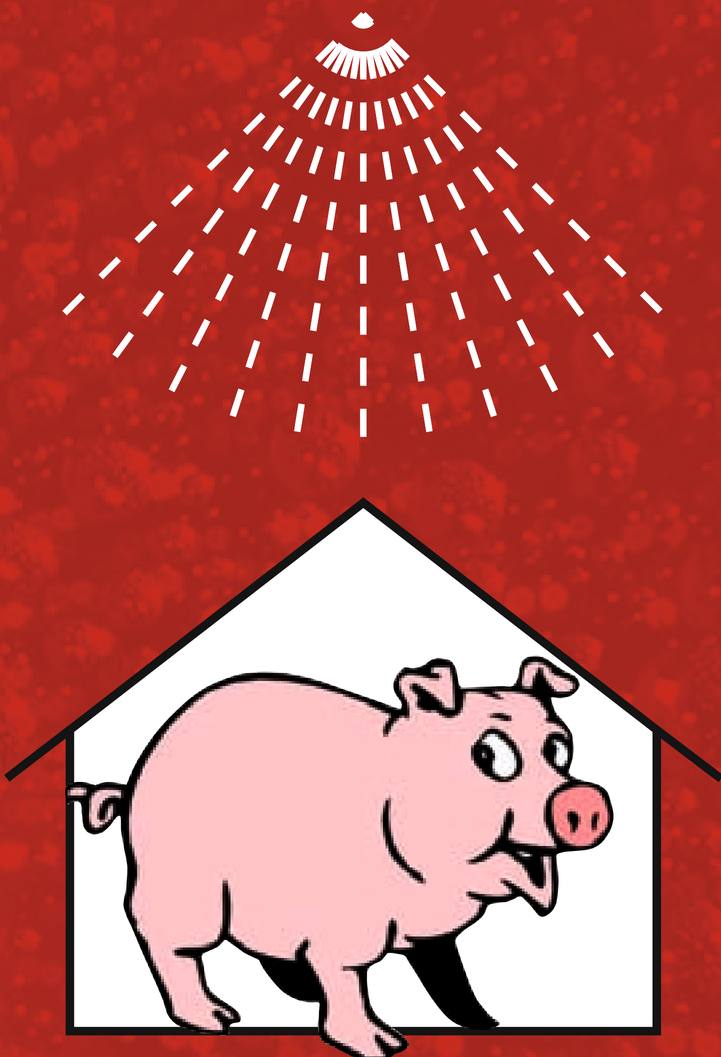
Air treatment techniques for abatement of emissions from intensive livestock production



Roland W. Melse

Air treatment techniques for abatement of emissions from intensive livestock production

Roland W. Melse

Thesis committee

Thesis supervisor

Prof Dr Ir W.H. Rulkens
Professor of Environmental Technology
Wageningen University

Thesis co-supervisor

Dr Ir N.W.M. Ogink
Senior scientist
Wageningen UR Livestock Research, Lelystad

Other members

Prof Dr Ir P.W.G. Groot Koerkamp	Wageningen University
Dr Ir G.J. Monteny	Monteny Milieu Advies, Renkum
Prof Dr E. Hartung	University of Kiel, Germany
Dr A. Feilberg	University of Aarhus, Denmark

Air treatment techniques for abatement of emissions from intensive livestock production

Roland W. Melse

Thesis
submitted in partial fulfilment of the requirements for the degree of doctor
at Wageningen University
by the authority of the Rector Magnificus
Prof Dr M.J. Kropff,
in the presence of the
Thesis Committee appointed by the Doctorate Board
to be defended in public
on Monday 26 October 2009
at 11 AM in the Aula.

Roland W. Melse
Air treatment techniques for abatement of emissions from intensive livestock production

Thesis Wageningen University, Wageningen, NL (2009)
With references, with summaries in Dutch and English

ISBN 978-90-8585-463-0

Abstract

Intensive livestock production is connected with a number of environmental effects, including emissions of ammonia (NH_3), greenhouse gases (CH_4 and N_2O), odour, and particulate matter (PM10 and PM2.5). Possible strategies for emission reduction from animal houses include feed management, adaptation of housing design, and the application of end-of-pipe air treatment, *viz* acid scrubbers and bioscrubbers. In order to comply with current and future regulations the implementation of air scrubbers is expected to expand in intensive livestock production areas across Europe. The aim of this thesis is to better understand and improve the performance of air scrubber systems in livestock operations. The objectives are to determine (1) how air scrubbers are performing at livestock farms with regard to ammonia and odour removal; (2) how the cost-efficiency of air scrubbers can be increased by applying a different treatment strategy; (3) if methane can be removed by biological air treatment systems; and (4) to discuss the drawbacks of current air scrubbing practices and make recommendations for improvement.

It was found that ammonia removal efficiency by scrubbers is relatively high (on average 96% for acid scrubbers and 70% for bioscrubbers), that odour removal is relatively low (on average 31% for acid scrubbers and 44% for bioscrubbers), and that especially bioscrubbers often show a large variation in odour removal efficiency. Furthermore, it was found that bioscrubbers often experience operational problems. Next a partial air cleaning approach has been presented in which air is bypassed at occasionally occurring high air flow rates. Model calculations show that a reduction of scrubber volume by 50% still enables treatment of 80 - 90% of the ammonia load. This strategy reduces investment and operational costs but hardly affects average ammonia emission levels. Furthermore, it was demonstrated that biological oxidation of methane is possible, although the low water solubility of methane limits the practical application of biofilters or bioscrubbers for methane removal. Finally, it is concluded that scrubbers applied at animal houses can still be improved and optimized. For example the process control and performance of bioscrubbers might be enhanced by measurement of electrical conductivity (EC) as a control parameter for water discharge.

Keywords: *Air treatment; Scrubber; Bioscrubber; Biofilter; Biotrickling filter; Ammonia;* NH_3*; Odour; Livestock production; Animal husbandry; Pig; Poultry.*

Contents

Chapter 1

Introduction

Contents

An adapted version of this chapter has been published in The Open Agriculture Journal, 2009, volume 3, pages 6-12 (Melse, R.W.; N.W.M. Ogink; W.H. Rulkens. Air treatment techniques for abatement of emissions from intensive livestock production) and is reproduced with permission from Bentham Science Publishers.

1.1 Environmental impact of livestock production

Intensive livestock production contributes substantially to the economies of many European countries in terms of employment and export of products. Pig production in Europe is concentrated in several regions characterised by large-scale intensive farms. Main pig producing areas can be found in the north (Denmark, the Netherlands, Belgium, Brittany in France, Niedersachsen in Germany) and in the south (Lombardy in Italy, Cataluña and Galicia in Spain) (EC, 2003). The Netherlands, with 16 million inhabitants and a population density of almost 400 inhabitants per km^2, houses 11 million pigs and 93 million chickens at approximately 10,000 and 3,000 farms, respectively, as per 2005 (CBS, 2007). The livestock operations are mainly concentrated in the eastern and southern part of the country where in the past opportunities for arable farming were limited by poor, sandy soils.

Intensive livestock production is connected with a number of environmental effects, which include emissions to the air, *e.g.* ammonia, odour, non-CO_2 greenhouse gases (methane (CH_4) and nitrous oxide (N_2O)) and particulate matter, and discharges to soils and surface waters (*e.g.* nitrogen, phosphorus, and heavy metals)[1]. The risks of ammonia emissions relate to acidification of soils and waters and high levels of nitrates found in drinking waters. High levels of nitrogen and phosphorus in surface waters may lead to eutrophication which involves excessive algal growth and can lead to potential adverse effects on biodiversity or human uses of waters (Heij and Erisman, 1995, 1997). The emission of greenhouse gases contributes to increasing global atmospheric concentrations of these gases, which global average net effect leads to global warming (UNFCCC, 1992, 1997; IPCC, 2007). In recent years, odour emissions from animal housing and land application of manure are being increasingly considered a nuisance in densely populated countries as the scale of livestock operations expands and an increasing number of rural residential developments are built in traditional farming areas (EC, 2003). Furthermore, the emission of particulate matter from animal houses has received attention since a few years as the inhalation of dust affects human health.

In order to reduce the environmental impacts of livestock production both national and international regulations went into effect that deal with these issues.

[1] Ammonia emission contributes minimally to odour emission because the odour threshold of ammonia is relatively high, *viz* 5 ppm (Amoore and Hautala, 1983), which is unlikely to be found outside animal houses. Therefore, emission of ammonia and odour are discussed as separate issues.

This thesis focuses on one of these issues: the abatement of gaseous emissions from from animal houses.

1.2 Gaseous emission abatement in livestock operations

Generally speaking, three different approaches can be distinguished in order to reduce the emission of gaseous compounds (such as ammonia, greenhouse gases, and odour) from animal houses to the atmosphere (EC, 2003):

(1) feed management
(2) adaptation of housing system design, including inside manure storage
(3) end-of-pipe air treatment

1.2.1 Feed management

For pigs and cows, where urine and faeces are excreted separately, the nitrogen excreted in the urine is predominant in the form of urea, which can easily be converted into ammonia and carbon dioxide by the enzyme urease that is present in faeces, thus resulting in emission of ammonia. Nitrogen excreted in faeces is mainly present as protein, which is less susceptible to decomposition into ammonia. For poultry, the urine and faeces are excreted together through the cloaca.

Feed management, or nutritional management, aims either to reduce the nitrogen excretion in faeces and/or urine by matching the amount and composition of feed more closely to animal requirements at various production stages, or by shifting nitrogen excretion from urine to faeces by increasing fibrous feedstuffs in the diet. Furthermore, adaptations of the diet may induce a decrease of urine and slurry pH. The use of these strategies can reduce the ammonia emission both for pigs (Cahn *et al.*, 1998a, 1998b, 1998c, 1998d; Kim *et al.*, 2004), poultry (Middelkoop and Harn, 1998; Angel *et al.*, 2008; Elwinger and Svensson, 1996) and dairy cattle (Smits *et al.*, 1995; Duinkerken *et al.*, 2005).

For pigs and poultry, feed management may reduce the emission of ammonia to the atmosphere up to a maximum of about 50% compared to standard feed composition. However, feed management for ammonia abatement may negatively affect the emission of methane and nitrous oxide during storage and after land application of the manure (Velthof *et al.*, 2005). For pigs it was shown that dietary approaches for ammonia emission reduction may not be effective for odour emission at the same time (Le, 2006). However, alteration of feed composition can be an effective tool for abatement of odour emission from pig manure (Le *et al.*, 2005, 2007); Le *et al.* (2007) reported an odour emission reduction of 80% after a drastic reduction of dietary crude protein feed levels.

For ruminants, feed management can also be used to reduce the emission of methane from the digestive system (Monteny *et al.*, 2006; Tamminga *et al.*, 2007).

1.2.2 Housing system

The design of a housing system, *i.e.* the combination of the floor-system, manure collection and the manure removal system, determines to a large extent the level of the emission of gaseous compounds, especially the emission of ammonia. Housing systems that have been developed to reduce ammonia emissions from pigs and cows basically involve one or more of the following abatement principles (Starmans and Hoek, 2007)[1]:

(1) reduction of emitting manure surface (Aarnink, 1997);
(2) fast and complete removal of the liquid manure from the pit to an external slurry storage (Groenestein and Montsma, 1993);
(3) applying an additional treatment, such as aeration, to obtain flushing liquid (Voermans *et al.*, 1990);
(4) cooling the manure surface (Groenestein and Huis in 't Veld, 1996);
(5) changing the chemical/physical properties of the manure, such as decreasing the pH (Hoeksma *et al.*, 1993).

For poultry, the main abatement principles that are applied are as follows (Starmans and Hoek, 2007):

(1) manure/litter management: regularly removing the excrements and litter to a closed storage area (Beurskens *et al.*, 2002; Scheer *et al.*, 2002);
(2) drying of manure (Kroodsma *et al.*, 1985; Reuvekamp and Niekerk, 1997; Huis in 't Veld *et al.*, 1999);

Housing systems that have been developed on the basis of these principles have proved to be able to reduce their ammonia emissions to the atmosphere by 30% to 80% when compared to conventional housings. In the Netherlands, animal housing

---

[1] For pig and cattle production most of these abatement technologies will result in a higher ammonia content of the liquid manure; this potentially leads to an increase of ammonia and nitrous oxide emission from storage and after land application of manure (Huijsmans, 2003; Huijsmans *et al.*, 2001, 2003; Velthof *et al.*, 2005).

systems and their ammonia emission levels are published in a regulatory list (VROM, 2002).

Brink *et al.* (2001) estimated for Europe that animal housing adaptations for ammonia abatement hardly affect the emission of methane but may increase nitrous oxide emissions significantly. The effect of animal housing adaptations on odour emission was demonstrated but is usually limited (Ogink and Lens, 2000; Mol and Ogink, 2002).

Furthermore, control of the indoor climate in terms of reducing air velocity at the manure surface, which decreases mass transfer at the manure-air interface (Aarnink and Elzing, 1998; Monteny *et al.*, 1999), and having relatively low indoor temperatures, which results in less fouling of floors, especially for pigs (Aarnink *et al.*, 2006), can reduce ammonia and odour emissions to the atmosphere even further, as emitting surface is reduced.

1.2.3 End-of-pipe air treatment

General description

Another approach for emission reduction is treatment of the ventilation air of a mechanically ventilated animal house. In such an end-of-pipe technique the house design and management inside the house remains essentially unaffected and is considered as a given emission source. End-of-pipe air treatment techniques that are applied nowadays for treatment of exhaust air in livestock operations include two types of air scrubbers: acid scrubbers and biotrickling filters. The main purpose of these scrubbers is ammonia abatement; the scrubber systems are commercially available and considered as off-shelf techniques in countries such as the Netherlands and Germany.

The following definitions are used throughout this thesis.

Definition:

Both the wordings "(bio)scrubber" and "(bio)trickling filter" are used as equivalent for describing a packed tower filter with an inert packing material that is usually continuously wetted. Such a system is illustrated in Figure 1.1 and can either be an acid scrubber or a bioscrubber. For a bioscrubber (or biotrickling filter) this implies that both absorption of the pollutant and biological conversion take place in the same compartment.

The wording "biofilter" is used to describe a system with an organic-based packing material and a low water flow, such as is presented in chapter 5.

A packed tower air scrubber, or trickling filter, is a reactor that has been filled with an inert or inorganic packing material (Figure 1.1). The packing material usually has a large porosity, or void volume, and a large specific area. Water is distributed on top of the packed bed which is consequently wetted. Contaminated air is introduced, either horizontally (cross-current) or upwards (counter-current), resulting in intensive contact between air and water enhancing mass transfer from gas to liquid phase. Usually a fraction of the trickling water is continuously recirculated; another fraction is discharged and replaced by fresh water (Groenestijn and Hesselink, 1993; Kennes and Thalasso, 1998; Devinny *et al.*, 1999; Burgess *et al.*, 2001; Kennes and Veiga, 2001; Sharefdeen and Singh, 2005).

For a given compound, the mass transfer rate from gas to liquid phase under equilibrium conditions is determined by several factors, that include the partition coefficient, the concentration difference between gas and liquid phase, the air and liquid flow rate, the size of the contact area between gas and liquid phase, and the contact time of gas and liquid phase (Coulson *et al.*, 1999; Richardson *et al.*, 2002; Riet and Tramper, 1991).

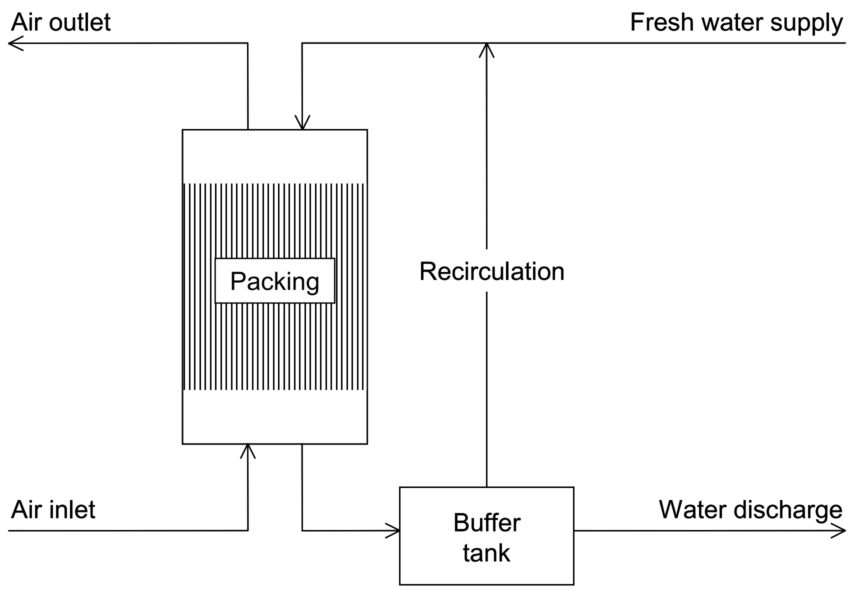


Figure 1.1 Schematic of a counter-current packed tower trickling filter.

For ammonia, the concentration in the liquid phase, NH_3 (aq), is mainly determined by the ammonia concentration in the gas phase and the pH driven dissociation into ammonium (NH_4^+) and hydroxide (OH^-) ions and, if applicable, by the transformation of ammonium into other compounds. In an acid scrubber the pH of the liquid phase is kept at low levels by addition of acid and the ammonium salt is removed from the system with the discharge water. In a biotrickling filter, ammonium is removed by bacterial conversion to nitrite (or nitrous acid) and nitrate (or nitric acid); this process is called nitrification (Focht and Verstraete, 1977; Prosser, 1986). The bacterial population, or biomass, in the system grows partly as a film on the packing material and is partly suspended in the water that is being recirculated. The accumulated nitrite and nitrate is removed with the discharge water.

The discharge water from a scrubber might be used for nitrogen fertilization of crops; sometimes the water is added to the liquid manure storage. The discharge water from a biotrickling filter might be treated in a denitrification process in order to decrease the nitrogen content which means that the water can be usually discharged at lower costs.

Weaknesses and strengths

In comparison with the emission reduction principles of feed management and adaptation of housing system design, which can be considered as source measures, an advantage of end-of-pipe technique air treatment techniques is that very high reductions of ammonia emission can be achieved, up to nearly 100% for acid scrubbers. This aspect makes application of air scrubbing systems suitable even for situations with very stringent emission limits. The application of air treatment techniques is limited to mechanically ventilated animal houses but as an end-of-pipe technique air treatment may also be applied in addition to feed management measures and housing design adaptations. However, increase of robustness and process stability, resulting in long-term reliable removal efficiencies, are aspects that still need further research and development, as is indicated by the long-term performance measurements reviewed and presented in chapters 3 and 6.

Investment and operational costs of scrubber systems for livestock operations (Melse and Willers, 2004; Arends *et al.*, 2008) are generally considered as high and therefore it is desirable to improve and further develop the currently commercially available techniques. The main elements determining the yearly costs of air scrubbing are fixed costs (depreciation, interest, maintenance) and energy use. The fixed costs are correlated to the size of the scrubber installation; the energy use is determined by the (continuously) running spraying pumps and the increased energy use of the mechanical ventilation system, which has to compensate for the additional backpressure caused by the scrubber system. For acid scrubbers the costs of chemical use, *i.e.* sulphuric acid, and for biotrickling filters water discharge costs are the next important cost factors (Melse and Willers, 2004) [1].

Besides ammonia, odorous compounds might also be removed by the air scrubber system to some extent. Generally speaking, the odour removal efficiency of an air treatment that has been designed for ammonia removal solely, is on average 30% for acid scrubbers and 45% for biotrickling filters (Ogink and Lens, 2001; Mol and Ogink, 2002), although reported ranges of individual measurements are wide. Especially odorous compounds that are well-soluble, easily biodegradable (in case of a biotrickling filter), or alkaline (in case of an acid scrubber) will be removed from the air relatively easily.

Furthermore, air scrubbing might partly remove dust or particulate matter (PM) from the air[2]. Especially the removal of PM10 and PM2.5[3] is relevant as the inhalation of these fractions affect human health. Recent measurements of particulate matter removal by air scrubbers treating animal house exhaust air showed an average removal efficiency ranging from 62 to 93% for PM10 and from 47 to 90% for PM2.5 (Aarnink *et al.*, 2007, 2008a, 2008b; Zhao *et al.*, 2008; Ogink and Hahne, 2007)[4]. These data suggest that end-of-pipe air treatment may be of major importance for compliance with current and future PM10 and PM 2.5 standards.

---

[1] The costs of scrubber systems are presented in detail in Table 3.5. The costs of water discharge, if any, largely depends on the local situation. Melse and Willers (2004) assume a discharge cost of EUR 12.60 per m^3 for the Netherlands, both for discharge water from biotrickling filters and acid scrubbers. For other countries different cost figures might apply.

[2] Dust from the ventilation air will partly accumulate in the scrubber and might cause channeling of air and an increase of pressure drop. In case of a biotrickling filter, the accumulation of solids might be further increased by bacterial growth (biomass). Usually a scrubber needs to be cleaned once or twice a year to ensure normal operation.

[3] PM10 (also called thoracic particles) represents the fraction of particles that have an aerodynamic diameter of 10 μm or less; PM2.5 (also called fine particles) is used to describe the particles fraction with an aerodynamic diameter of 2.5 μm or less. The aerodynamic diameter is the diameter of a spherical particle having a density of 1 kg/m^3 that has the same terminal settling velocity in the gas as the particle of interest.

[4] These measurements were carried out on multi-stage air scrubbers. Whereas the common single-stage acid and biological air scrubbers have been designed for ammonia removal, multi-stage air scrubbers also aim to achieve significant emission reduction of odour and particulate matter (PM 2.5 and PM 10). Multi-stage scrubbers can be considered as a new generation of air scrubbers and research and development in this field has only started recently (Ogink and Bosma, 2007; Arends *et al.*, 2008; Melse *et al.*, 2008). Data on PM removal by single-stage acid scrubbers and biological air scrubbers that have been designed just for ammonia removal is currently not available.

In the past, also biofilters with organic-based packing materials were tested and applied for ammonia and odour removal from exhaust air from livestock operations in several European countries. However, at air residence times that are normally applied in biofilter systems, packing life span is limited due to the relatively high NH_3 and dust concentrations of this air, and uniform humidification of the packing is difficult. That is why nowadays mostly trickling systems with inorganic packing are applied for treatment of exhaust air from animal houses in Europe. However, after ammonia has been removed from the air, biofiltration can be effectively used as a polishing step for effective odour removal.

Because of the low water solubility of methane, usually scrubber systems do not affect the methane concentration of the treated exhaust air. At high air residence times, however, significant removal of methane from this air might be achieved by bacterial oxidation in a biofilter system (Park *et al.*, 2002; Streese and Stegmann, 2003). Furthermore, some nitrous oxide (N_2O) might be formed as a by-product of nitrification and denitrification in biological air treatment systems (Rogers and Whitman, 1991; Trimborn, 2006).

1.3 Objectives and outline of this thesis

With ever stricter emission limits the successful application of end-of-pipe air treatment technologies in livestock farming is of increasing importance, as alternative emission reduction principles (such as feed management and adaptation of housing system design) are in some cases not sufficient. However, current applications of scrubber technology in livestock operations seem to be unsatisfactory with regard to long-term process robustness, removal efficiency, and cost-efficiency.

The aim of this thesis is to better understand and improve the performance of air scrubber systems in livestock operations where they are applied at mechanically ventilated animal houses. The following objectives are defined:

(1) to determine how air scrubbers are performing at livestock farms, with regard to ammonia and odour removal (chapters 3, 6, and 7);

(2) to determine how the cost-efficiency of air scrubbers at livestock farms can be increased by applying a different treatment strategy (chapter 4);

(3) to determine if, in addition to ammonia and odour, methane can be removed by biological air treatment systems (chapter 5);

(4) to discuss the problems and shortcomings related to current air scrubbing practices and make recommendations for possible improvement and future research (chapter 8).

Below a short summary is given of the content of each chapter.

Chapter 2 ("Policy on emission control related to livestock production") gives an overview of emission regulations and emission targets for gaseous compounds that are relevant for livestock production and emission abatement. These regulations are the driving force for application and research and development in the field of air treatment techniques as livestock operations must comply with increasingly stringent emission standards.

