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Biological recirculating air treatment system to reduce ammonia pollution in pig farms: potential and cost estimation

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High levels of ammonia and aerosol concentrations in barns for pigs can lead to health problems for animals and humans, reduce performance and pollute the environment as emissions. Therefore, the aim was to develop and evaluate a process for air cleaning in pig barns in which a partial airflow is extracted from the house, cleaned in a multi-stage cleaning process and then returned to the barn with the intention to improve the quality of the air in the barn. Ammonia is washed out of the raw gas and then converted into atmospheric nitrogen via a nitrification and a denitrification unit. During an experiment in the technical centre in which only the ammonia reduction was examined with the recirculating air treatment system designed for 16 animal places, the nitrification (0.21 ± 0.06 g NO_X-N l^{-1} d^{-1}) and denitrification performance (1.11 ± 0.37 g NO_X-N l^{-1} d^{-1}) were low in relation to the system design. Secondary gas emissions were low at 1.3 % of the N dosage. A comparative cost calculation with a conventional chemical air treatment system revealed significantly higher operating costs, meaning that the experimental system is not yet ready for practical application. Measurements to improve the air quality in the barn could not be carried out.

Keywords
recirculating air treatment system, ammonia reduction, nitrification, denitrification, secondary trace gases, process costs

In the Federal Republic of Germany, 537,000 t of ammonia (NH_3) were emitted in 2020. Of this, 512,300 t of NH_3 came from agriculture (Vos et al. 2022). Since 2020, Germany is obliged to reduce its NH_3 emissions by 5 % per year compared to 2005, and by 29 % per year from 2030 onwards (Directive (EU) 2016/2284). Of the 512,300 t of NH_3, 84,700 t of NH_3 (16.5 %) came from pig farming, of which the main share (67.5 %) was attributable to barns (Vos et al. 2022). From 1 January 2029 at the latest, all pig fattening barns with 1,500 fattening places or more must achieve an emission reduction of at least 40 % in relation to the reference value of 2.91 kg NH_3 per animal place (AP) a^{-1} with a crude protein and phosphorus-adapted multiphase feed (BImSchV 2021, TA Luft 2021). As most pigs are kept in the 1,000–1,999 pigs per farm category, this regulation affects the majority of farms (Statistisches Bundesamt 2023). This makes it clear that there is great pressure to act. A more precise statement on how many animals are kept in the category of 1,500–1,999 pigs per farm was not possible due to the lack of subdivision at the Federal Statistical Office of Germany (Statistisches Bundesamt).

Increased ammonia and particle concentrations can have negative effects on animal health and welfare. NH_3 has a pungent odour and an irritant effect on the mucous membranes of the respiratory tract and the conjunctiva (Kliche and Mehlhorn 1979). A significant correlation between the in-

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crease in NH_3 concentration and increasing pneumonia findings in fattening pigs was determined by Richter (2015). Donham (1991) states that pneumonia and pleurisy findings correlate with the NH_3 content and the respirable particle content of the barn air. The legal limit value for NH_3 is 20 ppm and since 2021 it must no longer be exceeded, not even for short periods (TierSchNutztV 2017, 2021). In addition to the negative effects of harmful gases and particles on the pigs, there are also negative impacts and possible respiratory tract diseases among farmers (Iversen et al. 1988, Larsson et al. 1994, Kölbeck et al. 2000, Radon et al. 2003).

Due to the problems described above, a new process was to be developed to eliminate ammonia and aerosols from the barn air, which would improve the air quality in the barn and thus animal welfare and working conditions for farmers. The NH_3 washed out of the barn air should be converted into environmentally friendly, gaseous nitrogen (N_2) by means of nitrification and denitrification. Furthermore, the recirculating air treatment system should also reduce the dust content of the barn air. An exhaust air treatment system was designed for this purpose, which was to be operated as a process of recirculating air in the compartment with partial flow purification at a maximum of twice the winter air rate. By using partial flow cleaning, the relative humidity in the compartment was to be kept at an acceptable level through humidification in the air washer. Partial flow cleaning should also reduce the exhaust air volume flow, as the fresh air supply in the compartment should be lower. Due to the lower volume flow, ammonia emissions are reduced. In addition, the reduced air exchange should result in energy savings, as less energy is required for ventilation and heating.

The aim was to test the basic suitability of the prototype, to determine the system performance including consumables and operational stability and to evaluate the operating costs in comparison with chemical exhaust air treatment systems. Only the effectiveness of the purification stages was analysed, as it was not possible to install the experimental system in a pig barn. Therefore, no statements about the improvement of the barn air can be made in this work.

Basics of the process

The ammonium (NH_4^+) dissolved in the wash water of the recirculating air treatment system is converted to nitrate (NO_3^-) via nitrite (NO_2^-) in the nitrification unit (Reaction equations 1 and 2). The oxidation of 1 mol NH_4^+ produces 2 moles of hydrogen ions (H^+). A buffer is used to keep the pH value stable (Reaction equation 3; Rennert 1981, Henze et al. 1995, Mudrack and Kunst 2003).

