Evaluation of the NH_3 Removal Efficiency of an Acid Packed Bed Scrubber Using Two Methods: A Case Study in a Pig Facility

F. Estellés, R. W. Melse, N. W. M. Ogink, S. Calvet

Abstract. *The use of air cleaning systems to reduce ammonia emissions from animal houses is increasing. These systems are normally used in order to comply with local or national regulations of ammonia emission. Therefore, accurate determination of the proportion of ammonia being removed by these systems is crucial. There are two main methods available to measure ammonia removal efficiency of scrubbers: air balance (based on the measurement of ammonia concentrations in air) and combined water-air balance (in which it is also necessary to determine the amount of nitrogen recovered in the liquid phase). The first method is simpler to establish, while the second method might provide deeper information about the processes occurring. The main aim of this work was to assess, in terms of the variability of the results, the use of these two methods to evaluate the efficiency of an acid packed bed scrubber on a pig farm. An acid packed bed scrubber (70% NH_3 removal) was monitored during ten complete 24 h cycles for ammonia concentrations, airflow rates, and nitrogen accumulation in the acid solution basin. The average efficiency calculated using the air balance method was 71% (±4%), close to the design value of 70%, while the average efficiency when using the combined water-air balance method was 255% (±53%). The accumulation and precipitation of ammonium salts in the packing material seem to be the main cause of the high variability and inaccuracy of the combined water-air balance method observed for this type of scrubber. According to these results, it is recommended to use the air balance method when determining the ammonia removal efficiency for acid packed bed scrubbers similar to the one studied here. According to the variability of the results observed in this work, at least 24 measurement days are needed in order to keep the relative error below 5% when using the air balance method to determine the ammonia removal efficiency of an acid packed bed scrubber.*

Keywords. *Acid scrubber, Ammonia, Efficiency, Methods, N balance.*

Livestock production is one of the major contributors to ammonia, odor, and particulate matter emissions in the agricultural sector. Recently, international commitments (e.g., Kyoto Protocol, European Ceilings Directive 2000/81 CE) are binding the participating countries to reduce total emissions of atmospheric pollutants. The use of techniques to reduce emissions is playing a key role in several areas across Europe where livestock farming facilities are concentrated.

A large variety of techniques is available for the reduction of ammonia emission from animal houses. According to Ndegwa et al. (2008), these techniques can be classified in four main groups: reduction of nitrogen excretion, reduction of volatile nitrogen, building design and manure management, and emissions capture and treatment. Techniques for treatment of exhaust air from farms are included in this last group. These systems, also called "end-of-pipe systems," have become off-the-shelf techniques for the reduction of NH_3 emissions from pig and poultry houses in countries like The Netherlands, Germany, and Denmark (Melse et al., 2009a).

A packed bed air scrubber is a reactor filled with an inert or inorganic packing material. This material is either continuously or intermittently sprayed with water to keep it wet. The exhaust air of the farm is driven through the scrubber. This process results in contact between the air and water, enabling a mass transfer from soluble gases to a liquid phase. A fraction of the trickling water is continuously recirculated, while another fraction is discharged and replaced by fresh water.

Packed bed air scrubbers can be classified in two main groups according to their operation principle: acid scrubbers and biotrickling scrubbers. Acid packed bed scrubbers are based on the capture of ammonia in an acid solution that is recirculated over the packed material. An ammonium salt is formed that is discharged with a certain frequency. Sulfuric acid is commonly used, and pH is kept between 2 and 4. Melse and Ogink (2005) reported an average ammonia removal efficiency of 96% in acid scrubbers (ranging from 40% to 100%). Biotrickling scrubbers work on the principle of the formation of a bacterial biofilm in the packing material. These bacteria degrade the water-soluble components of the air that have been trapped in the water. Due to this bacterial

Submitted for review in January 2011 as manuscript number SE 9019; approved for publication by the Structures & Environment Division of ASABE in August 2011.

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  ISSN 2151-0032

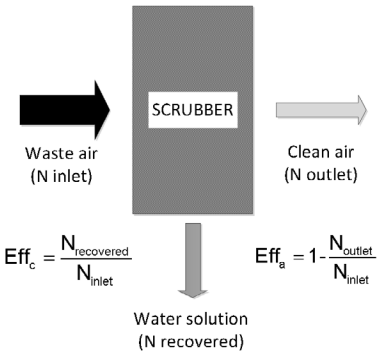

Figure 1. Types of mass balance for the determination of ammonia removal efficiency in air scrubbers, with Eff_c representing the combined water-air balance and Eff_a the air balance.

activity, ammonia is converted into nitrites and nitrates. Nitrogen concentrations in the water are kept below inhibiting levels by regular discharge of the recirculation liquid. The average ammonia removal in these filters has been estimated at 70% (ranging from 0% to 100%) by Melse and Ogink (2005). One of the negative aspects of biological air treatment systems may be the risk of N_2O emission as a consequence of the nitrification-denitrification (NDN) processes (Hahne and Vorlop, 2004). Besides ammonia, scrubbers also remove odor and particulate matter from the air to some extent (Melse and Ogink, 2005; Melse et al., 2010, 2011).

