AIR SCRUBBING TECHNIQUES FOR AMMONIA AND ODOR REDUCTION AT LIVESTOCK OPERATIONS: REVIEW OF ON-FARM RESEARCH IN THE NETHERLANDS

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ABSTRACT. *Acid scrubbers and biotrickling filters have been developed for ammonia (NH_3) removal at pig and poultry houses in The Netherlands over the last 20 years to prevent acidification and eutrophication of soils. Because of growing suburbanization, odor removal is increasingly considered important as well. In this review, we report the results of the on-farm research on full-scale operated scrubbers for treatment of exhaust air from animal houses with regard to NH_3 and odor removal in The Netherlands. The NH_3 removal of acid scrubbers ranged from 40% to 100% with an overall average of 96%. The NH_3 removal of biotrickling filters ranged from −8% to +100% with an overall average of 70%. Minimum empty bed air residence times (EBRTs) were 0.4 to 1.1 s. For acid scrubbers, process control with pH measurement and automatic water discharge is sufficient to guarantee sufficient NH_3 removal. For biotrickling filters, however, improvement of process control is necessary to guarantee sufficient NH_3 removal. The odor removal of acid scrubbers ranged from 3% to 51% with an overall average of 27%. The odor removal of biotrickling filters ranged from −29% to +87% with an overall average of 51%. Minimum EBRTs were 0.5 to 2.3 s. Further research is necessary to explain this variation and to improve the odor removal efficiency of both acid scrubbers and biotrickling filters.*

Keywords. *Air cleaning, Ammonia, Biofilter, Biotrickling, NH_3, Odor, Pig, Poultry, Scrubber, Veal calves.*

Pig and poultry production contributes substantially to the economies of many Western European countries in terms of employment and export of products. Pig production in Western Europe is concentrated in several regions characterized by large-scale intensive farms. The Netherlands, with 16 million inhabitants and a population density of about 400 inhabitants per km^2, houses 13 million pigs at approximately 13,000 farms (CBS, 2002). These farms are mainly concentrated in the eastern and southern part of the country where opportunities for arable farming are limited by poor sandy soils. Since 1980, the emission of ammonia (NH_3) from livestock farming has become a major environmental concern because NH_3 is one of the three main causes of soil acidification and eutrophication of natural soils in The Netherlands (Heij and Erisman, 1995, 1997). Considerable efforts were put into the development of NH_3 abatement techniques in animal operations. In 2000, the NH_3 emission from livestock farming, however, still accounted for about 50% of the total emission of acidifying compounds (Koch et al., 2003). This focus on NH_3 abatement has resulted in the development of a large variety of low-emission livestock housing systems that include systems for treatment of exhaust air from animal houses, namely, acid scrubbers and biotrickling filters.

Most publications of the research and experiences that have been gathered in The Netherlands in the field of treatment of exhaust air from animal houses have been in Dutch and cannot be easily accessed by the international research community. Therefore, in this review, we summarize and discuss these results and experiences. First, we describe the programs in which the research was conducted. Next, we describe the methods used for NH_3 and odor determination and the principles of air scrubbing. Finally, we present the results of on-farm research on full-scale acid scrubbers and biotrickling filters, followed by a discussion of the results and some concluding remarks.

RESEARCH PROGRAMS ON AMMONIA AND ODOR

Since 1990, an NH_3 research program has been carried out in The Netherlands to investigate the NH_3 emission for various animal categories from both conventional housing systems and systems designed for low NH_3 emission, including air scrubbers (Mosquera et al., 2004). The NH_3 emission rates that have been found are used for regulatory purposes and are published on a regular basis as the “Regeling Ammoniak en Veehouderij” [Regulation on Ammonia and Livestock] (VROM, 2002). The NH_3 emission rates for conventional housing systems are presented in table 1, which lists the main animal categories for which air scrubbers are applied. This focus on NH_3 emission has resulted in the development of acid scrubbers and biotrickling filters for application in pig and poultry houses, which are now commercially available and considered as off-the-shelf techniques.

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Table 1. Average ammonia (NH_3) and odor emission rates of conventional housing systems for some animal categories (Mol and Ogink, 2002; Ogink, 2005).

Animal Category	Emission Rates NH_3 (kg animal place^{-1} year^{-1})	 Odor (OU_E [a] animal place^{-1} s^{-1})
Dry and pregnant sows	4.2	20.3
Farrowing sows (incl. piglets until weaning)	8.3	26.5
Weaned piglets	0.6	7.8
Growing-finishing pigs	2.5	23.0
Rearing pullets (aviary housing)	0.045	0.18
Layers (cage housing)	0.100	0.37
Broilers	0.080	0.22

[a] OU_E = European odor unit (CEN, 2003).

More recently, besides NH_3, the removal of odor compounds is increasingly considered important because of growing suburbanization. From 1996 to 2002, an odor research program was carried out in The Netherlands to investigate odor emission from both conventional animal housing systems and systems designed for low NH_3 emission, including air scrubbers (Ogink et al., 1997; Ogink and Klarenbeek, 1997; Ogink and Groot Koerkamp, 2001; Ogink and Lens, 2001; Mol and Ogink, 2002). The results from this research have been used to set up a new regulatory framework for odor control in the livestock industry. The odor emission rates that were found for conventional housing systems are presented in table 1.

