Validation Methods for the Ammonia Removal of an Air Scrubber on a Poultry House Using the Acid Use and the Process Water Nitrogen Balance

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Highlights

- Acid use in an acid air scrubber predicts the ammonia removal of an air scrubber.
- Nitrogen balance of process and discharge water in an air scrubber predicts ammonia removal of an air scrubber.
- Long-term monitoring of acid use and/or the nitrogen balance can validate the ammonia reduction of an air scrubber.
- The two validation methods can support local authorities in granting environmental permits and enforcement.

Abstract. *Ensured ammonia reduction by novel technologies and guaranteed low ammonia emission from livestock houses is essential for the granting and monitoring of legal permits. This study focuses on methods to underpin the long-term effectiveness of an air scrubber and was done in a commercial poultry house. The measured ammonia removal by an air scrubber with ammonia sensors (method 1) was compared to the acid use for ammonia binding (method 2) and the nitrogen balance over the process water (method 3). Also, the effect of the length of the time period used for the comparison was evaluated, as was the effect of the spatial variation of the ammonia concentration in the house. The overall weekly data showed a clear linear relation between method 1 on the one hand and methods 2 and 3 on the other hand, with regression coefficients close to 1 and a variation of +/- 20% on a 4-weekly basis. The 4-weekly data showed a clearly reduced variation compared to weekly data, due to leveling out of the impact of irregular time events like acid dosage and water discharge. The barn emission assessed by method 1 was significantly (12%) influenced by the spatial aerial concentration differences of ammonia in the house. Taking this into account, method 2 overestimated the emission reduction of method 1 with 16.8% and method 3 was in good agreement with method 1. For method 2, a revised value for acid use per kg of ammonia binding needs to be substantiated in further research. Possible measurement errors influenced calculated ammonia removal with a maximum of +/-2%, but the relative effect on remaining emissions was larger with +/- 15% for method 3. This study shows that monitoring acid use and/or the nitrogen balance can validate the ammonia reduction of air scrubbers and the remaining ammonia emission and support local authorities in granting environmental permits and their enforcement.*

Keywords. Acid, Air scrubber, Ammonia, Nitrogen balance, Poultry.

Aerial nitrogen losses from agriculture can lead to nitrogen deposition in nature areas, resulting in loss of biodiversity, eutrophication, and pollution of drinking water (de Vries et al., 2021). The majority (87%) of the total yearly ammonia emission in the Netherlands in 2021 (122 million kg of ammonia), was caused by agriculture, of which 54% originated from cattle/dairy production, followed by pigs (17%) and poultry (11%) (CBS, 2023). In many European countries, measures have been taken to decrease ammonia and, thus, nitrogen emissions from livestock buildings (Melse et al., 2009). Examples of techniques and management measures used for poultry houses to reduce ammonia emissions are drying of manure on belts (both inside and outside the house), manure removal by belts, air scrubbers, and heat exchangers (Santonja et al., 2017; Shepperd et al., 2017; Van der Heyden et al., 2015; Hahne, 2022).

Measurement protocols are developed by research institutes, and these are used to validate the actual emission from housing systems with emission reduction techniques in practice (Melse et al., 2009). In measurement protocols for air

scrubbers, in some cases, besides the gaseous reduction of ammonia concentration, the nitrogen balance over the process water has to be monitored. For example, the German test institute for livestock technology (DLG) requires system approval that at least 70% of the reduced gaseous ammonia must be retrieved in the process water (DLG, 2022).

When emission reduction systems are applied in commercial livestock production, the actual emission can deviate by more than 25% from the measured emission during an official test (Dekock et al., 2009). The nutrient content of the diet and the type of bedding, which are generally not prescribed in emission reduction systems – are known to affect the ammonia emission, respectively, up to 37% (Rahman et al., 2012) and 47% (Van Harn et al., 2012). Also, the stocking density affects the ammonia emission per bird (Mendes et al., 2012). These could be reasons why Van Bruggen and Geertjes (2019) found that manure from poultry houses with low emission techniques, such as frequent manure removal with manure belts and manure drying techniques in the poultry house, did not, in all cases, result in the expected higher nitrogen content of the manure. This raised doubts about the effectiveness of the technologies to reduce ammonia emission.

The ammonia removal efficiency of air scrubbers - as commonly assessed under both controlled test as well as practical farm conditions by measuring the aerial ammonia concentration before and after passing the scrubber - is typically 80% or higher for acid air scrubbers when managed according to guidelines (Melse, 2009; Hadlocon et al., 2014; Mostafa et al., 2020). However, it is known that some pollutants as aerosols, can pass through the air scrubber (Aarnink et al., 2011). These aerosols can contain droplets from the recirculated acid process water, and therefore nitrogen dissolved in these aerosols can be lost from the system without noticing this in the measured gas concentration of the outgoing air. Especially at high ventilation rates, micron sized particles are less effectively separated than bigger waterdrops (Brouwers et al., 2012). This could happen under normal practical conditions, but especially when so-called droplet catchers are not properly designed or installed. Earlier studies showed that between 82.1% and 97% of the expected nitrogen was found in the acid process water (DLG, 2016a,b). Especially the lower retrieval number/values could indicate that more nitrogen/ammonia is lost from the system than measured by the aerial ammonia sensors.

Also, a lack of maintenance resulting in downtime for pumps and other technical parts can influence the effectiveness of the ammonia scrubbers (Melse, 2009). In response to these operating failures, additional measurements (of pH and conductivity of the process water and air pressure drop over the filter) and control and data logging systems were introduced and implemented at farms in the Netherlands (Infomil, 2018). This data can be used by local authorities to validate the environmental permits of farmers. However, these additional measurements are all related to parameters describing the conditions of the air scrubber process, not the ammonia emission itself or the reduction achieved. So, the real ammonia emission or ammonia reduction is still unknown, and downtime will affect the real environmental emissions.

In the Netherlands, this has resulted in a broad debate about the effectiveness of ammonia-reducing techniques in the entire animal husbandry, with impacts on the validity of, for example, environmental permits for livestock farms and the legal underpinning of national emission factors. So, there is an urgent need for reliable and robust barn mitigation techniques that ensure ammonia reduction effectiveness (Sutton et al., 2015).

In this paper, we focus on technological methods to underpin the effectiveness of air scrubber technology under commercial livestock circumstances, with a poultry housing system as an example. The measured ammonia emission reduction, or ammonia removal by air scrubber technology, was compared to the acid use for binding ammonia in the process water and the nitrogen balance of the process water. This was done for a poultry house with an ECO Air Care ventilation system.

ECO Air Care is a new technology for ventilation of poultry houses (that combines preheating or cooling of all ingoing air) and acid scrubbing of all outgoing air and consists of one or more air treatment units. Similar technology, where air scrubbing and heat recovery are combined, was tested in a laboratory pilot by Krommweh and Büscher (2021) and was also described by Deeken et al. (2023) with the idea that the heat recovery can compensate for increased operating costs due to the exhaust air purification system.

In this study, the poultry house had four air treatment units. The goal of this study was to compare three different methods for determining the ammonia removal by an air scrubber system under commercial livestock conditions. The three methods, including their sensors and equipment, were relatively cheap and could easily be applied in commercial situations, enabling multiple year monitoring. The three methods are:

1. The ammonia removal based on the ammonia concentrations in the ingoing and outgoing air flow of the air treatment unit.
2. The ammonia removal based on the acid use for the chemical reaction between ammonia and dissolved sulfuric acid.
3. The ammonia removal based on the increased nitrogen concentration of the process and discharged water (nitrogen balance).

Secondly, the effect of the length of the time period used for the balances was evaluated. These periods should be long enough to level out the effect of the dynamics of acid dosing and process water discharge from the system; in this study, a one week period and a four-week period were evaluated. Thirdly, the effect of spatial variation on the ammonia concentration in the house and consequently the number of air treatment units where ammonia concentrations were measured for method 1 was evaluated; we compared the effects of measuring on 1 and 2 air treatment units. Finally, a sensitivity analysis was done to assess the effect of measurement errors on both the ammonia removal and the remaining ammonia emission for all three methods.

MATERIALS AND METHODS

POULTRY HOUSE DESCRIPTION AND ANIMAL PRODUCTION

The poultry house had 18,500 broiler breeder places and was located in Hoogeloon, The Netherlands. The dimensions of the house were 105 x 24 m. In the house (see fig. 1 for plan view and figs. 2 and 3 for a cross section) there were two rows of laying nests. Next to these nests, wooden slats were positioned on both sides over the full length of the house. Under these slats, the manure was stored in a pit during the entire production period of 40 weeks. A scratching area (also used for foraging and dustbathing by the birds) is present over the entire floor area except under the nests and the slats. Above the slats and in the scratching area, there is

a drinking system, feeding pans for the roosters, and feeding lines for the hens. The stocking density was 7.3 birds per m² at the start of the flock. Table 1 presents some characteristics of the animal production of the flock during which the research was done, reflecting a good production flock for broiler breeders (Ross, 2016).

Table 1. Main production characteristics of the birds during the experimental flock.

