Aus dem Institut für Technologie und Biosystemtechnik

Jochen Hahne
Klaus-Dieter Vorlop

Treatment of waste gas from piggeries with nitrogen recovery

Manuskript, zu finden in www.fal.de

Published in: Landbauforschung Völkenrode 51(2001)3, pp. 121-130

Braunschweig
Bundesforschungsanstalt für Landwirtschaft (FAL)
2001

Treatment of waste gas from piggeries with nitrogen recovery

Jochen Hahne and Klaus-Dieter Vorlop [1]

Abstract

A piggery waste air treatment process with a combination of a wet scrubbing with sulfuric acid and a subsequent biofiltration is described. The scrubber liquid is concentrated up to the chemical saturation grade of ammonium sulfate. Trace gases as ammonia, nitric oxides, nitrous oxide, methane, carbon monoxide, carbon dioxide, hydrogen sulfide as well as total organic carbon were measured in a two-year period under practical conditions at different scrubber and biofilter gas loading rates. Above that the composition of the scrubber product and some analyses concerning the biofilter irrigation water and the biofilter material are presented. Ammonia is separated with an efficiency of at least 78 % during scrubbing and amounts to 93 % in the total process at least. The odour reduction efficiency is depending on both the raw gas odour concentration and the biofilter loading rate. At raw gas odour concentrations of more than 915 OU/m³ the total process odour reduction amounted to 85 % and more, even at biofilter loading rates of 220 m³/(m² h). Lower odour concentrations in the raw gas resulted in a lower odour reduction because of self odour emissions from the biofilter material. Above that the hedonic odour tone is significantly improved by the process. The high automation grade causes an electricity consumption of at least 2 kWh/1000 m³ waste gas. Around 11 kg of sulfuric acid are necessary to absorb the annual ammonia emissions from one pig place. The ammonium sulfate production amounts from 30 to 40 l per pig place and year. Apart from this liquid fertilizer there is no waste or waste water production.

Keywords: Biofilter, scrubber, waste air, treatment, odour, ammonia, trace gases, piggery

Zusammenfassung

Behandlung von Abluft aus Mastschweineställen mit Stickstoffrückgewinnung

Es wird ein Abluftreinigungssystem, bestehend aus einer Kombination einer Nasswäsche mit Schwefelsäure und einer nachgeschalteten Biofiltration, beschrieben. Die Wäscherflüssigkeit wird bis zur chemischen Sättigungsgrenze für Ammoniumsulfat konzentriert. Spurengase wie Ammoniak, Stickoxide, Lachgas, Methan, Kohlenmonoxid, Kohlendioxid, Schwefelwasserstoff sowie Gesamtkohlenstoff wurden über zwei Jahre unter praktischen Bedingungen bei verschiedenen Wäscher- und Biofiltergasbeladungen gemessen. Darüber hinaus werden einige Analysenergebnisse von der Wäscherflüssigkeit, dem Biofilterberegnungswasser sowie Biofiltermaterialanalysen vorgestellt. Ammoniak wird bei der Abluftwäsche mit einem Wirkungsgrad von mindestens 78 % und im Gesamtprozess mit mindestens 93 % abgeschieden. Der Geruchsminderungsgrad ist abhängig von der Geruchstoffkonzentration im Rohgas und der Biofiltergasbeladung. Bei Rohgaskonzentrationen von mehr als 915 GE/m³ betrug der Geruchsminderungsgrad des Gesamtverfahrens 85 % und mehr, auch bei Biofiltergasbeladungen von 220 m³/(m² h).Geringere Geruchstoffkonzentrationen im Rohgas führten wegen des Eigengeruchs des Biofiltermaterials zu sinkenden Wirkungsgraden. Darüber hinaus wird die hedonische Wirkung der Abluft durch die Abluftbehandlung signifikant verbessert. Der hohe Automatisierungsgrad führt zu einem Mindestverbrauch an elektrischer Energie in Höhe von 2 kWh/1000 m³ Rohgas. Etwa 11 kg konzentrierte Schwefelsäure sind erforderlich, um die Ammoniakemissionen eines Schweinemastplatzes im Jahr zu absorbieren. Die Ammoniumsulfatproduktion beträgt zwischen 30 und 40 l je Mastplatz und Jahr. Neben diesem Flüssigdünger werden weder Abfälle noch Abwässer produziert.

Schlüsselworte: Biofilter, Wäscher, Abluft, Behandlung, Geruch, Ammoniak, Spurengase, Schweinehaltung

[1] Institute for Technology and Biosystems Engineering of the Federal Agricultural Research Centre (FAL), Bundesallee 50, 38116 Braunschweig, Germany

1 Problems and objectives

The number of animals in Germany amounted to 14.9 million cattle, 26.2 million pigs and 103 million poultry in 1999 [1]. Intensive animal production results in comparatively low prices for the consumers but causes environmental problems also. This applies to fact that especially the extent of pig and poultry husbandry is not orientated to the minimum area per livestock unit. One of the essential environmental problems of this kind of livestock is the local manure surplus production. The surplus manure application does not fit to the plant requirements and consequently causes problems as phosphate, zinc and copper accumulation in soils and leaching of potassium and nitrate into the groundwater. Another considerable problem of animal husbandry is the emission of ammonia, odourants and dust. Ammonia puts pressure on the environment in variied forms by its acidification and nutrient potential with far reaching consequences. The acidification potential is based on the microbial oxidation of ammonia to nitrite and nitrate. This nitrification process leads to a decrease of the pH value in non or poor buffered ecosystems, which is the reason for a change of the phytocoenosis. Under extreme conditions these sensitive ecosystems may suffer a die off. The input of ammonia to ecosystems leads to an increased growing of biomass. Surplus ammonia inputs drive out nitrogen limited ecosystems as highmoors, oligotrophic seas and lean grass biotops. Beside this the eutrophication of oligotrophic seas results in an increased silt up whose aerobic degradation causes a corresponding oxygen requirement. Aerobic organisms may suffer a die off by a lack of oxygen. Nitrogen induced nutrient imbalances on the one hand and acidification by ammonia oxidation on the other are jointly responsible for damages to forests. Dust emissions from animal husbandries lead to affections of the human and animal lung function, in some cases, to diseases of the pig farmers respiratory tract [2].