The NH_3 and odor emission rates in table 1 are subject to considerable seasonal variation. Ogink and Lens (2001) reported coefficients of variations of odor emissions at different sampling days that ranged from 45% to 60% for conventional pig housing systems and from 50% to 80% for conventional poultry housing systems. For NH_3 emission, Mosquera et al. (2004) reported coefficients of variation for fattening pigs and pregnant sows of 45% and 22%, respectively, reflecting variations between day-to-day values.

When a scrubber system is installed for treatment of exhaust air from an animal house, the NH_3 and odor loading rate of the scrubber system equal the emission of a conventional housing system without air cleaning. The temperature of animal house exhaust air is about 18°C to 30°C and has a relative humidity of about 50% to 90%. The NH_3 and odor emission reduction that are achieved by air scrubbing will be discussed later.

METHODS FOR AMMONIA AND ODOR DETERMINATION

AMMONIA MEASUREMENT

Three different techniques are used for determination of the NH_3 concentration in the exhaust air of animal houses: an impinger method, a chemiluminescence method, and a photoacoustic gas analyzer.

In the impinger method, a fraction of the exhaust air is continuously drawn at a fixed flow rate, which is controlled by a critical orifice (usually 1 L min^{-1}) through a pair of impingers (0.5 L each) containing a strong acid solution (usually nitric acid, 0.03 to 0.2 M) and connected in series (Van Ouwerkerk, 1993). NH_3 is trapped by the acid and accumulates in the bottles until they are replaced, usually twice a week. Fluctuations in the NH_3 concentration of the sampled air are thus time-averaged. The values of the sampling flow rate and nitric acid concentration are chosen so that the second impinger, which serves as a control, does not contain more than 5% of the amount of NH_3 trapped in the first impinger. All sampling tubes are made of Teflon, insulated, and heated with a coil of resistance wire to approximately 20°C higher than ambient to prevent condensation of water and subsequent adsorption of NH_3. Finally, the NH_3 concentration of the air is calculated from the nitrogen content of the acid solution in the bottles, which is determined spectrophotometrically (NNI, 1998), and the given air sampling flow rate.

In the research program, the impinger method was mainly used for measuring scrubber efficiencies for NH_3 removal, as it can deal more easily with the water-saturated air from the scrubber outlet, as compared to the two methods described below. These methods were only used when more frequent continuous measurements, i.e., on a 1 to 5 min sampling basis, were required for the scrubber inlet air.

In the chemiluminescence method (Mosquera et al., 2002), the exhaust air is continuously sampled at a fixed flow rate, which is controlled by a critical orifice (0.5 L min^{-1}) and led to an NH_3 converter. In the converter, the sampled air is heated to 775°C in order to achieve catalytic conversion of NH_3 into NO (catalyst: stainless steel). The converter efficiency is calibrated regularly. After oxidation, the heated air is led to an NO_x analyzer (Monitor Labs, models 8840 and 42I) that measures the concentration of NO using the chemiluminescence principle at a temperature of 50°C. The NH_3 concentration is averaged over 1 min intervals and recorded by a datalogger. The NO_x analyzer is calibrated regularly. All sampling tubes are made of Teflon, insulated, and heated with a coil of resistance wire to approximately 20°C higher than ambient to prevent condensation of water and subsequent adsorption of NH_3.

In the photoacoustic gas analyzer method (Mosquera et al., 2002), the same sampling approach is used as for the chemiluminescence method. However, the concentration of NH_3 in the sample air is determined with a photoacoustic gas analyzer (Brüel and Kjaer; Multi-gas analyzer 1302). The NH_3 measurements are corrected for temperature and interference with H_2O and CO_2.

The accuracies of the three techniques described above, as expressed by the standard error under repeatability conditions, show levels that are within the 1% to 3% range (Mosquera et al., 2002; Ogink, 2005).

ODOR MEASUREMENT

For odor measurement, an air sample is collected in an initially evacuated Teflon odor bag (60 L). The bag is placed in an airtight container, the inlet of the bag is connected to the sampling port of the air inlet or air outlet of the scrubber, and the bag is filled by creating an underpressure in the surrounding airtight container by means of a pump. The air sampling flow rate is controlled by a critical orifice (0.5 L min^{-1}), and the odor bag is thus filled in 2 h. In this way, fluctuations in the composition of the air sample are time-averaged over 2 h. A filter (pore diameter: 1 to 2 μm) at the inlet of the sampling tube prevents the intake of dust that would otherwise contaminate the olfactometer. The sampling system is equipped with a heating system to prevent condensation in the bag or in the tubing. An odor bag remains in the container until analysis in the odor laboratory, which has to take place within 30 h after sample collection. Odor concentrations are determined in compliance with European olfactometric standard EN13725 (CEN, 2003) and the preceding Dutch olfactometric standard NVN2820/1A (NNI, 1996) that has been incorporated into the European standard. In both standards, the sensitivity of the odor panel is based on the 20 to 80 ppb n-butanol range. The odor concentrations are expressed in European odor units per m^3 air ($OU_E\ m^{-3}$) (CEN, 2003).

The accuracy of the sensory-based odor measurements is much lower than the accuracy of analytical NH_3 measurements. From an analysis on the accuracy of odor measurements, using olfactometric standards that comply with the EN13725 standard (Ogink et al., 1995), standard errors can be calculated for single odor measurements under repeatability conditions that range between 15% and 20%. It is therefore of importance in the design of measurement strategies that odor measurements are repeated sufficiently often. In the odor research program described here, measurements were taken on ten different sampling days to deal both with olfactometric measurement error and performance fluctuations.

