

# Odour and ammonia removal from pig house exhaust air using a biotrickling filter

R. W. Melse\* and G. Mol\*\*

\* IMAG, Institute of Agricultural and Environmental Engineering, P.O. Box 43, 6700 AA, Wageningen, Netherlands (E-mail: [roland.melse@wur.nl](mailto:roland.melse@wur.nl))

\*\* IMAG, Institute of Agricultural and Environmental Engineering. Current address: Alterra, P.O. Box 47, 6700 AA, Wageningen, Netherlands (E-mail: [gerben.mol@wur.nl](mailto:gerben.mol@wur.nl))

## Abstract

Odour from agricultural activities, such as the spreading of manure and the housing of animals, is increasingly being considered a nuisance in densely populated countries like the Netherlands. The objective of this research was to study the odour removal from pig house exhaust air by a biotrickling filter that had been implemented for ammonia abatement. At a regular pig production farm, the performance of a running full-scale biotrickling filter was studied for 72 days. Ammonia and odour removal efficiency were on average 79% and 49% respectively. Ammonia removal appeared to be based on an unintended accumulation of ammonium and nitrite in the system instead of on production and discharge of nitrate. The odour removal efficiency showed a large variation that was for a major part (about 80%) caused by the performance of the biotrickling system. This large variability was probably caused by variations in the composition of the air that were not completely reflected by the olfactometrically measured odour concentration; the many different components that make up the odour each have different removal characteristics. It seemed that the biotrickling filter was operated below its maximum absolute odour removal capacity, which means that the absolute odour removal [ $OU_E/(m^3 \text{ filter})/s$ ] will probably increase at higher loads. It was, however, not possible to distinguish between the influence of either the odour load or the odour concentration on the odour removal because of a positive correlation between the odour concentration and the air flow. To increase the relative odour removal, the design of the filter has to be optimized for both the well and poorly water-soluble odour components.

## Keywords

Odour, ammonia, removal, biotrickling filter, pig, air, olfactometry

## INTRODUCTION

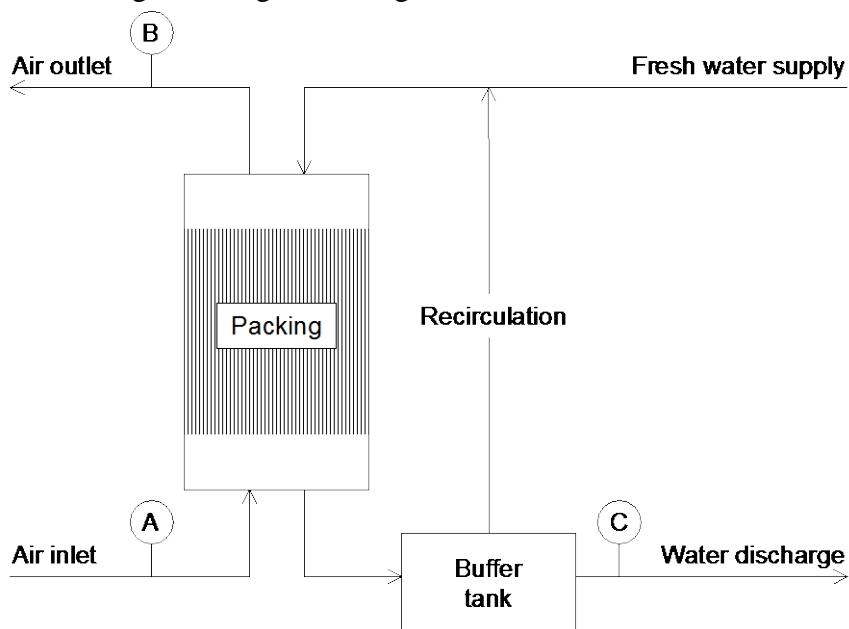
Pig production contributes substantially to the economies of many Western European countries in terms of employment and export of products. Pig production in Western Europe is concentrated in several regions characterised by large-scale intensive farms. The Netherlands, with 16 million inhabitants and a population density of 386 inhabitants per  $km^2$ , houses 13 million pigs at approximately 13,000 farms (CBS, 2002). The pig farms are mainly concentrated in the eastern and southern part of the country where opportunities for arable farming are limited by poor sandy soils. From the 1980's the emission of ammonia ( $NH_3$ ) from livestock farming has become a major environmental concern because ammonia emission is one of the three main source of soil acidification in the Netherlands (Heij and Erisman, 1995; Heij and Erisman, 1997). In 2000, the ammonia emission from livestock farming still accounted for about 50% of the total emission of acidifying compounds (CCDM, 2002). This focus on ammonia emissions has resulted in the development of a large variety of ammonia abatement techniques. An example of such a technique is the use of a biotrickling filter for treatment of exhaust air from animal houses. In recent years, odour emissions from animal housing and from land application of manure are increasingly considered a nuisance because of growing suburbanisation. It is unclear, however, whether ammonia abatement techniques also decrease odour emission, and if so, to what extent.