Concrete emission data for ammonia from livestock are available for ammonia [3]. In 1996 the ammonia emission amounted to 533 kt, which corresponds to 82 % of the German ammonia emission. The national nitrogen fertilizer consumption amounted to 1848 kt in 1996/97 and consequently the ammonia losses from livestock represent 24 % of the nitrogen requirement of the agricultural land. The accelerated change of structure in animal production aggrevates the situation in view of an environmental - friendly and sustainable animal husbandry. A view on the structure of animal production in Lower Saxony, Germany, illustrates the current development. In 1994 only 10.1 % of the pigs with a liveweight of more than 50 kg lived on farms with more than 1000 heads in Lower Saxony [4]. The portion of pigs in this farm scale was raised to 28.1 % in 1995 [5] and in 1999 more than 51 % of the pigs were kept on farms with more than 1000 heads [6]. Within five years the number of pigs on farms with more than 1000 heads increased nearly tenfold from 296,447 to 2,725,801. This change of structure, which reduces the production costs, was facilitated and accelerated by the national legislation.

There are a lot of strategies available to resolve the surplus nitrogen and acidification problems caused by livestock. Among other things the nitrogen losses are depending on species and number of animals, their production capacity (ratio of product per emission), the feeding strategy, the housing system, the manure storing and application system. Gaseous emissions of ammonia and other trace gases can be significantly reduced by modern manure application systems, by covering of storage tanks and also by the treatment of waste air from piggeries and poultry houses. The latter aspect becomes more important because of increasing odour nuisance complaints on the one hand and the current change of animal structure on the other. Although the animal-specific loss of gaseous nitrogen and trace gases may be lower in large piggeries than in small ones, the total release is significant. The ammonia losses of ventilated pig houses with manure storing beneath the slats amounted from 2.8 to 3.7 kg per fattening pig place and year [7]. Above that the emission of methane ranges between 2.8 and 4.5 kg per fattening pig place and year. With about 0.3 kg per fattening pig place and year the release of hydrogen sulfide can‘t be ignored [7]. The average and extrapolated gaseous ammonia loss of a pig house with 2,000 heads is theoretically sufficient for the nitrogen requirement of 35 hectars arable land in view of the German fertilizer guideline. Together with the manure nitrogen (1.5 m^3 manure per pig and year, total nitrogen concentration: 6 kg/m^3) the required area would raise to 141 hectars.

On the basis of this situation a waste air treatment system for piggeries has been realized. With the treatment process the following objectives should be fulfilled:

- Separation of dust as a carrier of endotoxins, microorganisms [8] and odourants,
- Separation of ammonia and recovery as a liquid fertilizer,
- Removal of typical livestock odourants and
- Degradation of other trace gases.

The process should not produce waste water or any wastes to avoid a secondary pressure on the environment.

2 State of the art

The treatment of waste air from husbandries is hardly used in practice. This situation is based on a lack of any emission limits for ammonia, dust, odourants or trace gases up to now. The approve of new animal husbandries is essentially determined by the keeping of minimum distances to the next residential area to keep the environmental precaution principle. These minimum distances have been fixed after some hundreds of farm inspections and are depending on the animal species and their numbers.

Beside this some other measures (manure storage capacity of six month at minimum, use of standardized ventilation systems) have to be fulfilled. Waste air treatment systems or other emission mitigating measures can only be demanded by the authorities if the minimum distances can not be kept. This often occures at expansions of the business.

The animal friendly use of standardized ventilation systems in pig houses requires air flow rates of 17 to 114 m³ per pig place and hour. This is necessary to keep the temperature and humidity levels within the range of the debit-values on the one hand and the indoor trace gas concentrations on the other. Waste gas from piggeries is, in so far, characterized by high and fluctuating air flow rates. These fluctuations are caused by a daily cycle as well as a yearly one and correspond essentially with the outdoor temperature shift. The air, which is warmed up in the stables, has a lack of humidity. The waste gas temperatures vary in a range from 16 to 32 °C. Above that the waste gas contains dust in mean concentrations between 0.38 mg/m³ (cattle), 2.19 mg/m³ (pigs) and 3.6 mg/m³ (laying hens) [8]. The dust concentrations in stables are depending on the general management (technical performance, cleanness), feeding technology (dry or liquid), housing system (littered or slatted floors) and the water management. Ammonia is a relevant compound in the waste gas from full slatted pig houses and occurs in concentrations between 4 and 30 ppm. Methane concentrations can be found in a similar range. Important is the fact that waste gas from piggeries contains comparatively low concentrations of easy biodegradable carbon sources and, in contrast to that, comparatively high concentrations of ammonia. Other trace gases are hydrogen sulfide (0 – 6 ppm), nitric oxides (0 – 2 ppm), nitrous oxide (0 – 3 ppm) and carbon monoxide (0 – 3 ppm) [9]. The odour concentrations, measured with the olfactometric method, normally range between 300 and 1500 odour units/m³.

Among of these basic conditions only a few technologies are available for the purpose of treating waste air from piggeries. Adsorption techniques with activated carbon are disadvantageous in so far that the recovery of nitrogen would need additional equipment (desorption and condensation). The incineration of loaded carbon would result in a loss of nitrogen. Biological operating systems are fundamentally useful for this waste gas treatment. Biofilters are kown to be a very successful and low cost systems to degrade odours in practice [10]. Problems arise if biofilters are loaded with an imbalanced nitrogen / carbon ratio. The waste gas is pressed or sucked through the biofilter bed and odourants as well as other waste air compounds are degraded by microorganisms which are immobilized on carriers as compost, peat or porous clay.

The use of biofilters for waste gas treatment from piggeries was investigated by Mannebeck and Hügle [11, 12]. The efficiency of odour and ammonia reduction was depending on the moisture of the organic biofilter material. Water contents of 48 to 62 % in the biofilter material result in an odour reduction efficiency of more than 50 %, while the ammonia reduction could be raised to 80 % at filter loading rates of about 1000 m³/(m² h) in a downstream process. Problems arise from the concentrating of microbial formed products as nitrites and nitrates in the biofilter. The authors concluded that the biofiltration is suited for this purpose if the reaction products will be removed from the biofilter. The use of this system is more likely limited by the operation costs. A more general problem is the maintenance of a regular air flow through the biofilter material, which significantly affects the biofilter efficiency.