An effective denitrification unit causes an increase in alkalinity, resulting from the release of hydroxide ions (OH^-) during the denitrification process (Reaction equation 4; Mudrack and Kunst 2003). This makes it possible to buffer approximately 50 % of the protons generated in the nitrification process by recycling the water from the denitrification unit to the nitrification unit (Van Rijn et al. 2006). The ratio of nitrogen dosing to buffer consumption is therefore approximately 1 : 1.

Reaction equation 1: $NH_4^+ + 1{,}5\ O_2 \rightarrow NO_2^- + 2\ H^+ + H_2O$

Reaction equation 2: $NO_2^- + 0{,}5\ O_2 \rightarrow NO_3^-$

Reaction equation 3: $H^+ + HCO_3^- \rightarrow CO_2 + H_2O$

Reaction equation 4: $6\ NO_3^- + 5\ CH_3OH \rightarrow 3\ N_2 + 5\ CO_2 + 7\ H_2O + 6\ OH^-$

Material and methods

Description of the test system

An experimental system consisting of three components was designed for air treatment in the barn. First, the ammonia from the barn air was to be bound in the air scrubber and transported into the nitrification system via the wash water. Oxidation to nitrite and nitrate then took place in the nitrification process. The final step was the conversion to N_2 gas in denitrification. Only nitrification and denitrification are described in more detail below, as the entire system was not the subject of the study (Figure 1). Due to the COVID-19 pandemic, it was not possible to test the entire system in a pig fattening house. Therefore, the experimental system was set up in the technical centre of the Thünen Institute of Agricultural Technology, Braunschweig, and operated without the air scrubber with an artificial nitrogen dosage in the form of urea. In terms of data documentation by means of an electronic operating logbook (Elektronisches Betriebstagebuch (EBTB)), similar requirements were placed on the experimental system to those placed on certified exhaust air treatment systems (Arends et al. 2006, Gramatte and Häuser 2006, DLG 2018, Vera 2018, TA Luft 2021).

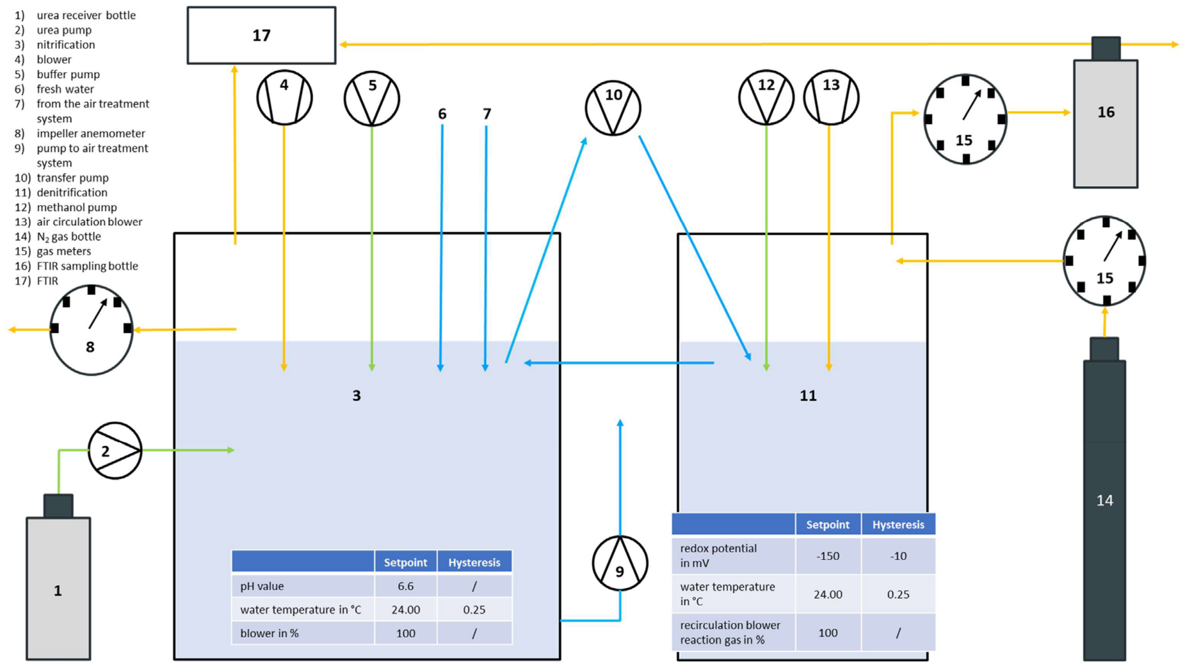
The experimental system was operated throughout the day, with water being exchanged between nitrification and denitrification only intermittently, in that the pump for transferring water from nitrification to denitrification was only switched on for eight hours during the day and then switched off again. This had proven to be optimal in preliminary tests. The methanol (CH_3OH) dosing was operated during these eight hours. The discontinuous operation of the system served to helped avoid the transfer of methanol from denitrification to nitrification and the associated disruption of the nitrification process. The experimental system was designed for twice the winter air rate in accordance with DIN 18910:2017-08 (2017) and 16 animal places.