The objective of this work was to study the odour removal from pig house exhaust air by a biotrickling filter that had been designed for ammonia removal. Both odour and ammonia removal data are presented.

## MATERIALS AND METHODS

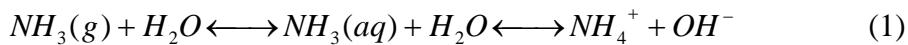
### Biotrickling filter

A biotrickling filter is a bioreactor that is filled with an inorganic packing material, or filter bed, on which a bacterial biofilm grows. Water is sprayed over the filter bed and the biofilm is wetted. The trickling water is partly recirculated and partly discharged and replaced by fresh water. Usually the water recirculation flow is much higher than the water discharge flow so that the composition of the discharge water equals the conditions in the whole filter bed. The air to be cleaned is forced through the filter bed resulting in an intensive contact between the air and the water. A schematic of a biotrickling filter is given in Figure 1.

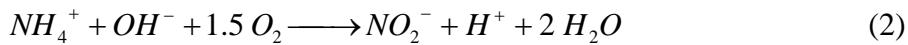


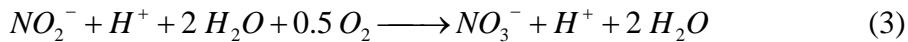
**Figure 1** Schematic of biotrickling filter.

If the air contains water-soluble components, these components are partly transferred to the wet biofilm and are available for bacterial degradation. In the case of ammonia, the mass transfer largely depends on the pH of the water due to the equilibrium between  $\text{NH}_3$  and  $\text{NH}_4^+$  as is shown in Equation 1:



Subsequent bacterial oxidation from ammonium to nitrite ( $\text{NO}_2^-$ ) and from nitrite to nitrate ( $\text{NO}_3^-$ ) is called nitrification and mainly carried out by *Nitrosomonas* and *Nitrobacter* species respectively. In Equation 2 and 3 these processes are shown:





A stable operating biotrickling filter usually is in a steady-state condition which means there is an equilibrium between the processes shown in Equation 1 through 3 and the amount of nitrogen and H<sup>+</sup> that is removed from the system by water discharge. This usually results in the following conditions: 6.5 < pH < 7.5, [NH<sub>4</sub><sup>+</sup>] < 0.4 g/L, and 0.8 < [NH<sub>4</sub><sup>+</sup>]/[NO<sub>2</sub><sup>-</sup> + NO<sub>3</sub><sup>-</sup>] < 3 on a molar basis (Den Brok *et al.*, 1997); usually full nitrification occurs so that [NO<sub>2</sub><sup>-</sup>]/[NO<sub>3</sub><sup>-</sup>] << 1.

Odour on the other hand, is a mixture of many different volatile compounds. Besides ammonia, the main odour components in exhaust air from animal houses are volatile fatty acids, *p*-cresol, indole, skatole, and diacetyl (O'Neill and Phillips, 1992); the sources of the odorous components are manifold (e.g. feed, animal, manure, bedding). Water solubility may vary from very poor to very good. Besides the nitrifying bacteria already mentioned, the microbial community in a biotrickling filter comprises bacteria that use odour components as a substrate thus resulting in reduction of odour emission. It is known that many odorous compounds can be biologically degraded although biodegradability may vary from very poor to very good.