Hartung et al. [13] measured odour reduction rates from 53 to 85 % at specific air flow rates of 808 - 814 m³/(m³ h). The biofilter material was a fresh mix of fibrous peat and coconut fibre. The ammonia reduction efficiency was negligible. It depends on the filter loading rate and high loading rates result in a breakthrough of ammonia. Hartung et al. concluded that biofiltration is – in relation to bioscrubbers – a economic and effective system only for odour reduction. In contrast to that ammonia separation is neither effective nor economic. Investment and operation costs of biofilters were estimated at 10 – 20 DM/fattening pig [11, (1993)] and 12 – 15 DM/fattening pig respectively [13, (1996)].

Siemers [14] investigated a combination of a water spraying system and a subsequent biofiltration to treat waste gas from piggeries. The dust separation efficiency amounted to 80 % and more and the odour reduction efficiency varied between 5.5 and 41 %. Ammonia was reduced about 90 % and more, if the biofilter material was irrigated with 50 l of water per square metre and day. But during fattening phase the ammonia reduction efficiency decreased. In the final fattening it was only 20 %.

Lais [15] investigated three different bioscrubbers in view of their use for waste gas treatment of piggeries. The fundamental principle of a bioscrubber is the separation of an absorption tower on the one hand and a aerated regeneration tank with active microorganisms on the other [16]. In trickle bed reactors absorption of pollutants and regeneration of polluted water take place simultaneously. If the absorption velocity is quite different from the microbial degradation velocity , the separation of both processes is useful. This situation applies to the absorption of ammonia and its microbial oxidation. Lais examined gas loading rates from 1200 to 11,000 m³ waste air per square metre packing material, the spinkling density was varied between 1.7 and 11.8 m³/(m² h). The pH values changed from 6.9 to 8.9. While the odour reduction efficiency was satisfying with 61 to 89 %, the ammonia reduction efficiency was low with 22 to 36 %. Lais concluded that the use of bioscrubbers for waste gas cleaning from piggeries can not be recommended because of the low ammonia reduction efficiency and the high treatment costs of more than 13 DM per fattening pig.

Chung et al. found that heterotrophic Arthrobacter oxydans, which was entrapped in calcium alginate, is well suited to degrade ammonia containing waste gas [17]. The maximum removal rate was 1.22 g of nitrogen per kg bead and day, the main products were nitrit (68 % of the removed ammonia) and organic bond nitrogen (25 %). The process could be maintained about 3 month by exchanging the basal medium periodically.

Investigations von Papen et al. showed that heterotrophic nitrification may result in a nitrous oxide production that is two times higher than with autotrophic ammonia oxidizers [18].

Kim et al. [19] tested the degradation of ammonia in a biofilter at loading rates from 2.4 – 22.5 g nitrogen per kg dry packing biofilter material (perlite) over a period of 61 days. They found a complete ammonia removal capacity of 18.6 g nitrogen per kg dry packing material and day in a lab biofilter after an inoculation with a marine and heterotrophic bacterium, Vibrio alginolyticus. This bacterium tolerates halophilic conditions and can live on several carbon sources as molasses, sucrose, fructose and glucose. The ammonia removal mechanism is not clear yet, but neither nitrite nor nitrate is formed.

Chung et al. [20] examined the ammonia oxidation in a biofilter with an autotrophic bacterium, Nitrosomonas europaea. It was entrapped in beads with calcium alginate. The maximum removal rate at ammonia inlet concentrations of 64 ppm was found to be 1.11 g nitrogen per kg filter bead and day. Main reaction products were nitrite (73 – 84 % of removed ammonia) and organic bond nitrogen (14 – 19 %).

Demmers [21] investigated the use of bioscrubbers for waste gas treatment from calf fattening. Ammonia was oxidized in the scrubber with a maximum velocity of 25 mg/(m² scrubber surface h). The main problem was the environmental use of the overflow with nitrate concentration of 2.5 g/l. Therefore Demmers took the denitrification and the water reuse into consideration. For denitrification processes easy degradable carbon sources are necessary that are not available in the waste gas.

Van Geelen and van der Hoek [22] found an odour reduction efficiency of 60 – 85 % in a simple bioscrubber during treatment of waste gas from poultry houses. Especially waste air odourants as phenol (43 μg/m³) and cresol (75 μg/m³) were degraded efficiently by microorganisms in the bioscrubber. Main problems during treatment were the clogging of spraying nozzles and the long time performance of the biomass in the scrubber. After a two month operation period the microorganisms in the scrubber were inactivated by unknown effects.

Backus et al. offered detailed costs calculations from three different bioscrubbers which were used for the waste gas treatment from piggeries [23]. The total costs varied between 42 and 54 DM per pig place and year without disposal costs of the overflow (1.3 m³ per pig place and year) which may change between 10 and 34 DM.

3 FAL waste gas treatment process

A sustainable animal production should not put pressure on the environment by high ammonia, dust and odour emissions. The treatment process, that avoids these environmental impacts, is a combined system consisting of a wet scrubber and a subsequent biofiltration (Fig. 1). During wet scrubbing with a diluted sulfuric acid solution ammonia gas is converted to liquid ammonium sulfate.

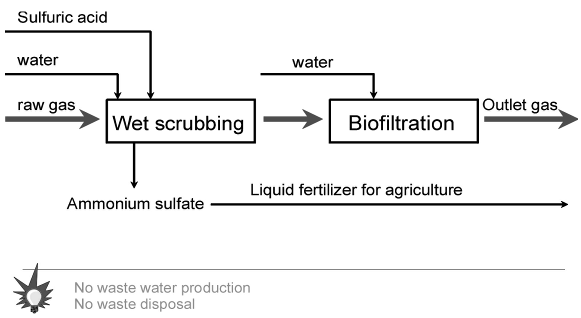

Fig. 1
FAL process for waste gas treatment from piggeries

Dust is separated from the waste gas and the humidity of the scrubber outlet gas is raised to saturation. Odourants, which are hardly soluble in water and therefore not separated during wet scrubbing, are oxidized during biofiltration. Problems as the release of greenhouse gases by nitrification/denitrification processes can be avoided strictly because ammonia is preseparated and concentrated in the wet scrubber. Because of the raw gas water steam deficiency and the according evaporation of scrubber liquid it is possible to concentrate ammonia to its saturation grade. The concentrated ammonium sulfate solution can be used as a combined nitrogen and sulfur liquid fertilizer.