Nitrification

The water volume was 129 l at standard fill level. The nitrification (3) contained 60 l of filling material (HEL-X BIOCARRIERS HXF12KLL, Christian Stöhr GmbH & Co. Elektro- und Kunststoffwaren KG, Marktrodach, Deutschland). The nitrification was a moving bed biofilm reactor (MBBR). The urea solution (1) was mixed ready for dosing and fed into the system using a dosing pump (2). A blower (4) continuously aerated the nitrification process with fresh air to supply oxygen (O_2). The air velocity (8) was measured manually at the air outlet of the nitrification unit. Sodium hydrogen carbonate at a concentration of 80 g l^{-1} was used as buffer solution (5). Dosing of sodium hydrogen carbonate was possible throughout and was carried out depending on the pH value of the water in the nitrification. The tank was equipped with sensors to determine the pH value, the water temperature and the oxygen content. A heating element was installed in the tank to reach the target temperature. The fresh water supply (6) for the entire system was provided by nitrification. The fill level was regulated via a fill level sensor. A peristaltic pump (10) supplied the denitrification with water from the nitrification.

Denitrification

The denitrification (11) had a water volume of 29 l at a standard filling level. The tank contained 12 l of the same type of filling material as in the nitrification. It was a so-called self-cleaning inherent gas denitrification reactor (SID reactor). The gas mixture in the tank was forced back into the water volume of the tank at intervals via a blower (13). Methanol was used as the carbon source (12) for the denitrifiers at a dilution of 1 : 5. The denitrification was actively gassed with nitrogen gas (14) in order to permanently provide a sufficient amount of gas volume for gas analysis by the Fourier transform infrared spectrometer (FTIR) (17). The gas flow rate was set to 1 l min^{-1}. Two drum gas meters (15) were used to measure the flow rates. A 50 l gas reservoir served as a gas collection tank (16) from which the FTIR took its samples. The denitrification system contained an identical pH sensor with integrated temperature sensor and the same heating element as in the nitrification system. A redox sensor was also installed. Excess water from the denitrification flowed back into the nitrification via a free return flow.



	Setpoint	Hysteresis
pH value	6.6	/
water temperature in °C	24.00	0.25
blower in %	100	/

	Setpoint	Hysteresis
redox potential in mV	-150	-10
water temperature in °C	24.00	0.25
recirculation blower reaction gas in %	100	/

Figure 1: Schematic illustration of nitrification (3) and denitrification (11) with the most important setpoint values

System support and data collection at the system

The system was checked several times a day and managed by two persons seven days a week according to a set protocol. The system was normally serviced once a week according to a maintenance schedule. If necessary, manual desludging was carried out or the sludge-laden water was removed, replacing a maximum of approximately 50 % of the tank contents, based on the standard fill level, with fresh water in the nitrification unit or with water from the nitrification unit in the denitrification unit. Desludging generally took place when the electrical conductivity in the nitrification unit increased to 20 mS cm^{-1} or to 25 mS cm^{-1} at the latest (Hahne et al. 2016, Hahne and Pfeiffer 2017).

Wet chemical analyses

The relevant nitrogen fractions, the electrical conductivity and the chemical oxygen demand (COD) were determined in the laboratory from water samples taken from the experimental system after filling to the standard level and rinsing the drain pipes (Table 1).

Table 1: Analysis methods for relevant water parameters

Parameter	Method of analysis
Ammonium-N (NH_4-N)	based on DIN 38406-5:1983-10 (1983) with 33 % technical sodium hydroxide solution
Nitrite-N (NO_2-N), Nitrat-N (NO_3-N)	DIN EN ISO 10304-1:2009-07 (2009)
Electrical conductivity	DIN EN 27888:1993-11 (1993)
Chemical oxygen demand (COD)	DIN ISO 15705:2003-09 (2003)

Determining the performance of the experimental system

The nitrification performance (NO_X-N) was calculated from the water samples taken at 08:00 a.m. and 04:00 p.m. and analysed for NO_2-N and NO_3-N, respectively. The nitrification performance was determined for 67 days for the nitrification unit and for 22 days for the degradation performance of the denitrification unit. From this data set, only those values were used for further evaluation for which the values from the EBTB for dissolved oxygen concentration, pH value, water temperature and redox potential in the nitrification or denitrification unit were in the optimum range (Table 2).

The evaluation scheme was derived from our own test results with the experimental plant and from literature references (Boon and Laudelout 1962, Forster 1974, Collins et al. 1975, Rennert 1981, Krüner and Rosenthal 1983, Ødegaard et al. 1994, Cheremisinoff 1995, Henze et al. 1995, Rusten et al. 1995, Lee et al. 2000, Mudrack and Kunst 2003, Vera 2018, Hahne 2019).