Chapter 3 ("Air scrubbing techniques for ammonia and odor reduction at livestock operations: Review of on-farm research in the Netherlands") summarizes and discusses research and experiences that have been gathered in the Netherlands for the last 20 years in the field of acid scrubbers and biotrickling filters for treatment of exhaust air from mechanically ventilated animal houses. Also available international literature is discussed. This chapter can be considered as a description of the state-of-the-art of air scrubbing techniques for livestock farms.

Chapter 4 ("Size reduction of ammonia scrubbers for pig and poultry houses: Use of conditional bypass vent at high air loading rates") aims to significantly increase the cost-effectiveness of air scrubbing for ammonia removal. A novel approach is presented in which scrubber design parameters are adapted and air loading control is introduced.

Chapter 5 ("Biofiltration for mitigation of methane emission from animal husbandry") deals with the subject of the negligible methane removal capacity that is usually experienced in scrubbing systems and presents the results of methane oxidation in a pilot-scale biofilter. Also the feasibility of this technique for application on farm-scale is discussed.

Chapters 6 and 7 are case-studies. Chapter 6 ("Odour and ammonia removal from pig house exhaust air using a biotrickling filter") describes and discusses the field performance of a specific biotrickling filter that is operated on a pig farm. Chapter 7 ("Evaluation of four farm-scale systems for the treatment of liquid pig manure") illustrates a broader range of application of ammonia scrubbers within agricultural practice by presenting several liquid manure treatment systems that comprise an ammonia scrubber. Furthermore, this chapter presents an evaluation of the technical performance and economics of the manure treatment systems, including gaseous emissions.

Finally, chapter 8 ("General discussion") summarizes the main findings of this thesis, discusses the problems and shortcomings related to current air scrubbing practices and makes recommendations for possible improvement and future research.

References

Aarnink, A.J.A. (1997). Ammonia emission from houses for growing pigs as affected by pen design, indoor climate and behaviour. Ph.D. thesis Agricultural University Wageningen, Wageningen, Netherlands; 175 pp.

Aarnink, A.J.A.; A. Elzing (1998). Dynamic model for ammonia volatilization in housing with partially slatted floors, for fattening pigs. Livest. Prod. Sci., Vol 53, No 3, pp 153-169.

Aarnink, A.J.A.; J.W. Schrama; M.J.W. Heetkamp; J. Stefanowska; T.T.T. Huynh (2006). Temperature and body weight affect fouling of pig pens. Journal of Animal Science, No 84, pp 2224-2231.

Aarnink, A.; Y. Zhao; T. van Hattum; A. Hol (2007). Reductie fijn stof emissie uit een varkensstal door de combi-wasser van Big Dutchman. Animal Sciences Group van Wageningen UR, Lelystad, Netherlands; ISSN 1570 8616.

Aarnink, A.; Y. Zhao; T. van Hattum; A. Hol (2008a). Reductie fijn stof emissie uit een varkensstal door de combi-wasser van Uniqfill. Confidential report. Animal Sciences Group van Wageningen UR, Lelystad, Netherlands.

Aarnink, A.; Y. Zhao; T. van Hattum; A. Hol (2008b). Reductie fijn stof emissie uit een varkensstal door de combi-wasser van Inno+/Siemers. Confidential report. Animal Sciences Group van Wageningen UR, Lelystad, Netherlands.

Amoore, J.E.; E. Hautala (1983). Odor as an aid to chemical safety. Odor thresholds compared with threshold limit values and volatilities for 214 industrial chemicals in air and water dilution. Journal of Applied Toxicology, Vol 3, No 6, pp 272-290.

Angel, R.; W. Powers; T. Applegate (2008). Diet impacts for mitigating air emissions from poultry. Proceedings of the 8th International Livestock Symposium (ILES VIII), August 31 - September 4, 2008, Iguassu Falls, Brazil.

Arends, F.; G. Franke; E. Grimm; W. Gramatte; S. Häuser; J. Hahne (2008). Exhaust Air Treatment Systems for Animal Housing Facilities, Techniques - Performance - Costs. KTBL-Schrift 464, KTBL, Darmstadt, Deutschland; ISBN 978-3-939371-60-1.

Beurskens, A.G.C.; J.M.G. Hol; G. Mol (2002). Onderzoek naar de ammoniak- en geuremissie van stallen LIII. Stal voor vleeskalkoenen met frequente strooiselverwijdering. Rapport 2002-14, IMAG, Wageningen, Netherlands.

Brink, C.; C. Kroeze; Z. Klimont (2001). Ammonia abatement and its impact on emissions of nitrous oxide and methane, Part 2: application for Europe. Atmospheric Environment, No 25, pp 6313-6325.

Burgess, J.E.; S.A. Parsons; R.M. Stuetz (2001). Developments in odour control and waste gas treatment biotechnology: a review. Biotechnol. Adv., No 19, pp 35-63.

Canh, T.T.; A.J.A. Aarnink; Z. Mroz; A.W. Jongbloed; J.W. Schrama (1998a). Influence of electrolyte balance and acidifying calcium salts in the diet of growing-finishing pigs on urinary pH, slurry pH and ammonia volatilisation from slurry. Livest. Prod. Sci., Vol 56, No 1, pp 1-13.

Canh, T.T.; A.J.A. Aarnink; J.B. Schutte; A. Sutton; D.J. Langhout; M.W.A. Verstegen (1998b). Dietary protein affects nitrogen excretion and ammonia emission from slurry of growing-finishing pigs. Livest. Prod. Sci., Vol 56, No 3, pp 181-191.

Cahn, T.T.; A.J.A. Aarnink; M.W.A. Verstegen; J.W. Schrama (1998c). Influence of dietary factors on the pH and ammonia emission of slurry from growing-finishing pigs. J. Anim. Sci., Vol 76, pp 1123-1130.

Cahn, T.T.; A.L. Sutton; A.J.A. Aarnink; M.W.A. Verstegen; J.W. Schrama, G.C.M. Bakker (1998d). Dietary carbohydrates alter the fecal composition and pH and the ammonia emission from slurry of growing pigs. J. Anim. Sci., Vol 76, pp 1887-1895.

CBS (2007). StatLine Databank; Statistics Netherlands website, http://www.cbs.nl.

Coulson, J.M.; J.F. Richardson; J.R. Backhurst; J.H. Harker (1999). Coulson & Richardson's Chemical Engineering: Fluid Flow, Heat Transfer and Mass Transfer. Chemical Engineering Vol. 1. Butterworth-Heinemann. ISBN 0-7506-4444-3.

Devinny, J.S.; M.A. Deshusses; T.S. Webster (1999). Biofiltration for air pollution control. Lewis Publishers, Boca Raton, ISBN 1-56670-289-5.

Duinkerken, van, G.; G. André; M.C.J. Smits; G.J. Monteny; L.B.J. Šebek (2005). Effect of rumen-degradable protein balance and forage type on bulk milk urea concentration and emission of ammonia from dairy cow houses. J. Dairy Sci., Vol 88, pp 1099-1112.

Elwinger, K.; L. Svensson (1996). Effect of dietary protein content, litter and drinker type on ammonia emission from broiler houses, Journal of Agricultural Engineering Research, Vol 64, No 3, pp 197-208.

EC (2003). Integrated Pollution Prevention and Control (IPPC). Reference Document on Best Available Techniques for Intensive Rearing of Poultry and Pigs; European Commission; http://ec.europa.eu/comm/environment/ippc/brefs/ilf_bref_0703.pdf.

Focht, D.D.; W. Verstraete (1977). Biochemical ecology of nitrification and denitrification. Advances in Microbial Ecology 1, pp 135-214.

Groenestein, C.M.; J.H.W. Huis in 't Veld (1996). Praktijkonderzoek naar de ammoniakemissie van stallen XIX. Vleesvarkensstal met koeling van mestoppervlak in de kelder. Rapport 96-1003, IMAG-DLO, Wageningen, Netherlands.

Groenestein, C.M.; H. Montsma (1993). Praktijkonderzoek naar de ammoniakemissie van stallen IIIa. Aanvullend onderzoek aan een biggenopfokstal met frequente en restloze mestverwijdering. Rapport 93-1001, IMAG-DLO, Wageningen, Netherlands.

Groenesteijn, van, J.W.; P.G.M. Hesselink (1993). Biotechniques for air pollution control. Biodegradation, No 4, pp 283-301.

Heij, G.J.; J.W. Erisman (eds) (1995). Acid Rain Research: Do We Have Enough Answers? Proceedings of a Speciality Conference, 's-Hertogenbosch, Netherlands, 10-12 October 1994. Studies in Environmental Science, 64; Elsevier, Amsterdam, Netherlands; 516 pp.

Heij, G.J.; J.W. Erisman (1997). Acid Atmospheric Deposition and its Effects on Terrestrial Ecosystems in the Netherlands. The Third and Final Phase (1991-1995); Studies in Environmental Science, 69; Elsevier, Amsterdam, Netherlands; 716 pp.

Hoeksma, P.; N. Verdoes; G.J. Monteny (1993). Two options for manure treatment to reduce ammonia emission from pig housing. In: Proceedings of the Congress on Nitrogen Flow in Pig Production and Environmental Consequences; M.W.A. Verstegen M W A, L.A. den Hartog, G.J.M. van Kempen, J.H.M. Metz (eds); EAAP Publication No. 69, Wageningen, Netherlands, pp 301-306.

Huijsmans, J.F.M. (2003). Manure application and ammonia volatilization. PhD thesis Wageningen University with summaries in English and Dutch, Wageningen, Netherlands; ISBN 90-5808-937-1; pp 160.

Huijsmans, J.F.M.; J.M.G. Hol; M.M.W.B. Hendriks (2001). Effect of application technique, manure characteristics, weather and field conditions on ammonia volatilization from manure applied to grassland. Netherlands Journal of Agricultural Science, No 49, pp 323-342.

Huijsmans, J.F.M.; J.M.G. Hol; G.D. Vermeulen (2003). Effect of application method, manure characteristiscs, atmosphere and field conditions on ammonia volatilization from manure applied to arable land. Atmospheric Environment , No 37, pp 3669-3680.

Huis in 't Veld, J.W.H.; P.W.G. Groot Koerkamp; R. Scholtens (1999). Onderzoek naar de ammoniakemissie van stallen XLVI. Voletage volièresysteem voor legouderdieren en een droogtunnel met geperforeerde mestbanden. Rapport 99-10, IMAG, Wageningen, Netherlands.

IPCC (2007). Climate change 2007. The physical science basis. Contribution of working group I to the fourth assessment report of the Intergovernmental Panel on Climate Change; Salomon, S. et al., eds.; Cambridge University Press: Cambridge, U.K.; ISBN 978-0-521-88009-1 ISBN 978-0-521-70596-7; 996 pp; http://www.ipcc.ch/ipccreports/ar4-wg1.htm.

Kennes, C.; F. Thalasso (1998). Waste gas biotreatment technology (review). J. Chem. Technol. Biotechnol., No 72, pp 303-319.

Kennes, C.; M.C. Veiga (eds) (2001). Bioreactors for waste gas treatment. Kluwer Academic Publishers, Dordrecht, Netherlands, ISBN 0-7923-7190-9.

Kim, I.B.; P.R. Ferket; W.J. Powers; H.H. Stein; T.A.T.G. van Kempen (2004). Effects of different dietary acidifier sources of calcium and phosphorus on ammonia, methane and odorant emission from growing-finishing pigs. Asian-Australas. J. Anim. Sci., Vol 17, No 8, pp 1131-1138.

Kroodsma, W.; J. Arkenhout; J.A. Stoffers (1985). New systems for drying poultry manure in belt batteries. IMAG Research Report 85-1, Wageningen, Netherlands, 27 pp.

Le, P.D. (2006). Odor from pig production: Its relation to diet. PhD thesis Wageningen University, Wageningen, Netherlands.

Le, P.D.; A.J.A. Aarnink; N.W.M. Ogink; P.M. Becker; M.W.A. Verstegen (2005). Odour from animal production facilities: its relation to diet. Nutrition Research Reviews, No 18, pp 3-30.

Le, P.D.; A.J.A. Aarnink; A.W. Jongbloed; C.M.C. Van der Peet-Schwering; N.W.M. Ogink; M.W.A. Verstegen (2007). Effects of dietary crude protein level on odour from pig manure. Animal, No 1, pp 734-744.

Melse, R.W.; H.C. Willers (2004). Toepassing van luchtbehandelingstechnieken binnen de intensieve veehouderij, Fase 1: Techniek en kosten; Report 029; Agrotechnology & Food Innovations, Wageningen UR, Wageningen, Netherlands. ISBN 90-6754-739-5.

Melse, R.W.; N.W.M. Ogink; B.J.J. Bosma (2008). Multi-pollutant scrubbers for removal of ammonia, odor, and particulate matter from animal house exhaust air. Proceedings of the Mitigating Air Emissions from Animal Feeding Operations Conference, May 19 - 21, 2008, Des Moines, Iowa (IA), USA.

Middelkoop, J.H. van; J. van Harn (1998). The influence of reduced protein levels in broiler feed on NH_3 emissions. Translation Silsoe Research Institute, Vol 66, No 34.

Mol, G.; N.W.M. Ogink (2002). Geuremissie uit de veehouderij II, Overzichtsrapportage 2000 -2002; Rapport 2002-09; IMAG, Wageningen, Netherlands.

Monteny, G.J.; A. Bannink; D. Chadwick (2006). Greenhouse gas abatement strategies for animal husbandry. Agriculture, Ecosystems and Environment , Vol 112, No 2-3, pp 163-170.

Monteny, G.J.; D.D. Schulte; A. Elzing; E.J.J. Lamaker (1999). A conceptual mechanistic model for the ammonia emissions from free stall cubicle dairy cow houses. Transaction of the ASAE, Vol 41, pp 193-201.

Ogink, N.W.M.; B.J.J. Bosma (2007). Multi-phase air scrubbers for the combined abatement of ammonia, odor and particulate matter emissions. International Symposium on Air Quality and Waste Management for Agriculture, Broomfield, Colorado, USA. CD-Rom Proceedings of the 16-19 September 2007. ASABE Publication Number 701P0907cd.

Ogink, N.W.M.; J. Hahne (2007). Removal of dust fractions by air scrubbers in livestock operations. Proceedings of DustConf 2007, International Conference in Maastricht, Netherlands, 23-24 April 2007; http://www.dustconf.com.

Ogink, N.W.M.; P. Lens (2001). Geuremissie uit de veehouderij, Overzichtsrapportage 1996-1999; Rapport 2001-14; IMAG, Wageningen, Netherlands.

Park, S.; K.W. Brown; J.C. Thomas (2002). The effect of various environmental and design parameters on methane oxidation in a model biofilter. Waste Manage Res., Vol 20, pp 434-444.

Prosser, I.J. (ed.) (1986). Nitrification. Society for General Microbiology special publication 20. IRL Press, Oxford, U.K., ISBN 1-85221-001-X and 1-85221-013-3.

Reuvekamp, B.E.J.; Th.G.C.M. van Niekerk (1997). Ammoniakemissie bij leghennen op batterijen bij drogen tot minimaal 55% drogestof en bij natte mest. PP-uitgave no. 63, Praktijkonderzoek Pluimveehouderij, Beekbergen, Netherlands.

Richardson, J.F.; J.R. Backhurst; J.H. Harker (2002). Coulson & Richardson's Chemical Engineering: Particle technology and separation processes. Chemical Engineering Vol. 2. Butterworth-Heinemann, ISBN 0-7506-4445-1.

Riet, van 't, K.; J. Tramper (1991). Basic bioreactor design. New York, U.S.A, Marcel Dekker Inc, ISBN 0-8247-8446-4.

Rogers, J.E. ; W.B. Whitman (eds) (1991). Microbial production and consumption of greenhouse gases: methane, nitrogen oxides, and halomethanes. American Society for Microbiology; ISBN 1555810357; 298 pp.

Sharefdeen, Z.; A. Singh (eds) (2005). Biotechnology for Odor and Air Pollution Control. Springer, Berlin/Heidelberg/New York, ISBN 3-540-23312-1.

Scheer, A.; J.M.G. Hol; G. Mol (2002). Onderzoek naar de ammoniak- en geuremissie van stallen LVI. Scharrelstal voor leghennen met frequente mest- en strooiselverwijdering. Rapport 2002-17, IMAG, Wageningen, Netherlands.
Smits, M.C.J.; H. Valk; A. Elzing; A. Keen (1995). Effect of protein nutrition on ammonia emission from a cubicle house for dairy cattle. Livestock Production Science, Vol 44, No 6, pp 147-156.
Starmans, D.A.J.; K.W. Van der Hoek (eds) (2007). Ammonia, the case for the Netherlands. Wageningen Academic Publishers, Wageningen, Netherlands; ISBN 978-90-8686-028-9; 201 pp.
Streese, J.; R. Stegmann (2003). Microbial oxidation of methane from old landfills in biofilters. Waste Manage., Vol 23, pp 573-580.
Tamminga, S.; A. Bannink; J. Dijkstra; R. Zom (2007). Feeding strategies to reduce methane losses in cattle. Report 34. Animal Sciences Group van Wageningen UR, Lelystad, Netherlands, pp 46.
Trimborn, M. (2006). Biofilter/Biowäscher an Tierhaltungslagen als relevante Quelle von Lachgas durch Ammoniakabscheidung?; Schriftenreihe des Lehr- und Forschungsschwerpunktes USL, No 138; 59 pp; Landwirtschaftliche Fakultät der Universität Bonn, Bonn, Deutschland.
UNFCCC (1992). United Nations Framework Convention on Climate Change. Convention text, UNEP/WMO Information Unit of Climate Change (IUCC) on behalf of the Interim Secretariat of the Convention; IUCC, Geneva, Switzerland.
UNFCCC (1997). Kyoto Protocol to the United Nations Framework Convention on Climate Change; FCCC/CP/L7/Add.1, 10 December 1997; UN, New York, NY, U.S.A.
Velthof, G.L.; J.A. Nelemans; O. Oenema; P.J. Kuikman (2005). Gaseous nitrogen and carbon losses from pig manure derived from different diets. J. Environ. Qual., Vol 34, pp 698-706.
Voermans, J.A.M.; J. Oosthoek; P. Hoeksma (1990). The NH_3-emission affected by flushing systems in pig houses. Agricultural and Food Processing Waste. Proceedings of the Sixth International Symposium of the ASAE, Chicago, Illinois, USA.
VROM (2002). Regeling Ammoniak en Veehouderij (RAV); 1 mei 2002; Staatsblad No 82; Ministry of Housing, Spatial Planning and the Environment, The Hague, Netherlands.
Zhao, Y.; A.J.A. Aarnink; P.W.G.G. Koerkamp; N.W.M. Ogink; M.C.M. de Jong (2008). Removal efficiency of dust and bacteria by multi-stage air scrubbers in pig houses Proceedings of the 8th International Livestock Symposium (ILES VIII), August 31 - September 4, 2008, Iguassu Falls, Brazil.

Chapter 2

Policy on emission control related to livestock production

Contents

An adapted version of this chapter has been published in Biosystems Engineering, 2009, volume 104, issue number 3, pages 289-298 (Melse, R.W.; N.W.M. Ogink; W.H. Rulkens. Overview of European and Netherlands' regulations on airborne emissions from intensive livestock production with a focus on the application of air scrubbers) and is reproduced with permission from Elsevier.

2.1 Ammonia

2.1.1 Gothenburg protocol

Since 1980, ammonia emitted from livestock production has become a major environmental concern in the Netherlands, because it was identified as one of the three main sources of soil acidification, together with nitrogen oxides (NO_x) and sulphur oxides (SO_x) (Heij and Erisman, 1995, 1997).

Besides emissions and depositions on a local scale, it has become clear that air pollutants, *e.g.* ammonia, can travel several thousands of kilometres before deposition and damage occurred. This implied that cooperation at the international level was necessary to solve problems such as acidification. In response to this, in 1979 the Convention on Long-range Transboundary Air Pollution (CLRTAP) was signed by 34 governments and the European Community. The convention was the first international legally binding instrument to deal with problems of air pollution on a broad regional basis. In 1999, the CLRTAP was extended by the Protocol to Abate Acidification, Eutrophication and Ground-level Ozone in Gothenburg, Sweden. At the beginning of 2007, this *Gothenburg protocol* was signed by 31 and ratified by 23 parties (UNECE, 2007).

The Gothenburg protocol sets emission ceilings for 2010 for four pollutants: sulphur, NO_x, VOCs and ammonia. These ceilings were negotiated on the basis of scientific assessments of pollution effects and abatement options. Once the Protocol is fully implemented, Europe's sulphur emissions should be cut by at least 63%, its NO_x emissions by 41%, its VOC emissions by 40% and its ammonia emissions by 17% compared to 1990 levels. The protocol requires that best available techniques are used to keep emissions down and describes specific measures that farmers will have to take to control ammonia emissions. For the European Union Member States, EU Directive 2001/81/EC sets the National Emission Ceilings (NEC) for these emissions (EC, 2001). The ammonia emission ceiling for the Netherlands has been set to 128 kton per year in 2010 by this directive.

The first review of the Gothenburg protocol was started in December 2005 and was completed by the end of 2007. This review might result in the addition of emission ceilings for other anthropogenic emissions to the protocol as well, *viz* emissions of greenhouse gases (CO_2, CH_4 and N_2O), carbon monoxide (CO), and primary particles (PM10 and PM2.5) (Sliggers and Bull, 2007; Sliggers, 2007).

2.1.2 EU directive on Integrated Pollution Prevention and Control (IPPC)

Integrated pollution control (IPC), as a principle of environmental protection and management, aims to minimize the overall environmental impact of human activities. IPC has been adopted by the European Union and been put in force as Integrated Pollution Prevention and Control (IPPC) by ratification of the EU Directive 96/61/EC (EC, 1996a). The IPPC directive sets common rules for permitting and controlling certain large-scale industrial and agricultural activities. Such activities need an operating permit that takes into account the whole environmental performance of the facility, covering *e.g.* emissions to air, water and land, generation of waste, use of raw materials, energy efficiency, local disturbance (odour, noise), prevention of accidents, and restoration of the site upon closure (EC, 2007a).

To gain a permit operators have to show that no significant pollution is caused and that the *Best Available Techniques* (BATs) are applied, taking into account the technical characteristics, geographical locations and local environmental circumstances of the installations concerned. "Best" means the most effective in achieving a high general level of protection of the environment as a whole. "Available" means those techniques that have been developed on a scale that allows implementation under economically and technically viable conditions, but does not necessarily mean that the technique has to be an industry standard or indeed widely available, as the intention of IPPC is to change practices to new, less polluting alternatives. "Techniques" includes both the technology used and the way in which the installation is designed, built, maintained, operated and decommissioned. This is in recognition of the fact that advanced technology can give poor results if managed poorly, just as good management can give acceptable results even if poor technology is used (Burton and Turner, 2003; EC, 1996a; Rostron, 2006). The environmental impacts of industry and agriculture can be significantly reduced through the use of new and more advanced techniques and technologies. Adoption of the BAT system aims to help to improve the management of material flows, increase energy-efficiency, and cut emissions.

The EU member states' authorities and industry cooperate on the development of so-called BAT reference documents (BREFs). A BREF assists the regulatory authorities and those applying for licences by describing reference techniques and reference levels for a specific economical sector. BREFs are meant to give information on achievable emission and consumption levels when using specified techniques and do not set legally binding standards. The application of techniques and the appropriate emission levels for any specific case will need to be determined taking into account the objectives of the IPPC directive and the local considerations

(EC, 2003a). For more than 30 different economical sectors BREFs have been published. One of the BREFs is dedicated to installations for intensive livestock production. This "Reference Document on Best Available Techniques for Intensive Rearing of Poultry and Pigs" (EC, 2003a) covers intensive livestock farms with more than 40,000 places for poultry, or 2,000 places for production pigs over 30 kg, or 750 places for sows. The BREF discusses good agricultural practice, nutritional management, housing systems, water and energy use, manure storage, manure processing, and land spreading of manure. The BATs that are described for intensive livestock production do currently not include end-of-pipe emission reduction techniques, such as air scrubbers and biofilters. These techniques were not considered BAT by the authors of this BREF because of economic (high running costs), ecologic (high energy consumption, chemical use of acid scrubbers, waste water production), and technical reasons (unstable performance of bioscrubbers) (VROM, 2007a; Hendriks, 2008). The BREFs are reviewed on a regular basis and updated if necessary. The BREF document for intensive rearing of poultry and pigs is expected to be updated from 2008 on.