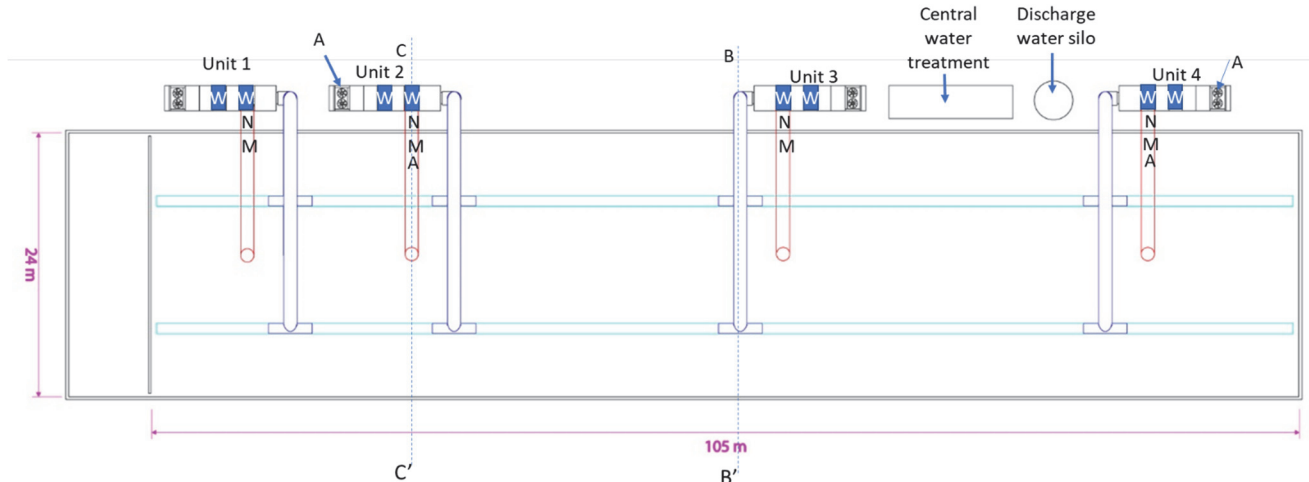|	
Number of animals at start of the flock	18,500
Breed	Ross 308
Average feed intake (g/day per bird)	157
Egg production @ 60 weeks (eggs per hen housed)	204
Relative number of floor eggs	0.4%
Cumulative mortality at 60 weeks	2.7%

Figure 1. Plan view of the poultry house with the four air treatment units: M = position of measuring fan, N = position of nozzle for spraying process water, W = section of the unit where process water is sprayed, A = ammonia concentration sensor. Cross sections B-B' with the air inlet system and C-C' with the air outlet system are presented in figures 2 and 3. PVC tubes for return water between the units, water treatment, and silo are left out of this plan view.
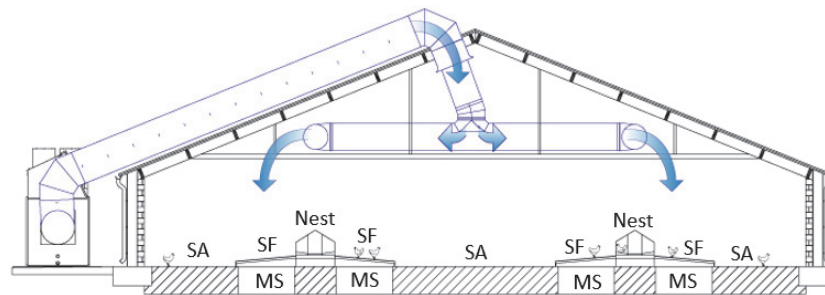
Figure 2. Cross section of the experimental house (B-B' in fig. 1). The arrows indicate the incoming fresh airflow, which is cooled in the summer and heated in winter due to the heat exchange in the air treatment unit; "Nest" indicates the laying nests; MS = manure storage; SA = scratching area with litter; SF = slatted floor.
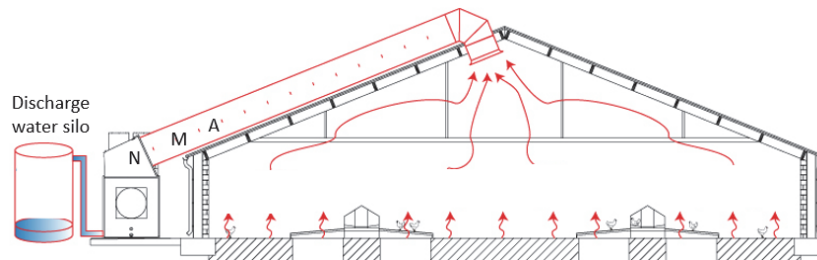

Figure 3. Cross section of the experimental house (C-C' in fig. 1). The arrows indicate the outgoing airflow with gaseous water vapor, ammonia and particulate matter: M = position of measuring fan, N = position of nozzle for spraying process water, and A = ammonia concentration sensor.

VENTILATION AND AIR SCRUBBER SYSTEM

Ventilation and air scrubbing were realized with an ECO Air Care system (Vencomatic, Eersel, the Netherlands), that was placed next to the poultry house. This system arranged all inlet and outlet ventilation flows of the house, there were no other air inlets or fans (see fig. 1 for a plan view). It consisted of four air treatment units that conditioned the ingoing air and scrubbed the outgoing air. Each unit had a maximum ventilation capacity of 30,000 m³/h. The central water treatment system consisted of a sedimentation tank (25.3 m³), a 'clean water' tank (with a volume between 10.7 and 15.7 m³), an acid storage and dosing unit, and a silo for saturated process water (90 m³). The air treatment units were connected to the central water treatment unit by 2 PVC tubes in the ground for supply and return of process water (volume 3.0 m³); the return water from the units was first collected in a collection pit (volume between 3.7 and 4.2 m³) and from there pumped into the sedimentation tank. The total amount of process water in the system varied between 42.3 and 47.6 m³. This variation was a result of the evaporation of process water or the condensation of water vapor in the process water. When the water level in the clean water tank reached below 10.7 m³, tap water was added to the system.

The first function of the air treatment units was to exchange energy between the outgoing and incoming air according to the counterflow heat exchange principle. This air treatment unit is the successor of the heat exchanger described by Bokkers et al. (2010), whose maximum thermal efficiency was determined at 85% by Goselink and Ramirez (2019). The ventilation rate of all four air treatment units was the same because they were controlled in parallel.

The second function of the air treatment units was to cool the ingoing air under warm outside conditions. The incoming air is cooled based on indirect evaporative cooling (Kabl, 2015). In order to cool the ingoing air, process water was sprayed in the outgoing exhaust air (at location N in figs. 1 and 3; continuously) and in the air treatment unit (per unit at 2 locations W in fig. 1; 1 minute per 5 minutes, alternating between the two locations). At locations designated 'W,' process water was sprayed with a volume of 11 L s⁻¹ and relatively low pressure (2 bar), which limits the formation of small droplets compared to the use of high-pressure water nozzles. This spraying wetted the outside of the heat exchanger panels, creating an exchange surface between outgoing air and the wet surface for evaporation. The cooling reduced the temperature of the ingoing air to 23°C or lower, also under outside conditions of 30°C or more. After the sprayer sections, the exhaust air traveled at least 4 m through the air treatment unit, resulting in this remaining part of the heat exchanger functioning as a droplet remover to prevent the emission of small droplets.

Because of indirect evaporative cooling, the ECO Air Care system has a substantial lower ventilation capacity, maximum 6.3 m³ h⁻¹ per bird, as compared to the two times higher ventilation capacity for conventional ventilation systems for broiler breeders in the Netherlands. The fresh conditioned air was distributed in the house with perforated air tubes above both rows of nests, as shown in figure 2. The exhaust air was sucked away through four openings in the roof (one per unit) in the center of the house, as shown in figure 3. Tubes over the roof for both ingoing and outgoing air connected the air treatment units to the house.

The third function of the units was to reduce ammonia emissions. The ammonia reducing principle of the ECO Air Care system was based on the use of acid in the process water, similar to the principle used in conventional acid air scrubbers (Melse and Willers, 2017; Van der Heyden et al., 2015). The wet outside of the heat exchange panels created a large exchange surface area for ammonia to dissolve in the process water.

Sulfuric acid was added to the process water, forming ammonium sulfate when bound with gaseous ammonia. The conductivity of the process water was measured with a Jumo CTI-500 conductivity sensor in the central water supply from the clean water tank towards the ECO Air Care units, and this indicated the concentration of ammonium sulfate. When this concentration reached a maximum level (EC value of 200 mS cm⁻¹), process water was discharged from the clean water tank into a silo, and tap water was added to the system. The excess condensation water, due to the cooling of outgoing air in the air treatment units, was also discharged to the silo by opening a valve, resulting in part of the waterflow towards the units flowing into the silo.

The pH of the process water was measured with a Jumo pH sensor (Article number 00300151) to control the acidity of the process water in the central water supply to the ECO Air Care units. The signal from this sensor was used to control the dosing pump, which added sulfuric acid to the process water. In this study, the pH was set at 3.0. The acid pump was activated when the pH level reached a value of 3.1, and stopped when the measured pH value of the process water was 2.9. The 98% concentrated sulfuric acid was stored in a 1,000 L vessel, and the acid was dosed into the process water with a dosing pump (Etatron PKX0102801).

MEASUREMENTS AND DATA PROCESSING

Research was done during four periods, each spanning four weeks, and spread out over the year to cover different climatic conditions due to seasonal changes. All these periods (see table 2) were during one flock that lasted a total of 40 weeks (from March to December 2022). The average outside temperature per period ranged from 4.1°C to 19.5°C, while the inside temperature was relatively constant between 22.6°C and 24.9°C (see table 2). All collected data were retrieved from sensors belonging to the control system of the installed air scrubbing/ventilation system; no specific research sensors were installed.

Method 1: Gaseous Ammonia Concentrations and Ventilation Rate

Gaseous ammonia concentration was measured with a Dräger Polytron C300 sensing head with an integrated NH_3-AL electrochemical ammonia sensor. The range of the sensor was 0-100 ppm. According to Melse et al. (2016), the detection limit of this sensor is 0.23 ppm. The accuracy is ± 0.11 ppm for the range of 0-1 ppm, and ± 0.17 ppm (equivalent to 0.12 mg/m³) for the range of 1-11 ppm. According to Melse et al. (2016), the Dräger sensor was found to be equivalent to the reference method described by Ogink et al. (2017) for ammonia concentrations > 1 ppm, and the Dräger

Table 2. Start and end dates of each 28-day period, including inside and outside climatic conditions.

Period	1	2	3	4
Start date	8-4-2022	1-7-2022	14-9-2022	21-11-2022
End date	5-5-2022	28-7-2022	11-10-2022	18-12-2022
Mean outside temperature (°C)	11.9	19.5	12.3	4.1
Mean outside relative humidity (%)	62	67	82	85
Mean inside temperature (°C)	23.0	24.9	23.6	22.6
Mean inside relative humidity (%)	46	52	50	47
Units with ammonia sensors	unit 2	unit 2	unit 2	units 2 and 4

sensor is applicable for indicative measurements for ammonia concentrations < 1 ppm.

