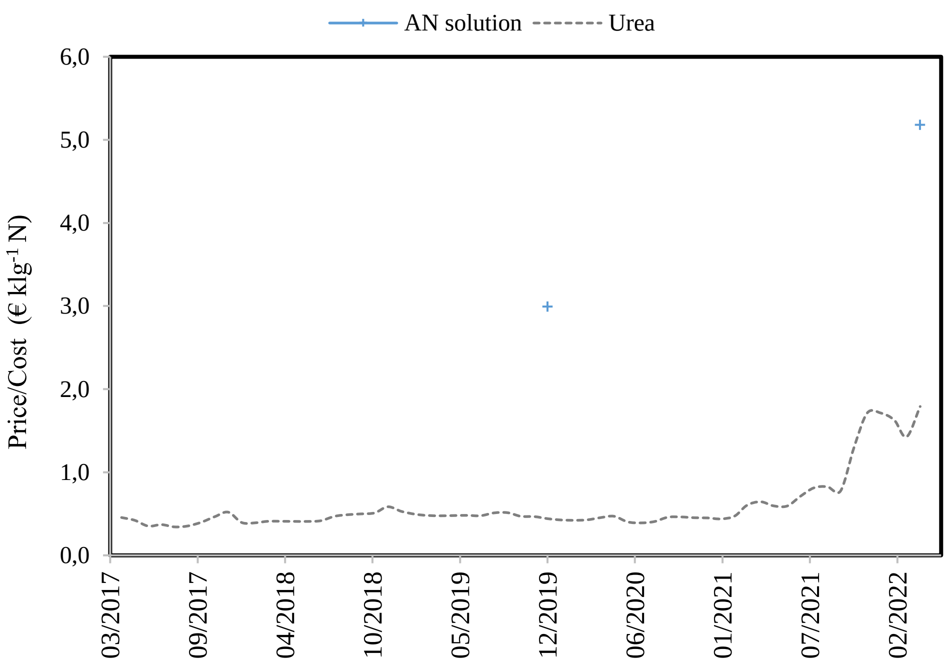

Figure S15. Trend of synthetic urea price in Eastern Europe from April 2017 until March 2022 and calculated cost of ammonium nitrate (AN) solution production over the monitoring period and in March 2022.