The waste gas from the stable (max: 1500 m³/h) is sucked from the bottom to the top through the scrubber which consists of two separated units and tanks of 560 l each (Fig. 2). In opposite direction the washing liquid is sprayed on the plastic filling bodies by nozzles and finally collected in separate tanks. Each unit (diameter: 0.6 m) is filled with 0.25 m³ spheric plastic hollow bodies which have a specific surface area of 98.4 m²/m³. The sprinkling density can be varied between 7 and 15 m³/(m² h). The scrubber is working pH controlled. Concentrated sulfuric acid of technical quality is pumped into tank 2 so far a debit pH value is realized. From tank 2 the sulfuric acid solution is pumped into tank 1 by pump 3, when the pH of the scrubber tank 1 exceeds a debit value. Because of the raw gas water steam deficiency water from tank 1 is evaporated and consequently ammonium sulfate is concentrated. The liquid level of both tanks is controlled and regulated by four switches. At position max max both the fresh water supply and the pump P3 are fundamentally stopped. The position max is the upper and min the lower

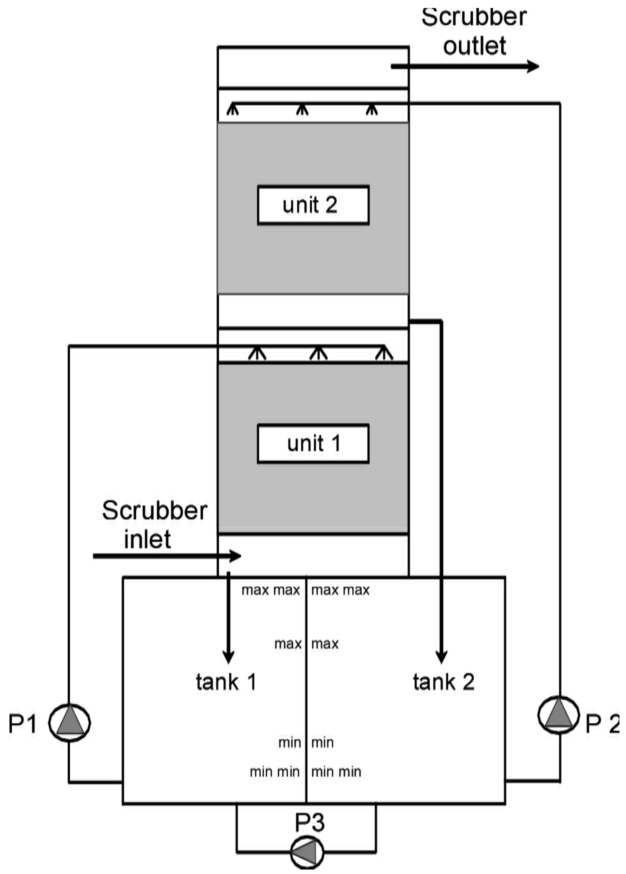

Fig. 2
Principle sketch of the wet scrubber used in the FAL waste air treatment process

fresh water filling level. At min min position in one tank the corresponding circulation pump (P1or P2) is stopped automatically controlled. The gas loading rates can be raised to 5400 m³/(m² h) at maximum which corresponds to an air velocity of 1.5 m/s.

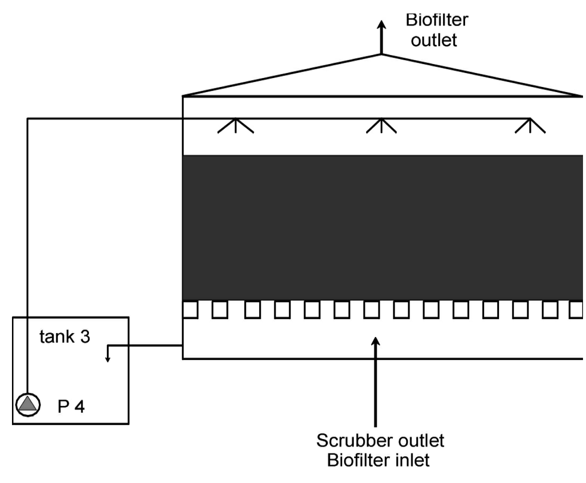

Fig. 3
Principle sketch of the biofilter used in the FAL waste air treatment process

The scrubber outlet gas is pressed through the closed biofilter unit (Fig. 3), which contains two layers of organic materials of 0.50 m each. For this purpose a ventilator with a frequency changer is used. While the lower layer (towards the biofilter inlet) consists of shreddered roots (length up to 30 cm, thickness: 2 – 4 cm) the upper layer consists of shreddered bark and wood chips (2 - 6 cm in diameter). The pressure drop of the biofilter ranges from 100 to 500 Pa and depends on the kind and the useful life of the biofilter material, particularly. Besides the humidification of the waste gas by passing through the scrubber, an additional water spraying is installed on the upper side of the biofilter material. Its use is necessary to compensate the evaporation losses by the ventilator caused gas warming up. Above that it is useful to moisten the material of the biofilter, which is located outdoor, during the warm summer month. The water spraying system works time controlled one or two times a day by the operation of pump P4. The reflux is collected and reused. Before pump P4 goes in operation, fresh water addition occurs automatically controlled and refills tank 3 from the min level to the max one. At position max max the fresh water addition is fundamentally stopped. If the water level falls to the min min position the pump is switched off automatically. With the upstream process the gas loading rate can be raised up to 300 m³/(m² h) which corresponds to minimum empty bed retention time of 12 s.