WORKING PRINCIPLE OF AMMONIA SCRUBBING

A packed tower air scrubber, or trickling filter, is a reactor that has been filled with an inert or inorganic packing material (fig. 1). The packing material usually has a large porosity, or void volume, and a large specific area. Water is sprayed on top of the packed bed and consequently wetted. Contaminated air is introduced, either horizontally (cross-current) or upwards (counter-current), resulting in intensive contact between air and water, and enabling mass transfer from gas to liquid phase. A fraction of the trickling water is continuously recirculated; another fraction is discharged and replaced by fresh water.

For a given compound, the mass transfer rate ($kg\ h^{-1}$) from gas to liquid phase is determined by the concentration gradient, the size of the contact area between gas and water phase, and the contact time of gas phase and liquid phase (Coulson et al., 1999; Richardson et al., 2002; Van 't Riet and Tramper, 1991).

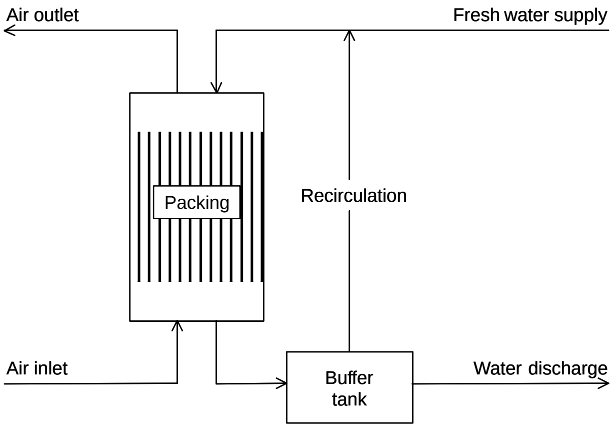

Figure 1. Schematic of a counter-current air scrubber.

Concentration Gradient

The rate of mass transfer of a compound is proportional to the concentration gradient between the gas phase and the liquid phase. For NH_3, the transfer to liquid phase and the dissociation in the water can be described as:

$$NH_3\ (g) + H_2O\ (l) \leftrightarrow NH_3\ (aq) + H_2O\ (l) \leftrightarrow NH_4^+\ (aq) + OH^-\ (aq) \qquad (1)$$

For exhaust air from an animal house, the NH_3 concentration in the gas phase, NH_3 (g), is given. The concentration in the liquid phase, NH_3 (aq), however, is determined by the water solubility, by the rate of water discharge and fresh water supply, by the pH-driven dissociation into ammonium (NH_4^+) and hydroxide (OH^-) ions (see "Experiences with Acid Scrubbers" section), and, if applicable, by the transformation of NH_3 into other compounds (see "Experiences with Biotrickling Filters" section). Because of the instantaneous dissociation of NH_3, the actual water solubility of NH_3 is not so much an issue.

Size of Contact Area between Gas and Water Phase

The rate of mass transfer of a compound is proportional to the contact area, which is determined by the specific surface area of the packing ($m^2\ m^{-3}$) and the degree of wetness of the packing material, which in turn is affected by means of wetting, such as trickling, spraying, and submerging, and the liquid flow rate.

Contact Time of Gas Phase and Liquid Phase

Mass transfer is only possible if the gas is in contact with the liquid for some duration of time. Usually this time is expressed as the empty bed air residence time (EBRT), which can be calculated by dividing the reactor volume (m^3) by the airflow rate ($m^3\ h^{-1}$). Especially for poorly water-soluble compounds, the gas residence time must be sufficiently long, as it directly determines the total mass transfer. Furthermore, a long gas residence time usually means that the ratio of liquid flow rate to gas flow rate is relatively high, which might promote mass transfer to the liquid phase for poorly water-soluble compounds.

Application of a packed tower scrubber requires that the animal house is equipped with a mechanical ventilation system, as the air is forced through the filter bed. Furthermore, the ventilation system must be able to yield the extra pressure drop.

In the past, biofilters with organic-based packing materials were used for treatment of exhaust air in animal houses in The Netherlands. Nowadays, only acid scrubbers and biotrickling filters are used for this purpose because biofilters cannot be operated well at the relatively high NH_3 and dust concentrations of the exhaust air (see "How to Increase Odor Removal" section).

EXPERIENCES WITH ACID SCRUBBERS

AMMONIA REMOVAL

In an acid scrubber, the pH is controlled, usually at a value below 4, by addition of acid to the recirculation water. The reaction equilibrium of equation 1 shifts to the right as the dissolved NH_3 is captured by the acid, forming an ammonium salt solution. For acid scrubbers that are applied in agriculture, Dutch regulations only allow sulfuric acid for this purpose, which results in the production of ammonium sulfate, or $(NH_4)_2SO_4$ solution. In a well-designed scrubber operating at a sufficiently low pH, NH_3 removal efficiencies of 90% to 99% can be achieved, as was demonstrated in a long-term measurement program that was carried out at five farm locations. Table 2 shows the results and the characteristics of the investigated scrubbers. The average NH_3 removal efficiency of all acid scrubbers was 96%. These results compare with average NH_3 removal efficiencies of over 90% that were found for an experimental pilot-scale acid scrubber treating pig house exhaust air (Hahne and Vorlop, 1998, 2001; Hahne et al., 2000). Current Dutch regulations require that implemented acid scrubbers achieve an average NH_3

Table 2. Characteristics and results of measurement programs on ammonia (NH_3) removal by full-scale acid scrubbers treating exhaust air of animal houses.