2.1.3 National regulations and emission levels for the Netherlands

In the Netherlands, about 1,700 intensive livestock farms, about 50% pig and 50% poultry farms, have to comply with the IPPC directive (VROM, 2007b). The IPPC directive is implemented mainly in framework of the Environmental Management Act (VROM, 1979) and the Ammonia and Livestock Farming Act (VROM, 2002a). Based on these acts, several general binding rules are in force regarding gaseous emissions from livestock production:

(a) Farmers are obliged to cover their outdoor slurry storage facilities in order to reduce emissions of ammonia and odour, (VROM, 1990).

(b) For dairy farming an agreement was made with the Dutch Farmers Union (LTO) that aims to reduce the nitrogen content of the feed and thus of the urine (as indicated by milk urea content) by 2010; this might substitute the obligation to build low-ammonia-emission housing systems for dairy cattle (VROM, 2003a). In 2008 the progress of the agreement is evaluated.

(c) In a zone of 250 m around nature areas that have been labelled as vulnerable ecosystems, new livestock farms are not allowed and existing livestock farms may only expand if housing systems are applied with a very high reduction of ammonia emission (VROM, 2002a).

(d) Surface spreading of animal manure was gradually banned by virtue of the Soil Protection Act (VROM, 1986) and the Decree on the use of Manures

(VROM, 1987). From 1995 nearly all animal manure has been applied using low-ammonia-emission techniques.

(e) Maximum emission levels are imposed for animal houses in order to reduce ammonia (Accommodation Order in Council; VROM, 2005a, 2007c).

According to this Accommodation Order in Council (in Dutch: *Besluit Huisvesting*) all pig and poultry operations must move to low-ammonia-emission housing systems at the latest in 2010; in some specific situations the transition period has been extended to 2012 or 2013. For every animal category maximum emission levels are listed, expressed as kg NH_3/animal place/year. The housing systems that may be implemented in order to reach these levels are published in a regulatory list, called the Ammonia and Livestock Farming Regulation (VROM, 2002b). This regulation lists all legally acknowledged low-ammonia-emission housing systems, including end-of-pipe techniques such as air scrubbers.

From 1993 farmers and industry in the Netherlands were financially encouraged to develop and implement low emission housing systems on a voluntary basis. Systems with an ammonia emission reduction of 50% or higher (as compared to traditional housing systems) received a so-called *Green Label* award and were taken up in the regulation. This resulted in the development of a large variety of low-emission livestock housing systems, including systems for end-of-pipe treatment of exhaust air from animal houses, *viz* air scrubbers. Over 50 Green Label awards were issued and the implementation of low-emission housing systems increased, especially in pig and poultry farming (Starmans and Van der Hoek, 2007). Later on the Green Label awarding system was terminated and replaced by the Ammonia and Livestock Farming Regulation (VROM, 2002b) and newly developed low-emission housing systems are still added to this regulatory list.

With regard to end-of-pipe treatment of exhaust air, the Ammonia and Livestock Farming Regulation distinguishes three types of low-ammonia-emission systems (VROM, 2002b):

(1) acid air scrubbers; assigned ammonia emission reduction is either 70%, 90%, or 95%;

(2) biological air scrubbers (or bioscrubber or biotrickling filter); assigned ammonia emission reduction is 70%;

(3) multi-stage air scrubbers (or combi-scrubber); assigned ammonia emission reduction is either 70%, or 85%.

The ammonia emission rate (kg NH_3/animal place/year) for a scrubber system is calculated by multiplication of the ammonia emission rate for a conventional housing system without a scrubber with the assigned reduction percentage for the scrubber system, as mentioned above.

When a manufacturer has developed a new scrubber system or redesigned an existing system and wants to market the system in the Netherlands, usually an application for inclusion in the regulatory list with low-ammonia-emission systems is submitted at the Ministry of the Environment (VROM). Usually the application is accompanied either by a theoretical assessment of the design and performance of the system, in case the application concerns a current scrubber design and the aimed ammonia removal efficiency is 70% or less (Melse and Willers, 2005), or a set of performance measurements according to a prescribed measurement protocol. A technical committee evaluates the application and advises the Ministry either to include the system in the regulatory list, to ask for additional measurements and information, or to reject the application.

In the last 25 years considerable efforts were put into the development of NH_3 abatement techniques in animal operations in the Netherlands. Since 1980 the ammonia emission from agriculture has been reduced by nearly 50%. In 2005 the total ammonia emission in the Netherlands was estimated at 133 kton (CBS, 2007) which is slightly higher than the NEC of 128 kton for 2010. The NH_3 emission from livestock production, however, still accounts for about 90% of the total ammonia emission and 50% of the total emission of all acidifying compounds (Koch *et al.*, 2003; EDC, 2007). The Dutch government aims to reduce the ammonia emission even further than the NEC in order to reduce the deposition of ammonia especially in nature areas. The Fourth National Environmental Policy Plan (NEPP4) lays down a target of 100 kton as the ammonia emission for 2010 (VROM, 2001a).

A recent study (Hettelingh *et al.*, 2007) that assesses the areas at risk of acidification and eutrophication in Europe and the potential evolution over time suggests that about 95% of ecosystems still at risk of acidification in 2010, could recover by 2030 if acid deposition is reduced in line with present legislation, *i.e.* implementation of the Gothenburg protocol and the NEC Directive.

2.2 Odour

2.2.1 National regulations and emission levels for the Netherlands

In the early 1990's, 23% of the Dutch population suffered odour nuisance from traffic and/or industry and 16% from agriculture (CBS, 2003)[1]. In 1998, the two major causes of odour nuisance from agriculture in the Netherlands were the application of manure and the emission from animal houses, respectively (Jong *et al.*, 2000). The policy objective in the Second National Environmental Policy Plan (NEPP2) (VROM, 1993) was that in 2000 no more than 12% of the Dutch population would suffer odour nuisance from traffic, industry and agriculture. However, in 2000 still 15% of the population suffered odour nuisance from traffic and/or industry and 10% from agriculture (CBS, 2003). In addition to the odour nuisance objective for 2000, another objective states that, in 2010, the Dutch population should no longer experience any severe odour nuisance[2]. No new objectives for odour nuisance have been set out since.

In the Netherlands, from 1972 until 2006 regulations have been in force to control odour emission from livestock buildings that were based on the use of specified setback distances between new or expanding livestock operations and odour-sensitive objects (VROM, 1985a, 1985b, 1996, 2002c, 2003b). Odour emissions from livestock farms were calculated on the basis of the number of animals present and so-called conversion values, which convert the odour emission for each species and category of livestock to a standard unit that corresponds to the odour emission of one fattening pig place per year. Next a distance chart gives the minimum allowable distance between the farm with its calculated odour emission and an odour-sensitive object, *e.g.* a house (Klarenbeek and Harreveld, 1995).

Since 2007 a new odour act is in force, the Odour Nuisance and Livestock Farming Act (VROM, 2006a). The act replaces the use of rigid setback distances by introduction of a prescribed odour dispersion model that calculates the odour

---

[1] Odour nuisance is defined as experiencing frequent or occasional nuisance from stench, in line with the questions asked in the "Ongoing Survey of Living Conditions" (CBS, 2003). Sources of odour included in the survey are road traffic, industry or business, agriculture and open fires/multi-burners.

[2] The definition of severe odour nuisance is based on the periodic nuisance survey conducted by TNO (Jong *et al.*, 2000). This examines to what extent people perceive a specific source in the residential environment to be a nuisance, based on a scale of 10 points ranging from 1 (no nuisance at all) to 10 (extreme nuisance). People giving answers in the 8 to 10 range are classified as experiencing "severe nuisance".

exposure of sensitive objects around the odour emission source as 98-percentiles[1]. Four different categories of odour-sensitive areas are distinguished on the basis of two location dependant criteria:

(1) Is the odour-sensitive object inside or outside the built-up area?
(2) Is the odour-sensitive object inside or outside a livestock concentration area?

For each of these four categories a maximum allowable odour exposure, expressed in 98-percentiles, is defined.

A crucial input parameter of the odour dispersion model is the calculation of the odour emission rate at the source. The emission rate at the source is calculated by multiplying the number of animal places by the corresponding standard odour emission rate for that specific livestock category and housing system.

For each animal species or category a standard odour emission rate has been defined which is expressed as (OU_E per animal place per second; OU_E stands for European Odour Units (CEN, 2003). The values of these standard odour emission rates have been estimated (Ogink, 2009) on the basis of available research on odour emission from animal housing systems in the Netherlands, including bioscrubbers and acid scrubbers (Ogink and Groot Koerkamp, 2001; Ogink and Lens, 2001; Mol and Ogink, 2002; Ogink and Aarnink, 2003). The list of standard odour emission rates is published as the Odour Nuisance and Livestock Farming Regulation (VROM, 2006b, 2007d) and updated on a regular basis.

The Odour Nuisance and Livestock Farming Regulation distinguishes for each animal category two clusters of housing systems: low-emission systems and conventional systems. Odour emission rates are assigned at cluster level, either low-emission or conventional, not at individual housing system level. An exception has been made for air scrubber systems, where odour emission reductions have been assigned as follows (VROM, 2006b, 2007d)[2]:

(1) acid air scrubbers; assigned odour emission reduction is 30%;
(2) biological air scrubbers, assigned odour emission reduction is 45%;

[1] The 98-percentile represents the odour concentration at a given point that is maximally reached during 98% of the hours during a year, and thus exceeded during 2% of that year.
[2] The odour emission rate (OU_E/animal place/year) for a scrubber system is calculated by multiplication of the odour emission rate for a conventional housing system with the assigned reduction percentage for the scrubber system.

(3) multi-stage air scrubbers, assigned odour emission reduction depends on measurements on concerning scrubber system; currently, odour emission reductions of 70%, 75%, and 85% have been assigned to multi-stage air scrubbers.

When a manufacturer has developed a new scrubber system or redesigned an existing system and wants to market the system in the Netherlands an odour emission reduction percentage needs to be assigned similar to what is described in section 2.1.3 for ammonia. A technical committee evaluates the new scrubber system design and advises the Ministry of the Environment either to assign either a standard odour emission reduction percentage, *viz* 30% for a standard acid scrubber or 45% for a standard biological scrubber, or to assign a specific odour emission reduction percentage to the system on the basis of measurements.

National legislation on odour has to comply with the IPPC directive (EC, 1996a) that, as described in section 2.1.2, covers the environmental performance of large-scale livestock farms, including local disturbance caused by emission of odorous compounds.

2.3 Greenhouse gases

2.3.1 United Nations Framework Convention on Climate Change

Since the industrial revolution the levels of carbon dioxide and other so-called greenhouse gases (GHGs) in the atmosphere have increased. Although the scientific community may not be unanimous about the effects of these elevated concentrations on climate change, binding agreements have been signed in order to reduce GHG emissions.

In 1992 the United Nations Framework Convention on Climate Change (UNFCCC) was formulated (UNFCCC, 1992) which entered into force in 1994. Its objective was "to achieve stabilization of greenhouse gas concentrations in the atmosphere at a low enough level to prevent dangerous anthropogenic interference with the climate system", which means that the global emission of GHGs in 2100 should be reduced by 40-50% with respect to 1990 levels. The 1997 Kyoto Protocol (UNFCCC, 1997) significantly strengthened the Convention by committing countries to individual, legally-binding targets to limit or reduce their greenhouse gas emissions. Many industrialized countries, including the Netherlands, have by now ratified the Kyoto protocol by which the developed countries commit to cut their emission of greenhouses gases by at least 5% during the years 2008-2012 with respect to 1990 levels (UNFCCC, 2007). The target for the European Union (EU-15) is a total reduction of 8%. On 16 February 2005 the Kyoto Protocol entered into force after the former Soviet Union had ratified the treaty. The Kyoto protocol will expire in 2012, and international talks began in May 2007 on a future treaty to succeed the current one.

The two gases that are the main contributors to the total greenhouse gas emission from livestock husbandry are methane (CH_4) and nitrous oxide (N_2O). These two gases substantially contribute to the enhanced greenhouse effect with a global warming potential (GWP) of 23 for methane and 296 for nitrous oxide, respectively, as compared to carbon dioxide (GWP = 1), based on a time horizon of 100 years (IPCC, 2007). Methane emission from livestock husbandry represents 5% of total GHG emissions in the EU-15 (EAA, 2004). This methane originates as 59% from enteric fermentation in ruminants, 12 and 13% from fermentation of manure in animal houses and manure storages, respectively, and 15% from grazing (Freibauer, 2002). Nitrous oxide emission from agriculture represents 5% of total GHG emissions in the EU-15. About 90% of this nitrous oxide emits from soils as a result of pasturage and application of manure and chemical fertilisers, and about 10% is emitted directly to the air from animal houses and manure storages (EAA, 2004).

2.3.2 European Union Emission Trading Scheme

In 2005 the European Union Emission Trading Scheme (EU ETS) commenced operation under the 2003 Emission Trading Directive (EC, 2003b) which aims to reduce CO_2 emissions. The EU ETS is a main pillar of EU climate policy and aims to help EU Member States achieve compliance with their commitments under the Kyoto Protocol (EC, 2005a).

Member States have drawn up national allocation plans (NAPs) for energy-intensive installations which give each installation a certain number of allowances free of charge. Companies that keep their emissions below the level of their allowances are able to sell their excess allowances ("CO_2-credits") to other companies that have higher emission levels at price determined by supply and demand at that time. The target of the cap-and-trade system is that emission reduction can be achieved at least costs (Ellerman and Buchner, 2007; EC, 2005a).

The EU ETS covers over 11.500 energy-intensive installations across the EU, which represent close to half of Europe’s emissions of CO_2. These installations include combustion plants, oil refineries, coke ovens, iron and steel plants, and factories making cement, glass, lime, brick, ceramics, pulp and paper; agricultural activities are not included (EC, 2005a). The current market price for a 1 ton CO_2 allowance is about € 24. (Point Carbon, 2007).

In future greenhouse gases other than CO_2 (*e.g.* CH_4 and N_2O) and other (industrial) sectors *(e.g.* agriculture) might be included in the trading scheme.

2.3.3 National regulations and emission levels for the Netherlands

By ratification of the Kyoto Protocol, the Netherlands have committed themselves to reduce GHG emissions by 6% in the period 2008-2012, compared to 1990 levels. In 1990, the annual GHG emission in the Netherlands was 215 Mton of CO_2-equivalents and it has been calculated that the GHG emission would rise to 239 Mton kg of CO_2-equivalents in 2010 if no climate policy was implemented (NER, 2004; NEAA, 2008). This means that the average annual emissions must be reduced by about 40 Mton of CO_2-equivalents. The first and second National Allocation Plan for the Netherlands (VROM, 2004a, 2007e) describe the approach and progress of national efforts towards GHG emission reduction.

The Dutch government aims to realize 50% of the emission reduction in the Netherlands and 50% abroad (VROM, 2004a, 2004b). The measures to achieve the domestic emission reduction have been formulated in the Climate Policy Implementation Memorandum (VROM, 1999) and include encouragement of energy conservation, use of renewable energy, and multi-year agreements for emission reduction with industry. The emission reduction that has to be made abroad is achieved by buying CO_2-credits from companies that achieve greenhouse-gas reductions in other countries, in accordance with the European Union Emission Trading Scheme (SenterNovem, 2007). Furthermore, the Dutch government has set a target of 70 - 100 Mton CO_2-equivalents for the annual domestic GHG emission in 2030 (VROM, 2001a), which is a 40 - 60% reduction as compared to 1990.

For carbon dioxide emissions, indicative targets have been established per (industrial) sector; for agriculture the 2010 target is a CO_2 emission of 8 Mton (VROM, 2006c)[1]. For methane and nitrous oxide emissions from agriculture the aim is to implement cost-effective measures for emission reduction but no quantitative targets have been set (VROM, 1999, 2005b).

The total greenhouse gas emission from agriculture has decreased by 15% from 26 Mton in 1990 to 22 Mton CO_2-equivalents 2002, mainly as a result of a fall in methane emission. The methane emission is steadily declining due to the shrinking of the dairy livestock herd during these years. The emission of N_2O from manure

[1] Horticultural activities, *viz* greenhouse farming, account for about 85% of the agricultural CO_2 emission (VROM, 2002d). For agriculture, CO_2 emission accounts for about 2% of the total greenhouse gas emission on a CO_2-eq. basis.

application, however, has increased since the early 1990's as an undesirable side effect of manure injection in order to reduce ammonia emissions (NEAA, 2008).

2.4 Particulate matter

2.4.1 Introduction

Although removal of particulate matter (PM 2.5 and PM 10) by scrubbers is not being investigated in this work, regulations with regard to particular matter emissions and exposure levels are also included here. Abatement of particulate matter emissions from animal houses is expected to become of major importance for the coming years and future research will have to address this aspect as well.

Particulate matter (PM) is the term for tiny solid or liquid particles that are suspended in the air and includes dust, dirt, soot, smoke, and liquid droplets. Particulate matter typically consist of a mixture of inorganic and organic chemicals, including carbon, sulphates, nitrates, metals, acids, and semi-volatile compounds. The size of PM in air ranges from approximately 0.005 to 100 μm in diameter. Sources of particulate matter can be anthropogenic or natural. Particulates may be either emitted into the atmosphere (primary particulates) or formed within the atmosphere itself (secondary particulates) as a result from chemical reactions of primary pollutants. Coarse particles are removed from the atmosphere by sedimentation and precipitation (the atmospheric residence time for particles > 20 μm is several hours while it is 2-4 days for 2-3 μm particles). Particles in the range 0.1-1 μm exhibit the longest lifetime in the atmosphere, ranging from days to a few weeks (CAFE, 2004; WHO, 2004).

An increasing number of studies have reported associations between the levels of PM in the air and adverse respiratory and cardiovascular effects in people. Scientists have observed these associations even at relatively low ambient levels that are prevalent in many urbanised countries. Research has shown that particle size is an important factor that influences how particles deposit in the respiratory tract and affect human health. Larger particles are generally filtered in the nose and throat and do not cause problems, but particulate matter smaller than about 10 micrometres, referred to as PM10, can settle in the bronchi and lungs and cause health problems. Fine particles (diameter of 0.1 to 2.5 μm) may penetrate to deep areas of the lung, and ultrafine particles (diameter of 0.1 μm or smaller) may pass through the lungs to affect other organs (MDH, 2008; WHO, 2006a)[1]. A large number of deaths and other health problems are associated with particulate pollution. PM emission is estimated to cause 348,000 premature deaths annually in Europe (WHO, 2006b).

2.4.2 EU Air Quality Framework Directive

The Air Quality Framework Directive on ambient air quality assessment and management (EC, 1996b) defines the policy framework for 12 air pollutants known to have a harmful effect on human health and the environment, including particulate matter. The limit values for the specific pollutants are set through a series of Daughter Directives. The first Daughter Directive (EC, 1999) sets limit values for, among other compounds, particulate matter in ambient air. The annual and 24-hour limit values in the EU for PM10 have been set to 40 $\mu g/m^3$ and 50 $\mu g/m^3$, respectively, which must be met on 1 January 2005. The 24-hour limit value is not to be exceeded more than 35 times a year. Recently a new Directive on ambient air quality has been adopted that sets an annual limit value for PM2.5 of 25 $\mu g/m^3$ to be reached between 2010 and 2015 (EC, 2008). World Health Organization (WHO) air quality guidelines (AQG) set annual and 24-hour limit values for PM2.5 on 10 $\mu g/m^3$ and 25 $\mu g/m^3$, respectively (WHO, 2006a).

In many urbanized regions in the world, both EU and WHO limit values are exceeded (Pandey *et al.*, 2006). Some 70% of European towns and cities with 250,000 inhabitants or more have reported exceeding the PM10 limits in at least part of their area (EC, 2007b). For regions with intensive agricultural activities, the emission of PM from animal houses may account for a large part of the total PM emission. Apart from the health hazards towards humans, emission of particulate matter is suspected to be a transmission way for disease spreading among farms (Seedorf and Hartung, 2002; Aarnink *et al.*, 2005).

2.4.3 National regulations and emission levels for the Netherlands

The European Air Quality Framework Directive (EC, 1996b) and its daughter directives were implemented in the Netherlands by the Dutch National Air Quality Decree (VROM, 2001c, 2005c). Currently, the Netherlands do not comply with the PM10 limit values for 2005 (Buijsman *et al.*, 2005); the European parliament,

---

[1] PM10 (also called thoracic particles) represents the fraction of particles that have an aerodynamic diameter of 10 µm or less; PM2.5 (also called fine particles) is used to describe the particles fraction with an aerodynamic diameter of 2.5 µm or less. The aerodynamic diameter is the diameter of a spherical particle having a density of 1 kg/m^3 that has the same terminal settling velocity in the gas as the particle of interest.

however, has decided to allow a three-year exemption to the PM10 limit under certain circumstances. The new Air Quality Act (VROM, 2007f), which replaces the National Air Quality Decree, aims to meet the PM10 limits with some delay and also sets limits for PM2.5 ambient air concentrations.

In 2004, total primary anthropogenic PM10 emission in the Netherlands was estimated 42 kton; 20% of this emission was estimated to originate from agricultural sources (NEAA, 2008). The major contributors to agricultural primary PM10 emissions (over 80%) are pig and poultry houses (Chardon and Hoek, 2002). The particulates emitted from animal houses comprise feather and hair fragments, faecal material, skin debris, feed particles, mould spores, bacteria, fungus fragments, and litter fragments (Anonymous, 1999; Aarnink and Ellen, 2007). Anthropogenic PM10 emission in the Netherlands consists for about 35% of primary particulates and for about 65% of secondary particulates; ammonia is a known precursor of secondary particulates so livestock farming also contributes indirectly to PM emission by the emission of ammonia.

End-of-pipe air treatment with air scrubbers may become of major importance for compliance with current and future PM10 and PM 2.5 emission standards.

2.5 Conclusion

In this chapter an overview has been given of emission regulations and emission targets for the gaseous compounds that are relevant in the scope of this thesis, *i.e.* air treatment of exhaust air from animal houses. Air treatment techniques have the potential to achieve a multi-pollutant removal (combined removal of ammonia, odour and particulate matter) in conjunction with high removal efficiencies, whereas other emission reduction options, such as adaptation of feed and housing system (see 1.2.1 and 1.2.2), seem to be unable to meet the combined emission standards that livestock operations must comply with. As such, the increasingly stringent emission regulations and targets can be seen as a driving force for research and development in the field of air treatment techniques.

The aim of this thesis is to better understand and improve the performance of air scrubber systems in livestock operations in order to meet the emission standards presented in this chapter at acceptable costs. In the next chapters (chapters 3 - 7) the research that was carried out to address these topics is presented, followed by a general discussion in chapter 8.

References

Aarnink, A.J.A.; H.H. Ellen (2007). Processes and factors affecting dust emissions from livestock production. Proceedings of DustConf 2007, International Conference in Maastricht, Netherlands, 23-24 April; http://www.dustconf.com.

Aarnink, A.J.A.; W.J.M. Landman; R.W. Melse; T. T. T. Huynh (2005). Systems for eliminating pathogens from exhaust air of animal houses. Proceedings of the 7th International Livestock Environment Symposium (ILES VII), May 18 - 20, 2005, Beijing, China; pp 239-244; ASAE, St. Joseph, MI, USA, ASAE Publication 701P0205, ISBN 1-892769-48-4.

Anonymous (1999). Management of dust in broiler operations. Poultry factsheet. Ministry of Agriculture and Food, British Columbia, Canada.

Buijsman, E.; J.P. Beck; L. van Bree; F.R. Cassee; R.B.A. Koelemeijer; J. Matthijsen; R. Thomas; K. Wieringa (2005). Fijn stof nader bekeken. De stand van zaken in het dossier fijn stof. Rapportnr. 500037008. Milieu- en Natuurplanbureau, Bilthoven, Netherlands; ISBN: 9069601249; pp 58.

Burton, C.H.; C. Turner (2003). Manure management. Treatment strategies for sustainable agriculture; 2nd editon; Silsoe Research Institute, Silsoe, U.K.; ISBN 0-9531282-6-1.

CAFE (2004). Second Position Paper on Particulate Matter Clean Air For Europe Working Group on Particulate Matter, December 20th, 2004; http://ec.europa.eu/environment/air/cafe/pdf/working_groups/2nd_position_paper_pm.pdf.