The ammonia sensors were initially only placed in the exhaust air flow of air treatment unit 2 (indicated with A in figs. 1 and 3), assuming this concentration was representative of air treatment units 1, 3, and 4. The first ammonia sensor was placed in the tube at the roof of the house, 2 meters before the air enters the air treatment unit, this was directly behind the position of the measuring fan (location M in fig. 1, estimated accuracy +/- 2% of measured value with air velocity > 0.4 m/s), which was present in the outgoing flow of all units. With these measuring fans, the ventilation rate was determined according to the measuring protocol which is used in the Netherlands to determine emissions from livestock housing systems (Ogink et al., 2017). The second ammonia sensor measured the ammonia concentration of the outgoing air after passing air treatment unit 2 before it entered the environment. During measurement period 4, two additional ammonia sensors were placed in the outgoing air flow of air treatment unit 4. Mean values of both ammonia concentrations and ventilation rates were logged every 5 minutes. All ammonia sensors were calibrated by the factory at the start and after period 4; the measuring fans were calibrated in a wind tunnel (Appendix 1).

The ammonia sensor measured the concentration in ppm. Converting the concentration in ppm to mg m^{-3} was done with equation 1:

$$Cg_{NH_3} = \frac{Cp_{NH_3} * M}{V_{normal}} \qquad (1)$$

where

Cg_{NH_3} = the ammonia concentration in g m^{-3}
Cp_{NH_3} = the ammonia concentration in ppm
M = the molar mass of ammonia (17.03 g mol^{-1})
V_{normal} = the specific molar volume in m^3 mol^{-1}.
V_{normal} was calculated with equation 2:

$$V_{normal} = R * \left(\frac{T}{p}\right) \qquad (2)$$

where

R = the gas constant (8.314 J K^{-1} mol^{-1})
T = the air temperature in kelvin
p = the ambient pressure in Pa.

To calculate V_{normal}, an average temperature of 295 K and an average pressure of 101,325 Pa (sea level) were used.

The ammonia reduction per period of 1 and 4 weeks was calculated with equation 3:

$$R_{NH3,gas} = \sum_{i=1}^{n} \left(Cghouse_{NH_3,i} - Cexhaust_{NH_3,i}\right)$$
$$\times \left(\varphi v_{1,i} + \varphi v_{2,i} + \varphi v_{3,i} + \varphi v_{4,i}\right) * \frac{1000}{12} \qquad (3)$$

where

$R_{NH3,gas}$ = the cumulative ammonia removal in kg
$Cghouse_{NH_3}$ = the ammonia concentration in the air before passing the air treatment unit in g m^{-3}
$Cexhaust_{NH_3}$ = the ammonia concentration after passing the air treatment unit in g m^{-3}
φv_{1-4} = the ventilation flow of air treatment unit 1 until 4 in m^3 h^{-1}
i = a 5 minute time interval
n = 2016 when calculating weekly values and 8064 when calculating 4-weekly/period average values.

The emission removal efficiency based on gaseous ammonia concentration measurements was calculated with equation 4:

$$\eta_{NH_3} = \frac{\left(\frac{\sum_{i=1}^{n} Cghouse_{NH_3,i}}{i}\right) - \left(\frac{\sum_{i=1}^{n} Cgexhaust_{NH_3,i}}{i}\right)}{\left(\frac{\sum_{i=1}^{n} Cghouse_{NH_3,i}}{i}\right)} * 100 \qquad (4)$$

where

η_{NH_3} = the ammonia removal efficiency in%
$Cgexhaust_{NH_3,i}$ = the ammonia concentration after passing the air treatment unit in g m^{-3}
$Cghouse_{NH_3,i}$ = the ammonia concentration in the air before passing the air treatment unit in g m^{-3}.

The concentration of ammonia in the fresh inlet air was assumed to be 0.

Method 2: Sulfuric Acid Usage

The level of liquid in the sulfuric acid storage was measured automatically with a wireless FMCW radar sensor placed on top of the storage vessel (type WLR05-2G, producer Staal Instruments; accuracy of +/- 10 L) and logged once every hour. The change in height was used to calculate the acid use in liters per day.

The theoretical binding capacity of sulfuric acid (98% concentrated) is 1.5 L to capture 1 kg of ammonia (NH$_3$) in air scrubbers (Melse and Willers, 2017) and was calculated with equation 5.

$$R_{NH3,acid} = \sum_{j=1}^{n}\left(AU_j * 1.5\right) \tag{5}$$

where

$R_{NH3,acid}$ = the cumulative ammonia reduction in kg

AU_j = the daily acid use in L

j = represents each day

n = 7 when calculating weekly values and 28 when calculating 4-weekly cumulative values.

Method 3: Nitrogen Content of the Process Water and Volume of the Discharged Water

The conductivity of the process water was continuously measured with a Jumo CTI-500 conductivity sensor (accuracy +/- 0.5% of measured value) in the central water supply from the clean water basin to the air treatment units; mean values over 5 minute intervals were logged. In a preliminary study, the relation between the conductivity and the nitrogen content of the process water was determined (see Appendix 2) and is given in equation 6.

$$C_N = 0.390\,E^{-5} * EC^3 - 0.670E^{-3} * EC^2 \\ + 0.225 * EC - 1.03 \tag{6}$$

where

C_N = the nitrogen content of the process water (kg L^{-1})

EC = the conductivity (mS cm^{-1}).

When the water level in the central water treatment reached a certain height, the water discharge valve was opened, water was automatically discharged into the silo, and the volume was measured with a flow meter (JUMO flowTRANS US W01, accuracy +/- 2.0% of the measured value). The level of the water in the clean water tank and the collection pit were measured with a water level meter (type PS3208, producer IFM, accuracy ≤ 0.2% of the measured value).

The cumulative amount of ammonia that was captured in the process water was calculated using equation 7:

$$R_{NH3,water} = A * \left(\left(C_{N,n} * V_{PW,n}\right) - \left(C_{N,0} * V_{PW,0}\right)\right) \\ + \sum_{k=1}^{m}\left(C_{N,k} * DW_k\right) \tag{7}$$

where

$R_{NH3,water}$ = the cumulative ammonia removal captured in the process water in kg

A = the conversion factor from nitrogen to ammonia based on the molecular weight (1.214 kg kg^{-1})

$C_{N,n}$ = the nitrogen concentration in the process water in kg L^{-1} at the end of a period (n was 2016 when calculating weekly values and 8064 when calculating 4-week average values)

$C_{N,0}$ = the nitrogen concentration in the process water in kg L^{-1} at the start of a period

$V_{PW,n}$ = the volume of the process water in L at the end of a period

$V_{PW,0}$ = the volume of the process water in L at the start of a period

$C_{N,k}$ = the nitrogen concentration in kg L^{-1} at moment of water discharge

DW_k = the amount of discharged water in L into the silo at moment k

m = the number of discharge moments varying between 0 and 9, depending on the selected period.

Statistical and Sensitivity Analysis

Logged data from 5-minute intervals—including concentrations (air and liquid), ventilation rates, electrical conductivity, and volumes of clean and process water—and hourly values for acid volume were used in Equations 1-7 to calculate weekly and period means (with MS Excel). These results are presented in tables and graphs. Linear models with intercept zero between $R_{NH3,gas}$ (as a dependent or predicted variable), and $R_{NH3,acid}$ and $R_{NH3,water}$ (x-variables) were made in R (R Core Team, 2022) with RStudio version 2023.06.0+421. The weekly data points were assumed to be time-independent.

Additionally, a sensitivity analysis was done on the dataset of period 4 (for method 1, data from units 2 and 4 were used) by de- and increasing each individual measurement ($Cghouse_{NH3}$, $Cexhaust_{NH3}$, φv_{1-4}, AU_j, $C_{N,n}$, $C_{N,0}$, $V_{PW,n}$, $V_{PW,0}$, $C_{N,k}$, DW_k) with the given sensor accuracies, and calculating the effect on the emission reduction ($R_{NH3,gas}$, $R_{NH3,acid}$, $R_{NH3,water}$) and on the remaining ammonia emission. The remaining emission based on method 1 was calculated as follows:

$$AE_{NH3,gas} = \sum_{i=1}^{n}\left(Cexhaust_{NH_3,i}\right) \\ \times \left(\varphi v_{1,i} + \varphi v_{2,i} + \varphi v_{3,i} + \varphi v_{4,i}\right) * \frac{1000}{12} \tag{8}$$

where $AE_{NH3,gas}$ = the ammonia emission based on the ammonia concentration in the exhaust air in kg.

To determine the remaining ammonia emission based on methods 2 and 3, first the ammonia production in the house was calculated (using the ammonia sensors in the house and the ventilation flow, analogous to method 1), and the calculated ammonia reductions were subtracted (eqs. 9 and 10):

$$AE_{NH3,acid} = \left(\sum_{i=1}^{n}\left(Cghouse_{NH_3,i}\right)\right. \\ \left. \times \left(\varphi v_{1,i} + \varphi v_{2,i} + \varphi v_{3,i} + \varphi v_{4,i}\right) * \frac{1000}{12}\right) - R_{NH3,acid} \tag{9}$$

where $AE_{NH3,acid}$ = the ammonia emission based on the ammonia production in the house minus the amount of ammonia binding based on the acid use in kg.

$$AE_{NH3,water} = \left(\sum_{i=1}^{n}\left(Cghouse_{NH_3,i}\right)\right. \\ \left. \times \left(\varphi v_{1,i} + \varphi v_{2,i} + \varphi v_{3,i} + \varphi v_{4,i}\right) * \frac{1000}{12}\right) - R_{NH3,water} \tag{10}$$

where $AE_{NH3,water}$ = the ammonia emission based on the ammonia production in the house minus the amount of ammonia measured in the process and discharge water in kg.