Since the ammonia removal efficiency of air scrubbing systems is used for regulatory purposes, the measurement technique for evaluating the removal efficiency plays a crucial role. Two main techniques, both based on a mass balance approach, can be used (Manuzon et al., 2007, Shah et al., 2008). The first technique (air balance) is based on measurement of the reduction of ammonia concentration in the exhaust air by determining the NH_3 concentration before and after the scrubbing process. This is the most common technique, and it is used worldwide since the early application of scrubbers. The second technique (combined water-air balance) consists of measuring the amount of nitrogen that has been recovered in the liquid phase and relating it to the total amount of ammonia entering the system. The combined water-air balance technique can be an additional requirement to the air balance measurements when certifying a scrubber, as it is in Germany. Figure 1 summarizes both measurement methods.

The main advantage of the air balance technique is its simplicity, because it only requires measurement of the ammonia concentration in the air, whereas the combined water-air balance technique requires measurement of the concentrations in air and water and determination of volumes and/or flows as well. However, the combined water-air balance technique can provide further information about the process and nitrogen fluxes, e.g., the occurrence of gaseous nitrogen emissions other than ammonia, or nitrogen accumulation in the system. This could lead to a better understanding of NDN processes in biological scrubbers, since the amount of nitrogen emitted as N_2O and N_2 could be assessed. It also may help in understanding the precipitation of ammonium salts in the system.

One of the main constraints of the combined water-air balance approach is related to its accuracy. It is expected that measuring more parameters (e.g., air and water volumes) introduces extra measurement errors as compared to the air balance approach. This may lead to a lower overall accuracy of the nitrogen balance and, consequently, of the determined ammonia removal efficiency. Estellés et al. (2011a,b), in a theoretical study of an acid packed bed scrubber, estimated that the uncertainty associated with the efficiency measurement increases between 3 and 50 times (depending on the measurement methods and expected efficiency) when using the combined water-air balance instead of the air balance.

Therefore, there is a need to test whether these theoretical findings may be applicable in practice. For the purpose of validating these results, the two balance methods for the determination of scrubber efficiency were tested in a particular case in an acid packed bed scrubber.

The main aim of this work is to assess the use of the air balance and the combined water-air balance to evaluate the efficiency of an acid scrubber installed on a pig farm in terms of the variability of the results.

Materials and Methods

General Approach

A single-stage acid scrubber was chosen in order to simplify the measurements and to avoid possible biological activity that could lead to the loss of nitrogen in the form of N_2O or N_2. All measurements were conducted during a three-week period, starting 9 June 2009, resulting in ten trials with each trial lasting 24 h. A graphical representation of the experiment is presented in figure 2.

Relative humidity and air temperature were recorded every 5 min at the air inlet and outlet using temperature and relative humidity (T/RH) sensors (Hygroclip-S, Rotronic Instrument Corp., Hauppauge, N.Y.). Data were collected using a data logger (CR10X, Campbell Scientific, Inc., Logan, Utah).

Description of the Scrubber

Measurements were taken from an acid scrubber (minimum required average NH_3 removal was 70% according to the manufacturer) installed in a fattening pig house (1,180 animals) with partially slatted floors in Schijndel, The Netherlands. The house had a single air exhaust in which a scrubber was installed (fig. 3), with a maximum capacity of 90,000 m^3 h^{-1} according to the manufacturer and a pressure drop that may vary from 50 to 115 Pa according to the

Figure 2. Time-line of the experiment. Gray bars indicate measuring periods, and white bars indicate non-measuring periods.

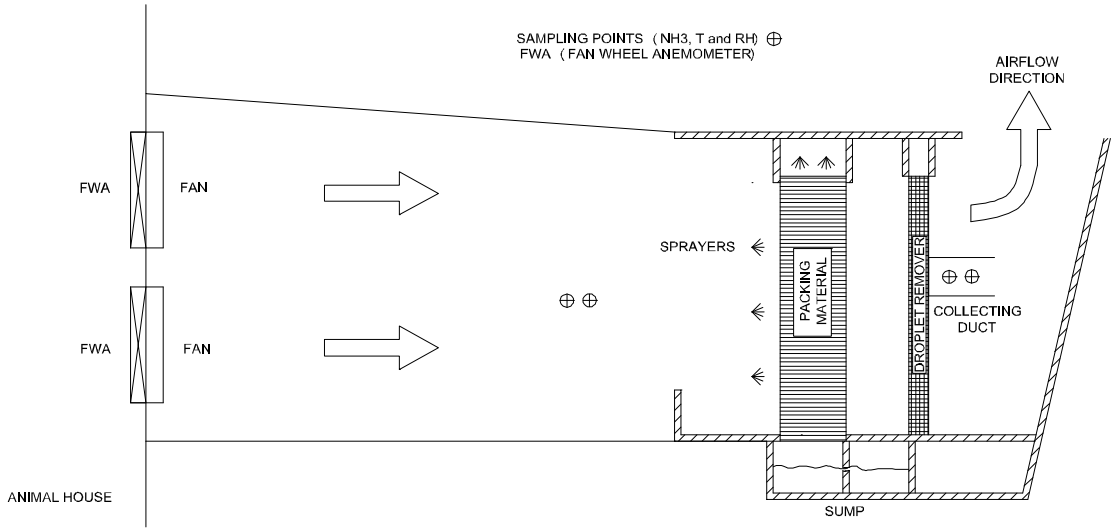

Figure 3. Scheme of the acid packed bed scrubber studied.