4 Analytical equipment

For determination of the gas composition a gas sampling system with 16 measuring points, a sample switch and different gas conditioning segments has been installed (Fig. 4). Besides oxygen, carbon dioxide and total organic carbon all relevant trace gases as ammonia, methane, carbon monoxide, nitrous oxide, nitric oxides (NO and NO_2) and hydrogen sulfide are measured quasi online. Any of the sixteen measuring points is sampled every three hours. After sampling the gas pipes and the sample switch are flushed with syntethic air and before next analysing the measuring points are presuckted with sample gas about five minutes. All measuring pipes are made of stainless

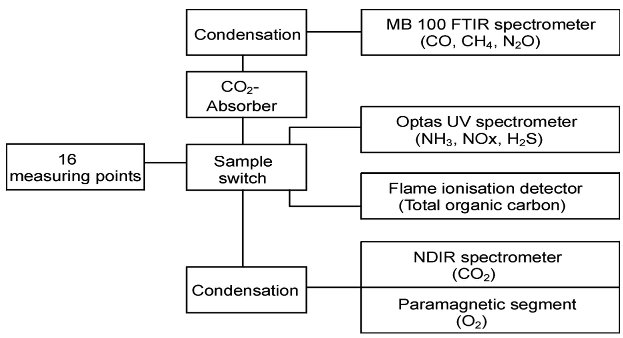

Fig. 4
FAL gas sampling and analyses system

Table 1
Raw and scrubber outlet concentrations of different trace gases at gas loading rates from 1,900 to 5,200 m³/(m² h) and a washing liquid pH value of 3.0

Scrubber Gas load [m³/(m² h]	Ammonia NH_3 [ppm]		Nitric oxides NO, NO_2 [ppm]		Nitrous oxide N_2O [ppm]		Total organic carbon*, Corg tot. [ppm]		Carbon monoxide CO [ppm]		Hydrogen sulfide H_2S [ppm]	
	In	Out	In	Out	In	Out	In	Out	In	Out	In	Out
1900	11.5	2.5	< 0.1	< 0.1	0.7	0.7	5.2	5.1	0.4	0.4	1.6	0.7
2200	5.0	0.8	< 0.1	< 0.1	0.1	0.1	2.1	2.0	< 0.1	< 0.1	0.2	0.3
2700	12.3	1.6	< 0.1	< 0.1	0.8	0.8	4.8	4.5	0.4	0.4	1.8	0.6
3100	9.5	0.2	0.6	0.1	0.6	0.6	4.8	5.1	0.3	0.3	0.4	0.4
3400	16.5	3.5	0.1	< 0.1	0.8	0.7	10.4	10.2	0.3	0.3	0.0	0.1
3700	7.8	0.6	< 0.1	< 0.1	0.6	0.6	3.1	2.9	0.4	0.4	0.2	0.2
3800	11.5	0.4	0.9	0.3	0.6	0.6	4.4	4.1	0.3	0.3	1.0	0.9
3900	14.6	2.7	< 0.1	< 0.1	0.7	0.7	10.3	10.1	0.3	0.3	< 0.1	0.2
4200	10.9	0.2	0.7	0.1	0.6	0.6	8.4	8.1	0.3	0.3	0.9	0.5
4300	14.9	0.9	< 0.1	< 0.1	0.7	0.7	4.4	4.2	0.4	0.4	1.9	0.4
4400	10.1	0.4	0.6	0.1	0.6	0.6	5.5	5.3	0.4	0.4	0.4	0.4
4500	12.6	0.3	< 0.1	< 0.1	0.6	0.6	11.2	10.7	0.4	0.4	0.6	0.4
5200	11.5	0.3	0.7	0.1	0.7	0.7	11.5	11.1	0.3	0.4	0.6	0.4

* including methane

steel, insulated and warmed up to 35 °C, permanently. For dust separation the measuring points are equipped with filter units. The pressure drop of each measuring point is controlled by a pressure gauge.

For the evaluation of the gas concentration values daily arithmetic means were calculated from the different gases. These daily arithmetic means were averaged equally to get results over the testing period.

Besides the gas analysis liquid samples are taken from the scrubber circulation pipes and from the biofilter water tank 3. Parameters as pH, electric conductivity, oxygen content, ammonia, nitrite, nitrate, organic nitrogen, COD (Chemical Oxygen Demand), BOD_5 (Biochemical Oxygen Demand in five days) and sulfate are analysed using the German standard methods for the examination of water, waste water and sludge [24]. The biofilter material is examined with regard to total solids, nitrogen fractions and sulfate.

5 Results

Scrubbing of piggery waste gas with a sulfuric acid solution at a pH value of 3.0 resulted, as expected, in a significant reduction of the scrubber outlet ammonia concentrations under practical conditions (Tab. 1). The efficiency of ammonia reduction was at least 78 %. Even at gas loading rates between 4200 and 5200 m³/(m² h) reduction efficiencies of more than 94 % could be maintained permanently at raw gas ammonia concentrations up to 15 ppm . The outlet ammonia mass flow was hardly affected by the raw ammonia mass flow under the tested conditions. If nitric oxides were detectable in higher concentrations in the raw gas, the scrubbing resulted in a reduction. Important is the fact that there was no production of nitric oxides during scrubbing on single time. Compared with the nitric oxides nitrous oxide concentrations were not increased during scrubbing. The efficiency of Total Organic Carbon (TOC) reduction (including methane) was low with less than 7 %. Carbon monoxide passed the scrubber unaffected. Concerning the reduction of hydrogen sulfide the scrubber effect is not quite clear. If high concentrations (> 1ppm) occured in the raw gas, the hydrogen sulfide reduction ranged between 56 and 79 %. At low concentrations the scrubber efficiency was negligible, sometimes negative in fact. It seems to be that apart from a slight reduction fluctuations of the raw gas concentrations were equalized in essence by the scrubber.

During biofiltration gas loading rates vom 19 up to 266 m³/(m² h) were investigated (Tab. 2). Ammonia that passed the scrubber in a concentration range from 0.2 to 3 ppm, was further reduced by biofiltration, as expected. Apart from biofilter loading rates of above 250 m³/(m² h) the outlet concentrations varied between 0 and 0.2 ppm. Nitric oxides passed the biofilter mainly unaffected, if the ammonia mass flow into the biofilter was low. Under oxygen limited conditions (water saturation of biofilter material, very low gas loading rates, process interruption) both the release of nitric oxides and nitrous oxide may raise significantly. The extent of this possible release is depending on the prior input of ammonia into the biofilter. Current investigations of an ammonia loaded biofilter show that at least 6 % of the ammonia nitrogen is converted to nitric oxides and more than 20 % results in nitrous oxide under oxygen reduced conditions. Without those limitations there was no increase of nitrous oxide concentrations during biofiltration (Tab. 2). TOC was hardly reduced by

Table 2
Biofilter inlet and outlet concentrations of different trace gases at loading rates from 19 to 266 m³/(m² h)