RESULTS AND DISCUSSION

AMMONIA CONCENTRATIONS AND OF AMMONIA REMOVAL EFFICIENCY

Table 3 shows the results of the ammonia concentration measurements before and after passing air treatment unit 2 for the 4 periods. The weekly mean ammonia concentration in the house ranged between 17.39 and 33.93 mg m^{-3}. These concentrations were quite stable and were in the same range as reported by Ellen et al. (2021). The ammonia concentration in the outgoing air was (excluding two weeks, see below) between 0.37 and 5.86 mg m^{-3}.

In week 3 of period 3, there was a very low reduction of ammonia (40%); this was caused by a failure in the acid pump, resulting in no acid being dosed into the process water and the pH of the process water rising from 3.0 up to 7.8 within a few days. Also, in week 3 of period 4, there was a decrease in ammonia removal efficiency (72%); this was caused by an empty acid storage vessel (a too late replacement), resulting in no acid being dosed into the system for three days. However, all results are used in the comparison of the three methods in this study because periods with lower ammonia removal efficiency will occur in reality and do not influence the comparison of the three methods.

The ammonia removal efficiency of the system was approximately 90% during the periods when the acid dosing system was working properly, as expected for acid air scrubbers (Melse, 2009; Hadlocon et al., 2014; Mostafa et al., 2020). This is also in line with an earlier study on a similar ventilation system and air scrubber system, where the reduction of ammonia was somewhat lower, with 77%. This can be explained by the somewhat higher pH value of 5.0 in the process water in that study (Goselink et al., 2023).

Table 4 shows the sulfuric acid use (AU) in the four periods. The total amount of acid used per period was of the same magnitude (between 930 and 1640 L), but the weekly data (excluding the two weeks without acid supply - weeks 3 of period 3 and 4) show relatively more variation in acid use, varying from 170 (period 1 week 2) to 410 (period 2 week 1) liters used per week.

Figure 4 shows the daily acid use for period 1; daily values ranged between 0 and 90 L d^{-1}, which is even a relatively bigger variation than on a weekly basis. On 24 April 2022, a lot of acid was dosed into the system, so much so that on 25 April 2022, no acid dosing was needed.

NITROGEN CONTENT OF THE PROCESS AND DISCHARGED WATER

Table 5 shows the concentration of nitrogen in the process water at the start (2.6 to 38.1 g L^{-1}) and end of each week (4.7 to 40.9 g L^{-1}), as well as the volume of the process water present, varying from 42.3 m^3 (in period 2 week 3) to 47.6 m^3 at the end of period 4. From these data, the calculated amount of ammonia that was absorbed in the process water is also shown in the table, as well as the amount of water that was discharged from the system with corresponding nitrogen and ammonia masses.

In period 1, there was no discharge of water; this was at the start of the flock, and the process water was not yet saturated with nitrogen (bound as ammonium sulfate). The increase in nitrogen mass in the process water during period 1 was 432 kg, corresponding to 524 kg of ammonia.

At the end of warm period 2, the nitrogen concentration in the discharged water was highest at 40.9 g L^{-1}; this was due to the evaporation of process water for indirect evaporative cooling in the air treatment units during this period. In the other cooler periods, the nitrogen concentrations in the discharged water were lower, indicating less water evaporation or even vapor condensation in the air treatment units; consequently, the volume of process water increased, activating the water to be discharged. Table 5 also shows that the water discharge was not activated in all weeks; for example, in period 3, the water discharge only occurred in weeks 3 and 4.

Table 3. Mean 4 weekly (wk1-4), weekly (week 1-4), and overall (period 1-4) ammonia concentrations in the house and exhaust, scrubbing efficiency, and ventilation rate measured on air treatment unit 2 for method 1.

	Period 1					Period 2					
	wk1-4	week 1	week 2	week 3	week 4	wk1-4	week 1	week 2	week 3	week 4	
$C_{ghouse\ NH3}$ (mg m^{-3})	24.61	19.24	23.40	29.11	26.70	20.82	23.57	21.56	17.39	20.75	
$C_{gexhaust\ NH3}$ (mg m^{-3})	0.92	0.37	0.65	1.59	1.07	2.09	4.10	1.65	1.41	1.21	
η_{NH3}(%)	96%	98%	97%	95%	96%	90%	83%	92%	92%	94%	
ϕv (m^3 h^{-1})	41652	37878	41612	42140	44978	80367	71515	82115	83563	81371	
	Period 3					Period 4					Period 1-4
	wk1-4	week 1	week 2	week 3	week 4	wk1-4	week 1	week 2	week 3	week 4	
$C_{ghouse\ NH3}$ (mg m^{-3})	23.12	24.14	22.42	23.13	22.78	29.54	27.01	28.49	28.96	33.93	25.10
$C_{gexhaust\ NH3}$ (mg m^{-3})	5.45	0.97	1.08	13.87	5.86	3.34	0.81	0.71	8.21	3.62	3.45
η_{NH3}(%)	76%	96%	95%	40%	74%	89%	97%	97%	72%	89%	86%
ϕv (m^3 h^{-1})	54699	55507	53221	54151	53979	40441	45695	40730	41686	32945	52077

Table 4. Mean 4- weekly (wk 1-4), weekly (week 1-4), and overall (period 1-4) acid use in the 4 periods.

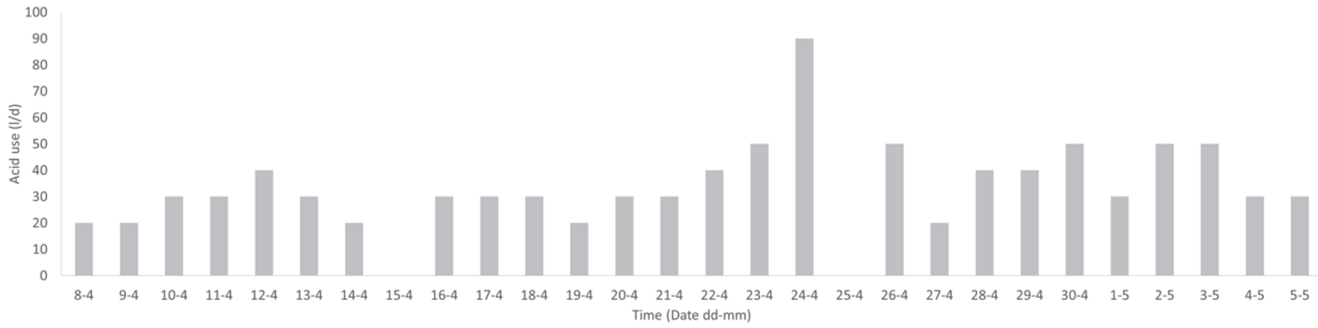|	Period 1					Period 2					
	wk1-4	week 1	week 2	week 3	week 4	wk1-4	week 1	week 2	week 3	week 4	
AU (L)	930	190	170	290	280	1640	410	330	280	620	
	Period 3					Period 4					Period 1-4
	wk1-4	week 1	week 2	week 3	week 4	wk1-4	week 1	week 2	week 3	week 4	
AU (L)	980	320	290	110	260	1081	340	288	184	269	4631

Figure 4. Total daily acid use in period 1.

Table 5. Mean 4- weekly (wk 1-4), weekly (week 1-4), and overall (period 1-4) nitrogen concentrations, process water volumes, increase in nitrogen content of the process water, and the volume and nitrogen content of the discharged water in the 4 periods (- indicates there is no value).

	Period 1					Period 2				
	wk1-4	week 1	week 2	week 3	week 4	wk1-4	week 1	week 2	week 3	week 4
$C_{N,0}$ (g L^{-1})	2.6	2.6	4.7	7.3	10.2	29.7	29.7	32.5	34.9	38.1
$C_{N,n}$ (g L^{-1})	12.5	4.7	7.3	10.2	12.5	40.9	32.5	34.9	38.1	40.9
$V_{PW,0}$ (m^3)	43.0	43.0	43.0	42.3	43.3	43.5	43.5	42.9	43.5	43.3
$V_{PW,n}$ (m^3)	43.2	42.9	42.3	43.3	43.2	43.4	42.9	43.5	43.3	43.4
Increase N mass in process water (kg)	432	91	109	134	98	479	101	122	132	124
Increase ammonia mass in process water (kg)	524	110	132	163	119	581	122	148	161	151
DW (L)	0	0	0	0	0	9000	2700	0	0	6300
$C_{N,k}$ (g L^{-1})	-	-	-	-	-	41	31	-	-	45
Mass of ammonia in discharged water (kg)	0	0	0	0	0	449	102	0	0	347

	Period 3					Period 4					Period 1-4
	wk1-4	week 1	week 2	week 3	week 4	wk1-4	week 1	week 2	week 3	week 4	
$C_{N,0}$ (g L^{-1})	12.3	12.3	15.6	17.5	18.5	31.2	31.2	32.5	35.6	35.0	18.9
$C_{N,n}$ (g L^{-1})	19.2	15.6	17.5	18.5	19.2	33.6	32.5	35.6	35.0	33.6	24.5
$V_{PW,0}$ (m^3)	43.2	43.2	43.5	44.4	43.1	44.2	44.2	44.0	43.8	45.2	43.5
$V_{PW,n}$ (m^3)	42.7	43.5	44.4	43.1	42.7	47.6	44.0	43.8	45.2	47.6	44.1
Increase N mass in process water (kg)	287	148	96	20	23	224	53	130	23	17	1421
Increase ammonia mass in process water (kg)	349	180	117	24	28	272	64	158	28	21	1726
DW (L)	6300	0	0	1800	4500	8100	3600	0	0	4500	23400
$C_{N,k}$ (g L^{-1})	19	-	-	19	19	34	34	-	-	35	40
Mass of ammonia in discharged water (kg)	142	0	0	41	101	339	148	0	0	191	930

COMPARISON OF THE THREE METHODS

For the comparison of the three methods, we assumed that method 1, which initially measured only on unit 2, is the gold standard as it is based on the reference method for assessing emissions from housing systems (Ogink et al., 2017).