Deutsche Landwirtschafts Gesellschaft certification (DLG, 2009).

The scrubber had a cross-flow configuration. The packing was formed by a stack of vertical ion-exchange fiber cloths, directed parallel to the airflow (460 cm width, 201 cm height and 50 cm thickness), over which the acid solution was sprayed intermittently (at a rate of 20 m^3 h^{-1}, about 1 min every 20 min). This means that the empty bed residence time at maximum airflow equals to 0.18 s. This packing material was cleaned using high-pressure water one week before the experiment started. A 15 cm wide plastic droplet remover, consisting of 2 cm side hexagonal cells parallel to the airflow, was also present. A water collection basin (460 cm width, 45 cm height, and 95 cm length) was installed below the packing unit. The acid solution was recirculated (1 m^3 h^{-1}), and pH was automatically controlled and kept below 4 and over 1.5 by means of addition of concentrated sulfuric acid (1 M) to the system. Fresh water was continuously added to the system to compensate for the volume of evaporated liquid. The acid solution in the basin was discharged weekly but never when a 24 h measurement trial was being carried out. After the discharge of the acid solution, fresh water and sulfuric acid were added automatically to the system in order to achieve normal process operations again. The scrubber under investigation is similar to the acid scrubber described by Aarnink et al. (2011).

It is important to remark that the operating principle of this specific scrubber may not be representative for all acid scrubber operations. The packing material of acid scrubbers is in most cases continuously wetted with an acid solution, which in this case happened intermittently according to the manufacturer's specifications. In addition, the packing material of the scrubber used in this work is formed by stacks of fiber cloths parallel to the airflow, while normally the packing material is formed by a structured plastic packed bed.

Air Balance

General Approach

To obtain the efficiency of the scrubber using the air balance method (Eff_a), the ammonia-N concentrations in the inlet ($[NH_3\text{-}N]_i$, mg m^{-3}) and outlet air ($[NH_3\text{-}N]_o$, mg m^{-3}) must be determined. The efficiency is calculated as follows:

$$Eff_a = 1 - \frac{[NH_3\text{-}N]_o}{[NH_3\text{-}N]_i} \quad (1)$$

Air Ammonia Concentrations

Average daily ammonia concentrations in the air were determined using active wet methods (Ni and Heber, 2008). Two sampling locations were defined (inlet air and outlet air). For each sampling location, duplicate sampling lines were installed. The sampling lines for the outlet air were placed inside a plastic collecting duct (30 cm diameter and 50 cm length) attached to the droplet remover in order to exclude interference with outside air and to obtain representative samples of outlet ammonia concentrations (fig. 3). Next, each sampling line was divided into two replicates, obtaining eight sampling points (four from the inlet and four from the outlet air). Two constant-flow pumps were installed for air sampling. Critical orifices (Louwers, Hapert, The Netherlands) were used in order to obtain constant airflows (1 L min^{-1}) through the wet traps (impingers). All sampling tubes were made of polytetrafluoroethylene (PTFE) in order to avoid ammonia adsorption (Philips et al., 1998). Two impingers containing a nitric acid solution (0.05 M) were located in series for each sampling point. The ammonia concentration in the air was calculated from the air sampling rate, the nitrogen content of the acid solution in the bottles, which was determined spectrophotometrically at 655 nm (NEN, 2006), and the weight of the bottles before and after the sampling period to correct for volume increase due to water condensation. The air sampling rate of the impingers was determined by measuring the airflow twice, at the beginning and at the end of each measuring period, with a flowmeter (Defender 510, BIOS International Corp., Butler, N.J., accuracy ±1% of reading).

Ammonia-N concentrations in the air (mg m^{-3}) were calculated as follows:

$$[NH_3\text{-}N] = \frac{NH_4^+\text{-}N \times IM}{IA \times t \times D \times 10^{-6}} \quad (2)$$

where NH_4^+-N is the ammonium-N concentration in the acid solution (mg L^{-1}), *IM* represents the impinger solution mass corrected for condensation (g), *IA* is the average airflow rate through the impinger (mL min^{-1}), *t* is the sampling time (min), *D* is the density of the acid solution (10^3 g L^{-1}), and 10^{-6} is the conversion factor from mL to m^3.

Combined Water-Air Balance

General Approach

For the determination of the efficiency using the combined water-air balance method (Eff_c), it is necessary to determine the ammonia flux coming in the scrubber through the air (ANH_3, kg N) and the amount of ammonia accumulated in the acid solution of the system (WNH_3, kg N). The efficiency of the system can be then calculated using equation 3:

$$Eff_c = \frac{WNH_3}{ANH_3} \quad (3)$$

Ammonia Flux in the Air

The amount of ammonia entering the scrubber through the air was determined by multiplying the average ammonia concentration determined using the impinger method ($[NH_3\text{-}N]_i$) by the airflow rate through the scrubber (*F*, m^3 h^{-1}) during the sampling period (*t*, min) using equation 4:

$$ANH_3 = [NH_3\text{-}N]_i \times 10^{-6} \times \int \frac{F}{60} \times dt \quad (4)$$

where 10^{-6} is the conversion factor from mg m^{-3} to kg m^{-3}.