Biofilter Gas load	Ammonia NH_3 [ppm]		Nitric oxides NO, NO_2 [ppm]		Nitrous oxide N_2O [ppm]		Total organic carbon*, Corg $_{tot}$.[ppm]		Methane CH_4 [ppm]		Carbon monoxide CO [ppm]		Hydrogen sulfide H_2S [ppm]	
[m³/(m² h]	In	Out	In	Out	In	Out	In	Out	In	Out	In	Out	In	Out
19	0.2	< 0.1	0.1	1.1	0.5	0.5	3.0	1.1	10.9	4.6	0.3	0.2	0.3	< 0.1
30	0.2	< 0.1	< 0.1	< 0.1	0.7	0.7	8.7	8.3	22.1	20.9	0.4	0.3	0.5	0.3
34	0.4	0.1	< 0.1	0.1	0.6	0.6	10.7	10.3	21.7	20.9	0.4	0.3	0.4	0.1
38	2.9	0.2	< 0.1	< 0.1	0.7	0.7	10.1	9.9	24.5	24.5	0.3	0.1	0.2	< 0.1
41	0.4	0.1	< 0.1	< 0.1	0.7	0.7	11.0	10.6	n. d.	n. d.	0.4	0.2	0.4	0.2
78	0.6	0.2	< 0.1	< 0.1	0.6	0.6	5.3	5.3	9.9	9.9	0.4	0.3	0.4	0.3
87	0.6	0.2	< 0.1	0.1	0.6	0.6	5.3	5.3	9.8	9.5	0.4	0.3	0.4	0.3
92	0.6	0.2	< 0.1	< 0.1	0.5	0.5	2.7	2.6	4.9	4.6	0.3	0.3	0.2	0.1
154	2.9	0.2	< 0.1	0.2	0.7	0.7	10.1	9.8	24.5	24.2	0.3	0.2	0.2	< 0.1
220	0.4	0.2	< 0.1	< 0.1	0.6	0.6	10.7	10.3	21.7	19.8	0.4	0.4	0.4	0.2
245	0.2	< 0.1	< 0.1	< 0.1	0.7	0.7	8.6	8.1	22.0	20.6	0.3	0.3	0.5	0.4
266	1.1	0.8	0.2	0.2	0.7	0.7	7.6	7.4	8.9	8.9	0.4	0.3	0.7	0.5

* including methane

biofiltration, apart from extremely low filter loading rates (19 m³/(m² h), where a degradation of methane and other scarcely water soluble organic compounds occured. Measurements of methane concentrations verifyed a significant degradation of more than 50 % only at those extremely low biofilter gas loading rates. Higher onces resulted in a methane degradation efficiency of less than 10 %. Carbon monoxide, a trace gas of lower importance in this context, was reduced with an efficiency of 25 to 33 % in most cases. The concentration of hydrogen sulfide, an important odourant, was generally reduced by biofiltration. The extent of reduction was depending on the gas loading rate. While loading rates of less than 220 m³/(m² h) caused a hydrogen sulfide reduction of 50 % in most cases, higher loading rates resulted in a significant efficiency drop to less than 30 %.

The efficiency of the whole gas treatment process was investigated at scrubber loading rates between 2700 and 4600 m³/(m² h) and biofilter loading rates between 37 and 220 m³/(m² h) (Tab. 3). Odour concentrations, which were measured with an olfactometer (Mannebeck T 06), showed a wide variation between 445 and 2187 OU/m³ in the raw gas. The odour concentrations correlated slightly with the hydrogen sulfide and with the TOC concentrations, but not with the ammonia concentrations. Scrubbing of the waste gas with a sulfuric acid solution at a pH value of 3.0 resulted in an odour reduction efficiency in a range from 24 to 74 %, depending on the dust and the raw gas odour concentration. On the basis of current results there is no correlation between odour reduction efficiency and gas loading rate under the test conditions. Ammonia was reduced in a range from 77 up to 99 % by scrubbing. Apart from high nitric concentrations in the raw gas (0.7 ppm), which were reduced, nitric oxides and nitrous oxide passed the scrubber unaffected. While TOC was reduced slightly, carbon monoxide was not separated. Carbon dioxide and methane concentrations were not influenced by this kind of waste gas scrubbing. Hydrogen sulfide concentrations were only reduced at higher raw gas concentrations.

The biofiltration resulted in a further odour reduction (Tab. 3), which was particularly depending on the odour concentration and the biofilter loading rate. Best results could be achieved at high scrubber outlet concentrations. Loading rates of 40 m³/(m² h) resulted in a mean odour reduction efficiency of 86 % at mean scrubber outlet concentrations of 537 OU/m³, whereas loading rates of 220 m³/(m² h) led to mean odour reduction efficiencies of only 63 %. The measured odour concentrations did not correlate with any single trace gas concentration, apart from hydrogen sulfide, where a slight correlation existed. With ammonia preseparation biofiltration did neither cause a increase of nitric oxides nor nitrous oxide. The TOC-reduction during biofiltration was low with less than 6 %. In most cases there was a carbon monoxide degradation during biofiltration, but the efficiency varied between 7 and 67 %. Carbon dioxide concentrations were slighty increased about less than 100 ppm by biofiltration. Methane degradation was negligible with less than 5 %. Hydrogen sulfide was reduced with an efficiency of 50 % at least.

At raw gas odour concentrations of more than 915 OU/m³ the total process odour reduction amounted to 85 % and more, even at biofilter loading rates of 220 m³/(m² h). Lower odour concentrations in the raw gas resulted in lower odour reduction efficiencies of 71 % (688 OU/m³ in the raw gas) and 42 % (445 OU/m³ in the raw gas). One reason of dropping odour reduction efficiencies at lower

Table 3
Odour and trace gas reduction during treatment of piggery waste air at different gas loading rates with the FAL process