Table 6 shows that the four-week average of the $R_{NH3,acid}$ varied between 97% and 120% of $R_{NH3,gas}$ and $R_{NH3,water}$ varied between 77% and 98% of $R_{NH3,gas}$. Over all 4 periods, $R_{NH3,acid}$ and $R_{NH3,water}$ amounted to 3087 and 2656 kg, respectively, being 104.7 and 90.1% of $R_{NH3,gas}$ (2948 kg). So, based on all data, method 2 gives a small (about 5%) overestimation and method 3 a small (about 10%) underestimation as compared to the initial reference method 1, with roughly a variation of +/- 20% per four week period.

Figure 5 shows the graphical relation between the weekly cumulative ammonia removal for the 3 methods (data from table 6). The shown regression lines were forced to go through the (0, 0) point in the graph, as we deemed this logic based on the physical and chemical background of the three methods.

Figure 5 shows that both methods 2 and 3 have predictive values for the ammonia emission reduction as measured by method 1. Details of the regression lines are presented in table 7.

$R_{NH3,acid}$ was generally higher than $R_{NH3,gas}$, as indicated by the regression coefficient for the weekly estimated ammonia removal based on the acid use (method 2) that was 0.900 and significantly different from y=x. Figure 5 clearly shows that the outlier at 400 kg greatly influenced this result

Table 6. Mean 4-weekly (wk1-4), weekly (week 1-4), and overall (period 1-4) cumulative ammonia removal and relative ammonia removal compared to method 1 for methods 1, 2, and 3 in the 4 periods.

	Period 1					Period 2				
	wk1-4	week 1	week 2	week 3	week 4	wk1-4	week 1	week 2	week 3	week 4
$R_{NH3,gas}$ (kg)	642 (100%)	114	150	190	188	962 (100%)	227	264	214	257
$R_{NH3,acid}$ (kg)	620 (97%)	127	113	193	187	1093 (120%)	273	220	187	413
$R_{NH3,water}$ (kg)	524 (82%)	110	132	163	119	1030 (98%)	224	148	161	498

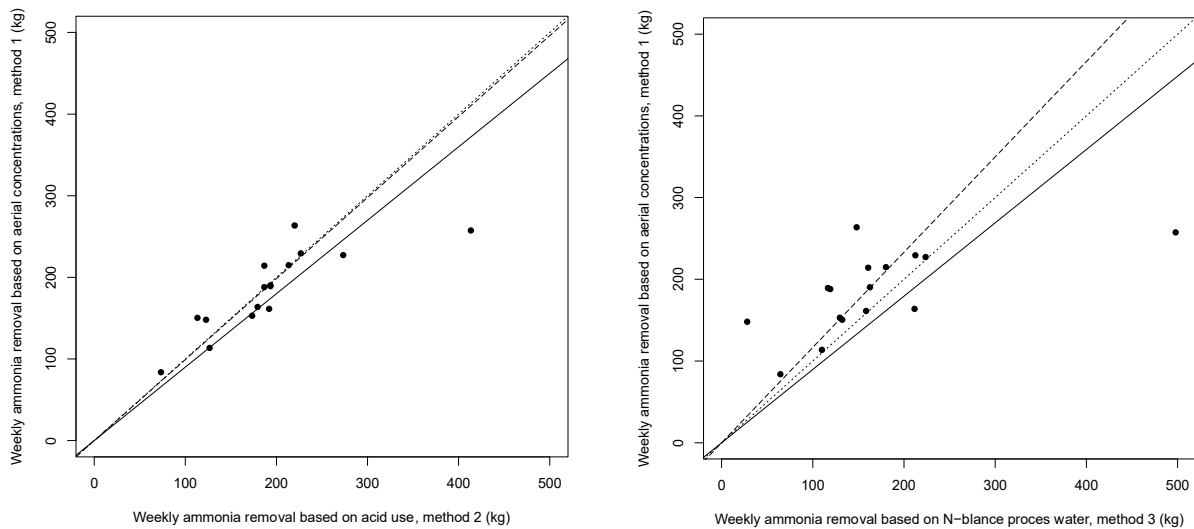|	Period 3					Period 4					Period 1-4
	wk1-4	week 1	week 2	week 3	week 4	wk1-4	week 1	week 2	week 3	week 4	
$R_{NH3,gas}$ (kg)	641 (100%)	215	189	84	153	702 (100%)	229	161	148	164	2948 (100%)
$R_{NH3,acid}$ (kg)	653 (102%)	213	193	73	173	721 (103%)	227	192	123	179	3087 (104.7%)
$R_{NH3,water}$ (kg)	491 (77%)	180	117	64	130	610 (87%)	212	158	28	212	2656 (90.1%)

Figure 5. Cumulative weekly ammonia removal according to method 2 (acid use; left) and method 3 (N-balance of the process water; right) against method 1 (aerial ammonia concentrations; y-axis). Measurement points (dots) and regression lines are shown for all four periods and four weeks (solid line), excluding the outliers at 400 kg and 500 kg (dashed line). The dotted line represents y=x.

Table 7. Numerical results of the linear regression of $R_{NH3,gas}$ with data of all weeks (16 data points) and without data of period 2, week 4: regression coefficient with standard error and the t-value for its difference from 1.000, as well as the R^2 adjusted and the residual standard error.[a]

Variable (method)	No. Data Points	Coefficient (s.e.)	t-value	R^2 Adjusted	Res. St. Error (kg)
$R_{NH3,acid}$ (2)	16	0.900 (0.051)	1.96 *	0.952	41.8
$R_{NH3,acid}$ (2)	15	0.992 (0.035)	0.23	0.981	24.9
$R_{NH3,water}$ (3)	16	0.897 (0.105)	0.98	0.820	80.8
$R_{NH3,water}$ (3)	15	1.166 (0.087)	1.91 *	0.923	51.3

[a] Significancy: * p<0.05.

as the estimation error of this data point weighed quadratically in the regression; leaving out this data point changed the regression coefficient to 0.992 (with higher R^2 and lower residual error) and was not significantly different from y=x. Leaving out this data point is not correct from the mass balance perspective. Based on the mass balance of four periods, a regression coefficient of 0.955 is expected (the inverse of 104.7% as compared to $R_{NH3,gas}$). Despite the overestimation, $R_{NH3,acid}$ (method 2), had a good linear correlation with $R_{NH3,gas}$ (method 1 – measuring only on unit 2) with a regression coefficient close to 1.

$R_{NH3,water}$ was generally lower than $R_{NH3,gas}$, as indicated by the regression coefficient for the weekly estimated ammonia removal based on the nitrogen balance of the process water (method 3) that was 0.897, which was not significantly different from y=x. Figure 5 clearly shows that the outlier at 500 kg greatly influenced this result as the estimation error of this data point weighed quadratically in the regression; leaving out this data point changed the regression coefficient to 1.166 (with higher R^2 and lower residual error) and was significantly larger than y=x. Leaving out of this data point is not correct from the mass balance perspective. Based on the mass balance of 4 periods, a regression coefficient of 1.110 is expected (the inverse of 90.1% as compared to $R_{NH3,gas}$), which means that method 3 ($R_{NH3,water}$) underestimated the ammonia removal as compared to method 1 when measuring aerial ammonia concentrations only at unit 2. Despite the underestimation $R_{NH3,water}$ had a good linear correlation with $R_{NH3,gas}$, with a regression coefficient close to 1.

COMPARISON OF WEEKLY VERSUS 4-WEEKLY PERIODS

The data on the weekly variation in acid use (shown in fig. 4) and the nitrogen balance of the process and discharge water (table 5, fig. 5) shows considerable variation due to irregular time events like acid dosage and water discharge. For example: In week 3 of period 4, there is hardly any ammonia removal based on method 3 (28 kg), while for methods 1 and 2, this was 148 and 123 kg in this week, respectively. However, in week 4 of period 4 this seems to be partly compensated, where method 3 removes 212 kg ammonia, and methods 1 and 2 show lower amounts with 164 and 179 kg. Something similar happened in weeks 1 to 3 of period 2: the ammonia removal based on method 3 was relatively low compared to methods 1 and 2. However, in week 4 of period 4, this seems to be compensated, where method 3 removed 498 kg ammonia, and methods 1 and 2 lower amounts with 257 and 413 kg. Table 6 shows that $R_{NH3,acid}$ and $R_{NH3,water}$ over 4-week instead of weekly time intervals clearly reduced the variation in the data for methods 2 and 3.

A possible explanation is that the process water was not always homogeneously mixed and that the location where the pH and EC were measured was not representative of the total volume of the process water. This can, for example, be caused by the sedimentation and floatation of certain components in the central water treatment. This could give an overestimation or underestimation of the pH and/or nitrogen content of the process water. Methods 2 and 3 are both prone to this. The effect of this nonhomogeneous mixing of the process water had a relatively bigger effect on weekly values

than on 4-weekly values because the impact of discharged nitrogen into the silo became more important for the total nitrogen balance.

COMPARISON OF MEASURING AMMONIA CONCENTRATION ON 1 VERSUS 2 UNITS

Table 8 shows the results of method 1 in period 4 for units 2 and 4. In period 4, we noticed a substantial spatial variation in ammonia concentration in the house; the air in unit 2 had a higher house concentration of ammonia (29.54 g m^{-3}) than in unit 4 (22.94 g m^{-3}). This spatial variation in source strength combined with the non-perfect mixing of air in livestock buildings, and the effect on emission measurements has been reported before (Van Buggenhout et al., 2009). By combining (averaging) the concentration measurements of both units 2 and 4, the ammonia reduction by method 1 ($R_{NH3,gas}$) for whole period 4 changed from 702 kg to 617 kg (12% lower), which is a considerable change. This amount was 104 kg lower and 7 kg higher than the ammonia reduction by methods 2 ($R_{NH3,acid}$) and 3 ($R_{NH3,water}$), respectively.

We expect that the spatial variation in ammonia concentrations also occurred in periods 1 to 3 and contributed to the differences in estimated ammonia reduction between the methods. This result shows (again) the need to take good care of spatial variation in ammonia concentrations in livestock houses when calculating absolute emissions and emission reductions.