	Vrielink et al., 1997	Verdoes and Zonderland, 1999	Hol and Satter, 1998	Hol et al., 1999	Wever and Groot Koerkamp, 1999
Design Characteristics					
Animal category and number	66 growing-finishing pigs	54 growing-finishing pigs	6,040 layer breeders	30,000 broilers	16 farrowing sows with piglets, and 240 dry and pregnant sows
Maximum airflow (m^3 h^{-1})	4,000	4,300	45,000 (3 scrubber units; 15,000 m^3 h^{-1} each)	75,000	14,500
Packing type	Structured packed bed	Structured packed bed	Structured packed bed	Structured packed bed	Stack of vertical ion-exchange fiber cloths, directed parallel to airflow; surface area of 250 m^2 m^{-3} (125 m^2 m^{-3} for each surface)
Specific surface area (m^2 m^{-3})	100	100	100	150	Not applicable
Packing volume (m^3)	1.0	0.6	NA	8.6	1.6
Minimum EBRT (s)[a]	0.9	0.5	NA	0.4	0.4
Max. superficial air velocity (m s^{-1})[b]	1.7	1.8	NA	NA	2.5
Flow configuration	Cross-current	Cross-current	Cross-current	Counter-current	Cross-current
Water recirculation	Continuous	Continuous	Continuous	Continuous	Intermittent; 2 min on, 18 min off
Maximum gas-to-liquid ratio	NA[c]	NA	NA	NA	NA
pH	1.3 to 4.4	4 (setpoint)	4 (setpoint)	3 to 5	0 to 3.5
Acid used	Sulfuric acid (96%)	Sulfuric acid (96%)	Sulfuric acid (96%)	Sulfuric acid (96%)	Sulfuric acid (96%)
Discharge water control	Time	Time	Time	Electrical conductivity	pH
NH_3 Measurement					
Measurement period	45 days	100 days	2 times 2 months (age 21 to 32 weeks and 42 to 50 weeks)	79 days	69 days
Measurement frequency	Continuous	Continuous	Continuous	Continuous	Continuous
Method	Inlet: photoacoustic gas analyzer; outlet: impinger, sampling time 3.5 days	Inlet: photoacoustic analyzer and chemiluminescence method; outlet: impinger, sampling time 3.5 days	Impinger, sampling time 3.5 days	Impinger, time-averaged over 3.5 days	Impinger, time-averaged over 3.5 days
Number of measurements	37	89	100	19	20
Average airflow (m^3 h^{-1})	2,200	1,600	18,900	48,000	7,300
Average inlet conc. (mg m^{-3})	5.7	10.9	20.1	13.1	7.7
Average removal efficiency (%)	91	99	90 (SEM = 0.91)[d]	95 (SEM = 1.5)	98 (SEM = 0.20)
Minimum removal efficiency (%)	77	90	40	76	96
Maximum removal efficiency (%)	97	100	99	100	100

[a] EBRT = empty bed air residence time.
[b] Based on an empty bed.
[c] NA = not available.
[d] SEM = standard error of the mean.

emission reduction of >90%. From table 2, it is concluded that acid scrubbers can meet this target.

A minimum water discharge rate is required to prevent unwanted precipitation of ammonium sulfate in the system; the ammonium sulfate concentration is usually controlled at a level of about 150 g L^{-1}, which is about 40% of the maximum solubility. At an NH_3 removal efficiency of 95%, the discharge water production is about 0.2 m^3 kg NH_3 $removal^{-1}$ $year^{-1}$, which equals a yearly amount of 70 L growing-finishing pig $place^{-1}$ or 2 L broiler $place^{-1}$.

ODOR REMOVAL

Odor is a mixture of many different volatile compounds. Besides NH_3, the main odor components in exhaust air from animal houses are volatile fatty acids, p-cresol, indole, skatole, and diacetyl (Aarnink et al., 2005). The efficiency of odor removal by an acid scrubber is the result of dissolution of the odorous compounds in the water phase and the water discharge rate. As the water solubility of odorous compounds may vary from very low to very high, odor removal efficiencies vary as well.

The odor removal efficiency of acid scrubbers was measured at two farm locations, as described in table 3 (measurements taken before 1995 are omitted because a different measurement protocol for odor concentration was in use, and the results cannot be converted to OU_E). The average odor removal efficiency of the acid scrubbers is 29% and 34%, respectively. These efficiencies are much lower than for NH_3, as most odorous compounds are not captured by the acid, as is the case with NH_3. The variation in the odor removal is high, with a minimum removal efficiency of 3% and a maximum of 51%. Hahne and Vorlop (2001) reported a higher average odor removal efficiency (45%, SEM = 8.4) for an experimental pilot-scale acid scrubber treating pig house exhaust air at EBRT from 1.4 to 2.4 s. However, the number of odor measurements was limited (n = 5), and the scrubber size (bed volume = 0.5 m^3) was much smaller than the scrubbers that are listed in table 3. The air sampling method used by these authors also differed from the method described above, as they used no dust filter for air sampling, in accordance with the protocols used in Germany. Because part of the odor may be associated with the presence of dust,

Table 3. Characteristics and results of measurement programs on odor removal by full-scale acid scrubbers and biotrickling filters treating exhaust air of animal houses.