CBS (2003). Statline. Permanent Onderzoek Leefsituatie (POLS). Statistics Netherlands, Voorburg/Heerlen, Netherlands.

CBS (2007). Monitor Mineralen en Mestwetgeving 2007; Statistics Netherlands, Voorburg/Heerlen, Netherlands; ISBN 978-90-357-1929-3.

CEN (2003). Air quality, Determination of odor concentration by dynamic olfactometry; European Committee for Standardization, Brussels, Belgium.

Chardon, W.J.; K.W. van der Hoek (2002). Berekeningsmethode voor de emissie van fijn stof uit de landbouw. Alterra rapport 682 / RIVM rapport 773004014. Alterra, Wageningen, Netherlands; ISSN 1566-7197; pp 36.

EAA (2004). Annual European Community greenhouse gas inventory 1990-2002 and inventory; Technical report No 2/2004; Environment Agency, Copenhagen, Denmark; ISBN: 92-9167-695-0; http://reports.eea.europa.eu/technical_report_2004_2/en/.

EC (1996a). Council Directive 96/61/EC of 24 September 1996 concerning integrated pollution prevention and control; Council of the European Union;

http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=CELEX:31996L0061:EN:NOT.
EC (1996b). Council Directive 96/62/EC of 27 September 1996 on ambient air quality assessment and management.
EC (1999). Council directive 1999/30/EC of 22 April 1999 relating to limit values for sulphur dioxide, nitrogen dioxide and oxides of nitrogen, particulate matter and lead in ambient air.
EC (2001). Directive 2001/81/EC of the European parliament and of the council of 23 October 2001 on national emission ceilings for certain atmospheric pollutants; Council of the European Union; http://europa.eu.int/eur-lex/pri/en/oj/dat/2001/l_309/l_30920011127en00220030.pdf.
EC (2003a). Integrated Pollution Prevention and Control (IPPC). Reference Document on Best Available Techniques for Intensive Rearing of Poultry and Pigs; European Commission; http://ec.europa.eu/comm/environment/ippc/brefs/ilf_bref_0703.pdf.
EC (2003b). Directive 2003/87/EC of the European Parliament and of the Council of 13 October 2003 establishing a scheme for greenhouse gas emission allowance trading within the Community and amending Council Directive 96/61/EC.
EC (2005a). EU action against climate change, EU emissions trading - an open scheme promoting global innovation; European Commission; ISBN 92-894-9866-8; 20 pp.
EC (2007a). Website of European Commission; http://www.ec.europe.eu/environment/.
EC (2007b). Air pollution: Commission takes action over levels of sulphur dioxide and PM10 in member states. Press Releases of the European Commission. Reference: IP/07/1537, 17/10/2007.
EC (2008). Directive 2008/50/EC of the European Parliament and of the Council of 21 May 2008 on ambient air quality and cleaner air for Europe; Council of the European Union; http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2008:152:0001:0044:EN:PDF.
EDC (2007). Environmental Data Compendium (EDC); http://www.mnp.nl/mnc/sitemap-en.html; MNP, Bilthoven, CBS, Voorburg, WUR, Wageningen, Netherlands.
Ellerman, A.D.; B.K. Buchner (2007). The European Union Emissions Trading Scheme: Origins, Allocation, and Early Results. Review of Environmental Economics and Policy, Vol. 1, No 1, pp 66-87; doi:10.1093/reep/rem003.

Freibauer, A. (2002). Biogenic Greenhouse Gas Emissions from Agriculture in Europe. Quantification and Mitigation; Ph.D. thesis; University of Hohenheim, Stuttgart, Germany.

Heij, G.J.; J.W. Erisman (eds) (1995). Acid Rain Research: Do We Have Enough Answers? Proceedings of a Speciality Conference, 's-Hertogenbosch, Netherlands, 10-12 October 1994. Studies in Environmental Science, 64; Elsevier, Amsterdam, Netherlands; 516 pp.

Heij, G.J.; J.W. Erisman (1997). Acid Atmospheric Deposition and its Effects on Terrestrial Ecosystems in the Netherlands. The Third and Final Phase (1991-1995); Studies in Environmental Science, 69; Elsevier, Amsterdam, Netherlands; 716 pp.

Hendriks, H.J.M. (2008). Personal communication. Member of the workgroup on the definition of the first BREF for Intensive Rearing of Poultry and Pigs.

Hettelingh, J.-P.; M. Posch; J. Slootweg; G.J. Reinds; T. Spranger; L. Tarrason (2007). Critical loads and dynamic modelling to assess European areas at risk of acidification and eutrophication. Water, Air & Soil Pollution: Focus, Vol. 7, No 1-3, pp 379-384.

IPCC (2007). Climate change 2007. The physical science basis. Contribution of working group I to the fourth assessment report of the Intergovernmental Panel on Climate Change; Salomon, S. et al., eds.; Cambridge University Press: Cambridge, U.K.; ISBN 978-0-521-88009-1 ISBN 978-0-521-70596-7; 996 pp; http://www.ipcc.ch/ipccreports/ar4-wg1.htm.

Jong, de, R.G.; J.H.M. Steenbekkers; H. Vos (2000). Hinder en andere zelf-gerapporteerde effecten van milieuverontreiniging in Nederland, Inventarisatie Verstoringen 1998. TNO-PG, Delft, Netherlands.

Klarenbeek, J. V.; A.P. van Harreveld (1995). On the regulations, measurement and abatement of odours emanating from livestock housing in the Netherlands. Proceedings of the International Livestock Odour Conference 1995, Ames, Iowa, U.S.A., pp 16-21.

Koch, W. W. R; A. K. Van Harmelen; P. W. H. G. Coenen; G. Van Grootveld (2003). Emission monitor for the Netherlands: 2000 and estimates for 2001; Report Series MilieuMonitor (environmental monitor) No 9; Coördinatiecommissie Doelgroepmonitoring (CCDM), The Hague, Netherlands; http://www.emissieregistratie.nl/erpubliek/documenten/Emissierapportages%20(Emission%20reports)/MM9%20Emission%20monitor%20data%202000%20and%20estimates%202001.pdf.

MDH (2008). Website of Minnesota Department of Health; http://www.health.state.mn.us/divs/eh/air/pm.htm.

Melse, R.W.; H.C. Willers (2005). Theoretical evaluation of ammonia scrubbers for governmental approval. Proceedings of the 7th International Conference on Construction, Technology and Environment in Livestock Farming, March 2-3, 2005, Braunschweig, Germany; pp 295-300, Kuratorium für Technik und Bauwesen in der Landwirtschaft e.V. (KTBL), Darmstadt, Germany, ISBN 3-7843-2185-2.

Mol, G.; N.W.M. Ogink (2002). Geuremissie uit de veehouderij II, Overzichtsrapportage 2000 -2002; Rapport 2002-09; IMAG, Wageningen, Netherlands.

NEAA (2008). Environmental Data Compendium; Netherlands Environmental Assessment Agency; http://www.mnp.nl/mnc/index-en.html.

NER (2004). Nederlands Emissieregistratie, definitieve cijfers; http://www.emissieregistratie.nl.

Ogink, N.W.M. (2009). Onderbouwing van geuremissiefactoren in de Regeling geurhinder en veehouderij op basis van geuremissie-onderzoek. In press. Animal Sciences Group van Wageningen UR, Lelystad, Netherlands.

Ogink, N. W. M; P. W. G. Groot Koerkamp (2001). Comparison of odour emissions from animal housing systems with low ammonia emissions. Water Science & Technology, Vol 44, No 9, pp 245-252.

Ogink, N.W.M.; P. Lens (2001). Geuremissie uit de veehouderij, Overzichtsrapportage 1996-1999; Rapport 2001-14; IMAG, Wageningen, Netherlands.

Ogink, N.W.M.; A.J.A. Aarnink (2003). Managing Emissions from Swine Facilities: Current situation in The Netherlands and Europe. Proceedings of the University of Illinois Pork Industry Conference, 11-12 December, 2003, Champaign, U.S.A., pp 285-302.

Pandey, K.D.; D. Wheeler; B. Ostro; U. Deichmann; K. Hamilton; K. Bolt (2006). Ambient Particulate Matter Concentrations in Residential and Pollution Hotspot areas of World Cities: New Estimates based on the Global Model of Ambient Particulates (GMAPS), The World Bank Development Economics Research Group and the Environment Department Working Paper, The World Bank, Washington DC, USA.

Point Carbon (2007). Point Carbon website, http://www.pointcarbon.com, visited on November 26th, 2007.

Rostron, J. (2006). Integrated pollution prevention and control: A review of English and European Union law and regulation. Environmental Quality Management, Vol. 16, No 2, pp 61-72. doi:10.1002/tqem.20121.

Seedorf, J.; J. Hartung (2002). Stäube und Mikroorganismen in der Tierhaltung. Kuratorium für Technik und Bauwesen in der Landwirtschaft, Darmstadt, KTBL-Schrift 393, Landwirtschaftsverlag, Münster, Germany.

SenterNovem (2007). SenterNovem website, http:// http://www.senternovem.nl/carboncredits/, visited on November 26th, 2007.

Sliggers, J.; K. Bull (2007). Ammonia and the UNECE convention on long-range transboundary air pollution. In: Ammonia emissions in agriculture, G.-J. Monteny and E. Hartung (eds), Proceedings of the International Conference on Ammonia in Agriculture: Policy, Science, Control and Implementation, March 19-21, 2007, Ede, Netherlands, pp 31-37; Wageningen Academic Publishers, Wageningen, Netherlands; ISBN 978-90-8686-029-6; 403 pp.

Sliggers, J. (2007). Mag het iets meer zijn? De Convention on Long-range Transboundary Air Pollution (9, slot). Milieu, Vol 13, No 7, pp 19-24.

Starmans, D.A.J.; K.W. Van der Hoek (eds) (2007). Ammonia, the case for the Netherlands; Wageningen Academic Publishers, Wageningen, Netherlands; ISBN 978-90-8686-028-9; 201 pp.

UNECE (2007). Website of UNECE on the Convention on Long-range Transboundary Air Pollution (CLRTAP); http://www.unece.org/env/lrtap/.

UNFCCC (1992). United Nations Framework Convention on Climate Change. Convention text, UNEP/WMO Information Unit of Climate Change (IUCC) on behalf of the Interim Secretariat of the Convention; IUCC, Geneva, Switzerland.

UNFCCC (1997). Kyoto Protocol to the United Nations Framework Convention on Climate Change; FCCC/CP/L7/Add.1, 10 December 1997; UN, New York, NY, U.S.A.

UNFCCC (2007). United Nations Framework Convention on Climate Change; website: http://unfccc.int/kyoto_protocol/items/2830.php.

VROM (1979). Wet van 13 juni 1979, houdende regelen met betrekking tot een aantal algemene onderwerpen op het gebied van de milieuhygiëne (Wet Milieubeheer); 13 juni 1979, Staatsblad No 442; Ministry of Housing, Spatial Planning and the Environment, The Hague, Netherlands.

VROM (1985a). Brochure Veehouderij en Hinderwet 1985; Ministry of Housing, Spatial Planning and the Environment, The Hague, Netherlands.

VROM (1985b). Rapport Beoordeling cumulatie stankhinder door intensieve veehouderijen; Rapport 85546/7-85; Publikatiereeks Lucht, No 46; Ministry of Housing, Spatial Planning and the Environment, The Hague, Netherlands.

VROM (1986). Wet van 3 juli 1986 (wet bodembescherming), Staatsblad No 374, Ministry of Housing, Spatial Planning and the Environment, The Hague, Netherlands.

VROM (1987). Besluit gebruik dierlijke meststoffen. Staatsblad No 114, Ministry of Housing, Spatial Planning and the Environment, The Hague, Netherlands.
VROM (1990). Besluit mestbassins milieubeheer; 31 december 1990; Staatsblad No 618; Ministry of Housing, Spatial Planning and the Environment, The Hague, Netherlands.
VROM (1993). Tweede Nationaal Milieubeleidsplan. Ministry of Housing, Spatial Planning and the Environment, The Hague, Netherlands.
VROM (1996). Richtlijn Veehouderij en Stankhinder 1996, 30 oktober 1996; Ministry of Housing, Spatial Planning and the Environment, The Hague, Netherlands.
VROM (1999). Uitvoeringsnota klimaatbeleid. Ministry of Housing, Spatial Planning and the Environment, The Hague, Netherlands.
VROM (2001a). Vierde Nationaal Milieubeleidsplan. Een wereld en een wil: werken aan duurzaamheid. Ministry of Housing, Spatial Planning and the Environment, The Hague, Netherlands.
VROM (2001c). Besluit luchtkwaliteit; 19 juli 2001; Staatsblad No 269, Ministry of Housing, Spatial Planning and the Environment, The Hague, Netherlands.
VROM (2002a). Wet ammoniak en veehouderij; 31 januari 2002; Staatsblad No 93; Ministry of Housing, Spatial Planning and the Environment, The Hague, Netherlands.
VROM (2002b). Regeling Ammoniak en Veehouderij (RAV); 1 mei 2002; Staatsblad No 82; Ministry of Housing, Spatial Planning and the Environment, The Hague, Netherlands.
VROM (2002c). Wet stankemissie veehouderijen in landbouwontwikkelingsgebieden; Staatsblad No 319; Ministry of Housing, Spatial Planning and the Environment, The Hague, Netherlands.
VROM (2002d). Evaluatienota Klimaatbeleid. De voortgang van het Nederlandse klimaatbeleid: een evaluatie bij het ijkmoment 2002. Tweede Kamer, vergaderjaar 2001-2002, 28 240, no 2; Sdu Uitgevers, The Hague, Netherlands.
VROM (2003a). Brief van de staatssecretaris van VROM en de minister van LNV aan de Tweede Kamer; Tweede Kamer, vergaderjaar 2002-2003, 24 445, no 65; Sdu Uitgevers, The Hague, Netherlands.
VROM (2003b). Regeling stankemissie veehouderijen in landbouwontwikkelings- en verwevingsgebieden. Staatscourant 28 april 2003, No 81; Ministry of Housing, Spatial Planning and the Environment, The Hague, Netherlands.
VROM (2004a). Allocatieplan CO_2-emissierechten 2005 t/m 2007; Nederlands nationaal toewijzingsplan inzake de toewijzing van broeikasgasemissierechten aan bedrijven, 20 augustus 2004; Ministry of Housing, Spatial Planning and the

Environment, The Hague, Netherlands; http://www.senternovem.nl/mmfiles/113920_Nederlands%20allocatieplan%20CO2%20emissiehandel1_tcm24-73088.pdf

VROM (2004b). Implementatiewet EG-richtlijn handel in broeikasgasemissierechten. 30 september 2004; Staatsblad No 511; Ministry of Housing, Spatial Planning and the Environment, The Hague, Netherlands.

VROM (2005a). Besluit ammoniakemissie huisvesting veehouderij (AMvB Huisvesting); 28 december 2005; Staatsblad No 675; Ministry of Housing, Spatial Planning and the Environment, The Hague, Netherlands.

VROM (2005b). Evaluatienota Klimaatbeleid 2005. Onderweg naar Kyoto. Kamerstukken II 2005-2006, 28 240, no 37; Ministry of Housing, Spatial Planning and the Environment, The Hague, Netherlands.

VROM (2005c). Besluit luchtkwaliteit; 23 juni 2005; Staatsblad No 316. Ministry of Housing, Spatial Planning and the Environment, The Hague, Netherlands.

VROM (2006a). Wet geurhinder en veehouderij (Wgv); 5 oktober 2006; Staatsblad No 531; Ministry of Housing, Spatial Planning and the Environment, The Hague, Netherlands.

VROM (2006b). Regeling geurhinder en veehouderij; Staatscourant 18 december 2006, No 246; Ministry of Housing, Spatial Planning and the Environment, The Hague, Netherlands.

VROM (2006c). Integrale afweging klimaatbeleid gericht op het halen van Kyoto. Brief aan de Tweede Kamer, 13 april 2006, KvI2006247406.

VROM (2007a). Oplegnotitie bij de BREF voor de intensieve pluimvee- en varkenshouderij. SenterNovem/InfoMil. Ministry of Housing, Spatial Planning and the Environment, The Hague, Netherlands.

VROM (2007b). Onderzoek implementatie IPPC-richtlijn in Nederland in 2006, VROM-Inspectie en de Inspectie Verkeer en Waterstaat, 26 januari 2007, No 7150; Ministry of Housing, Spatial Planning and the Environment, The Hague, Netherlands.

VROM (2007c). Ontwerpbesluit houdende wijziging van het Besluit ammoniakemissie huisvesting veehouderij (interne saldering); 27 maart 2007; Staatscourant No 61; Ministry of Housing, Spatial Planning and the Environment, The Hague, Netherlands.

VROM (2007d). Wijziging Regeling geurhinder en veehouderij; Staatscourant 18 juli 2007, No 136; Ministry of Housing, Spatial Planning and the Environment, The Hague, Netherlands.

VROM (2007e). Nederlands nationaal toewijzingsplan broeikasgasemissierechten 2008-2012, 16 mei 2007; Ministry of Housing, Spatial Planning and the

Environment, The Hague, Netherlands; http://www.senternovem.nl/mmfiles/NAP-II%2016%20mei%202007_tcm24-223591.pdf.

VROM (2007f). Wet luchtkwaliteit; 15 november 2007; Staatsblad No 434. Ministry of Housing, Spatial Planning and the Environment, The Hague, Netherlands.

WHO (2004). Health aspects of air pollution results from the WHO project "Systematic review of health aspects of air pollution in Europe”. WHO, Copenhagen, Denmark.

WHO (2006a). Air quality guidelines. Global update 2005. Particulate matter, ozone, nitrogen dioxide and sulfur dioxide. WHO, Copenhagen, Denmark; ISBN 92 890 2192 6; pp ix + 484, http://www.euro.who.int/Document/E90038.pdf

WHO (2006b). Health risks of particulate matter from long-range transboundary air pollution. Joint WHO/Convention Task force on the Health Aspects of Air Pollution. European Centre for Environment and Health Bonn Office, http://www.euro.who.int/document/E88189.pdf.

Chapter 3

Air scrubbing techniques for ammonia and odor reduction at livestock operations: Review of on-farm research in the Netherlands

R.W. Melse; N.W.M. Ogink

Wageningen University and Research Centre, Agrotechnology & Food Innovations, Wageningen, Netherlands.

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 mechanically ventilated 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%. 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 31%. The odor removal of biotrickling filters ranged from -29% to +87% with an overall average of 44%. Minimum empty bed air residence times (EBRT's) were 0.4 to 2.3 s. Further research is necessary to explain the large variation in odor removal for biotrickling filters and to increase the odor removal efficiency of both acid scrubbers and biotrickling filters.

***Key words**: Air cleaning, Ammonia, NH_3, Odor, Scrubber, Biotrickling, Biofilter, Pig, Poultry, Veal calves.*

3.1 Introduction

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 mechanically ventilated animal houses, *viz* 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 can not be easily accessed by the international research community, however. In this review, we summarize and discuss these results and experiences. Firstly, we describe the programs in which the research was conducted. Secondly, we describe the methods used for NH_3 and odor determination and the principles of air scrubbing. Finally we present the results of the on-farm research on full-scale acid scrubbers and biotrickling filters, followed by a discussion of the results and some concluding remarks.

3.2 Research programs on ammonia and odor

Since 1990, an NH_3 research program has been carried 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 3.1, 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 at mechanically ventilated pig and poultry houses, which are now commercially available and considered as off-shelf techniques.

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 has been 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 are 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 3.1 which lists the main animal categories for which air scrubbers are applied.

Table 3.1 Average ammonia (NH_3) and odor emission rates of conventional housing systems for some animal categories (Mol and Ogink, 2002; Ogink, 2005).

Animal category	NH_3 emission rate (kg animal place^{-1} year^{-1})	Odor emission rate (OU_E animal place^{-1} s^{-1})[a]
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 odour unit (CEN, 2003).

The NH_3 and odor emission rates in Table 3.1 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 a mechanically ventilated 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 - 30°C and has a relative humidity of about 50 - 90%. The NH_3 and odor emission reduction that are achieved by air scrubbing will be discussed later on.

3.3 Methods for ammonia and odor determination

3.3.1 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 an strong acid solution (usually nitric acid, 0.03 - 0.2 M), 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 have been made of Teflon, are isolated, and heated with a coil of resistance heating wire to a temperature that is approximately 20 °C higher than the ambient temperature 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, mainly the impinger method was 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 minutes 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 a NO_x-analyzer (Monitor Labs; model 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 minute interval and recorded by a datalogger. The NO_x analyzer is calibrated regularly. All sampling tubes have been made of Teflon, are isolated, and heated with a coil of resistance heating wire to a temperature that is approximately 20 °C higher than the ambient temperature 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. The concentration of NH_3 in the sample air, however, is determined with a photoacoustic gas analyzer (Brüel & 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 above described techniques, as expressed by the standard error under repeatability conditions, show levels that are within the 1 - 3% range (Mosquera *et al.*, 2002; Ogink, 2005).

3.3.2 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 two hours time. In this way fluctuations in the composition of the air sample are time-averaged over two hours. A filter (pore diameter: 1 - 2 µm) at the inlet of the sampling tube prevents the intake of dust that otherwise will 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 hours after sample collection. Odor concentrations are determined in compliance with the European olfactometric standard EN13725 (CEN, 2003) and the preceding Dutch olfactometric standard NVN2820/1A (NNI, 1996) that has been incorporated in the European standard. In both standards, the sensitivity of the odor panel is based on the 20 - 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 at ten different sampling days to deal both with olfactometric measurement error and performance fluctuations.

3.4 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 (Figure 3.1). 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 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.

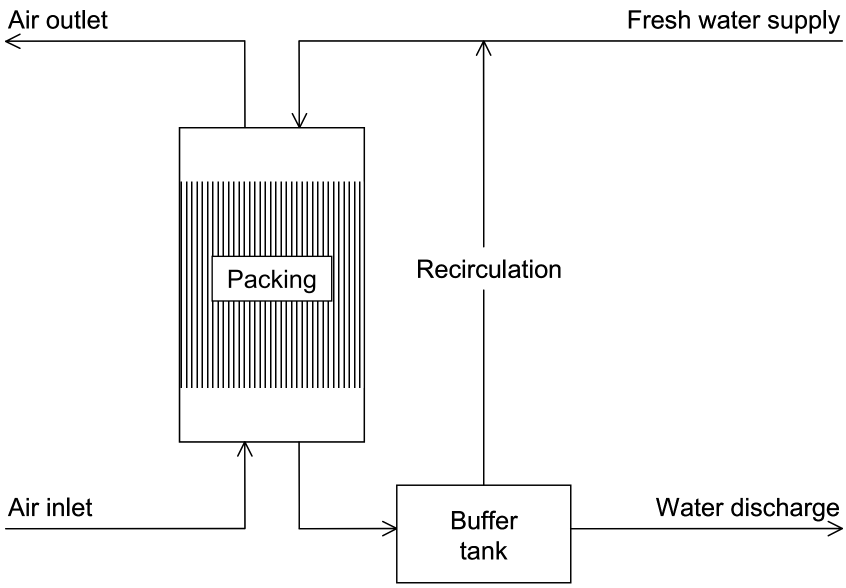


Figure 3.1 Schematic of a counter-current air scrubber.

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).

Concentration gradient

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

$$NH_3\,(g) + H_2O\,(l) \;<\!\!-\!\!-\!\!-\!\!>\; NH_3\,(aq) + H_2O\,(l) \;<\!\!-\!\!-\!\!-\!\!>\; NH_4^+\,(aq) + OH^-\,(aq) \qquad \text{[Eq. 3.1]}$$

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 section 3.5), and, if applicable, by the transformation of NH_3 into other compounds (see section 3.6). Because of the instantaneous dissociation of NH_3, the actual water solubility of NH_3 is not so much an issue.

Size of the contact area between gas and water phase

The rate of mass transfer of a compound is proportional to the contact area, which is determined is determined by the specific surface area of the packing (m^2 m^{-3}) and the degree of wetness of the packing material, which 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 air flow 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 systems must be capable to yield the extra pressure drop.

In the past, biofilters with organic-based packing materials were used for treatment of exhaust air of animal houses in the Netherlands. Nowadays, only acid scrubbers and biotrickling filters are used for this purpose because biofilters could not be operated well at the relatively high NH_3 and dust concentrations of the exhaust air (see section 3.8.3).