Table 8 shows that method 2 ($R_{NH3,acid}$) overestimated the emission reduction even more than in the previous comparison (increasing from 104.7% in table 6 to 116.8%). This could be explained by the possibility that the acid also reacts with other chemical components in the process water—such as alkanoic odor components—causing this method to overestimate the actual ammonia removal. Another possibility is that small drops of water are emitted from the system, which causes losses of acid and ammonia dissolved in these small droplets (Brouwers et al., 2012); these ammonia losses are not detected by the gaseous ammonia concentration measurements.

Table 8 also shows that the underestimation of method 3 ($R_{NH3,water}$) reduced to 98.9% (what was 90.1% in table 6). For this period, methods 1 and 3 were in good agreement, and the small difference indicates that the emission of small droplets, causing non observed losses of nitrogen from the system, were small.

Although methods 1 and 3 were in good agreement, the assumed relationship between the ammonium sulfate concentration and the conductivity of the process water as reported in Appendix 2 also needs attention. The relationship between the conductivity and the nitrogen content of the process water that is used in this research was determined with clean water. But the actual process water also contained other chemical components and dust particles, which could influence the conductivity and therefore affect the nitrogen balance. The relation between the EC and the ammonium-sulfate content should in the future also be determined in 'dirty' process water from the units.

SENSITIVITY ANALYSIS ON AMMONIA REDUCTION AND REMAINING EMISSION

The effect of possible measurement errors (given by the accuracy of the sensors used) on the calculated emission reduction and remaining ammonia emission is presented in table 9.

The effect of measurement errors on the ammonia removal of method 1 (R_{NH3gas}) was relatively small (max. 2.0%); the biggest influence was observed by a measurement error on the ventilation rate, giving a possible variation in ammonia removal between 604 and 629 kg. However, on the remaining ammonia emission according to method 1 ($AE_{NH3,gas}$), a possible measurement error on the ammonia concentration in the house had obviously no effect on the remaining emission.

As discussed, the ammonia removal by binding ammonia with acid ($R_{NH3,acid}$) was higher than according to method 1, even higher than the calculated production of ammonia in the house, resulting in a negative remaining ammonia emission. For this reason, the measurement errors had a very large relative effect (up to 43 and 49%) on the calculated remaining

Table 8. Mean 4-weekly (wk1-4), weekly (week 1-4) ammonia concentrations measured on air treatment units 2 and 4 in period 4, and the calculated effect on the cumulative ammonia removal of the system according to method 1, in comparison to the ammonia removal of methods 2 and 3.

		wk1-4	week 1	week 2	week 3	week 4
Unit 2	$C_{ghouseNH3}$ (mg m^{-3})	29.54	27.01	28.49	28.96	33.93
	$C_{gexhaustNH3}$ (mg m^{-3})	3.34	0.81	0.71	8.21	3.62
	η_{NH3}(%)	89%	97%	97%	72%	89%
	ϕ_v (m^3 h^{-1})	40441	45695	40730	41686	32945
	$R_{NH3,gas}$(kg)	**702**	**229**	**161**	**148**	**164**
Unit 4	$C_{gexhaustNH3}$ (mg m^{-3})	22.94	20.34	20.96	22.69	27.84
	$C_{ghouseNH3}$ (mg m^{-3})	3.23	0.93	0.87	7.96	3.17
	η_{NH3}(%)	86%	95%	96%	65%	89%
	ϕv (m^3 h^{-1})	40441	45695	40730	41686	32945
	$R_{NH3,gas}$ (kg)	**531**	**170**	**118**	**107**	**136**
Mean of unit 2 and 4	$C_{gexhaustNH3}$ (mg m^{-3})	26.24	23.67	24.72	25.83	30.88
	$C_{ghouseNH3}$ (mg m^{-3})	3.29	0.87	0.79	8.08	3.39
	η_{NH3}(%)	87%	96%	97%	69%	89%
	ϕ_v (m^3 h^{-1})	40441	45695	40730	41686	32945
	$R_{NH3,gas}$ (kg)	**617**	**200**	**140**	**128**	**150**
	$R_{NH3,acid}$ (kg)	721	227	192	123	179
	Relative to $R_{NH3,gas}$ (%)	116.8	113.5	137.1	96.1	119.3
	$R_{NH3,water}$ (kg)	610	212	158	28	212
	Relative to $R_{NH3,gas}$ (%)	98.9	106	112.9	21.9	141.3

Table 9. Emission reductions (R_{NH3}) and remaining emission (AE_{NH3}) in kg and the relative change of these values for the three methods, variable values (values based on data from period 4 and for method 1 data of both units 2 and 4) and the sensor accuracy.

Method	Variable	Mean Value	Accuracy	Range (kg)	Relative Change (%)	Range (kg)	Relative Change (%)
1				$R_{NH3,gas}$		$AE_{NH3,gas}$	
	C_{ghouse} (mg m^{-3})	26.2	+/-0.12 mg m^{-3}	614 / 620	+/- 0.6%	89 / 89	+/- 0.0%
	$C_{gexhaust}$ (mg m^{-3})	3.3	+/-0.12 mg m^{-3}	620 / 614	+/- 0.5%	86 / 93	+/- 3.6%
	ϕ_v (m^3 h^{-1})	40441	+/- 2.0%	604 / 629	+/- 2.0%	88 / 91	+/- 2.0%
2				$R_{NH3,acid}$		$AE_{NH3,acid}$	
	C_{ghouse} (mg m^{-3})	26.2	+/-0.12 mg m^{-3}	721 / 721	+/- 0.0%	-12 / -19	+/- 22%
	ϕ_v (m^3 h^{-1})	40441	+/- 2.0%	721 / 721	+/- 0.0%	-23 / 5	+/- 49%
	AU (L)	1081	+/-10 L	714 / 727	+/- 0.9%	-9 / -22	+/- 43%
3				$R_{NH3,water}$		$AE_{NH3,water}$	
	C_{ghouse} mg m^{-3})	26.2	+/-0.12 mg m^{-3}	610 / 610	+/- 0.0%	92 / 98	+/-3.4%
	ϕ_v (m^3 h^{-1})	40441	+/- 2.0%	610 / 610	+/- 0.0%	81 / 109	+/- 15%
	$C_{N,0}$ (g L^{-1})	31.2	+/- 0.5%	619 / 602	+/- 1.4%	87 / 103	+/- 8.8%
	$C_{N,n}$ (g L^{-1})	33.6	+/- 0.5%	601 / 620	+/- 1.6%	105 / 85	+/- 10.2%
	$V_{PW,0}$ (m^3)	44.2	+/- 0.2%	614 / 607	+/- 0.5%	92 / 98	+/- 3.5%
	$V_{PW,n}$ (m^3)	47.6	+/- 0.2%	606 / 614	+/- 0.6%	99 / 91	+/- 4.1%
	DW (L)	8100	+/- 2.0%	604 / 617	+/- 1.1%	102 / 88	+/- 7.1%

ammonia emission. When a revised value for acid use per kg of ammonia binding (eq. 5) can be substantiated by further research, method 2 can be valuable and used without resulting in physically impossible negative emission values.

Also, the effect of measurement errors on the ammonia removal of method 3 ($R_{NH3,water}$) was relatively small (max. 1.6%). The relative effect on the calculated remaining emissions was much larger, up to 15%, due to an error in the ventilation rate sensor (φv_{1-4}). This can be explained by the high level of ammonia removal by the air scrubber system and the relatively low remaining emissions. Although there is variation in the calculated remaining emissions for method 3 ($AE_{NH3,water}$), ranging from 81 to 109 kg, the values are all of the same magnitude and most likely provide a fair indication of the actual remaining ammonia emissions.

Based on these findings, an emission monitoring system for livestock houses with air scrubbers, based on method 2 (with revised acid use per kg of ammonia binding) or method 3 is proposed. Such a monitoring system is deemed to be technically and economically feasible, can be relatively easily controlled and enforced, and also takes into account periods where the scrubber technology is not working as efficiently as expected. The resulting yearly net emission of ammonia can be used to validate the environmental permit of farmers and prevent higher emissions to the environment than allowed.

This work as a whole was carried out in close collaboration between the Vencomatic Group and Wageningen University. This public-private cooperation fits in with the trend and societal and legal need that companies take responsibility for the proper functioning of their systems and, consequently, the long-term realized emission reduction.

CONCLUSION

The ammonia emission reduction by the air scrubber system, under commercial livestock circumstances, is compared on a 1 weekly and 4 weekly basis with three methods: aerial ammonia concentration measurements (method 1), acid use (method 2), and process water measurements

(method 3). The overall weekly data showed a good linear relationship between the methods, with regression coefficients close to 1 and a variation of +/- 20% based on a four week period. The 4-weekly data showed a clearly reduced variation compared to the weekly data due to leveling out the impact of the scrubbing process-related events like acid dosage and water discharge. The result of method 1 was significantly (12%) influenced by spatial concentration differences of ammonia in the house. Taking this into account, method 2 overestimated the emission reduction of method 1 with 16.8%, and method 3 was in good agreement with method 1. For method 2, a revised value for acid use per kg of ammonia binding needs to be substantiated in further research. Possible measurement errors influenced the calculated ammonia removal by +/- 2%, but the relative effect on remaining emissions was bigger, up to +/- 15% for method 3.

This study shows that monitoring acid use and/or the nitrogen balance can validate the ammonia reduction of air scrubber technologies under regular commercial circumstances. Also, these methods can give insight into the remaining emissions and thereby support local authorities in granting environmental permits and enforcing them.

DECLARATION OF INTEREST

The first author is an employee of the Vencomatic Group; he declares that this research was conducted according to the Netherlands Code of Conduct for Research Integrity. The research focuses on methods to validate ammonia removal by air scrubber technology, which is of scientific and societal importance, whereas the actual measurements for this study were carried out with equipment integrated in the air treatment units of Vencomatic Group.

Independent author 2 was involved in the research setup, data acquisition, analysis, and writing of this paper and takes

responsibility for the scientific quality of the paper; he declares to have no financial or other interests.