Table 2: Evaluation scheme for the system data of nitrification and denitrification as part of the performance determination

	O_2 content in mg l^{-1}	pH value	Water temperature in °C
Nitrification	< 0.5	< 6.0	< 12
	0.5–0.9	6.0–6.4	12–15
	1.0–9.0	6.5–7.5	> 15
	pH value change between 08:00 a.m. and 04:00 p.m.	**Redox potential at the end (04:00 p.m.) in mV**	**Water temperature in °C**
Denitrification	< 0.1	< -400	< 12
	/	/	12–15
	≥ 0.1	≥ -400	> 15

red = functionality of the system no longer guaranteed
yellow = tolerance range, but no longer in the optimum range
green = optimum range

The N emission factor for fattening pigs was based on 3 kg AP^{-1} a^{-1} with forced ventilation using the liquid manure method with partially or fully slatted floors without any emission reduction methods (TA Luft 2021). The experimental system was designed for the double winter air rate according to

DIN 18910:2017-08 (2017) (33.8 m^3 AP^{-1} h^{-1}) and 16 animal places. The average annual air rate is approximately 50 % of the maximum air rate according to Hahne and Pfeiffer (2017) and was assumed for the calculation of the system design with complete cleaning of the barn air. The test system was therefore able to clean a maximum of 68 % of the N emissions in relation to a complete air cleaning. The maximum N emission reduction of the experimental system was therefore 2,038 g N AP^{-1} a^{-1} (68 % of 3,000 g N AP^{-1} a^{-1} according to TA Luft (2021)).

The system performance in grams per day was related to the tank volume in litres. In a final step, the percentage system performance was calculated in relation to the system capacity.

The methanol dosing was carried out independently of the redox potential with fixed quantities, which are shown in Table 3. Only those days were taken into calculation that were included in the evaluation by means of the assessment scheme in Table 2 and the plausibility check.

Table 3: Methanol dosing on the days that were included in the evaluation to determine the performance of denitrification

N-dosage g d^{-1}	Number of measured values	Amount of methanol solution (1 : 5) g d^{-1}
15	6	90
15	4	150
30	2	350
60	2	900

Determination of the production of secondary trace gases and recording of the consumption of operating resources for the calculated evaluation of the overall process

The period from 24 August 2020 08:00 a. m. to 16 November 2020 07:59 a. m. was chosen to determine the nitrogen emissions via secondary trace gases and to record the consumption of operating resources. The N dosage was 30 g d^{-1} over the 84 test days.

Secondary trace gases

A FTIR with heated sample gas lines (sample gas cell and lines set at 80 °C) was used to record the secondary trace gases. The measuring points for outside air (for analysing the fresh air), nitrification (directly at the nitrification tank) and denitrification (at the gas collection tank) were sampled via a measuring point changeover switch. The first two were sampled alternately, while the denitrification measuring point was sampled approximately every 90 minutes. A zero-point measurement with pure nitrogen gas was carried out at 01:00 a. m. at night.

For the nitrification unit, the measured values for the outside air were subtracted from the measured concentrations. The measured values determined for the nitrification and denitrification units were multiplied by the respective standard volume flow (at 273.15 K) to calculate the emission loads. The N losses determined for N_2O, NH_3, NO and NO_2 from two hourly averages were summed separately for each tank over the observation period. In addition, the N emission was related to the N dosage in order to determine the percentage of nitrogen losses. For this purpose, the daily recorded nitrogen doses were added. For each of the four trace gases, the percentage of the total discharge was determined separately for each tank to measure the loss for each gas separately.

Consumables

The consumables for the experimental system were calculated as follows:

Water:

The water consumption of the experimental system was made up of the water losses via the blowdown and the water steam losses via the volume flow.

Blowdown volumes recorded by weight during manual water changes, were summed. Water losses by evaporation were recorded by the FTIR. The total water loss was calculated from this data and converted to the consumption per animal place and year.

Buffer and methanol:

Initially, methanol was dosed at a constant rate of approximately 350 g methanol (1 : 5) per day. Later, the methanol was automatically dosed according to the redox potential, with up to 350 g of methanol (1 : 5) dosed per day.

The values recorded daily were totalled for the period under review, then converted to the pure sodium hydrogen carbonate or methanol quantity and related to animal place and year.

The consumption volumes for all three media were then extrapolated to 100 % system capacity. The consumables were then calculated for the following three variants:

Variant 1: 2,038 g N TP^{-1} a^{-1} = 68 % N emission reduction (2-fold winter air rate, maximum capacity of the experimental system)

Variant 2: 1,019 g N TP^{-1} a^{-1} = 34 % N emission reduction (1-fold winter air rate)

Variant 3: 957 g N TP^{-1} a^{-1} = 40 % N emission reduction (emission reduction as from 2029 based on 2.91 kg NH_3 AP^{-1} a^{-1} according to TA Luft 2021)

The cost calculation was based on the media consumption extrapolated to 100 % system performance for the three variants and the prices listed in Table 4.