	Acid Scrubber		Biotrickling Filter		
	Ogink and Lens, 2001; Hol and Ogink, 2005	Klarenbeek et al., 1998	Melse and Mol, 2004[a]	Mol and Ogink, 2002; Hol and Ogink, 2005	
Design characteristics					
Source of exhaust air	250 growing-finishing pigs	Layer breeders	Growing-finishing pigs	Pregnant sows	560 growing-finishing pigs
Maximum airflow (m^3 h^{-1})	15,000	15,000	20,000	NA	48,000
Packing type	NA[b]	Structured packed bed	Vertical bundle of plastic tubes (4 cm diameter)	NA	Reticulated polyurethane foam
Specific surface area (m^2 m^{-3})	100	100	NA	NA	500
Packing volume (m^3)	2.5	NA	3	NA	30
Minimum EBRT (s)[c]	0.6	0.6	0.5	NA	2.3
Max. superficial air velocity (m s^{-1})[d]	NA	NA	2	NA	0.5
Flow configuration	Cross-current	Cross-current	Counter-current	NA	Counter-current
Water recirculation	Continuous	Continuous	Continuous	NA	Continuous
Maximum gas-to-liquid ratio	NA	NA	NA	NA	NA
pH	4 (setpoint)	4 (setpoint)	7.3 to 7.6	NA	NA
Discharge water control	Time	Time	Time[e]	NA	Time
Odor measurements					
Measurement period (days)	186	165	72	238	125
Measurement frequency	Incidental	Incidental	Incidental	Incidental	Incidental
Number of measurements	10	10	15	10	10
Average airflow (m^3 h^{-1})	NA	NA	NA	NA	NA
Average inlet concentration (OU_E m^{-3})	3,200	560	1,600	1,500	3,600
Average removal efficiency (%)	29 (SEM = 5)[f]	34 (SEM = 5)	49 (SEM = 8)	48 (SEM = 11)	37 (SEM = 7)
Minimum removal efficiency (%)	3	15	−29	−24	−10
Maximum removal efficiency (%)	50	51		83	63

[a] The NH_3 removal by this biotrickling filter is presented in table 4.

[b] NA = not available.

[c] EBRT = empty bed air residence time.

[d] Based on an empty bed.

[e] The high NO_2^- and NO_3^- content of the trickle water indicated that no water had been discharged for a long time, however. The accumulation of $N\text{-}NO_2^-$ and $N\text{-}NO_3^-$ in the trickle water during the measurement period equaled the removal of $N\text{-}NH_3$ from the air.

[f] SEM = standard error of the mean.

and a large part of the dust is removed in a scrubber, using a dust filter for air sampling, as is the case for the measurements in table 2, may decrease the apparent odor removal efficiency of a scrubber because the odor concentration of the inlet air could be underestimated. However, recent investigations (Willers, 2005) indicate that using a dust filter hardly influences the odor measurements because dust particles are removed from the sampled air, by settling in and attachment to the sample bags and tubing, prior to the olfactometrical analysis.

Installation of a scrubber increases electricity use, with about 57 W per 1,000 $m^3 h^{-1}$ of installed ventilation capacity or about 50 kWh per growing finishing pig place per year (Vrielink et al., 1997). Currently, some scrubber manufacturers claim to have drastically reduced the extra energy use associated with air scrubbing by optimization of the humidification system and by reduction of the pressure drop over the scrubber.

In The Netherlands, about 160 acid scrubbers are in operation for treatment of exhaust air from pig and poultry houses (Melse and Ogink, 2004); four companies are manufacturing these scrubbers. In Germany and Denmark, acid scrubbers of similar design are manufactured for this application.

EXPERIENCES WITH BIOTRICKLING FILTERS

AMMONIA REMOVAL

In a biotrickling filter, the reaction equilibrium of equation 1 shifts to the right as the dissolved NH_3 is removed by bacterial conversion. The bacterial population, or biomass, in the system grows as a film on the packing material and is suspended in the water that is being recirculated. The dissociated NH_3 is available for bacterial oxidation to nitrite (NO_2^-) and subsequently from nitrite to nitrate (NO_3^-). This oxidation process is called nitrification and is mainly carried out by *Nitrosomonas* and *Nitrobacter* species, respectively (Focht and Verstraete, 1977; Prosser, 1986). Equations 2 and 3 describe these processes:

$$NH_4^+ (aq) + OH^- (aq) + 1.5\, O_2 (g) \rightarrow NO_2^- (aq) + H^+ (aq) + 2\, H_2O (l) \qquad (2)$$

$$NO_2^- (aq) + H^+ (aq) + 2\, H_2O (l) + 0.5\, O_2 \rightarrow NO_3^- (aq) + H^+ (aq) + 2\, H_2O (l) \qquad (3)$$

A minimum water discharge rate is required to prevent unwanted accumulation of nitrogen in the system, as both free NH_3 and free nitrous acid (HNO_2^-) inhibit the nitrification process (Anthonisen et al., 1976). A well-designed and stable biotrickling filter is in a steady-state condition, which means there is an equilibrium between the processes shown in equations 1 through 3 and the amount of nitrogen and H^+ that is removed from the system by water discharge. This normally results in the following conditions for the recirculation water (Scholtens, 1996): $6.5 < pH < 7.5$, $1 < [N\text{−total}] (g\ L^{-1}) < 4$, and $0.8 < [NH_4^+]/[NO_2^- + NO_3^-] < 1.2$ on a molar basis.