The biotrickling filter that was studied is a commercially available system that had been installed at a pig farm in the Netherlands in order to remove ammonia from part of the exhaust air from 650 fattening pigs. The pig house was ventilated by two fans that were frequency controlled on the basis of the temperature in the pig house; therefore the air flow could not be changed for research purposes. The air outlet of one of the two fans was connected to the air inlet of the biotrickling filter. The biotrickling filter contained a square based packed filter bed with a volume of 3 m<sup>3</sup> and a height of 1 m and had been designed for a maximum air flow of 20,000 m<sup>3</sup>/hour. The packing consisted of a vertical bundle of plastic tubes (diameter: 4 cm) that were glued together. Water was sprayed on top of the packing and collected in a buffer tank (35 m<sup>3</sup>) and then partly discharged or recirculated. Both air inlet (Figure 1, point A) and air outlet (Figure 1, point B) tubes were equipped with an air sampling point and a sensor for measurement of temperature [°C] and relative humidity [%]. The air flow [m<sup>3</sup>/s] was calculated from the frequency [Hz] of the fan and the specifications of the manufacturer. The discharge water was sampled at point C (Figure 1). The research took place between 8th October (day 0) and 19th December 2001 (day 72).

### Ammonia measurements

Measurements of the ammonia concentration in the air inlet and air outlet were done on 5 days. The ammonia concentration in the air was determined by leading air (120 L/hour) from the air sampling point through two gas washing bottles connected in series and filled with sulphuric acid (0.02 M) during two hours (Van Ouwerkerk, 1993). Ammonia was captured by the acid and fluctuations in the ammonia concentration of the sampled air were time-averaged over two hours. Tubing was made of Teflon to prevent adsorption of ammonia. The ammonia concentration of the original air sample was calculated from the nitrogen content of the acid solution that had been determined with a wet-chemical method (NEN, 1998). Ammonia emission [g/s] was calculated by multiplying ammonia concentration [g/m<sup>3</sup>] by air flow [m<sup>3</sup>/s].

### Odour measurements

Air samples for odour measurement were taken from the air inlet and air outlet on 12 days. The samples were taken by using a so-called lung method. In this method, air samples were collected in Teflon odour bags (60 l) that were placed in airtight containers. The inlet of the, initially vacuum, odour bags was connected to the sampling tube from the inlet or outlet air, and filled by creating an underpressure in the surrounding airtight container by means of a pump. The odour bags were thus filled in two hours time, the sampling rate (0.5 L/minute) was controlled by a critical orifice. In this

way fluctuations in the composition of the air sample are time-averaged over two hours. A filter (1-2  $\mu\text{m}$ ) at the inlet of the sampling tube prevented the intake of dust that could contaminate the olfactometer. The sampling system was equipped with a heating system to prevent condensation in the bag or in the tubing. The odour bags remained in the container until analysis in the odour laboratory, which had to take place within 30 hours after sample collection. Odour concentrations were determined in compliance with the Dutch olfactometric standard method NVN2820/1A (NNI, 1996) that is based on the earlier NVN2820 (NNI, 1995). In the NVN2820/A1 standard, the sensitivity of the odour panel is based on the 20 - 80 ppb n-butanol range. The odour concentrations are expressed in odour units per  $\text{m}^3$  air ( $\text{OU}_E/\text{m}^3$ ) (CEN, 1998). Odour emission [ $\text{OU}_E/\text{s}$ ] was calculated by multiplying odour concentration [ $\text{OU}_E/\text{m}^3$ ] by air flow [ $\text{m}^3/\text{s}$ ].