Table 4: Price overview (net) for the consumables used in the experimental system

Cost item	Net price in EUR	Reference
Water in EUR m^3 (price over 10,001 m^3 consumption per year)	2.14	(BS ENERGY, Braunschweiger Versorgungs-AG & CO. KG 2021)
Sodium hydrogen carbonate in EUR kg^{-1}	0.33	(KTBL 2020)
Methanol HP in EUR 760 kg^{-1}	706.80	(Klein 2023)

In the final step, the consumables of the experimental system were compared with those of a chemical air treatment system in terms of quantity and costs. For the consumables and the costs of the chemical air treatment system reference was made to the literature by Hahne (2006) and KTBL (2020). As the test system was a prototype and the chemical air treatment system is a system that is already certified and in use, a cost degression of 50 % was assumed for the experimental system due to scaling effects. In addition, the consumables determined for the buffer and methanol was compared with the stoichiometrically calculated buffer and methanol consumption for the experimental system. The water consumption of the experimental system remained unchanged, as the nitrogen is removed from the water during denitrification and the water can therefore be reused.

Results

Service and support requirements

Following the start-up of the experimental system, continuous operation was briefly tested, with the nitrification and denitrification units continuously connected to each other. However, repeated carryover of methanol from the denitrification unit to the nitrification unit meant that this mode of operation was considered impractical and the experimental system was switched to discontinuous operation. Methanol carryover inhibited nitrification and led to a drop in performance. Even in discontinuous operation, these carryovers could not be completely prevented and were often detected only after a delay, which affected the stability of the system.

Depending on the amount of methanol load transferred, the restoration of nitrification performance required a few days or even one to two weeks and was associated with a significant increase in working hours.

Secondary trace gases

In the experimental phase to determine the extent of secondary trace gas production, a total of 2,710 g N was introduced into the experimental system. The process conditions that existed during this phase in nitrification and denitrification are shown in Table 5. The losses via secondary trace gases amounted to 0.5 % in nitrification and 0.8 % in denitrification of the N input (Figure 2). In nitrification, the N losses via N_2O were the highest with a relative share of 90 %, while in denitrification, NH_3 contributed the most to the N losses with 45 %.

Table 5: Operating conditions of nitrification and denitrification during the determination of secondary trace gases and the calculative evaluation of the overall process. Data are presented as median with minimum and maximum in brackets.

Process stage	**Parameter**	**Measured value**	**Number of measurements**
Nitrification	pH value	6.6 (6.1–7.7)	3,984
	O_2 content in mg l^{-1}	5.2 (0.1$^{1)}$–7.9)	3,984
	Water temperature in °C	24.2 (20.9–28.0)	3,984
	Air speed in m s^{-1}	5.1 (4.4–5.4)	43
Denitrification	pH value	8.6 (7.0–9.4)	3,984
	Redox potential in mV	-169.7 (-498.7–57.1)	3,984
	Water temperature in °C	24.3 (22.4–27.3)	3,984
	Volume flow in m^3 d^{-1}	1.4 (0.0$^{2)}$–1.6)	85

All measured values, except air speed and volume flow, were taken from the logged half-hourly average values of the experimental system.

1) Sensor covered with biomass for a few hours on one day.

2) 2 days without active nitrogen fumigation due to tests on the FTIR.

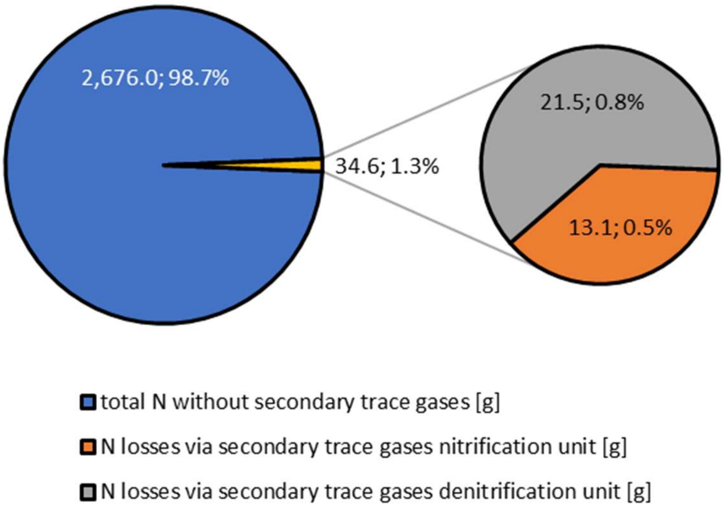


Figure 2: N losses of secondary trace gases from nitrification unit and denitrification unit in relation to the N input of 2,710.6 g via urea dosing (= 100 %); total N without secondary trace gases = N in the blowdown water of both tanks + N accumulation in the water of both tanks + emitted nitrogen gas

Determining the performance of the system

As part of the performance determination, the process conditions shown in Table 6 were determined for both tanks from the half-hourly average values.

Table 6: Experimental system parameters during performance determination for nitrification and denitrification. Data are presented as median with minimum and maximum in brackets.

Process stage	**Parameter**	**Measured value**	**Number of measurements (half-hourly averages)**
Nitrification unit	pH value	6.6 (6.4–8.5)	400
	O_2 content in mg l^{-1}	6.1 (0.1[1]–8.8)	400
	Water temperature in °C	25.7 (21.0–28.2)	400
Denitrification unit	pH value	9.2 (7.9–9.5)	224
	Redox potential in mV	-113.7 (-392.2–33.8)	224
	Water temperature in °C	25.0 (22.1–28.0)	224

[1] Oxygen sensor disturbed on one day, manual measurements resulted in values between 2.6–3.2 mg l^{-1}, n = 12.