A long-term measurement program that was carried out at six farm locations showed average NH_3 removal efficiencies ranging from 35% to 90%, with an overall mean of 70%. Table 4 shows the results and the characteristics of the investigated biotrickling filters. These results compare with average NH_3 removal efficiencies in a range from 54% to 73% that were reported by others for both a full-scale operated (Schirz, 2004, as cited by Van Groenestijn and Kraakman, 2005) and experimental pilot-scale biotrickling filters treating pig house exhaust air (Dong et al., 1997; Hahne and Vorlop, 2004). However, considerably lower average NH_3 removal efficiencies of 22% to 36% were reported by Lais (1996) for three experimental biotrickling filters (bed sizes from 2.2 to 18.1 m^3). Current Dutch regulations require that implemented biotrickling filters achieve an average NH_3 emission reduction of >70%. From table 4, it is concluded that biotrickling filters can meet this target but occasionally do not due to inadequate process control.

The discharge water from a biotrickling filter results in a yearly discharge water production of 790 L growing-finishing pig $place^{-1}$ or 25 L broiler $place^{-1}$ at an average nitrogen content of 2 g L^{-1}. This amount of discharge water is about 10 times higher than for an acid scrubber.

ODOR REMOVAL

In a biotrickling filter, a microbial community is present that comprises, besides nitrifying bacteria, bacteria that use odorous compounds as a substrate. As for acid scrubbers, the first step in odor removal by a biotrickling filter is dissolution of the odorous compounds in the water phase. In the second step, bacterial conversion of some or all of these compounds takes place, which results in odor removal. Either the first step of mass transfer from gas to liquid phase or the second step of bacterial conversion may be rate limiting. Low water solubility of compounds results in low concentrations in the biofilm and thus low conversion rates (Deshusses and Johnson, 2000). More information on biological treatment of waste air can be found for example in reviews by Van Groenestijn and Hesselink (1993), Kennes and Thalasso (1998), and Burgess et al. (2001).

The odor removal efficiency of biotrickling filters was measured at three farm locations (table 3). The average odor removal efficiency of the three biotrickling filters was 44%. The variation of the odor removal was high, with a minimum removal efficiency of −29% and a maximum of +87%. Higher average odor removal efficiencies for biotrickling filters treating pig house exhaust air were found by others. Lais (1996) found average odor removal efficiencies of 61% (SEM = 9.3), 89% (SEM = 2.3), and 85% (SEM = 1.1), respectively, for three experimental biotrickling filters with respective bed sizes of 2.2, 3.6, and 18.1 m^3 and minimum EBRTs of 0.5, 0.4, and 2.2 s. Schirz (2004), as cited by Van Groenestijn and Kraakman (2005), reported an average odor removal efficiency of 84% (SEM = 2.7) for a full-scale biotrickling filter (bed volume = 17.5 m^3) treating pig house exhaust air with a minimum EBRT of 1.35 s. The presence of appropriate process control, thus preventing accumulation of NH_3 and nitrite (NO_2^-) in the system, might explain why these authors found higher average odor removal efficiencies than in this study. It is well known that nitrifying bacteria are inhibited by accumulation of NH_3 and NO_2^- (Anthonisen et al., 1976). However, the bacterial population responsible for the removal of other odor compounds might also be inhibited by accumulation of NH_3 and/or NO_2^-.

Table 4. Characteristics and results of measurement programs on ammonia (NH_3) removal by full-scale biotrickling filters treating exhaust air of animal houses.

	Scholtens et al., 1988		Van de Sande-Schellekens and Backus, 1993a; Uenk et al., 1993a			Van Middelkoop, 1995	Melse and Mol, 2004[a]
Design characteristics							
Animal category and number	80 growing-finishing pigs	60 veal calves	63 growing-finishing pigs	63 growing-finishing pigs	160 growing-finishing pigs	4,950 broiler breeders	Growing-finishing pigs
Maximum airflow ($m^3\ h^{-1}$)	6,000	8,000	6,000	6,000	18,000 (2 scrubber units; 9,000 $m^3\ h^{-1}$ each)	48,000 (6 scrubber units; 8,000 $m^3\ h^{-1}$ each)	20,000
Packing type	Randomly packed bed	Randomly packed bed	Randomly packed bed	Randomly packed bed	Structured packed bed	Randomly packed bed	Vertical bundle of plastic tubes (4 cm diameter)
Specific surface area ($m^2\ m^{-3}$)	125	125	125	125	170	125	NA
Packing volume (m^3)	0.8	1.1	0.9	0.8	5.4	6.7	3
Minimum EBRT (s)[b]	0.5	0.5	0.5	0.5	1.1	0.5	0.5
Max. superficial air velocity ($m\ s^{-1}$)[c]	1.1	1.4	1.4	1.1	1.1	1.4	2
Flow configuration	Cross-current	Cross-current	Counter-current	Cross-current	Counter-current	Cross-current	Counter-current
Water recirculation	Continuous	Continuous	Continuous	Continuous	Continuous	Continuous	Continuous
Maximum gas-to-liquid ratio	2500	2400	NA	NA	NA	NA	NA
pH	6.8 to 8.3	6.6 to 7.8	6.2 to 8.5	6.5 to 9[d]	6.5 to 8.7	NA	7.3 to 7.6
Discharge water control	Time	Time	Time	Time		Time	NA
NH_3 measurements							
Measurement period	4 months	8 months	20 months	20 months	20 months	4 weeks	16 days
Measurement frequency	Incidental	Incidental	NA[e]	NA	NA	NA	Incidental
Method	Impinger, sampling time 20 min	Impinger, sampling time 20 min	Impinger	Impinger	Impinger	NA	Impinger, sampling time 2 h
Number of measurements	18	43	31	29	42	NA	8
Average airflow ($m^3\ h^{-1}$)	2,725	5,056	NA	NA	NA	NA	9,000
Average inlet conc. ($mg\ m^{-3}$)	4.8	4.4	NA	NA	NA	NA	4.3
Average removal efficiency (%)	65[f] (SEM = 6.7)[g]	35[h] (SEM = 3.6)	78	65	90	83[i]	79 (SEM = 5.7)
Minimum removal efficiency (%)	11	−8	33	5	26	NA	44
Maximum removal efficiency (%)	94	82	100	98	99	NA	94