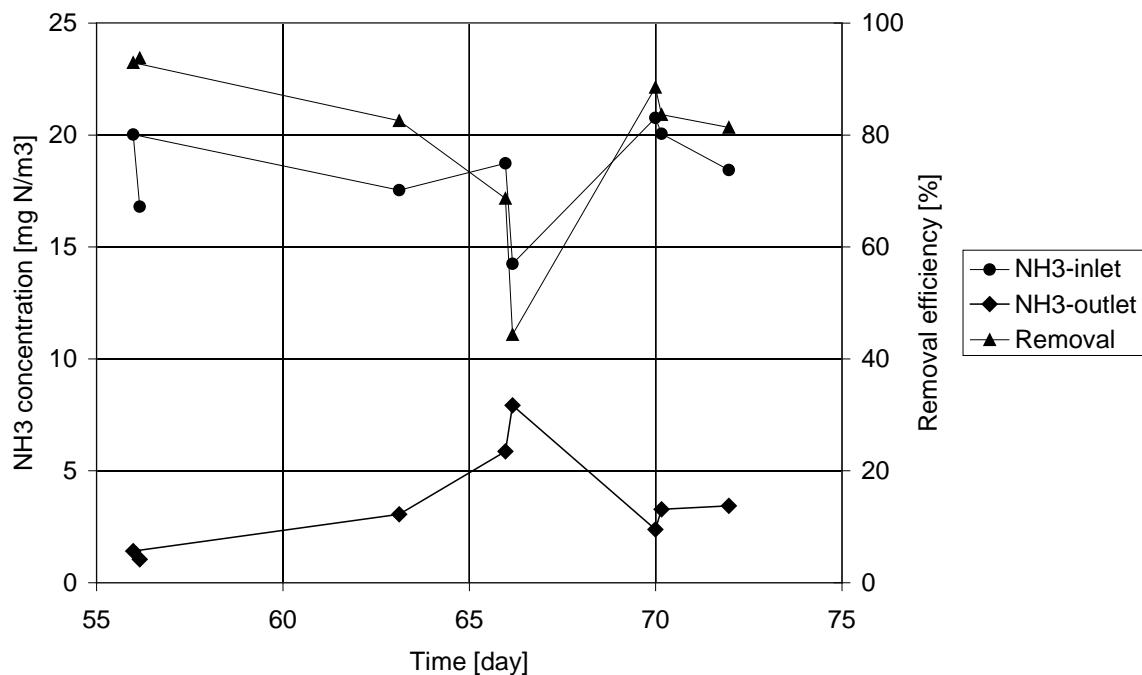
### Water analysis

Each time the air inlet and outlet was sampled for ammonia measurement, a sample (1 L) was taken from the discharge water and pH, ammonium, nitrite, and nitrate content were determined with standard methods (NEN, 1998).

## RESULTS AND DISCUSSION

### Ammonia removal

The results of the ammonia measurements are given in Figure 2. The ammonia concentration of the inlet air of the biotrickling filter varied from 14 to 20 mg N/ $\text{m}^3$  while the ammonia odour removal efficiency varied from 41 to 94% (79% on average). It is unclear why the removal efficiency drops at day 66. The air flow through the filter varied from 7,600 to 10,900  $\text{m}^3/\text{hour}$ .



**Figure 2** Ammonia removal by biotrickling filter treating exhaust air from a pig house.

The characteristics of the discharge water from the biotrickling filter (Table 1) indicate that the biotrickling filter is not in stable operation because accumulation of ammonia and nitrite takes place in time and concentrations strongly differ from steady-state concentrations that normally occur (see above). *Nitrosomonas* seems to be partially inhibited as  $[\text{NH}_4^+] \gg 0.4 \text{ g/L}$ ; *Nitrobacter* seems to be