The airflow rate was determined using four fan-wheel anemometers (Fancom BV, Panningen, The Netherlands) attached to the four exhaust fans (90 cm dia.) of the building. The anemometers were calibrated in a wind tunnel by the DLG in Germany (Ref 09-566). Airflow rates were recorded every 5 min using a data logger (CR10X, Campbell Scientific, Inc., Logan, Utah).

Ammonia Recovered in the Water

The amount of ammonia recovered in the water was calculated as the difference between the amount of ammonia present in the acid solution basin before and after the measuring period using equation 5. To obtain these values, the ammonium concentrations in water (WNH_4^+-N, kg N m^{-3}) as well as the water volume in the acid solution basin (*V*, m^3) at the beginning (*i*) and end (*f*) of the experiment were determined:

$$WNH_3 = \left(WNH_4^+ - N \times V\right)_f - \left(WNH_4^+ - N \times V\right)_i \quad (5)$$

Duplicate water samples (100 mL) were taken from the acid solution basin at the beginning and end of each sampling period. These samples were later analyzed for ammonium concentration using spectrophotometric methods (NEN, 2006). EC and pH determinations were conducted as well (APHA, 2005). For consecutive sampling periods (trials 3 to 5 and trials 6 to 10), it must be considered that the final sample of a period equals the initial sample of the next period.

The volume of the liquid in the acid solution basin was determined by multiplying the acid solution basin surface times the water height. The surface of the acid solution basin was calculated using the manufacturer's measurements. The measurement of the water height at the beginning and end of each sampling period was determined as follows: first, the recirculation pump of the scrubber was manually stopped, if running, to allow stabilization of the water level. After 5 min without recirculation, a calibrated ruler was introduced into the acid solution basin, and the water height was measured in triplicate. As explained earlier, for consecutive sampling periods, the final volume of a period equals to the initial volume of the next period. In order to check whether the water volume measurements were correct, a psychometric balance was also developed. Air humidity and temperature data, as well as the volume of compensation water added in each balance, were used. Water volume measurements were considered valid only if the difference between direct measurements and the psychometric balance remained below 5%, which happened for all measurement periods presented in this work.

Time Scale

In order to evaluate the effect of the time scale of the measurements, the data obtained from daily measurements were assessed in two ways. First, the daily variability of the air balance was calculated. This value was used as an estimator to determine the minimum number of daily measurements needed to obtain the average removal efficiency of the system with a relative error below 5%. The following expression was used:

$$N \geq \left(\frac{t_\alpha}{2}\right)^2 \times \left(\frac{s}{\delta}\right)^2 \quad (6)$$

where $t\alpha_{/2}$ is the upper critical values from a *t* distribution, α is the risk of rejecting a true hypothesis (0.05), *s* is the standard deviation of the sample (the ammonia removal efficiency in percentage), and δ is the maximum relative expected error (5%).

As a second step, the time scale of the combined water-air balance was evaluated. This balance was calculated on both a daily basis and during a cycle, considering that a cycle comprises consecutive measurements between water discharge processes. Therefore, this balance was developed using the data from the last five trials (trials 6 to 10).

Statistical Analysis

The effect of inlet ammonia concentration on outlet ammonia concentration was tested through linear regression. In addition, in order to test the effect of average pH and EC, inlet airflow rates, inlet ammonia concentrations, and environmental conditions (inlet temperature and relative humidity) on the ammonia removal efficiency calculated using the air balance, a simple linear regression with each of the parameters was developed. The REG procedure of the statistical software SAS (2001) was used, following the model below:

$$Eff_a = \alpha + \beta \times X + \varepsilon \quad (7)$$

where α is the intercept of the regression, β is the slope, *X* is the considered parameter (average pH, average EC, airflow rates, inlet ammonia concentrations, inlet *T*, and inlet RH), and ε is the error of the model.

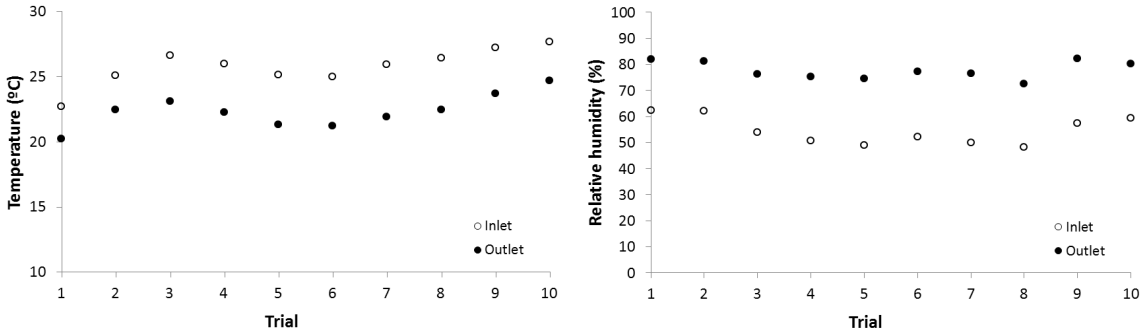

Figure 4. Average daily (*N* = 278) temperature and relative humidity registered during the experimental period in the inlet and outlet air.