[a] The odor removal by this biotrickling filter is presented in table 3.
[b] EBRT = empty bed air residence time.
[c] Based on an empty bed.
[d] pH neutralization took place by addition of a slowly dissolving mineral containing $CaCO_3$ and MgO.
[e] NA = not available.
[f] At EBRTs > 0.7 s, the average removal efficiency increased from 65% to 80%, which indicates that the scrubber had been overloaded with NH_3.
[g] SEM = standard error of the mean.
[h] This relatively low NH_3 removal efficiency appeared to be caused by inhibition of nitrifying bacteria due to accumulation of NH_3 and/or NO_2^-, as was indicated by analysis of the discharge water.
[i] The average NH_3 removal was measured separately for each of the six scrubber units and ranged from 78% to 88% for the individual units.

The increase in energy use of a biotrickling filter installation is generally the same as for an acid scrubber, with about 57 W per 1,000 $m^3\ h^{-1}$ of installed ventilation capacity or about 50 kWh per growing finishing pig place per year (Vrielink et al., 1997). Currently, some scrubber manufacturers claim to have drastically reduced the extra energy use associated with air scrubbing by optimization of the humidification system and by reduction of the pressure drop over the scrubber.

Table 5. Investment and operational costs[a] of acid scrubber and biotrickling filter for ammonia (NH_3) removal for newly built production facility (\$ animal place^{-1}, excluding value-added tax) (Melse and Willers, 2004).

	Acid Scrubber 95% NH_3 Removal		Biotrickling Filter 70% NH_3 Removal	
Cost category	Growing-Finishing Pigs	Broilers	Growing-Finishing Pigs	Broilers
Investment costs[b]	42	1.3	45	1.45
Operational costs (year^{-1})				
Depreciation (10%)	4.16	0.13	4.54	0.15
Maintenance (3%)	1.25	0.04	1.36	0.04
Interest (6%)	1.25	0.04	1.36	0.04
Electricity use (\$ 0.11 kWh^{-1})	5.50	0.18	5.50	0.18
Water use (\$ 1.0 m^{-3})	0.48	0.02	1.52	0.05
Chemical use (\$ 0.6 L^{-1} H_2SO_4, 98%)	2.18	0.07	n/a[c]	n/a
Total operational costs (year^{-1})[d]	14.82	0.47	14.29	0.46

[a] Excluding possible water discharge costs.

[b] The investment costs for growing-finishing pigs are based on a maximum ventilation capacity of 60 m^3 animal place^{-1} h^{-1}. For broilers, all costs are calculated using the ratio between the yearly NH_3 emission rates of broilers and growing-finishing pigs (see table 1).

[c] n/a = not applicable.

[d] The total operational costs of a scrubber increase the production costs per animal with 4% for Dutch conditions.

In The Netherlands, about 45 biotrickling filters are in operation for treatment of exhaust air from pig and poultry houses (Melse and Ogink, 2004); four companies are manufacturing these filters. In Germany and Denmark, biotrickling filters of similar design are manufactured for this application.

COSTS OF AIR SCRUBBING

Melse and Willers (2004) calculated the investment and operational costs of both acid scrubbers and biotrickling filters for treatment of exhaust air of newly built animal production facilities (table 5), based on recent quotations from manufacturers.

Table 5 shows that the operational costs of an acid scrubber with 95% NH_3 removal equal those of a biotrickling filter with 70% NH_3 removal, as long as the disposal costs of the discharge water are not taken into account. However, the amount and characteristics of the discharge water differ greatly, as has been described above. The local situation determines what costs, if any, are charged for the disposal of discharge water.

GENERAL DISCUSSION

AMMONIA REMOVAL

From the results presented in table 2, it can be concluded that acid scrubbing significantly reduces the NH_3 emission from animal houses (average removal efficiency >90%). As long as the pH in the scrubber is low and the water discharge flow has been set high enough, the NH_3 removal efficiency is guaranteed. Both pH and water discharge rate can be automatically controlled relatively simply with standard equipment, so that acid scrubbing can be considered as a stable and reliable measure for NH_3 emission reduction.

The NH_3 removal by biotrickling filters is significantly lower (removal efficiency on average 50% to 90%) than for acid scrubbers. Analyses of the discharge water indicated that decreased NH_3 removal efficiencies was caused by inhibition of nitrifying bacteria due to high NH_3 and/or nitrite concentrations. Usually this situation develops when the biotrickling filter is overloaded or when the discharge water flow rate is set too low. Because biotrickling filters produce a relatively large amount of discharge water, about 10 times as much as for an acid scrubber, in some cases manufacturers or end-users tend to decrease the discharge flow in order to reduce water disposal costs. Current disposal costs in The Netherlands are about \$ 7.50 per m^3 if application on the user's own land is not possible, which is very often the case, but the discharge water must be applied to arable land of a third party. In order to guarantee successful NH_3 removal, process control and monitoring of biotrickling filters need to be improved. This might be done by the installation of an electrical conductivity (EC) meter that controls the water discharge flow rate; accumulation of salts can be noticed and prevented in this way.