3.5 **Experiences with acid scrubbers**

3.5.1 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 3.1 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 - 99% can be achieved, as was demonstrated in the long-term measurement program that was carried out at five farm locations. Table 3.2 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 emission reduction of >90%. From Table 3.2 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}.

3.5.2 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, and is described in Table 3.3 (measurements that were carried out before 1995 were omitted because until then a different protocol for odor concentration measurements had been used, which results can not 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 of 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, *viz* 45% (sem = 8.4) for an experimental pilot-scale acid scrubber treating pig house exhaust air at EBRT from 1.4 - 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 3.3. It is stated that the air sampling method used by these authors differs from the one described above, as they used no dust filter for air sampling in accordance with the protocols that are used in Germany. As part of the odor may be associated with the presence of dust 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 3.2, might 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 anyway 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, in 2004 about 160 acid scrubbers were in operation for treatment of exhaust air from pig and poultry houses, manufactured by four companies. In Germany and Denmark acid scrubbers of similar design are manufactured for this application.

Table 3.2 Characteristics and results of measurement program on ammonia (NH_3) removal by full-scale acid scrubbers treating exhaust air of animal houses.[a]

	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 air flow (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 air flow; 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)[b]	0.9	0.5	NA	0.4	0.4
Maximum superficial air velocity (m s^{-1})[c]	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 minutes on, 18 minutes off
Maximum gas-to-liquid ratio (m^3 m^{-3})	NA	NA	NA	NA	NA
pH (-)	1.3 - 4.4	4 (setpoint)	4 (setpoint)	3 - 5	0 - 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

(table continues on next page)

(Table 3.2 continued)

	Vrielink *et al*., 1997	Verdoes and Zonderland, 1999	Hol and Satter, 1998	Hol *et al*., 1999	Wever and Groot Koerkamp, 1999
NH_3 measurement					
Measurement period	45 days	100 days	2 times 2 months (from age of 21 to 32 weeks, and from 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, sampling time 3.5 days	impinger, sampling time 3.5 days
Number of measurements	37	89	100	19	20
Average air flow (m^3 h^{-1})	2,200	1,600	18,900	48,000	7,300
Average inlet concentration (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)[d]	98 (sem = 0.20)[d]
Minimum removal efficiency (%)	77	90	40	76	96
Maximum removal efficiency (%)	97	100	99	100	100

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

Table 3.3 Characteristics and results of measurement program on odor removal by full-scale acid scrubbers and biotrickling filters treating exhaust air of animal houses.[a]

	Acid scrubber		Biotrickling filter		
	Ogink and Lens, 2001; Hol and Ogink, 2005	Klarenbeek *et al*., 1998	Melse and Mol, 2004[b]	Mol and Ogink, 2002; Hol and Ogink, 2005	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 air flow (m^3 h^{-1})	15,000	15,000	20,000	NA	48,000
Packing type	NA	structured packed bed	vertical bundle of plastic tubes (diameter: 4 cm)	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
Maximum 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 (m^3 m^{-3})	NA	NA	NA	NA	NA
pH (-)	4 (setpoint)	4 (setpoint)	7.3 - 7.6	NA	NA
Discharge water control	time	time	time[e]	NA	time

(table continues on next page)

(Table 3.3 continued)

	Acid scrubber		Biotrickling filter		
	Ogink and Lens, 2001; Hol and Ogink, 2005	Klarenbeek *et al*., 1998	Melse and Mol, 2004[b]	Mol and Ogink, 2002; Hol and Ogink, 2005	Mol and Ogink, 2002; Hol and Ogink, 2005
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 air flow (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)[f]	49 (sem = 8) [f]	48 (sem = 11) [f]	37 (sem = 7) [f]
Minimum removal efficiency (%)	3	15	-29	-24	-10
Maximum removal efficiency (%)	50	51	87	83	63

[a] NA = not available.

[b] The NH_3 removal efficiency of this biotrickling filter is presented in Table 3.4.

[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. The accumulation of N-NO_2^- and N-NO_3^- in the trickle water during the measurement period equaled the removal of N-NH_3 from the air.

[f] sem = standard error of the mean.

3.6 Experiences with biotrickling filters

3.6.1 Ammonia removal

In a biotrickling filter, the reaction equilibrium of Equation 3.1 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). In Equation 3.2 and 3.3 these processes are schematically described:

$$NH_4^+ (aq) + OH^- (aq) + 1.5\ O_2 (g) \dashrightarrow NO_2^- (aq) + H^+ (aq) + 2\ H_2O (l) \qquad \text{[Eq. 3.2]}$$

$$NO_2^- (aq) + H^+ (aq) + 2\ H_2O (l) + 0.5\ O_2 \dashrightarrow NO_3^- (aq) + H^+ (aq) + 2\ H_2O (l) \qquad \text{[Eq. 3.3]}$$

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 Equation 3.1 through 3.3 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): $6.5 < pH < 7.5$, $1 < [NH_4\text{-}N]\ (g\ L^{-1}) < 4$, 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 3.4 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 - 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 - 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 3.4 it is concluded that biotrickling filters can meet this target but not always do 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.

3.6.2 Odor removal

In a biotrickling filter, a microbial community is present which 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 result 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 3.3). The average odor removal efficiency of the three biotrickling filters was 44%. The variation of the odor removal is 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 EBRT's 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^- (Lee *et al.*, 2000; Rowe *et al.*, 1979; Yarbrough *et al.*, 1980).

Table 3.4 Characteristics and results of measurement program on ammonia (NH_3) removal by full-scale biotrickling filters treating exhaust air of animal houses.[a]

	Scholtens *et al*., 1988		Van de Sande-Schellekens and Backus, 1993a; Uenk *et al*., 1993a			Van Middelkoop, 1995	Melse and Mol, 2004[b]
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 air flow (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 (diameter: 4 cm)
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)[c]	0.5	0.5	0.5	0.5	1.1	0.5	0.5
Maximum superficial air velocity (m s^{-1})[d]	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	continu-ous	continu-ous	continu-ous	continu-ous	continu-ous	continu-ous	continuous
Maximum gas-to-liquid ratio (m^3 m^{-3})	1000	1300	NA	NA	NA	NA	NA
pH (-)	6.8 - 8.3	6.6 - 7.8	6.2 - 8.5	6.5 - 9[e]	6.5 - 8.7	NA	7.3 - 7.6

(table continues on next page)

(Table 3.4 continued)

	Scholtens *et al*., 1988		Van de Sande-Schellekens and Backus, 1993a; Uenk *et al*., 1993a			Van Middelkoop, 1995	Melse and Mol, 2004[b]
Design characteristics							
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	NA	NA	NA	incidental
Method	impinger, sampling time 20 minutes	impinger, sampling time 20 minutes	impinger	impinger	impinger	NA	impinger, sampling time 2 hours
Number of measurements	18	43	31	29	42	NA	8
Average air flow (m^3 h^{-1})	2,725	5,056	NA	NA	NA	NA	9,000
Average inlet concentration (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)[g]	78	65	90	83[i]	79 (sem = 5.7)[g]
Minimum removal efficiency (%)	11	-8	33	5	26	NA	44
Maximum removal efficiency (%)	94	82	100	98	99	NA	94

[a] NA = not available. [b] The odor removal by this biotrickling filter is presented in Table 3.3. [c] EBRT = empty bed air residence time. [d] Based on an empty bed. [e] pH neutralization took place by addition of a slowly dissolving mineral containing $CaCO_3$ and MgO. [f] At EBRT's > 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 6 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.

In the Netherlands, in 2004 about 45 biotrickling filters were in operation for treatment of exhaust air from pig and poultry houses, manufactured by four companies. In Germany and Denmark biotrickling filters of similar design are manufactured for this application.

3.7 Costs of air scrubbing

The investment and operational costs were calculated for both acid scrubbers and biotrickling filters for treatment of exhaust air of newly built animal production facilities, based on quotations from manufacturers (Table 3.5).

Table 3.5 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 VAT[b]).[c]

Cost category	Acid scrubber 95% NH_3 removal		Biotrickling filter 70% NH_3 removal	
	growing-finishing pigs	broilers	growing-finishing pigs	broilers
Investment costs[d]	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[e]	n/a[e]
Total operational costs (year^{-1})[f]	14.82	0.47	14.29	0.46

[a] Excluding possible water discharge costs.
[b] VAT = value-added tax.
[c] More details can be found in Melse and Willers, 2004 and in Melse and Ogink, 2004.
[d] 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 3.1).
[e] n/a = not applicable.
[f] It was calculated that the total operational costs of a scrubber increase the production costs per animal place with 4% for Dutch conditions.

Table 3.5 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. The amount and characteristics of the discharge water largely differ, however, as has been described above. It depends on the local situation what costs, if any, are charged for the disposal of discharge water.

3.8 General discussion

3.8.1 Ammonia removal

From the results presented in Table 3.2, it can be concluded that acid scrubbing significantly reduces the NH_3 emission from mechanically ventilated 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 simple 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 - 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 to a too low value. 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-user 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 own land is not possible, what is very often the case, and 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.

3.8.2 Odor removal

Both acid scrubbers and biotrickling filters are capable of odor removal with an average removal efficiency of 27% and 43% respectively, as is shown in Table 3.3. It is striking, however, that individual odor removal efficiency measurements strongly vary, *viz* 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 for about 20% to the total variance of the odor removal efficiency measurements, whereas the actual performance of the scrubber system contributes for 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 each 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, the measured odor removal efficiency will increase. If, on the other hand, an odor component is difficult to remove, 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 can currently not be verified.

3.8.3 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 increases the availability of the odor component to the bacteria and thus increases biodegradation rates (*e.g.* Césario, 1997; Van Groenestijn and Lake, 1999; Davidson and Daugulis, 2003). An increase of the air residence time will usually improve the uptake of odor components as well but also means higher investment and operational cost per volume of air treated.

Another possibility is, after having passed the air through the acid scrubber or biotrickling filter first, 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, 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 generally considered economically unfeasible.

Finally, the odor removal performance of a biotrickling filter might be improved by appropriate process control and monitoring as, besides the nitrifying bacteria, also the bacterial population responsible for the removal of odor compounds might be inhibited by accumulation of NH_3 and/or NO_2^- in the system.

3.8.4 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 air flow rate. Ventilation air of animal facilities contains dust that for a part 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 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.

3.9 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 31% and 44%, 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 an electrical conductivity (EC) measurement.
- Further improvement of the odor removal capacity of air scrubbers.
- Analysis of the large performance differences in odor removal efficiency, that were found for both acid scrubbers and biotrickling filter, by combining olfactometric methods using a human panel with advanced analyses of individual compounds by gas chromatography - mass spectrometry (GC-MS).

Acknowledgements

We thank Annemieke Hol for her help on gathering the data of the previously carried out experimental studies. This work was supported by the Dutch Ministry of Agriculture, Nature and Food Quality (LNV).

References

Aarnink, A.J.A., P.D. Le, N.W.M. Ogink, P.M. Becker, and M.W.A. Verstegen. 2005. Odor from animal production facilities: its relationship to diet. *Nutr. Res. Rev.* 18(1): 3-30.

Anthonisen, A.C., R.C. Loehr, T.B.S. Prakasam, and E.G. Srinath. 1976. Inhibition of nitrification by ammonia and nitrous acid. *Journal Water. Poll. Contr. Fed.* 48: 835-852.

Asseldonk, M.M.L., and J.A.M. Voermans. 1989. The use of biofilters for pig husbandry (in Dutch, with English summary). Report 1.47. Rosmalen, Netherlands: Proefstation voor de Varkenshouderij.

Burgess, J.E., S.A. Parsons, and R.M. Stuetz. 2001. Developments in odour control and waste gas treatment biotechnology: a review. *Biotechnol. Adv.* 19: 35-63.

CBS. 2002. StatLine. Statistics data bank for the Netherlands. Available at: http://www.cbs.nl.

CEN. 2003. Air quality - Determination of odour concentration by dynamic olfactometry (EN13725). Brussels, Belgium: European Committee for Standardization.

Césario, M.T. 1997. Water-immiscible solvents for the biological treatment of waste gases. PhD diss. Wageningen, Netherlands: Landbouwuniversiteit Wageningen. ISBN 90-5485-685-8.

Coulson, J.M., J.F. Richardson, J.R. Backhurst, and J.H. Harker. 1999. Coulson & Richardson's Chemical Engineering: Fluid Flow, Heat Transfer and Mass Transfer. Chemical Engineering Vol. 1. Butterworth-Heinemann. ISBN 0 7506 4444 3.

Davidson, C.T., and A.J. Daugulis. 2003. Addressing biofilter limitations: A two-phase partitioning bioreactor process for the treatment of benzene and toluene contaminated gas streams. *Biodegradation* 14: 415-421.

Demmers, T.G.M., and G.H. Uenk. 1996. Experiments with a small-scale biofilter (in Dutch). Report 96-37. Wageningen, Netherlands: DLO Instituut voor Milieu- en Agritechniek (IMAG-DLO).

Deshusses, M.A., and C.T. Johnson. 2000. Development and validation of a simple protocol to rapidly determine the performance of biofilters for VOC treatment. *Environ. Sci. Tehnol.* 34(3): 461-467; DOI: 10.1021/es9909172.

Dong, L., A.J. Heber, J.A. Patterson, B.R. Stobel, D.D. Jones, and A.L. Sutton. 1997. Bioscrubber for removing ammonia from swine house exhaust air. In *Proc. Int. Symp. on Ammonia and Odour Control from Animal Production Facilities*, 529-532. J.A.M. Voermans and G.J. Monteny, eds. Vinkeloord, Netherlands, 6-10 October. Rosmalen, Netherlands: NVTL. ISBN 90-9011059-3.

Eggels, P.G., and R. Scholtens. 1989. Biofiltration of ammonia containing exhaust air at intensive livestock operations. Phase 3 (in Dutch). Ref. 89-107. Wageningen/Apeldoorn, Netherlands: Instituut voor Mechanisatie, Arbeid en Gebouwen (IMAG) / TNO Hoofdgroep Maatschappelijke Technologie (MT-TNO).

Focht, D.D., and W. Verstraete. 1977. Biochemical ecology of nitrification and denitrification. *Advances in Microbial Ecology* 1: 135-214.

Hahne, J., W. Asendorf, and K.-D. Vorlop. 2000. Abluftreinigung – eine realistische Maßnahme zur Reduzierung von Ammoniak- und Geruchsemissionen? (Waste gas cleaning - A realistic measure for reduction of ammonia and odor emission?) In *Bewertung von Geruchsbelastungen aus der Landwirtschaft,* KTBL Sonderveröffentlichung 031, 105-114.

Hahne, J., and K.-D. Vorlop. 1998. Behandlung von Abluft aus Schweinemastanlagen (Treatment of waste gas from pig facilities). *Entsorgungspraxis* 16(12): 45-49.

Hahne, J., and K.-D. Vorlop. 2001. Treatment of waste gas from piggeries with nitrogen recovery. *Landbauforsch Völkenrode* 51(3): 121-130.

Hahne. J., and K.-D. Vorlop. 2004. Sind Abluftwäscher zur Minderung von Ammoniakemissionen geeignet? (Are scrubbers suitable for reduction of ammonia emission?) *Landtechnik* 59(2): 106-107.

Heij, G.J., and J.W. Erisman. 1995. *Acid Rain Research: Do We Have Enough Answers?* Proceedings of a Speciality Conference, 's-Hertogenbosch, Netherlands, 10-12 October 1994. Studies in Environmental Science, Vol. 64. Amsterdam, Netherlands: Elsevier Science. ISBN 0-444-82038-8.

Heij, G.J., and J.W. Erisman. 1997. *Acid Atmospheric Deposition and its Effects on Terrestrial Ecosystems in the Netherlands. The Third and Final Phase (1991-1995).* Studies in Environmental Science, Vol. 69. Amsterdam, Netherlands: Elsevier Science. ISBN 0-444-82037-X.

Hol, J.M.G., and N.W.M. Ogink. 2005. Personal communication. Wageningen, Netherlands: Agrotechnology & Food Innovations, Wageningen UR.

Hol, J.M.G., and I.H.G. Satter. 1998. Acid scrubbing of exhaust air from free-range layer breeders (in Dutch). Report 98-1002. Wageningen, Netherlands: DLO Instituut voor Milieu- en Agritechniek (IMAG-DLO).

Hol, J.M.G, A.C. Wever, and P.W.G. Groot Koerkamp. 1999. Acid scrubbing of exhaust air from broilers (in Dutch). Report P 99-23. Wageningen, Netherlands:DLO Instituut voor Milieu- en Agritechniek (IMAG-DLO).

Kennes, C., and F. Thalasso. 1998. Waste gas biotreatment technology (review). *J. Chem. Technol. Biotechnol.* 72: 303-319.

Klarenbeek, J.V., N.W.M. Ogink, J.M.G. Hol, and I.H.G. Satter. 1998. Odor reduction by acid scrubbing of exhaust air from free-range layer breeders (in Dutch). Internal report. Wageningen, Netherlands: DLO Instituut voor Milieu- en Agritechniek (IMAG-DLO).

Koch, W.W.R, A.K. Van Harmelen, P.W.H.G. Coenen, and G. Van Grootveld. 2003. Emission monitor for the Netherlands: 2000 and estimates for 2001. Report Series MilieuMonitor (Environmental monitor), No 9. The Hague, Netherlands: Coördinatiecommissie Doelgroepmonitoring (CCDM). Available at http://dm.milieumonitor.net/en/.

Lee, S.-M., J.-Y. Jung, and Y.-C. Chung. 2000. Measurement of ammonia inhibition of microbial activity in biological wastewater treatment process using dehydrogenase assay. *Biotechnol. Lett.* 22: 991-994.

Lais, S. 1996. *Untersuchungen zur Reduzierung der Ammoniak- und Geruchsemissionen aus der Schweinehaltung durch biologische Abluftwäscher* (Investigation into reduction of ammonia and odor emissions from pig husbandry using biotrickling filters). Forschungsbericht Agrartechnik des Arbeitskreises Forschung und Lehre der Max-Eyth-Gesellschaft Agrartechnik im VDI, Vol. 293. PhD diss. University of Hohenheim, Germany. ISSN 0931-6264.

Melse, R.W., and N.W.M. Ogink. 2004. Treatment of exhaust air of animal houses. Phase 2: Possibilities for cost reduction (in Dutch). Report 271. Wageningen, Netherlands: Agrotechnology & Food Innovations, Wageningen UR. ISBN 90-6754-835-9.

Melse, R.W., and G. Mol. 2004. Odour and ammonia removal from pig house exhaust air using a biotrickling filter. *Water Sci. Tech.* 50(4): 275-282.

Melse, R.W., and H.C. Willers. 2004. Treatment of exhaust air of animal houses. Phase 1: Techniques and costs (in Dutch, with English abstract). Report 029. Wageningen, Netherlands: Agrotechnology & Food Innovations, Wageningen UR. ISBN 90-6754-739-5.

Mol, G, and N.W.M. Ogink. 2002. Odour Emissions from Animal Husbandry II. General report 2000–2002 (in Dutch). Report 2002-09. Wageningen, Netherlands: Instituut voor Milieu- en Agritechniek (IMAG), Wageningen UR.

Mosquera, J., P. Hofschreuder, J.W. Erisman, E. Mulder, C.E. van 't Klooster, N. Ogink, D. Swierstra, and N. Verdoes. 2002. Methods for measurement of gaseous emissions from animal husbandry (in Dutch). Report 2002-12. Wageningen, Netherlands: Instituut voor Milieu- en Agritechniek (IMAG), Wageningen UR. ISBN 90-5406-216-9.

Mosquera, J., J.M.G. Hol, and N.W.M. Ogink. 2004. Analysis of ammonia emissions from pig husbandry (1990-2003) (in Dutch, with English abstract). Report 312. Wageningen, Netherlands: Agrotechnology & Food Innovations, Wageningen UR. ISBN 30-6754-874.

NNI. 1996. NVN 2820/A1. Luchtkwaliteit, sensorische geurmetingen met behulp van een olfactometer, Wijzigingsblad NVN 2820/A1 (Dutch olfactometric standard). Delft, Netherlands: Nederlands Normalisatie-instituut.

NNI. 1998. Overview of standards for analysis of water and sludges (in Dutch). Delft, Netherlands: Nederlands Normalisatie Instituut.

Ogink, N.W.M., 2005. Unpublished rdata. Agrotechnology & Food Innovations, Wageningen UR, Wageningen, Netherlands.

Ogink, N.W.M., C. ter Beek, and J.V. Klarenbeek. 1997. Odor emissions from traditional and low-emitting swine housing systems. ASAE Annual International Meeting, Minneapolis, Minn., 10-14 August. ASAE Paper No. 974036. St. Joseph, Mich.: ASAE.

Ogink, N.W.M., and P.W.G. Groot Koerkamp. 2001. Comparison of odour emissions from animal housing systems with low ammonia emission. *Water Sci. Tech.* 43(11): 245-252.

Ogink, N.W.M., A. Keen, and J.V. Klarenbeek. 1995. Simulation as a research tool for improving accuracy and precision in olfactometry. In *Proc. Int. Livestock Odor Conference '95*, 35-39. Iowa State University of Science and Technology, Ames, Iowa, 16-18 October.

Ogink, N.W.M., and J.V. Klarenbeek. 1997. Evaluation of a standard sampling method for determination of odour emission from animal housing systems and calibration of the Dutch pig odour unit into standardized odour units. In *Proc. Int. Symp. on Ammonia and Odour Control from Animal Production Facilities*, 231-238. J.A.M. Voermans and G.J. Monteny, eds. Vinkeloord, Netherlands, 6-10 October. Rosmalen, Netherlands: NVTL. ISBN 90-9011059-3.

Ogink, N.W.M., and P.N. Lens. 2001. Odour Emissions from Animal Husbandry I. General report 1996–1999 (in Dutch). Report 2001-14. Wageningen, Netherlands: Instituut voor Milieu- en Agritechniek (IMAG), Wageningen UR.

Prosser, I.J. (ed.) (1986). Nitrification. Society for General Microbiology special publication 20. Oxford, U.K.: IRL Press. ISBN 1-85221-001-X and 1-85221-013-3.

Richardson, J.F., J.R. Backhurst, and J.H. Harker. 2002. Coulson & Richardson's Chemical Engineering: Particle technology and separation processes. Chemical Engineering Vol. 2. Butterworth-Heinemann. ISBN 0 7506 4445 1.

Rowe, J.J., J.M. Yarbrough, J.B. Rake, and R.G. Eagon. 1979. Nitrite inhibition of aerobic bacteria. *Curr. Microbiol.* 2: 51-54.

Schirz, S. 2004. Personal communication. Fachhochschule Münster, Münster, Germany.

Scholtens, R. 1996. Inspection of air treatment systems for mechanically ventilated pig houses (in Dutch). Internal report. Wageningen, Netherlands: Instituut voor Milieu- en Agritechniek (IMAG).

Scholtens, R., J.V. Klarenbeek, and M.A. Bruins. 1988. Control of ammonia emissions with biofilters and bioscrubbers. In *Volatile emissions from livestock farming and sewage operations*, 196-208. V.C. Nielsen, J.H. Voorburg and P. L'Hermite, eds. Barking, Essex, U.K.: Elsevier Applied Science. ISBN 1-85166-277-8.

Takai, H., S. Pedersen, J.O. Johnson, J.H.M. Metz, P.W.G. Groot Koerkamp, G.H. Uenk, V.R. Phillips, M.R. Holden, R.W. Sneath, J.L. Short, R.P. White, J. Hartung, J. Seedorf, M. Schröder, K.H. Linkert, and C.M. Wathes. 1998. Concentrations and emissions of airborne dust in livestock buildings in Northern Europe. *J. Agric. Engng Res.* 70: 59-77.

Uenk, G.H., G.J. Monteny, T.G.M. Demmers, and M.G. Hissink. 1993a. Reduction of ammonia emission from pig houses using bioscrubbers (in Dutch, with

English summary). Report 93-27. Wageningen, Netherlands: DLO Instituut voor Milieu- en Agritechniek (IMAG-DLO). ISBN 90-5406-060-3.