fully inhibited as  $[\text{NO}_3^-] \approx 0$ . It is known that both ammonium and nitrite, in the forms of  $\text{NH}_3$  (aq) and  $\text{HNO}_2$  (aq), are inhibitive to nitrifying bacteria. Inhibition of *Nitrosomonas* by  $\text{NH}_3$  (aq) starts at concentrations of 10 - 150 mg/L, whereas *Nitrobacter* is already inhibited by  $\text{NH}_3$  (aq) at concentrations from 0.1 to 1 mg/L; moreover, *Nitrobacter* is inhibited by  $\text{HNO}_2$  (aq) starting at concentrations of 0.2 - 2.8 mg/L (Anthonisen *et al.*, 1976). Assuming equilibrium, the  $\text{NH}_3$  (aq) and  $\text{HNO}_2$  (aq) concentrations in the discharge water are calculated to be 17 - 46 mg/L and 0.3 - 0.4 mg/L respectively at the pH that was measured. Therefore it is likely that the inhibition of *Nitrosomonas* and *Nitrobacter* is caused by the high  $\text{NH}_4^+$  and  $\text{NO}_2^-$  concentrations that were found in the discharge water resulting in accumulation of both ammonium and nitrite.

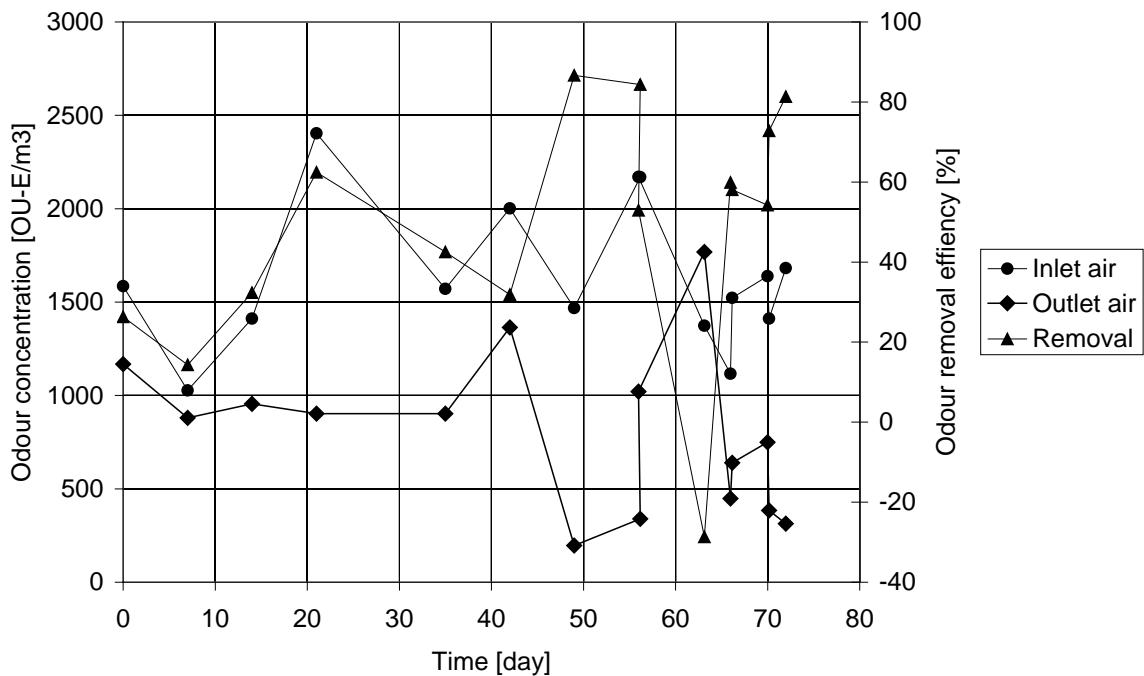
**Table 1.** Characteristics of discharge water of biotrickling filter treating exhaust air from a pig house.

Sample day	$\text{NH}_4\text{-N}$ [g/l]	$\text{NO}_2\text{-N}$ [g/l]	$\text{NO}_3\text{-N}$ [g/l]	pH [-]
56	1.19	1.22	< 0.01	7.3
63	1.32	1.42	< 0.01	7.6
66	1.55	1.59	< 0.01	7.7
70	1.59	1.61	< 0.01	7.6
72	1.63	1.65	< 0.01	7.6

Although it follows that the biotrickling filter does not function well from a microbiological point of view, still the ammonia removal efficiency is 79% on average. In fact it can be calculated that the amount of ammonia that is removed from the air equals the accumulation of ammonium and nitrate in the buffer tank whereas in a normally operating biotrickling nitrogen does not accumulate but leaves the system as nitrate with the discharge water. Measurements of the composition of the water that enters the filter bed after addition of fresh water (data not presented), show that the composition is equal to the composition of the discharge water indicating that hardly any fresh water is added to the system. In the long run however, biological activity will further decrease and accumulation of ammonium will proceed until hardly any ammonia will be removed from the air because the equilibrium of Equation 1 will shift to the left more and more. In order to stop the accumulation of nitrogen in the system, the fresh water supply must be increased.