Treatment unit	Gas load [m³/(m² h)]	Measuring point	Odour [OU/m³]	NH_3 [ppm]	NO_x [ppm]	N_2O [ppm]	C_{tot}* [ppm]	CO [ppm]	CO_2 [Vol-%]	CH_4 [ppm]	H_2S [ppm]
		Raw gas	1960	9.9	< 0.1	0.8	9.9	0.3	0.12	23.5	0.8
Scrubber	2700	Scr. outlet	1302	0.3	0.3	0.7	9.6	0.3	0.12	23.2	0.6
Biofilter	87	Biof. outlet	66	0.1	0.3	0.7	9.3	< 0.1	0.12	22.5	0.1
		Raw gas	915	16.1	< 0.1	0.8	10.9	0.4	0.16	30.8	< 0.1
Scrubber	3900	Scr. outlet	504	3.7	< 0.1	0.8	10.4	0.3	0.15	30.0	0.1
Biofilter	42	Biof. outlet	47	0.4	0.1	0.8	10.1	0.1	0.16	30.0	< 0.1
Biofilter	220	Biof. outlet	88	0.4	0.1	0.8	9.9	0.1	0.15	30.1	< 0.1
		Raw gas	688	10.0	0.3	0.3	n. d.	1.3	0.08	15.6	< 0.1
Scrubber	3800	Scr. outlet	356	0.1	0.2	0.4	n. d.	1.4	0.08	14.8	< 0.1
Biofilter	154	Biof. outlet	201	< 0.1	0.2	0.3	n. d.	1.3	0.08	14.1	< 0.1
		Raw gas	2187	11.5	0.7	0.6	10.7	0.4	n. d.	25.3	0.6
Scrubber	4600	Scr. outlet	569	0.2	< 0.1	0.7	10.8	0.4	n. d.	26.0	0.4
Biofilter	220	Biof. outlet	330	< 0.1	< 0.1	0.6	10.3	0.2	n. d.	25.5	0.2
Biofilter	37	Biof. outlet	109	< 0.1	< 0.1	0.6	10.3	0.2	n. d.	25.5	0.2
		Raw gas	445	8.3	< 0.1	0.7	3.3	0.4	0.08	6.8	0.2
Scrubber	4200	Scr. outlet	336	0.3	< 0.1	0.7	2.8	0.5	0.08	7.1	0.2
Biofilter	110	Biof. outlet	260	0.6	< 0.1	0.6	3.2	0.5	0.09	7.2	< 0.1

n. d.: not detected, C_{tot}: Total organic carbon, including methane

raw gas odour concentrations was the self odour emission from the biofilter material. Apart from the reduction of the odour intensity the hedonic odour tone was improved significantly. A test with 83 randomly selected persons showed that the raw gas was felt as pleasant (3 test persons), neutral (3), unpleasant (41), very unpleasant (29) and extremely unpleasant (7). The scrubber outlet concentration was felt as pleasant (10), neutral (13) unpleasant (58) and very unpleasant (2). A significant improvement of the hedonic odour tone was achieved by biofiltration. The biofilter outlet air was felt as pleasant (14), neutral (67), unpleasant (1) and very unpleasant (1). Although dust, ammonia and the odour concentration were reduced by the scrubbing, the hedonic odour tone was felt only improved slightly. Ammonia was reduced about 93 % at least in the total process, but is most cases the efficiency was more than 98 %. Nitric oxides and nitrous oxide were not produced in the process because of the preseparation of ammonia. TOC including methane was reduced less than 10 % in the total process. Carbon dioxide was slightly produced in the process and carbon monoxide was oxidized in some cases. The methane degradation was less than 10 %. Hydrogen sulfide was oxidized especially during biofiltration and the degradation efficiency amounted from 50 to 87 %.

Ammonia, water soluble compounds and dust particles were separated from the waste air and concentrated in the scrubber liqiud. Dust and other biodegradable carbon sources were oxidized by microorganisms, which developed in the scrubber depending on the pH value. Less and not biodegradable compounds as well as salts were concentrated. The normal composition of the concentrated lquid product from the scrubbing is shown in Tab. 4. Besides the high and desired salt contents only low COD, BOD_5 and carbon concentrations were detected in the liquid scrubber product. With annual ammonia nitrogen emissions of 3.0 kg per pig place around 11 kg of pure sulfuric acid are necessary for ammonia separation. Depending on the concentration grade 30 - 40 l ammonium sulfate with a nitrogen content of at least 7 % (m/m) are obtained per pig place and year.

The composition of the biofilter irrigation water that was collected in tank 3 (Fig. 3) and which was topped up with fresh water if necessary showed only low concentrations of nitrogen compounds (Tab. 5). The scrubber passing ammonia nitrogen was slowly oxidized to nitrate without a significant accumulation of nitrite in the biofilter irrigation water. On average 58.5 % of total nitrogen in the irrigation water was ammonia, 0.06 % nitrite, 32.9 % nitrate and 8.4 % was organic bond nitrogen. The mean sulfate concentration was four times the concentration of the used drinking water for filling up. In view of carbon sources the analyses of the biofilter irrigation water showed a mean COD:BOD_5 ratio of 31, that corresponds with a poor biodegradability. During a two-year period there was only a very slight concentration increase of the different parameters apart from the pH value which dropped slowly from 7.7 to 6.2.

The biofilter material analyses showed apart from an increase of ammonia and nitrate concentration neither a

Table 4
Composition and range of the scrubber product

Parameter	Range of concentration
pH	2.6 - 3.4
Electric conductivity	400 - 680 mS/cm
Specific gravity	1.1 - 1.3 g/ml
Ammonia nitrogen	66 - 79 g/kg
Nitrate	0.1 - 0.2 g/kg
Sulfate	180 - 340 g/kg
Chemical oxygen demand (COD)	1.8 - 2.7 g/kg
Biochemical oxygen demand (BOD5)	0.05 - 0.3 g/kg
Total salt content	380 - 550 g/kg
Total carbon content	30 - 40 g/kg TS

TS: Total solids

Table 5
Composition and range of the biofilter irrigation water

Parameter	unit	Range of concentration	Mean*
pH	[ ]	5.8 - 7.7	6.8
Electric conductivity	[mS/cm]	0.8 - 3.5	1.9
Ammonia nitrogen	[mg/kg]	0 - 304	69
Nitrite nitrogen	[mg/kg]	0 - 3.7	0.7
Nitrate nitrogen	[mg/kg]	6 - 94	39
Total nitrogen	[mg/kg]	11 - 316	79
Sulfate	[mg/kg]	173 - 1370	669
COD	[mg/kg]	13 - 655	188
BOD_5	[mg/kg]	0 - 16	6

* n=57

reduction of total solids (TS) nor a reduction of the carbon content in a two-year period. Ammonia increased from 0.06 g/kg TS to 1.4 g/kg TS and nitrate from 2.7 g/kg TS to 29.8 g/kg TS. The pH value of the biofilter material dropped from 6.5 to 5.6 during that time. Nitrite was not detected.