Uenk, G.H., G.J. Monteny, T.G.M. Demmers, and M.G. Hissink. 1993b. Reduction of ammonia emission from pig houses using biofilters (in Dutch, with English summary). Report 93-28. Wageningen, Netherlands: DLO Instituut voor Milieu- en Agritechniek (IMAG-DLO). ISBN 90-5406-059-X.

Van de Sande-Schellekens, A.L.P., and G.B.C. Backus. 1993a. Experiences with air scrubbers on PROPRO farms with fattening pigs b(in Dutch, with English summary). Report P 1.93. Rosmalen, Netherlands: Proefstation voor de Varkenshouderij.

Van de Sande-Schellekens, A.L.P., and G.B.C. Backus. 1993b. Experiences with biofilters on PROPRO farms with fattening pigs (in Dutch, with English summary). Report P 1.99. Rosmalen, Netherlands: Proefstation voor de Varkenshouderij.

Van Groenestijn, J.W., and N.J.R. Kraakman. 2005. Recent developments in biological waste gas purification in Europe. *Chem. Eng. J.* 113(2-3): 86-91.

Van Groenestijn, J.W., and M.E. Lake. 1999. Elimination of alkanes from off-gases using biotrickling filters containing two liquid phases. *Environ. Prog.* 18(3): 151-155.

Van Groenesteijn, J.W., and P.G.M. Hesselink. 1993. Biotechniques for air pollution control. *Biodegradation* 4: 283-301.

Van Middelkoop, J.H. 1995. Air scrubbing: an effective but costly measure (in Dutch). *Praktijkonderzoek* 95(1): 21-23. Beekbergen, Netherlands: Praktijkonderzoek Pluimveehouderij.

Van Ouwerkerk, E.N.J. 1993. Methods to measure NH_3-emission from animal houses (in Dutch). Onderzoek inzake de mest- en ammoniakproblematiek in de veehouderij 16. Wageningen, Netherlands: DLO.

Van 't Riet, K., and J. Tramper. 1991. Basic bioreactor design. New York, U.S.A: Marcel Dekker Inc. ISBN 0 8247 8446 4.

Verdoes, N., and J.J. Zonderland. 1999. The effect of a chemical air scrubber on ammonia emission from a growing-finishing pig house (in Dutch, with English summary). Report P 4.39. Rosmalen, Netherlands: Praktijkonderzoek Varkenshouderij.

Vrielink, M.G.M., N. Verdoes, and J.P.B.F. Van Gastel. 1997. Reducing the ammonia emission with a chemical air scrubber (in Dutch, with English summary). Report P 1.178. Rosmalen, Netherlands: Praktijkonderzoek Varkenshouderij.

VROM. 2002. Regeling Ammoniak en Veehouderij (Regulation on Ammonia and Livestock). *Staatscourant*, 82, 1 mei 2002. The Hague, Netherlands: Sdu Uitgevers.

Wever, A.C., and P.W.G. Groot Koerkamp. 1999. Acid scrubbing of exhaust air from farrowing, dry, and pregnant sows (in Dutch, with English summary). Report 99-126. Wageningen, Netherlands: Instituut voor Milieu- en Agritechniek (IMAG).

Willers, H.C. 2005. Unpublished data. Agrotechnology & Food Innovations, Wageningen UR, Wageningen, Netherlands.

Yarbrough, J.M., J.B. Rake, and R.G. Eagon. 1980. Bacterial inhibitory effects of nitrite—inhibition of active-transport, but not of group translocation, and of intracellular enzymes. Appl. Environ. Microbiol. 39:831–834.

Chapter 4

Size reduction of ammonia scrubbers for pig and poultry houses: Use of conditional bypass vent at high air loading rates

R.W. Melse[1]; A.V. van Wagenberg[2]; J. Mosquera[1]

[1]Wageningen University and Research centre, Agrotechnology & Food Innovations, Wageningen, Netherlands;[2]Wageningen University and Research centre, Animal Sciences Group, Lelystad, Netherlands.

ABSTRACT In the Netherlands, both acid and biological air scrubbers are used for removal of ammonia from exhaust air at mechanically ventilated pig and poultry houses. Current regulations require that scrubbers are dimensioned for treating the maximum air flow rate that may occur, so on average these systems are overdimensioned and underloaded. A new approach is introduced that is based on bypassing air flow peaks untreated. As a result, the air loading rate (m^3 [air] m^{-3} [scrubber] h^{-1}) and ammonia loading rate (kg [NH_3] m^{-3} [scrubber] h^{-1}) of the scrubber are more constant in time and average loading rates increase. By model calculations and analyses of measurement datasets it was demonstrated that the application of such a scrubber significantly decreases the required scrubber size while ammonia emission levels are only slightly increased (*e.g.* where the bypass is operated at 50% of the maximum ventilation rate and the scrubber volume is reduced by 50%, the bypass venting systems only allows 10 - 20% of the total ammonia load to be vented untreated). As a result, both the efficiency of scrubber utilization (kg [NH_3 removal] m^{-3} [scrubber volume]) and the cost-effectiveness of air scrubbing for ammonia removal (kg [NH_3 removal] €$^{-1}$) are increased.

Reproduced with permission from Biosystems Engineering, volume 95, issue number 1, pages 69-82. Copyright 2006 IAgrE.

4.1 Introduction

4.1.1 Animal husbandry and ammonia abatement

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 characterised by large-scale intensive farms. The Netherlands, with 16 million inhabitants and a population density of about 400 inhabitants per km^2, houses 11 million pigs at approximately 10,000 farms (CBS, 2004).

From the 1980's onwards, the emission of ammonia (NH_3) from livestock farming has become a major environmental concern because ammonia emission is one of the three main sources of soil acidification and eutrophication of natural soils in the Netherlands (Heij and Erisman, 1995, 1997). Therefore considerable efforts have been put into the development of ammonia abatement techniques in animal operations. This focus on ammonia abatement has resulted in the development of a variety of low emission livestock housing systems that are applied today. These systems include end-of-pipe systems for treatment of the exhaust air from pig and poultry houses, *viz* acid scrubbers and bioscrubbers or biotrickling filters. However, investment and operational costs of scrubber systems are generally considered as high.

4.1.2 Working principle of ammonia scrubbers

An air scrubber, or trickling filter, is a reactor that has been packed with an inert packing material. The packing material usually has a large porosity, or void volume, and a large specific area. The packed bed is wetted by spraying water on top. Exhaust air from an animal house, containing ammonia, is introduced either in a cross-current or counter-current direction which results in intensive contact between air and water enabling mass transfer from air to liquid phase. A schematic of a trickling filter is given in Figure 4.1.

In an acid scrubber, the pH of the recirculation water is kept below 4 by addition of acid, usually sulphuric acid. The ammonia dissolves in the liquid phase and is captured by the acid forming an ammonium salt solution which is discharged on a regular basis and replaced with fresh water. The average ammonia removal efficiencies of acid scrubbers may vary from 40 - 100% (overall average of 96%) as was demonstrated in on-farm research at three pig and two poultry sites (Melse and Ogink, 2005).

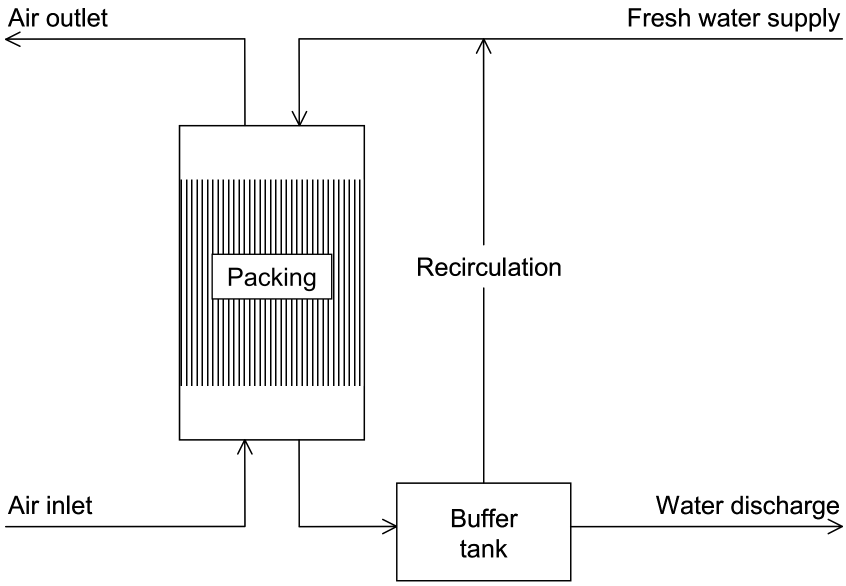


Figure 4.1 Schematic of an air scrubber; from Melse and Ogink (2005).

In a biotrickling filter, bacteria are present partly as a biofilm on the packing material and partly suspended in the water that is being recirculated. The dissolved ammonia is converted into nitrite (NO_2^-) and subsequently into nitrate (NO_3^-) by nitrification which is mainly carried out by *Nitrosomonas* and *Nitrobacter* species, respectively (Focht and Verstraete, 1977; Prosser, 1986). The liquid phase, containing nitrite and nitrate, is discharged on a regular basis and replaced with fresh water. The average ammonia removal efficiencies of biotrickling filters may vary from negative values to 100% (overall average of 70%) as was demonstrated in on-farm research at five pig sites, one poultry, and one veal calve site (Melse and Mol, 2004; Melse and Ogink, 2005).

In the Netherlands about 160 acid scrubbers and 45 biotrickling filters are in operation for ammonia removal from ventilation air of animal houses, mainly at pig and poultry farms (Melse and Ogink, 2005; Melse and Willers, 2005).

4.1.3 Air scrubber dimensioning

For mechanically ventilated pig houses, ventilation rates vary from 10 to 90 m^3 fattening pig^{-1} h^{-1}, depending on animal production stage and weather conditions (Ogink and Lens, 2001; Seedorf *et al.*, 1998). The average year-round ventilation rate for fattening pigs is about 35 m^3 $animal^{-1}$ h^{-1} at Dutch weather conditions, *i.e.* a moderate maritime climate. For mechanically ventilated broiler houses, ventilation rates usually vary from 1 to 3.6 m^3 kg^{-1} [liveweight] h^{-1} (ASG, 2004; Seedorf *et al.*, 1998), which equals 0.04 to 9 m^3 $broiler^{-1}$ h^{-1}, depending on weather conditions. The average year-round ventilation rate for broilers is about 3 m^3 $animal^{-1}$ h^{-1}. Minimum empty bed air residence times (EBRT) of ammonia scrubbers, *i.e.* scrubber volume in m^3 divided by maximum air flow rate in m^3 s^{-1}, have typically values of 0.5 - 1.0 s, which equals air loading rates from 7,200 to 3,600 m^3 m^{-3} [scrubber volume] h^{-1}.

Pig houses with separated room ventilation systems are usually equipped with a central ventilation system if an air scrubber system is applied. In a central ventilation system, one or more central fans withdraw air from the rooms into a central ventilation duct, so air that exits the central ventilation duct and subsequently enters the scrubber system is a mixture of the exhaust air from all rooms. The air flow rate and ammonia concentration vary from room to room if in each room a batch is started at a different date. The use of a central ventilation system, which mixes the exhaust air from several rooms that each contain pigs of a different age, will reduce these variations and result in a more constant loading rate of both air in m^3 m^{-3} [scrubber volume] h^{-1} and ammonia in kg [NH_3] m^{-3} [scrubber volume] h^{-1}. The more constant air loading rate results in a lower average ventilation rate per animal place, thus the installed ventilation capacity will be lower than for a ventilation system per room. Consequently, the required scrubber size for treating all air will be lower. The foregoing is not the case for a pig house in which all rooms are operated simultaneously and where all pigs in the house have the same age.

4.1.4 Air scrubber with bypass vent

Although air scrubbing systems for ammonia removal from animal house exhaust air are commercially available in the Netherlands and considered as off-the-shelf techniques today, investment and operational costs are relatively high as compared to other available housing systems with emission reduction techniques. Current regulations in the Netherlands require that ammonia scrubbers that are applied at animal houses at all times treat the entire exhaust air flow and meet the required minimum removal efficiency. Therefore conventional scrubbers are designed for treating the maximum exhaust air flow rate, even while this maximum air flow rate only occurs for a short period of time. So most of the time these scrubbers are oversized and underloaded, even if they are connected to a central ventilation system.

However, a significant reduction of the investment and operational costs of air scrubbing is achieved if required scrubber volumes can be decreased so that scrubbers are dimensioned for lower maximum ventilation rates. This could be achieved by combining an air scrubber with an air bypass system that conditionally allows a part of the exhaust air to bypass the scrubber and be vented untreated. In this way a scrubber can be designed for the average air flow as peaks in the air flow will be bypassed. In such a system, the bypass vent is set to operate whenever the actual air flow rate exceeds the maximum air flow rate the scrubber has been designed for, *i.e.* the bypass setpoint air flow rate. Only the part of the air that exceeds this setpoint is bypassed, so the scrubber is operated just at its designed air flow rate. It is not clear beforehand to what extent the ammonia emission rate will be increased if part of the exhaust air is vented untreated, as the ammonia concentration of the air varies.

4.1.5 Objectives and approach

The first aim of this paper is to analyse how ventilation rate and ammonia emission from pig and poultry houses are interrelated and how the conditional bypassing of part of the exhaust air of the scrubber system affects the total ammonia emission. The second aim is to determine how conditional bypassing affects the required scrubber size, because a reduction of scrubber size will increase the economic feasibility of ammonia removal by air scrubbing.

In order to achieve these aims, previously gathered experimental datasets of ammonia emission and ventilation rate during batches of broiler and fattening pigs (Table 4.1) are analysed. For fattening pigs, a model is developed for year-round simulations of the ammonia emission when using a scrubber with a bypass vent, both for a central ventilation system and for a ventilation system per room.

4.2 Materials and methods

4.2.1 Ammonia emission datasets for fattening pigs and broilers

From previously carried out research, six datasets were analysed that contained continuous measurements of the ventilation rate and ammonia concentration of exhaust air from fattening pigs (dataset 1 - 4) and broilers (dataset 5 and 6) in conventional housing systems. All houses were equipped with a mechanical ventilation system that is controlled by the temperature of the room air; ventilation and temperature setpoints, heat production by the animals and outside conditions vary in time during a batch, resulting in a varying ventilation rate. In Table 4.1 some details are given for these datasets.

For each dataset, the ventilation rate in $m^3\ h^{-1}$ was measured continuously by a calibrated ventilation rate sensor in the ventilation shaft. The exhaust air was

Table 4.1 Datasets of hourly averaged ventilation rate and ammonia emission measured at conventional housing systems for pigs and poultry.[a]

Dataset no.	Source	Measuring period	No. of animals present	Room temperature, min.-max. (average), °C	Ventilation rate, min.-max. (average), m^3 $(animal)^{-1}$ h^{-1}	Ammonia emission, min.-max. (average), mg $(animal)^{-1}$ h^{-1}
1	Satter *et al.*, 1997	Batch 1: 13 March - 9 July, 1996	130 fattening pigs	19.1-34.9 (23.1)	5.3-57.5 (28.8)	45-640 (218)
		Batch 2: 17 July - 18 November, 1996	130 fattening pigs	19.2-31.0 (22.9)	7.5-53.7 (27.6)	108-514 (277)
2	Groenestein and Huis in 't Veld, 1996	15 July - 9 November, 1995	110 fattening pigs	17.6-31.4 (22.7)	8.5-65.0 (42.9)	32-672 (330)
3	Hol and Groenestein, 2005	Batch 1: 4 June - 10 October, 2002	80 fattening pigs	19.3-33.1 (24.3)	22.9-66.8 (49.6)	155-551 (402)
		Batch 2: 23 October, 2002 - 16 January, 2003	80 fattening pigs	17.7-25.6 (20.7)	14.6-43.6 (24.0)	180-628 (393)
4	Huis in 't Veld and Groenestein, 1995	1 June - 28 September, 1994	64 fattening pigs	16.1-33.5 (23.3)	11.6-112.4 (65.8)	62-562 (252)
5	Wever *et al.*, 1999	Batch 1: 22 July - 31 August, 1998	41,040 broilers	15.9-33.7 (26.5)	0.13-3.44 (1.67)	0.0-42.7 (11.9)
		Batch 2: 16 October - 23 November, 1998	40,630 broilers	12.1-34.0 (26.5)	0.08-1.99 (0.79)	0.0-41.0 (8.8)
6	Hol and Groot Koerkamp, 1998	Batch 1: 11 July - 20 August, 1997	11,925 broilers	21.2-32.8 (26.7)	0.14-4.82 (1.80)	0.1-12.7 (4.1)
		Batch 2: 2 September - 13 October, 1997	10,900 broilers	18.3-36.7 (25.4)	0.10-5.25 (1.48)	0.1-28.5 (7.1)
		Batch 3: 25 October - 5 December, 1997	11,000 broilers	18.7-33.9 (25.0)	0.10-2.06 (0.88)	0.1-12.8 (5.8)
		Batch 4: 24 July - 2 September, 1998	10,865 broilers	10.6-32.9 (25.7)	0.10-4.35 (1.64)	0.1-65.6 (15.5)

[a] All farms were located in the Netherlands.

continuously sampled using a vacuum pump at a fixed flow rate which was controlled by a critical orifice (0.5 *l* minute^{-1}) and led to a NH_3 converter/NO_x-analyser system in which determination of the ammonia concentration in mg m^{-3} took place (Mosquera *et al.*, 2002). All sampling tubes had been made of Teflon, were isolated, and heated with a coil of resistance wire to a temperature of approximately 20°C above the ambient temperature to prevent condensation of water and subsequent adsorption of ammonia. The data of the continuously measured ventilation rate and ammonia concentration were averaged over one-hour intervals and the resulting datasets were analysed in this study.

All datasets were gathered during rather short periods (several months) with the specific weather conditions at that time and place. Furthermore, each dataset for fattening pigs contains the measurements of the ventilation rate and ammonia concentration of a single pig room. In practice, however, scrubber systems are usually installed at pig houses that are equipped with a central ventilation system.

In order to estimate the effect of the use of a scrubber system with a bypass vent on the year-round emission of ammonia from a fattening pig house without these drawbacks, both for a central ventilation system and for a ventilation system per room, model calculations were done of ventilation rate and ammonia emission.

4.2.2 Modelling of the ventilation rate of a pig house

Model calculations of ventilation rates and ammonia emission were done to describe the year-round emission pattern of a fattening pig house. This pattern was used to estimate the effect of the use of a scrubber system with a bypass vent on the ammonia emission.

For calculating the ventilation rate as a function of outside temperature and animal production stages (day of batch), relationships were used that had been calculated by Van Wagenberg and Vermeij (2001a, 2001b) with the simulation model ANIPRO (Van Ouwerkerk, 1999); these relationships are shown in Figure 4.2. The ANIPRO model has been based on earlier detailed studies on heat production of pigs (Bruce and Clark, 1979; Sterrenburg and Van Ouwerkerk, 1986a and 1986b). The conditions used as input for the ANIPRO model were based on a conventional situation that is representative for pig houses in the Netherlands, *i.e.* mechanically ventilated and insulated. It is assumed that the fattening pigs have a weight of 23 kg at the start and 110 kg at the end of a batch of 110 days.

For the outside air temperature, *i.e.* the temperature of the inlet air of the ventilation system, a meteorological reference year was used which is representative for Dutch weather conditions and contains year-round hourly temperature values (Lund, 1984). The temperature profile of this reference year is shown in Figure 4.3.

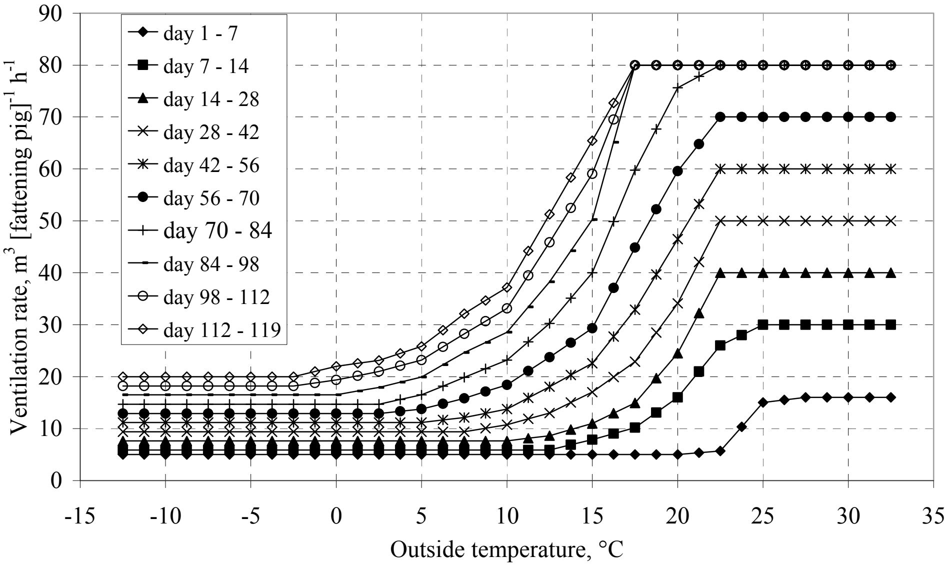


Figure 4.2 Ventilation rate for fattening pigs, depending on production stage (day of batch) and outside temperature; from Van Wagenberg and Vermeij (2001a).

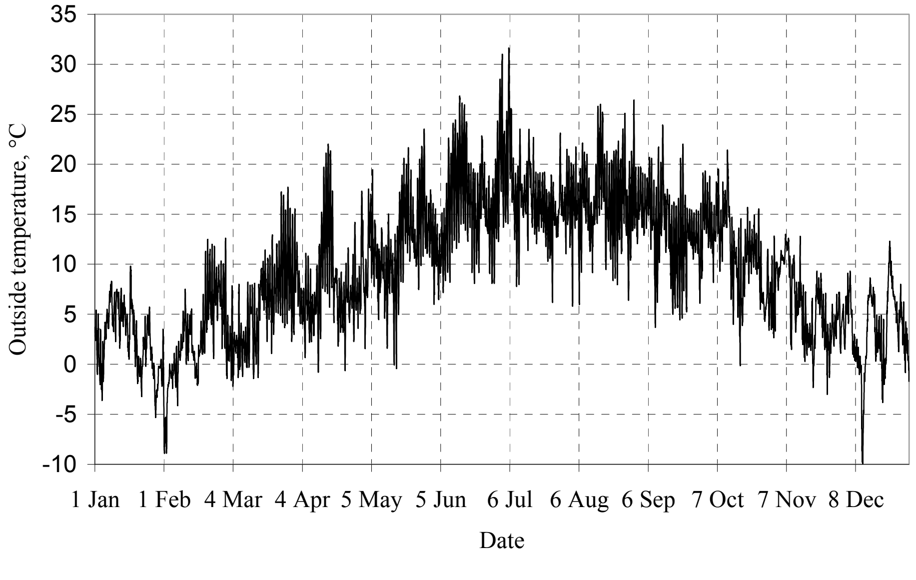


Figure 4.3 Temperature pattern of a meteorological reference year for the Netherlands (hourly averages); from Lund (1984).

Simulations of the ventilation rate were done both for a central ventilation system and for a ventilation system per room. For the pig house with the central ventilation system, it is assumed that the house contains 12 rooms which are operated non-simultaneously, *i.e.* the animals differ 9 days in age from room to room, so every 9 days a new batch is started in one of the rooms.

4.2.3 Modelling of the ammonia emission from a pig house

The ammonia emission rate in kg [NH_3] h^{-1} of pig houses is correlated to the animal production stage as the ammonia emission rate depends on the amount of manure that has been accumulated inside the animal house (Ni *et al.*, 2000; Mosquera *et al.*, 2005). Mosquera *et al.* (2005) analysed 34 ammonia emission datasets that were gathered at 19 different pig farm locations in the Netherlands. The datasets contained hourly-averaged values of continuous measurements of the ventilation rate and ammonia concentration of exhaust air from fattening pigs in both conventional housing systems and housing systems with emission reduction techniques, *viz* manure flushing, manure cooling, or reduction of emitting manure surface. The average number of animals included in the datasets was 95 fattening pigs and the average duration of the measurement period was 91 days.

Ammonia emission, calculated as the average of all datasets, linearly increases during a batch (Figure 4.4). The equation of Figure 4.4 was used to model the ammonia emission year-round as Mosquera *et al.* (2005) could not distinguish a seasonal effect on the ammonia emission pattern.