### Odour removal

The results of the odour measurements are given in Figure 3. The odour concentration of the inlet air of the biotrickling filter varied from 1,000 to 2,400  $\text{OU}_E/\text{m}^3$  while the odour removal efficiency varied from -29 to 87% (49% on average).



**Figure 3** Odour removal by biotrickling filter treating exhaust air from a pig house.

Other research (Ogink and Lens, 2001; Mol and Ogink, 2002) shows that it is a common phenomenon for both biotrickling filters and acid air scrubbers to have a much higher variation in the odour removal efficiency than in the ammonia removal efficiency. This variation could in principle be caused by the functioning of the filter itself or by the olfactometric method that was used, or by a combination of both. We had a closer look at the possible sources of the variation because the laboratory variances of the olfactometric method is generally higher than that of standard (chemical) analytical methods. By comparing the pooled variance of the odour concentrations in the inlet air and the outlet air with the laboratory variance associated with olfactometric analyses by the same panel, we could establish that the olfactometric method contributes about 20% to the total variance of the odour removal efficiency measurement whereas the functioning of the biotrickling system contributes about 80%.

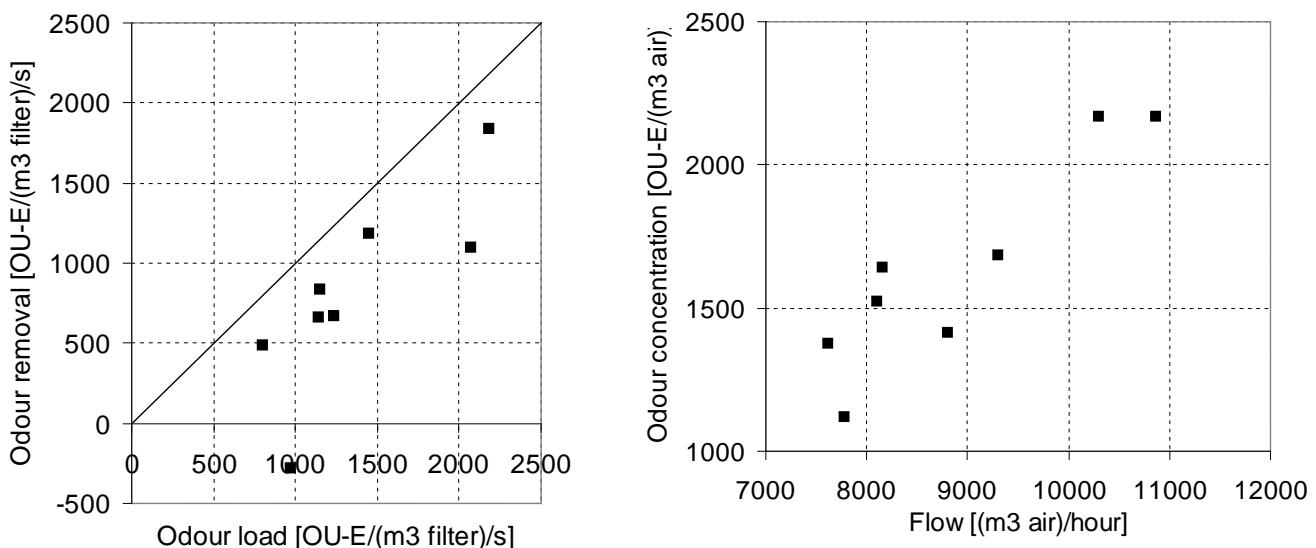
The reason for the variation of the odour removal efficiency being higher than for ammonia is probably that changes in the odour composition are not fully reflected in the odour concentration values. In contrast with the removal of ammonia, which is easily transferred to the liquid phase and easily biodegraded, the measured removal of odour is the sum of the removal of many separate odour components that each have different characteristics with regard to mass transfer, i.e. water solubility, and biodegradability. If, at a constant odour load, the concentration of an easily removable odour component increases in comparison with the other odour components, the measured odour removal efficiency will increase. If, on the other hand, an odour component is difficult to remove, a relative increase of this component will result in a decrease of the measured removal efficiency at the same odour load. As the odour components could not be measured separately, the phenomenon described here may explain the relatively large variation that was found for the odour removal efficiency.