The oxidation performance of nitrification was determined at seven different N doses. It was thereby shown that an oxidation performance was measurable even in the absence of N dosing. In denitrification, the degradation performance was analysed at three different N doses. The average C : N ratio was 0.69 : 1 and the methanol : N ratio was 1.85 : 1. In both tanks, strong variations in oxidation and degradation performance were observed within the respective N doses (Figure 3). Based on the oxidation and degradation performance, an average experimental system performance of 33 % was determined for the overall system in relation to the system capacity (Table 7).

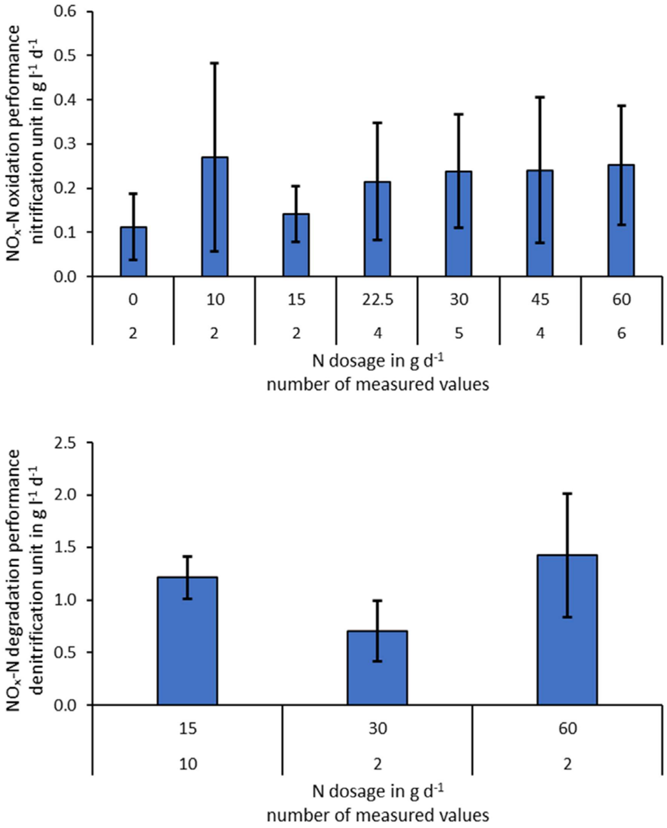


Figure 3: Oxidation performance of nitrification (top) and degradation performance of denitrification (bottom) at different N doses in relation to the respective tank volume (mean value ± standard deviation)

Table 7: Determination of performance for nitrification and denitrification as well as the total system performance in relation to the maximum system capacity.

Nitrification unit		**Denitrification unit**	
Number of measurements	25	Number of measurements	14
Oxidation performance NO_X-N in g l^{-1} d^{-1} (mean value ± standard deviation)	0.21 ± 0.06	Degradation performance NO_X-N in g l^{-1} d^{-1} (mean value ± standard deviation)	1.11 ± 0.37
Oxidation performance NO_X-N in g d^{-1} (mean value ± standard deviation)	27.12 ± 7.72	Degradation performance NO_X-N in g d^{-1} (mean value ± standard deviation)	32.34 ± 10.70
Oxidation performance NO_X-N in g a^{-1}	9,898.76	Degradation performance NO_X-N in g a^{-1}	11,802.34
N emissions[1)] in g AP^{-1} a^{-1}	2,038.19	N emissions[1] in g AP^{-1} a^{-1}	2,038.19
Oxidation performance NO_X-N in AP^{-1}	4.86	Degradation performance NO_X-N in AP^{-1}	5.79
Capacity of the system in AP^{-1}	16	Capacity of the system in AP^{-1}	16
Performance of the system[2] in %	30.35	Performance of the system in %	36.19
Average **total performance** from nitrification and denitrification[2)] in %		**33.27**	

AP = animal place

[1)] Base: 3,000 g N AP^{-1} a^{-1} (TA Luft 2021); 2,038.19 g N AP^{-1} a^{-1} correspond to the maximum system capacity for the experimental system (double winter air rate).

[2)] System performance in relation to the maximum system capacity of 16 fattening pig places.

Calculated evaluation of the process

The water consumption was made up of the sludge volumes of 129 l (nitrification) and 11 l (denitrification unit) as well as water evaporation losses of 29 l in nitrification and 12 l in denitrification.

The consumption of sodium hydrogen carbonate (pure substance) totalled 10.7 kg. The buffer consumption was thus four times as high as the dosed N quantity of 2.7 kg.

The methanol consumption (pure substance) was 0.546 kg. The C : N ratio was 0.08 : 1.

The consumption values shown in Table 8 were calculated based on the measured consumption. Considering a cost degression of 50 %, the experimental system performed worse in the cost calculation, with costs almost twice as high as for a chemical air treatment system (Table 9). If the stoichiometric consumption under optimum conditions is assumed for the buffer and methanol consumption, the experimental system performs with slightly lower costs than a chemical air treatment system (Table 8 and 9).