REFERENCE LIST

Aarnink, A. J., Landman, W. J., Melse, R. W., Zhao, Y., Ploegaert, J. P., & Huynh, T. T. (2011). Scrubber capabilities to remove airborne microorganisms and other aerial pollutants from the exhaust air of animal houses. *Trans. ASABE, 54*(5), 1921-1930. https://doi.org/10.13031/2013.39833

Bokkers, E. A., van Zanten, H. H., & van den Brand, H. (2010). Field study on effects of a heat exchanger on broiler performance, energy use, and calculated carbon dioxide emission at commercial broiler farms, and the experiences of farmers using a heat exchanger. *Poultr. Sci., 89*(12), 2743-2750. https://doi.org/10.3382/ps.2010-00902

Brouwers, J. J., van Kemenade, H. P., & Kroes, J. P. (2012). Rotational particle separator: An efficient method to separate micron-sized droplets and particles from fluids. *Filtration, 12*(1), 49-60.

Centraal Bureau voor de Statistiek (CBS). (2023). Stikstofemissies naar lucht. Retrieved from https://www.cbs.nl/nl-nl/dossier/dossier-stikstof/stikstofemissies-naar-lucht

de Vries, W., Schulte-Uebbing, L., Kros, H., Voogd, J. C., & Louwagie, G. (2021). Spatially explicit boundaries for agricultural nitrogen inputs in the European Union to meet air and water quality targets. *Sci. Total Environ., 786*, 147283. https://doi.org/10.1016/j.scitotenv.2021.147283

Deeken, H. F., Lengling, A., Krommweh, M. S., & Büscher, W. (2023). Improvement of piglet rearing's energy efficiency and sustainability using air-to-air heat exchangers — a two-year case study. *Energies, 16*(4), 1799. https://doi.org/10.3390/en16041799

Dekock, J., Vranken, E., Gallmann, E., Hartung, E., & Berckmans, D. (2009). Optimisation and validation of the intermittent measurement method to determine ammonia emissions from livestock buildings. *Biosyst. Eng., 104*(3), 396-403. https://doi.org/10.1016/j.biosystemseng.2009.07.003

DLG. (2016a). Chemischer Abluftwäscher "Pura aer II" für die Legehennenhaltung und Junghennen Aufzucht - DLG-Prüfbericht 6406. Retrieved from https://pruefberichte.dlg.org/filestorage/6406.pdf

DLG. (2016b). 1-stufiger Chemowäscher mit Tropfenabscheider Inno+ Pollo-L - für die Legehennenhaltung. DLG-Prüfbericht 6344. Retrieved from https://pruefberichte.dlg.org/filestorage/6344.pdf

DLG. (2022). DLG-Prüfrahmen "Abluftreinigung in der Tierhaltung". Retrieved from https://www.dlg-testservice.com/fileadmin/Redaktion/downloads/landwirtschaft/flyer/DLG-Pruefrahmen-Abluftreinigung.pdf

Ellen, H. H., Goselink, Y., Nijeboer, G., Winkel, A., & van Emous, R. (2021). Ammoniakemissie van een mestbeluchtingssysteem voor vleeskuikenouderdieren met beluchtingsbuizen op de vloer van de mestput (Ammonia emission of a manure aeration system for broiler breeders with aeration pipes at the floor of the manure pit). WLR report 1334. Wageningen Livestock Research. https://doi.org/10.18174/553812

Goselink, Y. S., & Ramirez, B. C. (2019). Characterization of an air-to-air heat exchanger for manure belt drying ventilation in an aviary laying hen house. *J. Appl. Poult. Res., 28*(4), 1359-1369. https://doi.org/10.3382/japr/pfz075

Goselink, Y., Ellen, H., & Nijeboer, G. (2023). Emissiemetingen ECO Zero stal: Onderzoek naar de emissies van ammoniak en fijnstof uit de ECO Zero stal en de reductie van deze emissies door de ECO Units (Emission measurements ECO Zero barn: Emission of ammonia and particulate matter from the ECO Zero barn and the reduction of emissions by the ECO units). WLR Report 1361. Wageningen Livestock Research. Retrieved from https://doi.org/10.18174/565366

Hadlocon, L. J., Zhao, L., Manuzon, R. B., & Elbatawi, I. E. (2014). An acid spray scrubber for recovery of ammonia emissions from a deep-pit swine facility. *Trans. ASABE, 57*(3), 949-960. https://doi.org/10.13031/trans.57.10576

Hahne, J. (2022). Langzeitmessungen zu Ammoniakemissionen aus der Lege-Elterntierhaltung und Maßnahmen zu deren Minderung. *LANDTECHNIK, 77*(3), 66-78. https://doi.org/10.15150/lt.2022.3279

Infomil. (2018). Activiteitenbesluit Milieubeheer Artikel 3.125. Retrieved from https://wetten.overheid.nl/BWBR0022762/2018-01-01/#Hoofdstuk3_Afdeling3.5_Paragraaf3.5.8_Artikel3.125

Kabl, M. (2015). Adiabatic cooling. Technical university of Liberec. https://doi.org/10.13140/RG.2.2.17728.94729

Krommweh, M. S., & Büscher, W. (2021). Heating performance of a laboratory pilot-plant combining heat exchanger and air scrubber for animal houses. *Sci. Rep., 11*(1), 6872. https://doi.org/10.1038/s41598-021-86159-5

Lide, D. R. (2007). *CRC handbook of chemistry and physics* (87th ed.). Boca Raton, FL: CRC. https://doi.org/10.1021/ja069813z

Melse, R. W. (2009). Air treatment techniques for abatement of emissions from intensive livestock production. PhD diss. Wageningen: Wageningen University and Research. https://doi.org/10.2174/1874331500903010006

Melse, R. W., & Willers, H. (2017). Toepassing van luchtbehandelingstechnieken binnen de intensieve veehouderij (Application of air treatment systems in the livestock industry). WLR report 29. Wageningen: Wageningen Livestock Research.

Melse, R. W., Ogink, N. W., & Rulkens, W. H. (2009). Overview of European and Netherlands' regulations on airborne emissions from intensive livestock production with a focus on the application of air scrubbers. *Biosyst. Eng., 104*(3), 289-298. https://doi.org/10.1016/j.biosystemseng.2009.07.009

Melse, R. W., Ploegaert, J. P., & Ogink, N. W. (2016). Laboratory test of Draeger Polytron 8000 with FL-6813260 sensor for NH3 measurement. WLR Report 972. Wageningen: Wageningen Livestock Research. https://doi.org/10.18174/388326

Mendes, L. B., Xin, H., & Li, H. (2012). Ammonia emissions of pullets and laying hens as affected by stocking density and manure accumulation time. *Trans. ASABE, 55*(3), 1067-1075. https://doi.org/10.13031/2013.41511

Mostafa, E., Selders, A., Gates, R. S., & Buescher, W. (2020). Pig barns ammonia and greenhouse gas emission mitigation by slurry aeration and acid scrubber. *Environ. Sci. Pollut. Res., 27*(9), 9444-9453. https://doi.org/10.1007/s11356-020-07613-x

Ogink, N. W., Mosquera, J., & Hol, A. (2017). Protocol voor meting van ammoniakemissie uit huisvestingssystemen in de veehouderij 2013a (Measurement protocol for ammonia emission from housing systems in livestock production 2013a). WLR report 1032. Wageningen: Wageningen Livestock Research. https://doi.org/10.18174/418425

R. Core Team. (2022). R: A language and environment for statistical computing. Vienna, Austria: R Foundation for Statistical Computing.

Rahman, S., Xin, H., Roberts, S. A., Arthur, J. A., Burns, R. T., Li, H.,... Bregendahl, K. (2012). Effects of laying-hen strain on manure properties and ammonia emission. *Trans. ASABE, 55*(3), 1059-1065. https://doi.org/10.13031/2013.41510

Ross. (2016). Performance objective Parent Stock. Retrieved from www.aviagen.com

Santonja G., G., Georgitzikis, K., Scalet, B. M., Montobbio, P., Roudier, S., & Delgado Sancho, L. (2017). Best available techniques (BAT) reference document for the intensive rearing

of poultry or pigs. EUR 28674 EN.
https://doi.org/10.2760/020485

Shepherd, T. A., Xin, H., Stinn, J. P., Hayes, M. D., Zhao, Y., & Li, H. (2017). Ammonia and carbon dioxide emissions of three laying-hen housing systems as affected by manure accumulation time. *Trans. ASABE, 60*(1), 229-236. https://doi.org/10.13031/trans.11860

Sutton, M., Dragosits, U., Geels, C., Gyldenkærne, S., Misselbrook T., H., & Bussink, W. (2015). Review on the scientific underpinning of calculation of ammonia emission and deposition in the Netherlands. Wageningen: Wageningen University & Research. Retrieved from https://edepot.wur.nl/357694

Van Bruggen, C., & Geertjes, K. (2019). *Stikstofverlies uit opgeslagen mest: Stikstofverlies berekend uit het verschil in verhouding tussen stikstof en fosfaat bij excretie en bij mestafvoer (Nitrogen losses based on differences in the nitrogen to phosphate ratio at moment of excretion and transport).* Den Haag, Heerlen, Bonaire: Centraal Bureau voor de Statistiek.

Van Buggenhout, S., Van Brecht, A., Eren Özcan, S., Vranken, E., Van Malcot, W., & Berckmans, D. (2009). Influence of sampling positions on accuracy of tracer gas measurements in ventilated spaces. *Biosyst. Eng., 104*(2), 216-223. https://doi.org/10.1016/j.biosystemseng.2009.04.018

Van der Heyden, C., Demeyer, P., & Volcke, E. I. (2015). Mitigating emissions from pig and poultry housing facilities through air scrubbers and biofilters: State-of-the-art and perspectives. *Biosyst. Eng., 134*, 74-93. https://doi.org/10.1016/j.biosystemseng.2015.04.002

Van Harn, J., Aarnink, A. J., Mosquera, J., van Riel, J. W., & Ogink, N. W. (2012). Effect of bedding material on dust and ammonia emission from broiler houses. *Trans. ASABE, 55*(1), 219-226. https://doi.org/10.13031/2013.41249

APPENDIX 1
CALIBRATION OF AMMONIA SENSORS AND MEASURING FANS

The ammonia sensors were tested after the test period, and the results are shown in table A1. The results show that the maximum deviation was -3.8% for the sensor used at the exhaust of unit 4, meaning that the real emission would be somewhat higher. The other sensors overestimated the concentration with circa 1%. These calibration data were not used for correction of the data presented in this paper.