4.3 Results and discussion

4.3.1 Analysis of ammonia emission datasets for fattening pigs and broilers

Fattening pigs

The ventilation rate varies in time as it is controlled by the temperature of the exhaust air which in turn depends on pig production stage and outside temperature. For datasets 1-4 (pig houses), the cumulative frequency distribution of the ventilation rate was determined, *i.e.* for every value of the ventilation rate it was calculated what percentage of the time the actual air flow had been below this value. In Figure 4.5, as an example, this relationship is plotted for dataset 2. From Figure 4.5 it can be seen that the maximum ventilation rate was 65 m^3 (fattening pig)$^{-1}$ h^{-1} [for 100% of the time the ventilation rate was below 65 m^3 (fattening pig)$^{-1}$ h^{-1}] and that, for example, for 50% of the time the actual ventilation rate was below 45 m^3 (fattening pig)$^{-1}$ h^{-1}.

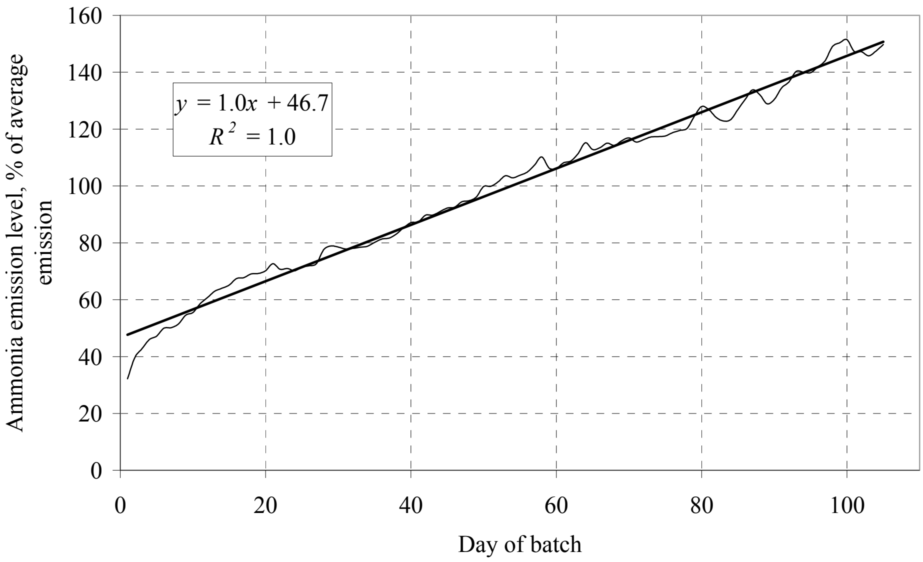


Figure 4.4 Ammonia emission pattern of fattening pigs during a batch (day 1 - 105), expressed as percentage of the average emission level (average of 34 emission datasets); the equation of the trendline and the coefficient of determination are given in the text box; from Mosquera *et al*. (2005); R^2, coefficient of determination.

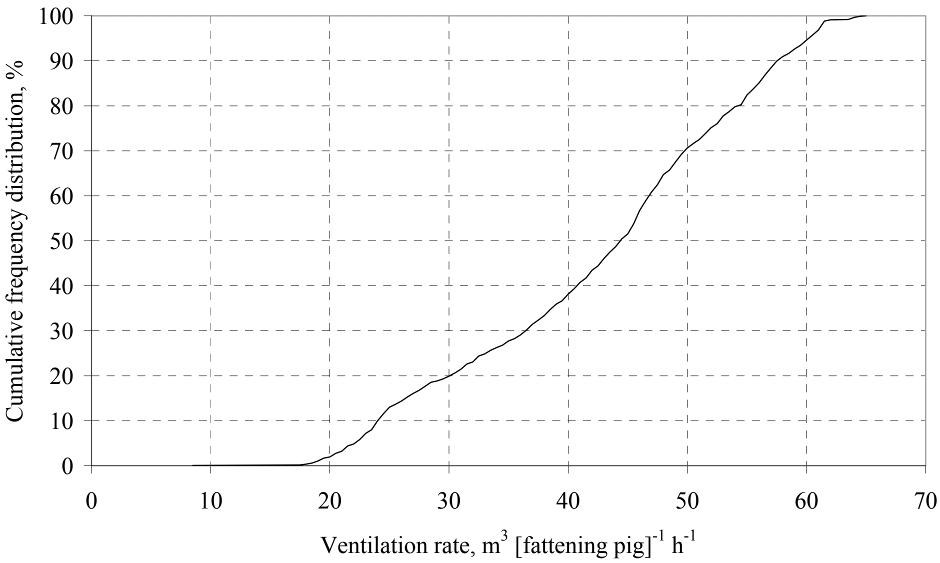


Figure 4.5 Cumulative frequency distribution of ventilation rate (single room with 110 fattening pigs, dataset 2).

If a scrubber with a bypass system had been installed, from the cumulative frequency distribution pattern it follows what percentage of the time a part of the air would be bypassed and vented untreated for a particular bypass setpoint air flow rate. There is a large variation in the ammonia concentration of the exhaust air for dataset 2 (Figure 4.6).

With regard to the unwanted emission of ammonia, however, it is necessary to know the amount of ammonia in kg h^{-1} that is emitted through the bypass. The amount of ammonia that would be vented untreated if a scrubber system with a bypass would be used, was calculated by combining the hourly measured ventilation rates and ammonia concentrations. In Figure 4.7 the bypassed ammonia load is plotted as a function of the bypass setpoint flow rate for all datasets. For dataset 1, 2, and 3 the trend of Figure 4.7 is similar and all have a maximum ventilation rate of about 60 m^3 (fattening pig)$^{-1}$ h^{-1}, which is a current design air flow rate for mechanical ventilation systems in pig houses. Dataset 4 has a maximum ventilation rate of over 100 m^3 (fattening pig)$^{-1}$ h^{-1} and is considered as a non-representative situation.

The average of dataset 1 - 3 clearly shows that in case of a bypass setpoint flow rate of 30 m^3 (fattening pig)$^{-1}$ h^{-1}, which is 50% of the maximum air flow rate of about 60 m^3 (fattening pig)$^{-1}$ h^{-1}, still 80% of the ammonia load enters the scrubber and only 20% is vented untreated.

Assuming that the maximum air loading rate of the scrubber in m^3 m^{-3} [scrubber volume] h^{-1} remains unchanged, the size of the scrubber with the bypass setpoint of 50% is only 50% of the size of a conventional ammonia scrubber without a bypass vent. Assuming the same relative ammonia removal (%) as for a conventional scrubber, the efficiency of scrubber utilization, expressed as kg [NH_3 removal] m^{-3} [scrubber volume], thus increases. The slopes of the tangent lines that can be drawn for the curves of Figure 4.7 represent this efficiency. For the curve that indicates the average of dataset 1 - 3, starting at the right end of the x-axis, the tangent line slope continually decreases (gets more negative) with decreasing x-as values down to a bypass setpoint of 20 m^3 (fattening pig)$^{-1}$ h^{-1}. This means the efficiency of scrubber utilization has its maximum value at a bypass setpoint of 20 m^3 (fattening pig)$^{-1}$ h^{-1} or lower. For dataset 4, a similar relationship exists.

Broilers

For the two datasets on ventilation rates and ammonia concentrations of broilers, similar calculations were done as for fattening pigs. In Figure 4.8 the ammonia load is plotted that would be bypassed if a scrubber with a bypass vent had been installed as a function of the bypass setpoint flow rate. As for Figure 4.7, starting at the right end of the x-axis, the tangent line slopes for the curves in

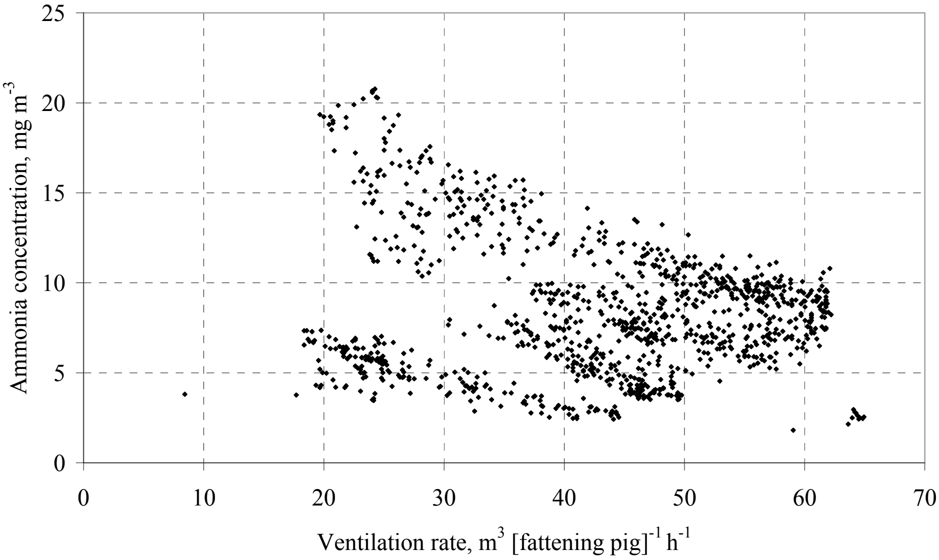


Figure 4.6 Plot of measured ammonia concentration versus ventilation rate (single room with 110 fattening pigs, dataset 2).

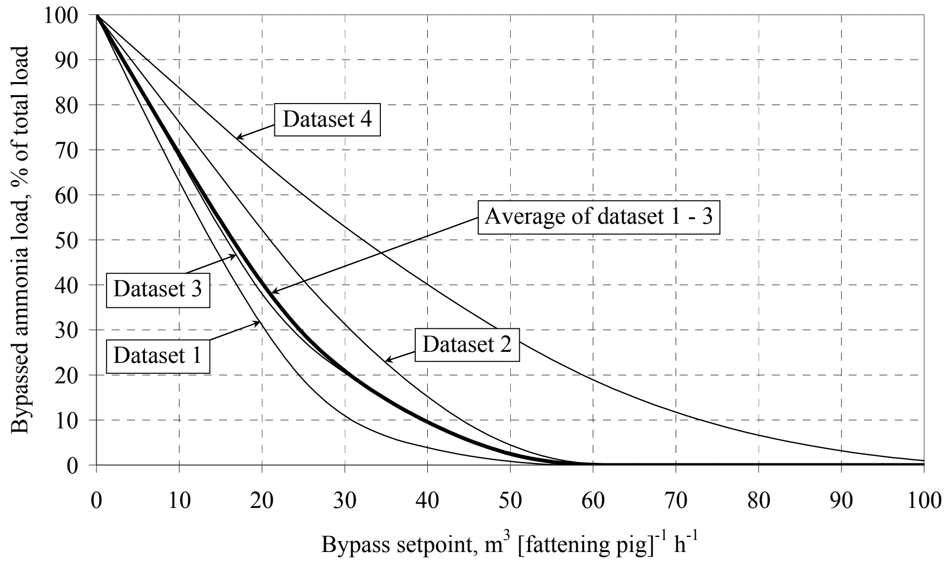


Figure 4.7 Amount of ammonia that would be bypassed if a scrubber with a bypass vent had been installed as a function of the bypass setpoint (fattening pigs, dataset 1 - 4).

Figure 4.8 continually decrease (get more negative) with a decreasing bypass setpoint flow rate. A decreasing tangent line slope means that the efficiency of scrubber utilization in kg [NH_3 removal] m^{-3} [scrubber volume] increases. The average of dataset 5 and 6 clearly shows that in case of a bypass setpoint flow rate of 1.75 m^3 (broiler)$^{-1}$ h^{-1}, which is 50% of the maximum air flow rate of about 3.5 m^3 (broiler)$^{-1}$ h^{-1}, still 85% of the ammonia load enters the scrubber and only 15% is vented untreated.

4.3.2 Model results of the ammonia emission from a scrubber with a bypass vent at a pig house

Ventilation rate

The ventilation rate was calculated for an individual pig room during one year. Every 112 days a new batch is started with a low ventilation rate at the start and a high ventilation rate at the end date (between batches, rooms are empty for 2 days); the maximum ventilation rate is 80 m^3 (fattening pig)$^{-1}$ h^{-1} (Figure 4.9).

In Figure 4.10 the calculated ventilation rate is shown for a central ventilation system with 12 rooms during one year. The ventilation rate is significantly higher in summer than in winter. The variation of the average ventilation rate for the whole building (Figure 4.10) is much smaller than for the ventilation rate of a single room (Figure 4.9) and batch start and end dates cannot be distinguished anymore; the maximum of the average ventilation rate for the whole building is 62 m^3 (fattening pig)$^{-1}$ h^{-1}. The average ventilation rate for both the simulation of a single room and the simulation of 12 rooms, is 27 m^3 (fattening pig)$^{-1}$ h^{-1}.

Ammonia emission

From 13 datasets containing continuous measurement of ventilation rate and ammonia concentrations, each gathered during several months at 9 different locations in the Netherlands, Mosquera *et al.* (2005) calculated that the average ammonia emission from fattening pigs in a conventional housing system was 2.9 kg animal^{-1} yr^{-1}. Using this value and Figure 4.4, the year-round ammonia emission was calculated for individual fattening pig rooms. In Figure 4.11, the ammonia emission that was calculated is shown for one of the rooms as an example; for every next room, the emission pattern is shifted by 9 days.

The calculated year-round ammonia emission, as shown in Figure 4.11 for a single room, is added for all 12 rooms and divided by the total ventilation rate of the pig house that was modelled (Figure 4.10). This results in a simulation of the year-round ammonia concentration of the exhaust air from the central ventilation system and is shown in Figure 4.12.

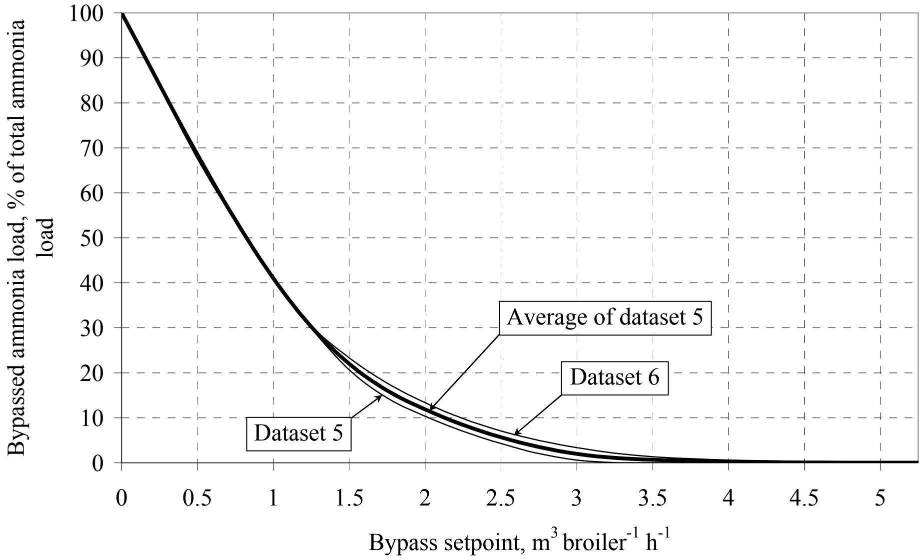


Figure 4.8 Amount of ammonia that would be bypassed if a scrubber with a bypass vent had been installed as a function of the bypass setpoint (broilers, dataset 5 and 6).

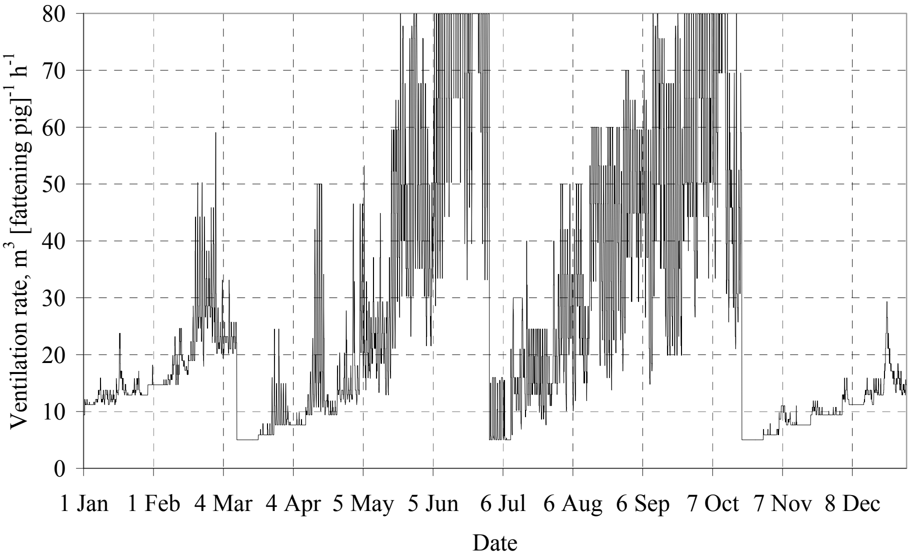


Figure 4.9 Model calculation of the ventilation rate of one fattening pig room for a meteorological reference year (hourly averages); every 112 days a new batch is started.

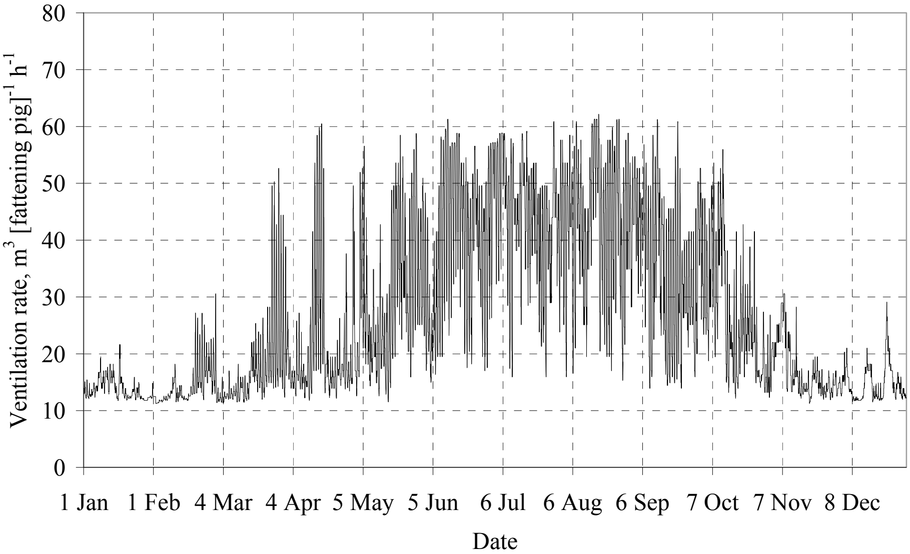


Figure 4.10 Model calculation of the ventilation rate of 12 fattening pig rooms with a central ventilation system for a meteorological reference year (hourly averages); every 9 days a new batch is started in one of the rooms.

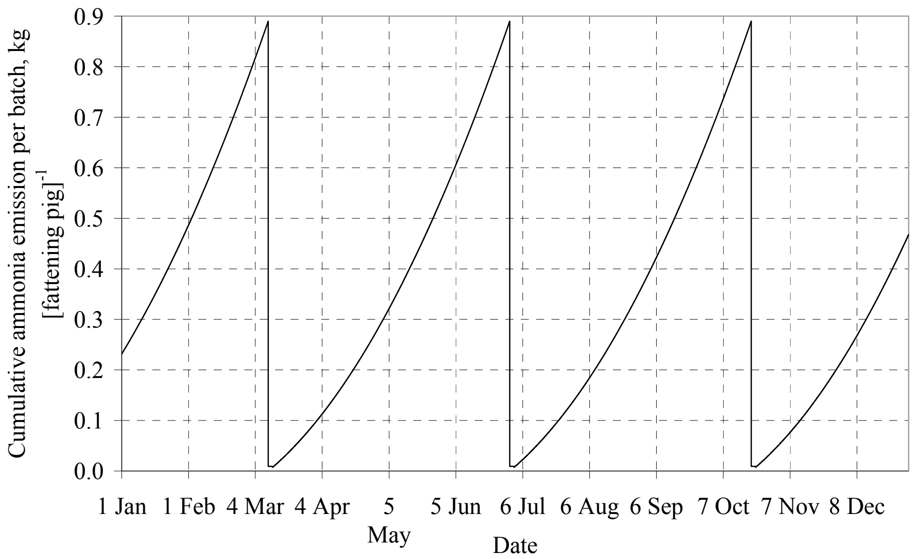


Figure 4.11 Model calculation of the ammonia emission from a fattening pig room, accumulated per batch.

Effect of the use of a scrubber with a bypass vent on the year-round ammonia emission of a pig house

Finally, from the modelled year-round ventilation rate and ammonia concentration data for the central ventilation system, the effect of the installation of an air scrubber with a bypass vent was determined (Figure 4.13), in the same way as was done for the datasets with the actual ammonium and ventilation rate measurements (Figure 4.7 and 4.8). In Figure 4.13, the ammonia load in kg h^{-1} that would be bypassed if an air scrubber with a bypass vent had been installed is plotted as a function of the bypass setpoint flow rate. As long as the tangent line slopes of the curves in Figure 4.13 decrease (get more negative) with a decreasing bypass setpoint flow rate, the efficiency of scrubber utilization in kg [NH_3 removal] m^{-3} [scrubber volume] increases, assuming that the same relative ammonia removal (%) is achieved and the same air loading rate of the scrubber in m^3 m^{-3} [scrubber volume] h^{-1} is used as for a conventional scrubber without a bypass vent.

For a ventilation system per room, the simulation of Figure 4.13 shows that in case of a bypass setpoint flow rate of 30 m^3 (fattening pig)$^{-1}$ h^{-1}, still 84% of the ammonia load enters the scrubber and only 16% is vented untreated. The datasets with the measured ammonia emissions showed that in this situation 20% of the ammonia was vented untreated (Figure 4.7, average of dataset 1 - 3), so the simulation of Figure 4.13, which is a year-round simulation using a meteorological reference year, indicates that the cost-reducing effect of using a bypass vent is even stronger than was shown by Figure 4.7. This can be explained by the fact that each dataset with measurement was gathered during one batch only with the specific weather conditions at that time and place, but could also by influenced by limitations of the model that was used.

Furthermore, Figure 4.13 shows that the use of a central ventilation system, as had been expected, further reduces the cost of ammonia removal by air scrubbing. In case of a bypass setpoint flow rate of 30 m^3 (fattening pig)$^{-1}$ h^{-1}, still 89% of the ammonia load enters the scrubber and only 11% is vented untreated, whereas 16% was vented untreated for the ventilation system per room. The efficiency of scrubber utilization has its maximum value at a bypass setpoint of 13 m^3 (fattening pig)$^{-1}$ h^{-1} or lower for the central ventilation system.

Both the investment and operational costs of air scrubbing will be reduced, as an increased scrubber utilization efficiency means that a relatively small scrubber needs to be operated in comparison with a conventional scrubber without a bypass venting system. For a bypass scrubber system, the investment cost per m^3 scrubber size will be slightly higher than for a conventional scrubber system because of the additional costs of the bypass itself.

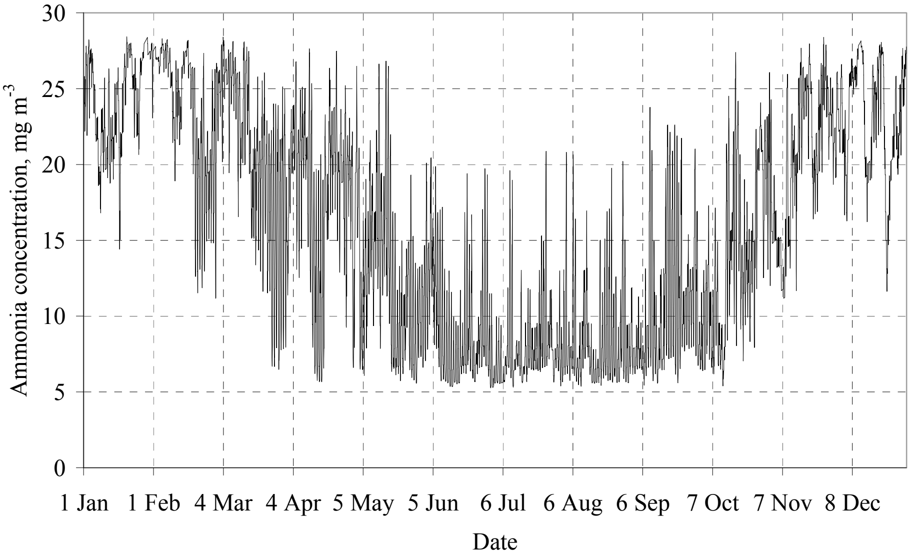


Figure 4.12 Model calculation of the ammonia concentration in the air from a central ventilation system for 12 rooms for a meteorological reference year (hourly averages); every 9 days a new batch is started in one of the rooms.