6 Benefits and disadvantages

Because of its high automation grade the FAL waste air treatment system for piggeries is working stable. Ammonia is separated with an efficiency of more than 93 % and concentrated in a liquid fertilizer which can be used in agriculture. Odour emissions from stables can be reduced significantly. The odour concentration are reduced by the process about 42 % (raw gas concentration: 445 OU/m³), 71 % (688 OU/m³) and 85 % (915 OU/m³). In addition to that the hedonic odour tone is improved clearly, as an evaluation with 83 test persons showed. Dust in raw gas concentrations less than 2 mg/m³ is separated in the scrubber and degraded by microorganisms without any technical problems. Hydrogen sulfide is oxidized in the biofilter in a considerable extent and carbon monoxide is sometimes oxidized. There is no secondary production of trace gases by the waste air treatment in form of nitric oxides or nitrous oxide.

The main disadvantage of the process is the high energy requirement which amounted from 2 to 4 kWh per 1000 m³ raw gas treated, depending on the scrubber and biofilter loading rate. Above that around 11 kg of pure sulfuric acid are necessary to separate the annual ammonia emission of one pig place. Waste gas scrubbing in this form produces saturated ammonium sulfate solution in a range from 30 to 40 l per pig place and year. The pure nitrogen content is 7 up to 8 % (m/m) and the mean sulfur content is in the same range.

7 Outlook

The process operation costs are particularly determined by the electricity consumption which have to be reduced even by reducing the stable ventilation rates. These ventilation rates are currently controlled and regulated by the tolerated difference between the indoor and outdoor temperature. The temperature drop can be maintained by using water evaporating and fresh air cooling systems. In addition to that stable shading by planting and use of soil heat exchangers might be a promising way. The rearrangement of ventilation systems should be taken into account in view of a regulation that is additionally based on trace gas concentrations in the stable. All these measures would reduce the current and very high ventilation rates which make the waste air treatment so expensive.

The biofilter, which is particularly used for odour reduction, causes the main pressure drop in the whole treatment process. In view of energy consumption the pressure drop has to be reduced without an odour reduction efficiency drop. Alternatively it should be taken into account to oxidize odourants chemically or biologically during a two step scrubbing that causes a lower pressure drop.

Literature

Backus G B C, van de Sande-Schellekens, Voermans J A M (1993) Economics of bioscrubbers to reduce ammonia emission from fattening pig farms. In: Nitrogen flow in pig production and environmental consequences. Wageningen, EAAPO publication 69 [23]

Chung Y-C, Huang C (1998) Biotreatment of ammonia in air by an immobilized Nitrosomonas europaea biofilter. Environmental Progress 17: 70-76 [20]

Chung Y-C, Huang C, Tseng C-P (1997) Biotreatment of ammonia from air by an immobilized Artrobacter oxydans CH8 biofilter. Biotechnol Prog 13: 794-798 [17]

Demmers T G M (1989) Adsorption und Nitrifikation von Ammoniak im Biowäscher. VDI-Berichte 735 [21]

Department of Environment (ed) Environment data Germany 1998 [03]

Hahne J, Hesse D, Vorlop K-D (1999) Trace gas emissions from pig fattening. Landtechnik 54 (3): 180-181 [07]

Hahne J, Vorlop K-D (2001) Cleaning of waste gas from piggeries with integrated nitrogen removal. Gefahrstoffe - Reinhaltung der Luft 61 (4): 139-143 [09]

Hartung E, Martinec M, Jungbluth T (1997) Reduzierung der Ammoniak- und Geruchsemissionen aus Tierhaltungsanlagen der Landwirtschaft durch biologische Abluftfilter. Forschungsbericht Universität Hohenheim [13]

Iversen M, Dahl R (2000) Working in swine-confinement buildings causes an accelerated decline in FEV1: a 7-yr follow-up of Danish farmers. Eur Respir J 16: 404 - 408 [02]

Kim N J, Sugano Y, Hirai M, Shoda M (2000) Removal characteristics of high load ammonia gas by a biofilter seeded with a marine bacterium, Vibrio alginolyticus. Biotechnology Letters 22: 1295-1299 [19]

Lais S (1996) Untersuchungen zur Reduzierung der Ammoniak- und Geruchsemissionen aus der Schweinehaltung durch biologische Abluftwäscher. Dissertation Hohenheim [15]

Lower Saxony Department of Food, Agriculture and forests (ed) Animal production in Lower Saxony 1995 [04]

Lower Saxony Department of Food, Agriculture and forests (ed) Animal production in Lower Saxony 1997 [05]

Lower Saxony Department of Food, Agriculture and forests (ed) Animal production in Lower Saxony 1999 [06]

Mannebeck D, Hügle T (1993) Biofilter als Bestandteil raumlufttechnischer Anlagen. Beiträge zur 1. Internationalen Tagung "Bau und Technik in der landwirtschaftlichen Nutztierhaltung. Gießen, pp 175-184 [12]

Mannebeck D, Hügle T (1993) Verminderungen von Emissionen aus der Tierhaltung. SchriftenR Agrarwiss Fak Univ Kiel 76: 115-123 [11]

N N (2001) German standards methods for the examination of water, waste water and sludge. Weinheim : Wiley-VCH [24]

N N: FAOSTAT DATABASE [01]

Papen H, Berg R von, Hinkel I, Thoene B, Rennenberg H (1989) Heterotrophic nitrification by Alcaligenes faecalis: NO_2-, NO_3-, N_2O and NO production in exponentially growing cultures. Appl Environ Microbiol 55: 2068-2072 [18]

Seedorf J et al (1998) Concentrations and emissions of airborne endotoxins and microorganisms in livestock buildings in Northern Europe. J agric Engng Res 70: 97-109 [08]

Siemers V (1996) Nassabscheider/Biofilterkombinationen zur Verminderung von Ammoniak-, Geruchs- und Staubemissionen aus Schweinemastställen. Dissertation Göttingen [14]

van Geelen M A, van der Hoek K W (1977) Odour control with biological air washers. Agriculture and Environment 3: 217-222 [22]

VDI - Guideline 3477 "Biofilters". Düsseldorf 1991 [10]

VDI - Guideline 3478 "Bioscrubbers and Trickle Bed Reactors". Düsseldorf 1996 [16]