Results and Discussion

Environmental Conditions, Airflow Rates, and Water Volumes

The average temperature and relative humidity registered during each trial are shown in figure 4. Average (±SE) values for temperature (25.8˚C ±2.3˚C) and relative humidity (54.5% ±7.3%) were in the ranges observed by Seedorf et al. (1998b) in fattening pig farms during summer in northern European buildings. As expected, an increase in the humidity of the air (on average 21.8% ±5.5%) and subsequently a reduction of the air temperature (averaging 3.2˚C ±0.9˚C) were observed. This fact is due to the "wetting effect" (adiabatic evaporation) of the scrubber, which causes an average (±SE) water content increase of the air of 1.9 ±0.4 g H_2O kg^{-1} air. The average (±SE) airflow rate for the whole experiment was 41.9 ±6.6 m^3 h^{-1} $animal^{-1}$. The daily average value for each trial is presented in table 1.

If expressed per animal place, the airflow rates were similar to those obtained for fattening pig buildings by Seedorf et al. (1998a) in summer conditions in The Netherlands, which were on average 42.7 m^3 h^{-1} $animal^{-1}$ (in this study, the average airflow rate was 42.0 ±0.1 m^3 h^{-1} $animal^{-1}$). The standard error of these measurements was 0.3% of the average value. This low error value is attributable to the high frequency of the measurements. This standard error must not be considered an indicator of the measurement accuracy, since it does not include any reference to the measurement device's accuracy, only its precision.

Average water volumes in the acid solution basin for each trial are also shown in table 1. The water volume always remained below the maximum level of the basin (1.97 m^3). The water level presented a low variability among different trials, varying from 1.3 to 1.6 m^3.

Ammonia and Ammonium Concentrations

In table 1, the average inlet and outlet ammonia concentrations in the air are shown for each measurement period. The average (±SE) inlet concentration for all measurement periods was 9.8 ±0.2 mg m^{-3}. The variability of the inlet concentrations within days was low during the experiment, ranging from 8.1 to 11.6 mg m^{-3}. The average value is within the range reported by Melse and Ogink (2005) for the same animal category, and it is also very close to the average NH_3 concentration of 10.4 mg m^{-3} observed by Groot Koerkamp et al. (1998) for the same animal category and management system in northern European facilities. The standard error was 1.6% of the average value, which is very close to the uncertainty calculated for this factor (1.2%) in the theoretical approach developed by Estellés et al. (2011a,b).

Outlet ammonia concentrations were on average (±SE) 2.9 ±0.2 mg m^{-3}, ranging from 1.2 to 5.3 mg m^{-3}. The variability of the data was higher as compared to the inlet, and a clearly decreasing tendency as the experiment advanced can be observed, which means that the removal efficiency increased. The standard error (7.11% of the average value) is similar to the uncertainty value (3.9%) determined by Estellés et al. (2011a,b) for this parameter in a theoretical framework. In this case, the relative error is higher due to the lower absolute concentrations observed. There was no statistical relationship between the inlet and outlet ammonia concentrations ($p > 0.09$).

Table 1. Average (±SE) air ammonia concentrations in the inlet ($[NH_3\text{-}N]_i$) and outlet ($[NH_3]_o$), ammonium-N concentrations in the water at the beginning (WNH_{4i}) and end (WNH_{4f}) of the trial, airflow rates, and initial (V_i) and final (V_f) water volumes in the acid solution basin for each 24 h trial (June 2009, Schijndel, The Netherlands).

Trial	$[NH_3\text{-}N]_i$ (mg NH_3 m^{-3})	$[NH_3\text{-}N]_o$ (mg NH_3 m^{-3})	Airflow (m^3 h^{-1})	WNH_{4i} (kg NH_4^+-N m^{-3})	WNH_{4f} (kg NH_4^+-N m^{-3})	V_i (m^3)	V_f (m^3)
1	9.5 ±0.1	4.3 ±0.3	37,589 ±240	20.9 ±1.8	35.3 ±0.9	1.28 ±0.00	1.40 ±0.00
2	8.9 ±0.1	3.3 ±0.3	41,858 ±218	26.6 ±4.9	33.2 ±4.1	1.36 ±0.00	1.60 ±0.00
3	7.4 ±0.0	3.0 ±0.1	55,046 ±318	43.0 ±2.7	41.9 ±0.2	1.36 ±0.01	1.58 ±0.00
4	7.7 ±0.0	2.9 ±0.1	52,616 ±391	41.9 ±0.2	6.90 ±0.0	1.58 ±0.00	1.60 ±0.01
5	8.1 ±0.2	2.6 ±0.2	48,177 ±295	6.90 ±0.0	40.6 ±4.2	1.6 ±0.01	1.47 ±0.00
6	8.5 ±0.0	2.0 ±0.3	44,695 ±376	23.2 ±0.2	39.8 ±2.9	1.55 ±0.00	1.57 ±0.00
7	7.7 ±0.0	1.5 ±0.0	51,737 ±503	39.8 ±2.9	20.3 ±1.3	1.57 ±0.00	1.41 ±0.00
8	6.1 ±0.0	1.9 ±0.0	55,593 ±178	20.9 ±1.3	35.3 ±0.7	1.41 ±0.00	1.40 ±0.01
9	7.6 ±0.0	1.2 ±0.0	53,062 ±313	35.4 ±0.7	42.8 ±0.1	1.4 ±0.01	1.53 ±0.00
10	8.2 ±0.1	1.0 ±0.1	53,288 ±230	42.8 ±0.1	53.3 ±0.2	1.53 ±0.00	1.50 ±0.00
Average (±SE)[a]	8.0 ±0.4	2.4 ±0.5	49,475 ±148	30.1 ±2.7	34.9 ±2.9	1.47 ±0.02	1.51 ±0.01