Table 8: Comparison of the calculated (stoichiometric) and the measured consumables for the experimental system and a chemical air treatment system according to literature data

Air treatment system	**Experimental system 1)**				**Chemical air treatment system**		
Data source	Calculated (stoichiometric) optimum conditions for buffer and methanol consumption 2)	Measured values			Values according to literature 3)		
Cleaning performance in g N AP^{-1} a^{-1}	2,038 = twice the winter air rate	2,038 = twice the winter air rate	1,019 = winter air rate	957 = reduction from 2029 on the basis of 2.91 kg NH_3 AP^{-1} a^{-1} 4)	3,000 4)	1,500	957 = reduction from 2029 on the basis of 2.91 kg NH_3 AP^{-1} a^{-1} 4)
Calculated emission reduction in %	68	68	34	40	100	50	40
Water consumption in l AP^{-1} a^{-1}	432.1	432.1	216.0	203.1	40.0	20.0	12.8
Buffer consumption 5) in kg AP^{-1} a^{-1}	2.0	25.6	12.8	12.0	/	/	/
Methanol consumption in kg AP^{-1} a^{-1}	3.9	1.3	0.7	0.6	/	/	/
Acid consumption 6) in kg AP^{-1} a^{-1}	/	/	/	/	10.9	5.5	3.5

AP = animal place

1) The data refer to a system performance of 100 % based on the actual system performance determined in Table 7.

2) For buffer consumption, a stoichiometric ratio of 1 g sodium hydrogen carbonate to 1 g dosed N was assumed. Stoichiometrically, 50 % of the buffer capacity is recovered from denitrification (van Rijn et al. 2006). For methanol consumption, a stoichiometric ratio of 2.32 g methanol per g nitrate was assumed. This corresponds to a C : N ratio of 0.71 : 1 (Sauthier et al. 1998).

3) HAHNE (2006).

4) TA LUFT (2021).

5) Buffer = sodium hydrogen carbonate.

6) Calculation basis: 3 kg acid per kg NH3 input (Hahne 2006) and 3.64 kg NH3 emissions AP-1 a-1 (TA Luft 2021).

Table 9: Comparison of the net costs for the calculated (stoichiometric) and the measured consumables for the experimental system and a chemical air treatment system according to literature data

Air treatment system	**Experimental system 1)**				**Chemical air treatment system**		
Data source	Calculated (stoichiometric) optimum conditions for buffer and methanol consumption 2)	Measured values			Values according to literature 3)		
Cleaning performance in g N AP^{-1} a^{-1}	2,038 = twice the winter air rate	2,038 = twice the winter air rate	1,019 = simple winter air rate	957 = reduction from 2029 on the basis of 2.91 kg NH$_3$ AP^{-1} a^{-1} 4)	3,000 4)	1,500	957 = reduction from 2029 on the basis of 2.91 kg NH$_3$ AP^{-1} a^{-1} 4)
Calculated emission reduction in %	68	68	34	40	100	50	40
Water in EUR AP^{-1} a^{-1}	0.93	0.93	0.46	0.44	0.48	0.24	0.15
Acid in EUR AP^{-1} a^{-1}	/	/	/	/	2.28	1.14	0.73
Buffer 5) in EUR AP^{-1} a^{-1}	0.67	8.44	4.22	3.97	/	/	/
Methanol in EUR AP^{-1} a^{-1}	3.61	1.21	0.61	0.57	/	/	/
Total costs in EUR AP^{-1} a^{-1}	5.21	10.58	5.29	4.97	2.76	1.38	0.88
Total costs with cost degression 6) in EUR AP^{-1} a^{-1}	2.61	5.29	2.65	2.49	2.76	1.38	0.88

All costs are net costs.

AP = animal place

1) The data refer to a system performance of 100 % based on the actual system performance determined in Table 7.

2) For buffer consumption, a stoichiometric ratio of 1 g sodium hydrogen carbonate to 1 g dosed N was assumed. Stoichiometrically, 50 % of the buffer capacity is recovered from denitrification (van Rijn et al. 2006). For methanol consumption, a stoichiometric ratio of 2.32 g methanol per g nitrate was assumed. This corresponds to a C : N ratio of 0.71 : 1 (Sauthier et al. 1998).

3) KTBL (2020).

4) TA Luft (2021).

5) Buffer = sodium hydrogen carbonate

6) A cost degression due to scaling effects of 50 % was assumed for the experimental system, as it was a prototype. The chemical air treatment system is a standard system with standard costs.

Discussion

Error-free operation of the experimental system could not be guaranteed at all times, even with a high level of supervision. Methanol was repeatedly carried over from the denitrification unit into the nitrification unit, resulting in a sustained inhibition of nitrification and thus a loss of performance. Demmers (1992) made a similar experience with a biological scrubber with connected denitrification regarding the inhibition of nitrification by methanol carryover and the necessary intensive system support.