In Figure 4 the odour removal of the system is plotted against the odour load showing a positive correlation. No data are shown for day 0-56 because no reliable air flow measurements are available for this period. In general, a biofiltration system has a maximum removal capacity, i.e., when the load exceeds the maximum removal capacity, the absolute removal will remain at the same level as the load further increases. As the odour removal in Figure 4 is still rising at high odour loads it can

be concluded that the biotrickling filter is running below its maximum absolute odour removal capacity and higher absolute removal levels can probably be achieved at higher loads. However, the odour load is the product of air flow and odour inlet concentration which are also positively correlated as is shown in Figure 5. Therefore it is, in this study, not possible to distinguish between the influence of either odour concentration or odour load on odour removal.

In biofiltration literature (e.g. Deshusses and Johnson, 2000), it is generally assumed that the removal of a component is determined by the load of the component, i.e. the product of the inlet concentration of the component and the air flow, and not by the concentration of the component as the mass transfer from the air to the liquid phase is usually not the rate limiting step in the removal process. However, if the component is very poorly water soluble, i.e. has a very high Henry-constant, mass transfer may be the rate limiting step so that the removal of the component is primarily determined by the air inlet concentration and not by the load of the component. As odorous air consists of both well and poorly water soluble compounds, this may also be the case in this study. Experiments with independent alteration of air flow and odour concentration should give a decisive answer about this matter. Such experiments were not possible at our research site as the air characteristics were determined by the automatic ventilation system of the animal house.

A final remark on the performance of the biotrickling filter concerns the relative removal of the



odour load that leaves the pig house. This relative removal now averages at about 50% removal of the incoming odour load. Because the most probable rate limiting step is the mass transfer of poorly soluble odour components to the water phase, the relative performance of filter can only be increased by changing some of the system characteristics that, together with the chemical characteristics of the components to be removed, determines the mass transfer. Some options in this respect are the dimensions of the filter (e.g. increased exchange surface), the retention time (e.g. a lower flow or a larger filter), and designing a two or even a multi-phase filter in which both polar and apolar components can be forced out of the air.

## CONCLUSIONS

The removal of ammonia from the exhaust air of the pig house appeared to be based on accumulation of ammonia and nitrite in the biotrickling system instead of on oxidation of ammonium to nitrate followed by removal with the discharge water. It is concluded that the fresh water supply of the system needs to be increased to achieve a stable and reliable ammonia removal system.

The efficiency of removal of odour from the exhaust air of the pig house showed a large variation that was mainly caused by the actual performance of the biotrickling filter. The variation in the

odour removal performance of the biotrickling filter is probably caused by variations in the composition of the odorous air that are not completely reflected by the olfactometrically measured odour concentration; the many different components that make up the odour each have different removal characteristics.

It is concluded that the biotrickling filter was operated below its maximum absolute odour removal capacity [ $\text{OU}_E/(\text{m}^3 \text{ filter})/\text{s}$ ] meaning that a higher odour load will probably result in a higher absolute odour removal. It was not possible, however, to distinguish between the influence of either odour concentration or odour load on odour removal. To increase the relative odour removal, the design of the filter has to be optimized for all odour components, both the well and poorly water-soluble components.

## ACKNOWLEDGMENTS

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