[a] SE = standard error of the mean.

Table 2. Values for each component of the nitrogen balance, initial and final pH and EC, inlet temperature, and calculated efficiencies for each trial.

Trial	ANH_3 (kg N-NH_3)	WNH_3 (kg N-NH_4)	pH_i	pH_f	EC_i (mS cm^{-1})	EC_f (mS cm^{-1})	Inlet T (°C)	Air Balance (Eff_a, %)	Water-Air Balance (Eff_c, %)
1	8.7	22.8	1.9	2.5	145.5	182.8	24.9	54.5	263.3
2	7.5	16.9	1.5	2.1	192.6	>200	22.7	62.9	225.0
3	9.7	7.8	2.9	1.9	>200	>200	25.1	59.1	80.4
4	9.6	-55.0	1.9	2.0	>200	59.1	26.6	62.5	-571.8
5	9.2	48.5	2.0	1.4	59.1	>200	26.0	68.1	525.4
6	9.0	26.5	1.9	1.5	104.5	>200	25.2	76.9	294.2
7	9.4	-34.0	1.5	1.5	>200	137.8	25.0	81.7	-363.1
8	8.9	20.8	1.6	1.5	137.8	196.7	25.9	71.7	235.6
9	9.6	16.0	1.4	1.5	196.7	>200	26.4	84.5	165.8
10	9.3	14.7	1.8	1.6	>200	>200	27.2	87.8	157.9
Average (±SE)[a]	9.1 ±0.2	8.5 ±9.6	1.8 ±0.2	1.8 ±0.1	163.6[b]	177.6[b]	25.8 ±1.5	70.9 ±3.6	254.5 ±52.5[c]

[a] SE = standard error of the mean.
[b] SE was not calculated due to the presence of values out of the equipment measuring range (>200 mS cm^{-1}).
[c] The average value and its SE have been calculated only for non-consecutive (i.e., independent) trials (trials 1, 2, 3, 5, 6, 8, and 10).

The measured ammonium concentrations in water during the experiment are shown in table 1. Ammonium concentrations varied from 6.9 to 53.3 g L^{-1}. It is not clear why WNH_{4i} of trial 5 is much lower than the other measurements. The variability of concentration in the basin for a given moment can be related to the standard error of the results, which represents 8.9% and 8.3% of the average value for the initial and final concentrations, respectively. This adds an extra error source with respect to the theoretical model developed by Estellés et al. (2011a,b).

The variability within days (deviation between replicated samples) ranged from 0.2% to 26% of the average observed value, being on average 8.7%. This is an indicator of the high variability of ammonium concentrations within different days in the acid solution basin. This high variability could be also caused by the operating principle of this specific scrubber, since the discontinuous water recirculation and the packing material with sheets parallel to the airflow could favor the formation of ammonium salts in the packing.

An increase in concentration over time was not observed, nor was a reduction in ammonium concentration observed after water discharges (before trials 2 and 6; fig. 2). This may have been caused by the partial (rather than full) discharge of the water contained in the acid solution basin. In addition, ammonium salts retained in the packing material may have dissolved in the fresh water, leading to an increase of the ammonium concentration in the acid solution after water discharge.

Removal Efficiencies Calculated from Air Balance and Water-Air Balance

The average (±SE) NH_3 removal efficiency calculated using the air balance was 70.9% ±11.4% over the inlet concentration, ranging from 54.5% to 87.9% (table 2). It can be considered that this average removal efficiency accomplishes the specifications of the manufacturer (70% reduction) and the Dutch regulations (Melse et al., 2009b). The variation of the system efficiency between days was reported during the certification of the system by the DLG (2009). The average pH of each balance presented a significant effect ($p < 0.01$) on the ammonia removal efficiency calculated using the air balance method, with an intercept $\alpha = 124.0 \pm 13.2$ ($p < 0.001$) and slope $\beta = -29.7 \pm 7.3$ ($p < 0.005$). The inlet temperature of the air also presented a significant effect ($p < 0.05$) on the system efficiency. Nevertheless, the R^2 of the regression obtained was below 45%. Thus, further research is needed to confirm this relationship. There was no statistical effect of average inlet ammonia concentration ($p > 0.3$), airflow ($p > 0.2$), or relative humidity ($p > 0.7$) on the calculated efficiency.