The operation of the experimental system showed only very low N losses via secondary trace gases of 0.5 % in nitrification and 0.8 % in denitrification in relation to the dosed N quantity. Together, 0.6 % N_2O emissions escaped from both tanks in relation to the N dosage. Compared to other systems, these values were very low and therefore very positive. The following values can be found in the literature:

- 4 % (average) N_2O emissions in relation to NH_3-N for a trickling filter (Melse et al. 2012)
- 3 % N_2O emissions in relation to the NH_3 input in a two-stage packed-bed air treatment system operated with water (Hahne and Vorlop 2004).
- 0.9–1.3 % N_2O emissions in relation to the nitrogen load in a deammonification process (Weissenbacher et al. 2012)
- 3 % in wastewater treatment plants (Kampschreur et al. 2009)

Dumont (2018) states that the use of bio air treatment systems to remove NH_3 is acceptable as long as less than 3 % N_2O are emitted in relation to the NH_3 input. N_2O emissions are particularly important, as N_2O is a climate-damaging gas with almost 300 times the activity of carbon dioxide over one hundred years (Weissenbacher et al. 2012).

The system performance was only 33 % in relation to the system capacity and showed the missing practical capability. The results of the oxidation performance in nitrification showed significant variations. A possible explanation for these large variations could lie in a very slow urea hydrolysis, so that the dosed urea was not converted directly into ammonium, but with a time delay and then possibly in larger quantities, and this was then oxidized to nitrate. Due to the delay in oxidation, the determined oxidation rates no longer matched the actual quantities of N dosed on the sampling days. The degradation performance of denitrification also fluctuated considerably and showed very high performance, particularly at the N dosage of 15 g d^{-1}. Here too, the delayed provision of nitrate by nitrification may have had an influence. However, poor conditions could be excluded at both stages of the process (Table 6). The C : N ratio of 0.69 : 1 and the methanol : N ratio of 1.85 : 1 were, however, at the lower end of the literature data when determining the performance. During the period for evaluating the imputed overall process, the C : N ratio of 0.08 : 1 was even lower and thus far from the literature data:

- 2.3 : 1 C : N (Torno et al. 2018)
- 1.5 : 1 C : N (Kaiser 1987)
- 1 : 1 C : N (Balderston and Sieburth 1976)
- 0.71 : 1 C : N stoichiometric for complete denitrification (Sauthier et al. 1998)
- 1.3 : 1 C : N (Lompe and Wiesmann 1991)
- 2–3 mg methanol per mg NO_3-N reduced to N_2 (Becker 1979)
- 1.90 g methanol per g nitrate-N plus 0.57 g methanol for bacterial growth, resulting in a total amount of 2.47 g methanol per g nitrate-N (Kadlec and Wallace 2009)
- 2.7 : 1 methanol to NO_3-N (Clifford and Liu 1993)

The low methanol consumption during the period for evaluating the calculated overall process resulted from the redox potential of -150 mV, which remained below the setpoint value. An effect of the N_2 gas from the active fumigation cannot be completely excluded. It is likely that the N_2 gas affected the measuring sensor on several days, resulting in lower redox potential readings, but the target value was not exceeded and therefore no methanol was added. This could also be an explanation for the sometimes extremely low measured values for the redox potential and the very low C : N ratio, as all other parameters showed no abnormalities (Table 5).

Contrary to theoretical principles, the consumption of sodium hydrogen carbonate was 4 times as high as the N dosage. The reason for this could be delayed urea hydrolysis, resulting in more NH_4^+ produced during the monitoring period than expected from the added nitrogen dosing. Nevertheless, the denitrification unit did not reach its full performance potential, which prevented sufficient production of pH-stabilising hydroxide ions. It should be emphasised that the general conditions in both tanks were optimal (Table 5).

When calculating the costs for the experimental system, the significantly higher costs for the consumables at EUR 5.29 AP^{-1} a^{-1} compared to a chemical air treatment system at EUR 2.76 AP^{-1} a^{-1} are striking. With a stoichiometric calculation of the buffer and methanol consumption, the costs fall just below the costs of the chemical air treatment system. However, it should be noted that the stoichiometric calculation represents a theoretical amount which would be higher in practice, as not all of the carbon from the methanol would be available for the denitrification process (Sauthier et al. 1998).

It became apparent that the experimental system in this configuration is not yet suitable for practical use in terms of labour input, reliability, consumables and costs. On the other hand, the very low formation of secondary trace gases was positive.

Conclusions

All things considered, recirculating air treatment system with nitrification and denitrification is a complex process that is not yet ready for practical application. However, some practical conclusions can be drawn:

1. Separation of the nitrification and denitrification process stages is necessary as unused methanol leads to significant losses in nitrification performance.
2. The possible methanol input into the nitrification stage must be prevented by improved process control.
3. The media consumption of the process must be significantly reduced for the practical implementation of the system, also in comparison with the chemical air treatment system.
4. As the recirculating air cleaning system is an interesting process from the point of view of environmental protection, animal welfare and working conditions, it would be useful to analyse partial air cleaning with a sulphuric acid operating mode.

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