ODOR REMOVAL

Both acid scrubbers and biotrickling filters are capable of odor removal, with an average removal efficiency of 27% and 43%, respectively, as shown in table 3. It is striking, however, that individual odor removal efficiency measurements strongly vary, namely, from −66% to +87%.

Melse and Mol (2004) investigated the possible effects of the relatively large measurement error of the olfactometric method on the total variation of the odor removal efficiency that was found between consecutive measurements for one scrubber. Calculations showed that the olfactometric method contributes about 20% to the total variance of the odor removal efficiency measurements, whereas the actual performance of the scrubber system contributes about 80%. Hence, the main variation in removal efficiency between consecutive measurements of one scrubber is caused by real performance differences.

Another explanation for the varying odor removal performance might be that changes in the odor composition are not fully reflected in odor concentration values. Odor removal is the sum of the removal of many separate odor components that all have different characteristics with regard to mass transfer from gas to liquid phase and biodegradability. If, at a constant odor load, the concentration of an easily removable odor component increases in comparison with the other odor components in the air, then the measured odor removal efficiency will increase. If, on the other hand, an odor component is difficult to remove, then a relative increase of this component will result in a decrease of the measured removal efficiency at the same odor load (Melse and Mol, 2004).

Finally, odor compounds can be produced inside a biotrickling system. In a biotrickling filter, the products of (partial) conversion of organic odor compounds can negatively affect the measured odor removal efficiency. This might explain the negative odor removal efficiencies that were sometimes measured for the biotrickling filters. As no individual odor components were identified in the presented studies, this hypothesis cannot currently be verified.

HOW TO INCREASE ODOR REMOVAL

The odor removal efficiency of air scrubber systems might be improved by adjustment of design and operational strategy. It is noted that the current design of acid scrubbers and biotrickling filters has been optimized for the removal of NH_3 only and that the removal of odor has been considered as an unintentional, but welcome, circumstance until now. Removal of poorly water-soluble odor components might be improved by addition of an organic solvent to the water phase, which would increase the availability of the odor component to the bacteria and thus increase biodegradation rates (e.g., Césario, 1997; Van Groenestijn and Lake, 1999; Davidson and Daugulis, 2003). An increase of the air residence time usually improves the uptake of odor components as well but also means higher investment and operational cost per volume of air treated.

Another possibility, after having passed the air through the acid scrubber or biotrickling filter first, is to pass the air through a biofilter. Although biofilters have been extensively tested for treatment of exhaust air from animal operations in The Netherlands (Scholtens et al., 1988; Asseldonk and Voermans, 1989; Eggels and Scholtens, 1989; Van de Sande-Schellekens and Backus, 1993b; Demmers and Uenk, 1996; Uenk et al., 1993b), they are not considered suitable for long-term treatment of exhaust air that is directly drawn from an animal house because this air has relatively high dust and NH_3 concentrations. The filter bed, normally a mixture of materials such as compost, wood bark, wood chips, peat, perlite, and organic fibers, usually suffers from clogging and preferential flow paths by accumulation of dust, quick acidification by nitric acid accumulation, and problems related with inhomogeneous humidification. However, if most of the NH_3 has been removed from the air by an acid scrubber or biotrickling filter first, then a biofilter is an efficient measure for further odor reduction.

Besides biofiltration, other techniques are available for polishing air, such as oxidative treatment with ozone, hydrogen peroxide, and ultraviolet radiation; however, due to the large exhaust airflows of animal houses, these techniques are considered economically unfeasible.

Finally, the odor removal performance of a biotrickling filter might be improved by appropriate process control and monitoring, as accumulation of NH_3 and NO_2^- in the system might not only inhibit the nitrifying bacteria but also the bacterial population responsible for the removal of other odor compounds.

CLEANING

A well-designed scrubber usually has an average pressure drop of about 50 Pa and a pressure drop of about 200 Pa at the maximum airflow rate. Ventilation air of animal facilities contains dust that accumulates in the scrubber and causes unwanted channeling of air and an increase of pressure drop. Total dust concentrations in exhaust air from pig and poultry houses are about 2.42 mg m^{-3} and 4.05 mg m^{-3}, respectively (Takai et al., 1998). In the case of a biotrickling filter, the accumulation of solids is further increased by bacterial growth as time passes. Although some solids will be removed from the system with the discharge water, both the packing of a scrubber and the buffer tank usually need to be cleaned once or twice a year to prevent clogging of the bed.

CONCLUSION

The following conclusions can be drawn from this review:

- Acid scrubbers showed average removal efficiencies of 91% to 99%. Process control with pH measurement and automatic water discharge appeared to be sufficient to guarantee sufficient NH_3 removal.
- Biotrickling filters showed average NH_3 removal efficiencies from 35% to >90%. It appears that process control should be improved to guarantee sufficient NH_3 removal.
- Acid scrubbers and biotrickling filters showed lower removal efficiencies for odor, with average odor removal efficiencies of 27% and 43%, respectively.

Suggestions for further research are:

- Development of a reliable and economically feasible system for process control of a biotrickling filter. Such a system might include electrical conductivity (EC) measurement.
- Further improvement of the odor removal capacity of air scrubbers.
- Analysis of the large performance differences in odor removal efficiency, which were found for both acid scrubbers and biotrickling filters, by combining olfactometric methods using a human panel with advanced analyses of individual compounds by gas chromatography − mass spectrometry (GC-MS).

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