The efficiency calculations on a daily basis using the combined water-air balance method produced unsatisfactory results, with an average (±SE) of 254.5% ±52.5% (calculated only for independent trials) and with values ranging from -571.8% to 525.4% (table 2). No relationship was observed among these results and the efficiencies calculated using the air balance method. The high variability observed for the calculated efficiencies based on the combined water-air balance (if compared with the air balance) may indicate a high random error for this method.

Considering that no water was discharged during the trials and the measurements of ammonia concentration in air, airflow rate, and water volume presented very low variability, these errors may arise from the measurements of ammonium concentrations in the acid solution. The high variability observed in these concentrations may arise from accumulation and precipitation processes of ammonium salts occurring in the packing material. The DLG (2009) reported an accumulation of ammonium-N in the packing material of 16.3% over the inlet ammonia-N load when testing an acid packed bed scrubber of the same model as the scrubber evaluated in this work. This accumulation and precipitation of ammonium salts may occur in all scrubbers (i.e., acid scrubbers, forming ammonium salts, and biological scrubbers, forming organic nitrogen floccules). Nevertheless, the characteristics of this model (with discontinuous wetting and sheets placed parallel to the airflow rate) make this process more frequent and significant.

Another explanation for the high variability of this specific type of scrubber is that, due to the on/off cycle of the pumps, the mixing of the water in the acid solution basin before sampling was not optimal, resulting in non-representative sampling.

Time Scale

Given the calculated variability of the removal efficiency using the air balance method (SD was 16% of the average value), and following equation 6, to obtain an average efficiency of the system with a relative error below 5%, at least 23 daily measurements must be performed ($p < 0.05$). This variability between trials is slightly higher than the variability reported by Melse and Ogink (2005) for different types of acid packed bed scrubbers (in that work, the standard deviation of the average efficiency ranged from 1% to 10% of the mean for different scenarios). Nevertheless, the number of measurements proposed in this work is within the range proposed by Melse and Ogink (2005) when monitoring the performance of acid packed bed scrubbers in The Netherlands (from 19 to 100 measurements per scrubber).

Regarding the time scale of the combined water-air balance method, during the period from trials 6 to 10 (consecutive measurements without water discharge), the total ammonium–N recovered in the water was 44.1 kg NH_4^+-N, and the ammonia–N at the inlet was 46.1 kg N-NH_3, which equals a removal efficiency of 95.50%. This value is still different from the calculated average for the whole period using the air balance method (80.4%). This means that the results obtained with the combined water-air balance method improve (considering the air balance method as a reference) when longer integration times are used, i.e., when a number of consecutive measurements is performed. Nevertheless, the accuracy of this method is low when compared to the air balance method.

Based on the results of this case study, the accuracy of the air balance method is higher than that of the combined water-air balance method. Because more parameters are involved when developing the combined water-air balance, the error sources also increase, leading to higher errors when determining the overall ammonia removal efficiency. These findings agree with the theoretical work developed previously by Estellés et al. (2011a,b).

Conclusions

Ten complete 24 h nitrogen balances were carried out in order to determine the ammonia removal efficiency of the acid air scrubber system. The results obtained show that:

- The average (±SE) ammonia removal efficiency calculated using the air balance method was 70.9% ±3.6%. This method is normally used to assess the efficiency of a scrubber. Using the combined water-air balance method, which also takes into account the nitrogen recovery from the water phase, the calculated efficiency (average ±SE) was 254.5% ±52.5%.
- The large variations that were found using the combined water-air balance method might be explained by accumulation and precipitation of ammonium salts in the packing material, as well as non-representative sampling of water due to incomplete mixing occurring in this type of scrubber (i.e., operating with discontinuous recirculation of water).
- 24 h air balances are simple and stable (due to the low variability observed) tools to assess the ammonia removal efficiency of acid scrubbers. However, 24 h combined water-air balances are not recommended for the type of scrubber that was investigated in this study due to the large variations that were found using this method.
- From the results of this study, it follows that in order to determine the removal efficiency using the air balance method with a relative error lower than 5% it is necessary to acquire at least 23 measurements with a time basis of 24 h.

Acknowledgements

We would like to thank the Netherlands Ministry of Economic Affairs, Agriculture, and Innovation and the Netherlands Ministry of Infrastructure and the Environment for financial support. We also acknowledge the owner of the facility where the study was conducted for allowing the access to the building and for help in many other ways.

References

Aarnink, A. J. A., W. J. M. Landman, R. W. Melse, and T. T. T. Huynh. 2011. Elimination of airborne microorganisms from exhaust air of animal houses. Submitted to *Trans. ASABE*.

APHA. 2005. *Standard Methods for the Examination of Water and Wastewater.* 21st ed. Baltimore, Md.: Centennial Edition.

DLG. 2009. Abluftreinigungsanlage “Chemowäscher (+)”. DLG-Prüfbericht 5880. Meijel, The Netherlands: Uniqfill Air BV. Available at: www.dlg-test.de/pbdocs/5880.pdf.