The measuring fan, as built into the tubes used in this study, was calibrated in a wind tunnel test. The results are shown in figure A1.

APPENDIX 2
RESULTS OF A PRELIMINARY STUDY ON THE RELATION BETWEEN CONDUCTIVITY AND NITROGEN CONTENT OF THE PROCESS WATER

The maximum solubility of ammonium sulfate in water is 744 g kg^{-1} at a temperature of 20°C (Lide, 2006), but no conductivity was known so far for this concentration. To verify this solubility and to determine the conductivity, ammonium sulfate was dissolved in steps of 1 g in 100 mL demi water at a temperature of 20°C until the ammonium sulfate no longer dissolved. During this experiment, the conductivity was measured every step with a Hanna HI98192 conductivity sensor.

Table A1. Calibration result of the four ammonia sensors after the test round when 100 ppm test gas and outside air (close to 0 ppm) was presented to the sensors.

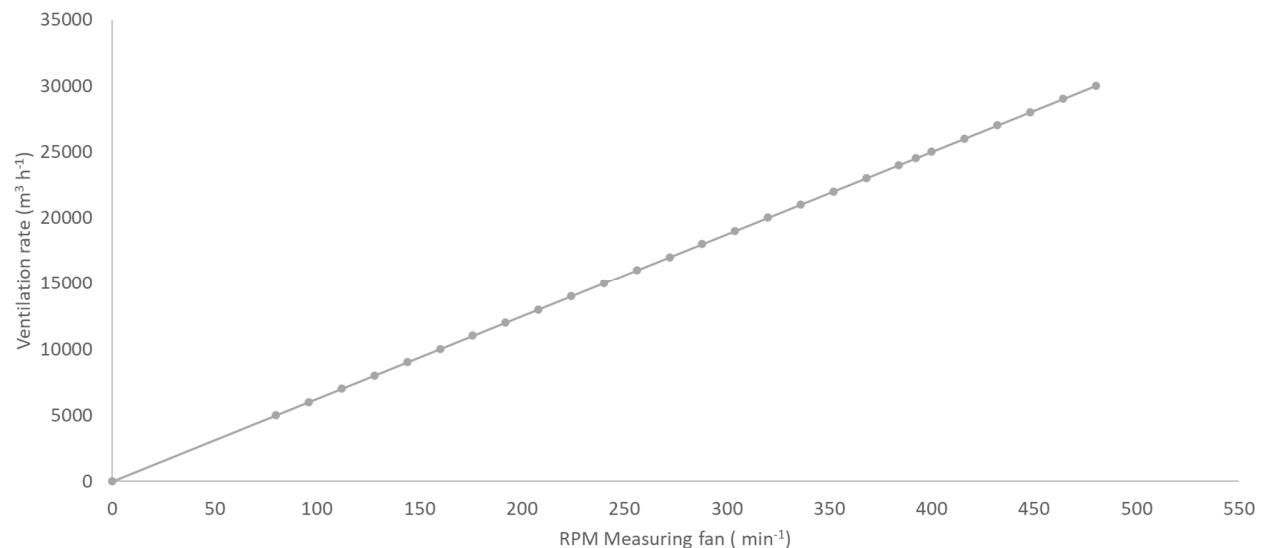| Sensor	Variable	Test Gas (100 ppm)		Outside Air (close to 0 ppm)	
		Signal (V)	Measured Ammonia Concentration (ppm)	Signal (V)	Measured Ammonia Concentration (ppm)
Unit 2 – house	$Cghouse_{NH_3}$	10.10	101.0	< 0.05	< 0.5
Unit 2 – exhaust	$Cexhaust_{NH_3}$	10.09	100.9	< 0.05	< 0.5
Unit 4 – house	$Cghouse_{NH_3}$	10.10	101.0	< 0.05	< 0.5
Unit 4 - exhaust	$Cexhaust_{NH_3}$	9.61	96.1	< 0.05	< 0.5

Figure A1. Relationship between the rotation frequency (RPM) of measuring fan in a tube of 1135 mm and ventilation flow rate as determined in wind tunnel.

The maximum solvability of ammonium sulfate was determined in a laboratory test. The experiment showed that 765 g of ammonium sulfate can be dissolved in 1 L of demi water, with a measured conductivity of 299 mS cm^{-1}. After this point ammonium sulfate did not dissolve anymore.

With the relation shown in figure A3, the concentration of ammonium sulfate was converted into the concentration of nitrogen. The conversion factor from ammonium sulfate (NH4)2SO4—containing 2 nitrogen atoms per molecule—to nitrogen N was done based on their molecular weights, which were 132 and 14, respectively.

$$Mass\ of\ N = \frac{2*14}{132}*Mass\ of\left(NH_4\right)_2 SO_4 \qquad (A1)$$
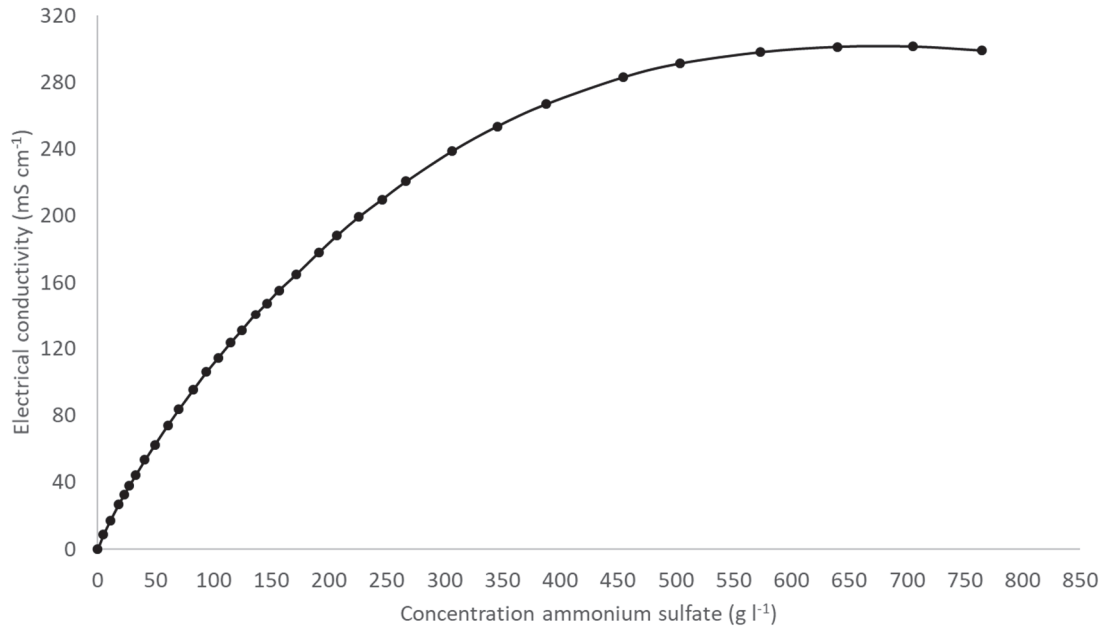

Figure A2. Relation between the concentration of ammonium sulfate and the conductivity of the process water; regression line through the dots representing measurements after stepwise increase of the concentration.
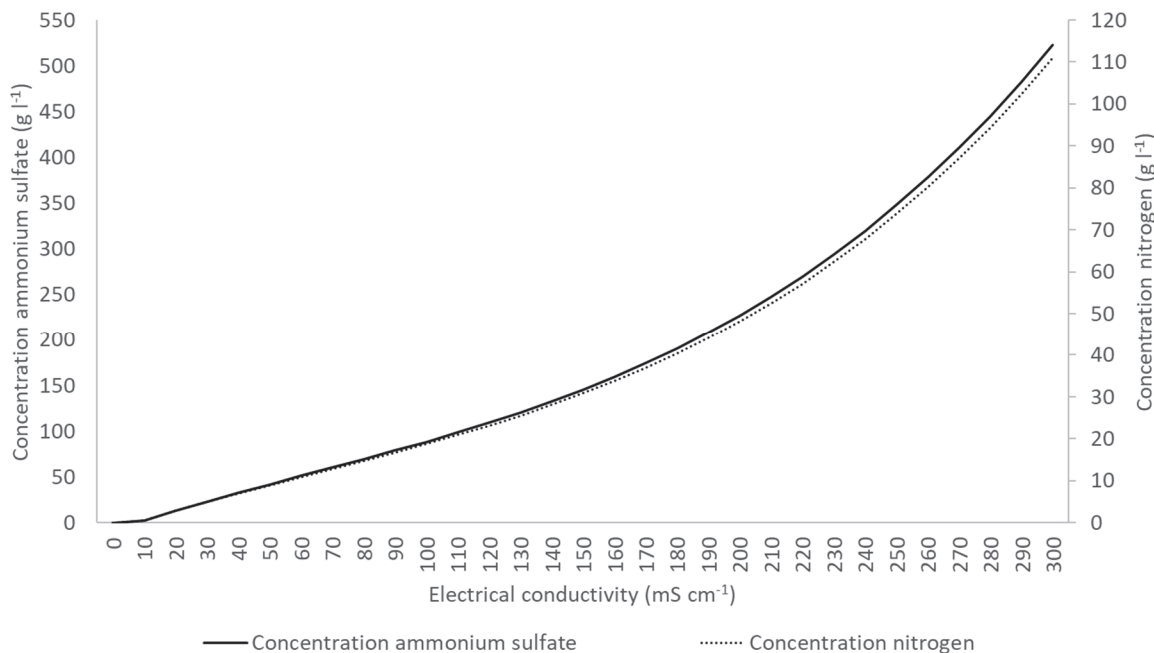

Figure A3. Regression lines to predict the ammonium sulfate (black solid line; left axis) and nitrogen concentration (dotted line; right axis) in process water based on the conductivity.