Estellés, F., S. Calvet, R. W. Melse, and N. W. M. Ogink. 2011a. Evaluation of N_2 and N_2O formation in ammonia scrubbers: Options for performance measurement. Submitted to *Environ. Eng. Sci*.

Estellés, F., S. Calvet, R. W. Melse, and N. W. M. Ogink. 2011b. Analysis of reactive nitrogen (N_r) balances as a tool for assessing the performance of air scrubbers at livestock facilities. Report 376. Wageningen, The Netherlands: Wageningen UR, Livestock Research.

Groot Koerkamp, P. W. G., J. H. M. Metz, G. H. Uenk, V. R. Phillips, M. R. Holden, R. W. Sneath, J. L. Short, R. P. White, J. Hartung, J. Seedorf, M. Schroder, K. H. Linkert, S. Pedersen, H. Takai, J. O. Johnsen, and C. M. Wathes. 1998. Concentrations and emissions of ammonia in livestock buildings in northern Europe. *J. Agric. Eng. Res.* 70(1): 79-95.

Hahne, J., and K. D. Vorlop. 2004. Are waste air scrubbers useful in reducing ammonia emissions from piggeries? *Landtechnik* 59(2): 106-107.

Manuzon, R. B., L. Y. Zhao, H. M. Keener, and M. J. Darr. 2007. A prototype acid spray scrubber for absorbing ammonia emissions from exhaust fans of animal buildings. *Trans. ASABE* 50(4): 1395-1407.

Melse, R. W., and N. W. M. Ogink. 2005. Air scrubbing techniques for ammonia and odor reduction at livestock operations: Review of on-farm research in The Netherlands. *Trans. ASAE* 48(6): 2303-2313.

Melse, R. W., N. W. M. Ogink, and W. H. Rulkens. 2009a. Air treatment techniques for abatement of emissions from intensive livestock production. *Open Agric. J.* 3: 6-12.

Melse, R. W., N. W. M. Ogink, and W. H. Rulkens. 2009b. Overview of European and Netherlands’ regulations on airborne emissions from intensive livestock production with a focus on the application of air scrubbers. *Biosystems Eng.* 104(3): 289-298.

Melse, R. W., P. Hofschreuder, and N. W. M. Ogink. 2010. Fine dust (PM_{10}) removal by air scrubbers treating animal house exhaust air. ASABE Paper No. 1009986. Pittsburgh, Pa.: ASAE.

Melse, R. W., P. Hofschreuder, and N. W. M. Ogink. 2011. Fine dust (PM_{10}) removal by air scrubbers at livestock facilities:

Results of measurement program on 24 farms. Submitted to *Trans. ASABE*.

Ndegwa, P. M., A. N. Hristov, J. Arogo, and R. E. Sheffield. 2008. A review of ammonia emission mitigation techniques for concentrated animal feeding operations. *Biosystems Eng.* 100(4): 453-469.

NEN. 2006. Water: Fotometrische bepaling van het gehalte aan ammoniumstikstof en van de som van de gehalten aan ammoniumstikstof en aan organisch gebonden stikstof volgens Kjeldahl met behulp van een doorstroomanalysesysteem. Delft, The Netherlands: Nederlands Normalisatie Instituut.

Ni, J. Q., and A. J. Heber. 2008. Sampling and measurement of ammonia at animal facilities. *Adv. in Agron.* 98: 201-269.

Philips, V. R., M. R. Holden, R. W. Sneath, J. L. Short, R. P. White, J. Hartung, J. Seedorf, M. Schröder, K. H. Linkert, S. Pedersen, H. Takai, O. J. Johnsen, P. W. G. Groot Koerkamp, G. H. Uenk, R. Scholtens, J. H. M. Metz, and C. M. Wathes. 1998. The development of robust methods for measuring concentrations and emission rates of gaseous and particulate air pollutants in livestock buildings. *J. Agric. Eng. Res.* 70(1): 11-24.

SAS. 2001. *SAS User's Guide: Statistics*. Ver. 6a. Cary, N.C.: SAS Institute, Inc.

Seedorf, J., J. Hartung, M. Schroder, K. H. Linkert, S. Pedersen, H. Takai, J. O. Johnsen, J. H. M. Metz, P. M. G. Groot Koerkamp, G. H. Uenk, V. R. Phillips, M. R. Holden, R. W. Sneath, J. L. Short, R. P. White, and C. M. Wathes. 1998a. A survey of ventilation rates in livestock buildings in northern Europe. *J. Agric. Eng. Res.* 70(1): 39-47.

Seedorf, J., J. Hartung, M. Schroder, K. H. Linkert, S. Pedersen, H. Takai, J. O. Johnsen, J. H. M. Metz, P. M. G. Groot Koerkamp, G. H. Uenk, V. R. Phillips, M. R. Holden, R. W. Sneath, J. L. Short, R. P. White, and C. M. Wathes. 1998b. Temperature and moisture conditions in livestock buildings in northern Europe. *J. Agric. Eng. Res.* 70(1): 49-57.

Shah, S. B., P. W. Westerman, R. D. Munilla, M. E. Adcock, and G. R. Baughman. 2008. Design and evaluation of a regenerating scrubber for reducing animal house emissions. *Trans. ASABE* 51(1): 243-250.