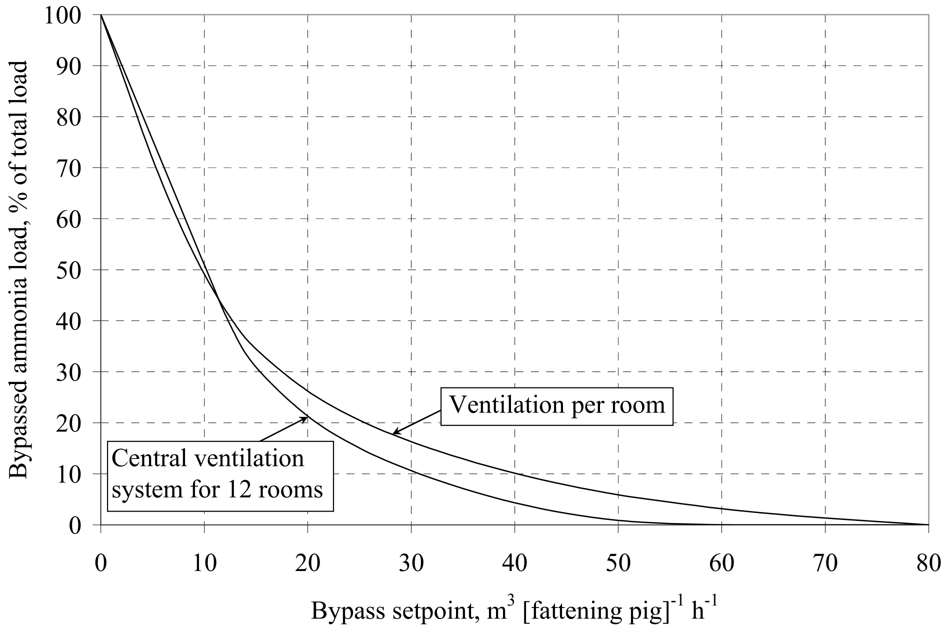


Figure 4.13 Amount of ammonia that would be bypassed if a scrubber with a bypass vent was installed as a function of the bypass setpoint, both for ventilation per room and for a central ventilation system; model calculations for a meteorological reference year.

4.3.3 General discussion

In Figure 4.7, 4.8, and 4.13, it was demonstrated that for the treatment of exhaust air from pig and poultry houses the use of a bypass vent increases the efficiency of scrubber utilization (kg [NH_3 removal] m^{-3} [scrubber volume]), as long as the maximum air loading rate of the scrubber (m^3 m^{-3} [scrubber volume] h^{-1}) remains unchanged and the scrubber size thus is reduced. This implies that the costs of air scrubbing will be reduced, as the investment and operational costs of air scrubbing are correlated to the scrubber size.

Bypassing peaks in the air flow results in an increase of the average air loading rate of the system, even when the maximum air loading rate of the scrubber is assumed to be unchanged. A conventional scrubber that has been designed for a maximum air flow of 60 m^3 (fattening pig)$^{-1}$ h^{-1} has an average air loading rate of 1,600 m^3 m^{-3} [scrubber volume] h^{-1} and a maximum air loading rate of 3,600 m^3 m^{-3} [scrubber volume] h^{-1} (minimum EBRT is 1.0 s). From the data for the central ventilation system presented in Figure 4.10, it can be calculated that the installation of a half-sized scrubber with a bypass vent at a bypass setpoint flow rate of 30 m^3 (fattening pig)$^{-1}$ h^{-1} increases the average air loading rate of the scrubber with 60% from 1,600 to 2,600 m^3 m^{-3} [scrubber volume] h^{-1}, assuming the same maximum air loading rate is applied. This increase of the average air loading rate, or air velocity in the scrubber bed, increases the pressure drop over the scrubber and thus increases the energy costs of the mechanical ventilation system. A conventional scrubber without a bypass vent in general has a maximum pressure drop of about 200 Pa at the designed maximum air flow rate and an average pressure drop of about 50 Pa; it was calculated that the average pressure drop will increase from 50 to about 100 Pa by the use of a smaller scrubber with a bypass vent.

As it is desirable to minimize the extra pressure drop in the scrubber, the spatial dimensions of the scrubber (length, width, height) can be changed to level down this increase of the pressure drop, even if the scrubber volume remains unchanged. When the surface area of the scrubber is increased by increasing the, in case of an upward air flow direction, width and length of the scrubber, the surface loading rate (m^3 [air] m^{-2} [scrubber area] h^{-1}) decreases and thus a lower air velocity is achieved in the bed. At the same bed volume, increasing the width and length means that the height of the bed is reduced. The pressure drop, which is proportional to the bed height and the square of the air velocity, can thus be levelled down. The model calculations show that the height of the bed needs to be reduced by 27% in order to reduce the average pressure drop of the scrubber with the bypass-system down to the average pressure drop of a conventional scrubber, assuming the same maximum air loading rate is applied for both systems.

Finally, the average ammonia load of the scrubber with a bypass vent, expressed as kg [NH_3] m^{-3} [scrubber volume] h^{-1}, is higher than for a conventional scrubber without a bypass vent. From the data for the central ventilation system in Figure 4.13, it can be calculated that the installation of a scrubber with a bypass vent at a bypass setpoint flow rate of 30 m^3 (fattening pig)$^{-1}$ h^{-1} increases the average ammonia loading rate of the scrubber from 20 to 35 g m^{-3} [scrubber volume] h^{-1}. As a result of the increased ammonia loading rate, the relative ammonia removal in % of the scrubber system might decrease in theory.

For an acid scrubber, it is expected that the relative removal efficiency will hardly be influenced by an increase of the average ammonia loading rate as the scrubber is operated at low pH so that the ammonia is captured by the acid very quickly and removed with the discharge water.

For a biotrickling filter, however, the relative removal efficiency might be influenced by an increase of the average ammonia loading rate. After its transfer to the liquid phase, the ammonia must be biologically converted and therefore a larger amount of nitrifying biomass is needed in the scrubber system at a higher average ammonia loading.

On the other hand, currently operating conventional ammonia scrubbers for treatment of exhaust air from animal houses, both acid scrubbers and biotrickling filters, have been designed to achieve sufficient ammonia removal under all operating conditions that may occur in practice, including occasionally high ammonia and air loading rates. Therefore these systems might be capable of successful ammonia removal at higher average ammonia and air loading rates without reduction of the relative ammonia removal.

Further experimental research is necessary to demonstrate if the design and cost calculations for air scrubber, based on the relationships of Figure 4.7, 4.8, and 4.13, need to be corrected for these aspects. Also it is necessary to gather practical experience in design, application and control of bypass venting system. Finally, further work is required to determine if the air scrubbing strategy described in this paper is applicable for mechanical ventilated houses for other animal species than pigs and poultry (*e.g.* veal calves) and how it influences odour emission from animal houses.

4.4 Conclusion

Conventional acid and biological air scrubbers that are used for ammonia removal from exhaust air at mechanically ventilated pig and poultry houses have the capacity for treating the entire air flow at all times. As a result of the fluctuating ammonia emission pattern of this air, *i.e.* the time course of air flow rate and ammonia concentration, for most of the time these air scrubbers are overdimensioned and underloaded.

By model calculations and analyses of measurements datasets it was demonstrated that the application of an air scrubber that bypasses peaks of the air flow, will significantly decrease the required scrubber size while ammonia emission levels are only slightly increased. In such a system, most of the air is treated by the scrubber but a part of the air is vented untreated at times that the actual air flow exceeds a certain setpoint (*e.g.* in case the bypass setpoint is set to 50% of the maximum air flow rate and the scrubber volume is reduced by 50%, only 10 - 20% of the total ammonia load is bypassed and subsequently emitted to the environment, for the analysed datasets and made assumptions). This means that the use of the bypass venting system increases both the efficiency of scrubber utilization in kg [NH_3 removal] m^{-3} [scrubber volume] and the cost-effectiveness of air scrubbing for ammonia removal in kg [NH_3 removal] €$^{-1}$.

Although the use of a scrubber with a bypass venting system decreases the maximum air in m^3 m^{-3} h^{-1} and ammonium loading rate in kg m^{-3} h^{-1} that needs to be treated, the average air and ammonium loading rate are increased in comparison with a conventional scrubber. Further experimental research on acid and biological scrubbers is necessary to demonstrate to what extent this might affect the performance of the scrubbers with regard to the relative removal efficiency in % and how this affects the total emission from scrubbers with bypass systems.

Acknowledgements

This work was supported by the Dutch Ministry of Agriculture, Nature and Food Quality (LNV). We thank N.W.M. Ogink for his suggestions on the manuscript.

References

ASG (2004). Handboek pluimveehouderij [Handbook of poultry farming]. Praktijkboek 36. Praktijkonderzoek van de Animal Sciences Group van Wageningen UR, Lelystad, Netherlands, 244 pp.

Bruce J M; Clark J J (1979). Models of heat production and critical temperature for growing pigs. Animal Production, 28(3), 353-369.

CBS (2004). StatLine. Statistics data bank for the Netherlands. Available at: http://www.cbs.nl.

Focht D D; Verstraete W (1977) Biochemical ecology of nitrification and denitrification. Advances in Microbial Ecology, 1, 135-214.

Groenestein C M; Huis in 't Veld J H W (1996). Praktijkonderzoek naar de ammoniakemissie van stallen XXVII. Vleesvarkensstal met koeling van mestoppervlak in de kelder. [Research into the ammonia emission from livestock housing systems XXVII: fattening pig house with slurry surface cooling]. Report 96-1003. IMAG-DLO, Wageningen, Netherlands.

Heij G J; Erisman J W (1995). Acid Rain Research: Do We Have Enough Answers? Proceedings of a Speciality Conference, 's-Hertogenbosch, Netherlands, 10-12 October 1994. Studies in Environmental Science, Vol. 64. Elsevier Science, Amsterdam, Netherlands, ISBN 0-444-82038-8.

Heij G J; Erisman J W (1997). Acid Atmospheric Deposition and its Effects on Terrestrial Ecosystems in the Netherlands. The Third and Final Phase (1991-1995). Studies in Environmental Science, Vol. 69. Elsevier Science, Amsterdam, Netherlands, ISBN 0-444-82037-X.

Hol J M G; Groot Koerkamp P W G (1998). Praktijkonderzoek naar de ammoniakemissie van stallen XXXX. Vleeskuikenstal met verwarming en koeling van de vloer met strooisel [Research into the ammonia emission from livestock housing systems XXXX: broiler house with heating and cooling of litter covered floor]. Report 98-1004. IMAG-DLO, Wageningen, Netherlands.

Hol J M G; Groenestein C M (2005). Unpublished data. Agrotechnology & Food Innovations, Wageningen UR, Wageningen, Netherlands.

Huis in 't Veld J H W; Groenestein C M (1995). Praktijkonderzoek naar de ammoniakemissie van stallen XXIV [Research into the ammonia emission from livestock housing systems XXIV: fattening pig house with manure dilution by collection in ammonia-free liquid]. Vleesvarkensstal met verdunning van mest door opvang in ammoniakvrije vloeistof. Report 95-1007. IMAG-DLO, Wageningen, Netherlands.

Lund H (1984). Test Reference Years (TRY). Weather data sets for computer simulations of solarenergy systems and energy consumptions. CEC Programme Solar Energy. Technical University of Denmark, Thermal Insulation Laboratory, Kongens Lyngby, Denmark.

Melse R W; Mol G (2004). Odour and ammonia removal from pig house exhaust air using a biotrickling filter. Water Science and Technology, 50(4), 275-282.

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

Melse R W; Willers H C (2005). Theoretical evaluation of ammonia scrubbers for governmental approval. In: Proceedings of the 7th International Conference on Construction, Technology and Environment in Livestock Farming, March 2-3, 2005, Braunschweig, Germany, pp 295-300, Kuratorium für Technik und Bauwesen in der Landwirtschaft e.V. (KTBL), Darmstadt, Germany, ISBN 3-7843-2185-2.

Mosquera J; Hofschreuder P; Erisman J W; Mulder E; Van 't Klooster C E; Ogink N; Swierstra D; Verdoes N (2002). Meetmethoden gasvormige emissies uit de veehouderij [Methods for measurement of gaseous emissions from animal husbandry]. Report 2002-12. IMAG-DLO, Wageningen, Netherlands, ISBN 90-5406-216-9.

Mosquera J; Hol J M G; Ogink N W M (2005). Analyse ammoniakemissieniveaus van praktijkbedrijven in de varkenshouderij (1990-2003) [Analysis of ammonia emission levels of pig farms (1990-2003)]. Report 312. Agrotechnology & Food Innovations, Wageningen UR, Wageningen, Netherlands, ISBN 30-6754-874.

Ni J-Q; Hendriks J; Vinckier C; Coenegrachts J (2000). Development and validation of a dynamic mathematical model of ammonia release in pig house. Environment International, 26, 105-115.

Ogink N W M; Lens P N (2001). Geuremissies uit de veehouderij I. Overzichtsrapportage 1996-1999 [Odour Emissions from Animal Husbandry I. General report 1996–1999]. Report 2001-14. IMAG-DLO, Wageningen, Netherlands.

Prosser I J (ed.) (1986). Nitrification. Society for General Microbiology special publication 20, IRL Press, Oxford, UK, ISBN 1-85221-001-X and 1-85221-013-3.

Satter I H G; Hol J M G; Huis in 't Veld J H W; Groenestein C M (1997). Praktijkonderzoek naar de ammoniakemissie van stallen XXXIV. Vleesvarkensstal met mestverwijdering door spoelen met dunne mestfractie via spoelgoten [Research into the ammonia emission from livestock housing systems XXXIV: fattening pig house with manure removal by a flushing-gutter system]. Report 97-1004. IMAG-DLO, Wageningen, Netherlands.

Seedorf J; Hartung J; Schröder M; Linkert K H; Pedersen S; Takai H; Johnsen J O; Metz J H M; Groot Koerkamp P W G; Uenk G H; Phillips V R; Holden M R; Sneath R W; Short J L; White R P; Wathes C W (1998). A survey of ventilation rates in livestock buildings in Northern Europe. Journal of Agricultural Engineering Research, 70, 39–47.

Sterrenburg P; Van Ouwerkerk E N J (1986a). Rekenmodel voor de bepaling van de thermische behaaglijkheidszone van varkens (BEZOVA) [Calculation model for

determination of the thermal comfort zone of pigs (BEZOVA)]. Report 78. IMAG-DLO, Wageningen, Netherlands.
Sterrenburg P; Van Ouwerkerk E N J (1986b). Tabellen van warmteproduktie en temperatuureisen van varkens [Tables with heat production and temperature requirements of pigs]. Report 79. IMAG-DLO, Wageningen, Netherlands.
Van Ouwerkerk E N J (1999). ANIPRO klimaat- en energiesimulatiesoftware voor stallen [ANIPRO climate and energy simulation software for animal houses]. Report V99-109. IMAG-DLO, Wageningen, Netherlands.
Van Wagenberg A V; Vermeij I (2001a). Energiegebruik en kosten van centrale afzuiging en afzuiging per afdeling in varkensstallen [Energy use and costs of ventilation using either central fans or one fan per pig room]. Report 218. Praktijkonderzoek Veehouderij, Lelystad, Wageningen UR, Netherlands, ISSN 0169-3689.
Van Wagenberg A V; Vermeij I (2001b). The use of a central extraction ventilation system for pig facilities, energy savings and economic consequences. ASAE Annual International Meeting, Sacramento, California, USA, July 30-August 1, 2001. ASAE Paper No. 01-4050.
Wever A C; Huis in 't Veld J W H ; Groot Koerkamp P W G (1999). Onderzoek naar de ammoniakemissie van stallen XLV. Vleeskuikenstal met isolatie en ventilatie volgens het VEA-concept [Research into the ammonia emission from livestock housing systems XLV: broiler house with insulation and ventilation according to the VEA concept]. Report 99-09. IMAG-DLO, Wageningen, Netherlands.

Chapter 5

Biofiltration for mitigation of methane emission from animal husbandry

R.W. Melse[1]; A.W. van der Werf[2]

[1]Wageningen University and Research centre, Agrotechnology & Food Innovations, Wageningen, Netherlands; [2]Bioclear, Groningen, Netherlands.

ABSTRACT Removal of methane from exhaust air of animal houses and manure storages has a large potential for the reduction of greenhouse gas emissions from animal husbandry. The aim of this study was to design a biofilter for methane removal at a full-scale livestock production facility. Air from the headspace of a covered 6 m^3 liquid manure storage (air flow: 0.75-8.5 m^3 m^{-3} $hour^{-1}$; CH_4: 500-5,500 mg m^{-3}), was treated in an experimental biofilter (160 L). The filterbed, a mixture of compost and perlite in a 40 : 60 (v/v) ratio, was inoculated with activated sludge that had shown a good methane oxidation rate as compared to pure cultures in preceding laboratory tests. Methane removal up to 85% could be achieved in the experimental biofilter. The methane removal (g m^{-3} h^{-1}) appeared to be proportional to the concentration (g m^{-3}) with k = 2.5 $hour^{-1}$. Relatively low methane concentrations and high air flows, as reported for the exhaust air of animal houses, would require very large biofilter sizes. Extrapolation of the results showed that treatment of air from a 1,000 m^3 liquid manure storage with a methane concentration of 22 g m^{-3} would require a 20 m^3 biofilter for a desired emission reduction of 50%. The costs for such a biofilter are USD 26 per t of CO_2 equiv reduction.

5.1 Introduction

Many industrialized countries have agreed to the Kyoto protocol (UNFCC, 1997) which commits them to cut the emission of greenhouses gases (GHG's) with 5% during the period 2008 to 2012 with respect to 1990 levels. Methane (CH_4) is one of the GHG's that substantially contributes to the enhanced greenhouse effect with a global warming potential (GWP) of 23 as compared to carbon dioxide (GWP=1) (IPCC, 2001). Methane emission from livestock husbandry is a significant GHG emission source that represents 2 and 4% of total GHG emissions in the U.S. and in the European Union (EU-15), respectively (EAA, 2002; EPA, 2004). Under European conditions, it originates for 59% from enteric fermentation in ruminants, for 12% and 13% from fermentation of manure in animal houses and manure storages, respectively, and for 15% from grazing (Freibauer, 2002). It follows that cleaning of exhaust air from animal houses and manure storages has a large potential for the reduction of GHG emissions.

Typical methane concentrations in animal houses are 5 to 100 mg m^{-3} at an average ventilation rate of 35 m^3 $hour^{-1}$ fattening pig^{-1} (mechanically ventilated) or 1,000 m^3 $hour^{-1}$ milking cow^{-1} (naturally ventilated). At a regular sized farm of 1,700 pig fattening places or 100 milking cows, the average exhaust air flow is 60,000 and 100,000 m^3 $hour^{-1}$, respectively.

For a covered liquid manure storage, the methane concentration in the headspace depends on the number of times the headspace air changes over with outside air. In theory, at an air-exchange rate of zero the headspace concentration equals undiluted biogas which contains about 425 g m^{-3} (65% v/v) methane. In practice, however, the cover contains some openings that permit ventilation which results in a methane concentration from 0.1 to 20 g m^{-3}. The methane production rate in a liquid manure storage depends on manure composition, inoculation, and temperature (Melse, 2003).

Treatment of this air in a biofilter aims at the aerobic conversion of methane to carbon dioxide (CO_2) and water by so-called methanotrophic bacteria (Hanson and Hanson, 1996). The bacterial oxidation of methane is a common phenomenon which has been studied extensively for methane-influenced habitats, such as peatlands, rice paddy soils, and landfill cover soils (Bender and Conrad, 1992; Jones and Nedwell, 1993; Kightley *et al.*, 1995; Boeckx *et al.*, 1996; Heipieper and De Bont, 1997; Humer and Lechner, 1999b; Stein and Hettiaratchi, 2001; De Visscher and Van Cleemput, 2003). Half-saturation constants (K_M) for methane oxidation in such habitats were reported from 0.29 to 8.6 μM CH_4 (Whalen *et al.*, 1990; Kigthly *et al.*, 1995; Krumholz *et al.*, 1995; Czepiel *et al.*, 1996; Bosse and

Frenzel, 1997; De Visscher *et al.*, 1999). Recently, Petersen *et al.* (2005) showed methanotrophic activity in naturally formed and artificially established liquid manure storage crusts. Oxidation of methane to carbon dioxide allows a significant reduction of the emission of greenhouse gases (GWP from 23 to 1). Although the produced CO_2 is a greenhouse gas as well, this CO_2 is part of the short-term organic carbon cycle so it does not contribute to the enhanced greenhouse effect.

Although much experience has been gathered with biofiltration for odor removal, currently in livestock farming there is no experience with biofilters that are designed for methane removal and research in this field is scarce (Hahne and Vorlop, 2001; Martinec *et al.*, 2001). In other areas, however, some experience has been gathered. Although most recent literature (Devinny *et al.*, 1999; Kennes *et al.*, 2001) suggests that bioreactors are only cost-effective up to pollutant concentrations of 5 to 10 g m^{-3}, for landfill-gas treatment biofilters have been developed and operated at various methane inlet concentrations up to 260 g m^{-3} (40% v/v) at empty bed air residence times (EBRT's) between 5 minutes and 5 hours (Figueroa, 1996; Kussmaul and Gebert, 1998; Dammann *et al.*, 1999; Straka *et al.*, 1999; Scharff *et al.*, 2001; Park *et al.*, 2002; Gebert *et al.*, 2003; Streese and Stegmann, 2003; Park *et al.*, 2004), whereas typical EBRT's are 25 seconds to over a minute for common biofilter applications (Devinny *et al.*, 1999). Also some papers were published on biofiltration of coalmine atmospheres that are usually controlled at a methane content of 1 to 10 g m^{-3} (Apel *et al.*, 1991; Sly *et al.*, 1993; Du Plessis *et al.*, 2003). An alternative technology for removal of high pollutant concentrations, such as activated carbon adsorption, is not suitable for treatment of methane containing air because of the low methane affinity for adsorbents. If the methane-air mixture contains 30 to 100 g CH_4 m^{-3}, however, incineration might be cost-effective as this mixture is flammable.

Biological conversion of methane in a biofilter is a slow process due to the low water solubility of methane (Henry's law constant = 1.5×10^{-3} M atm^{-1}) and this is why such long EBRT's are applied. Previous work by Streese and Stegmann (2003) showed first-order removal kinetics for methane inlet concentrations up to 16 g CH_4 m^{-3} in an operated biofilter. One report by Gebert *et al.* (2003) is known which mentions a K_M value for methanotrophs in a biofilter, *viz* 15.1 μM CH_4; this value is higher than most reported K_m values for methane-influenced habitats. Assuming equilibrium gas and liquid phase concentrations, a gas phase concentration of 16 g CH_4 m^{-3} equals a liquid phase concentration of 37μM CH_4, which is about twice as high as the K_m value that was reported by Gebert *et al.* Thus probably mass transfer of methane from the gas to the liquid phase is rate limiting and results in first-order kinetics at high methane concentrations. At low methane inlet concentrations and high air flows, as is the case for exhaust air from animal houses, first-order kinetics will lead to very large sized biofilters. At high methane inlet concentrations and low air flows, however, as is the case for exhaust air from liquid manure storages, the necessary biofilter size is expected to be much smaller.

Besides methane, the exhaust air of a manure storage or animal house usually contains some ammonia (NH_3) and dihydrogen sulfide (H_2S). In a biofilter, NH_3 is converted to nitrous and nitric acid by nitrification and H_2S is converted to sulfuric acid. At high NH_3 and H_2S loading rates, methanotrophs may be inhibited by high ammonium and nitrite concentrations (Bedard and Knowles, 1989; Bronson and Mosier, 1993; King and Schnell, 1994; Boeckx *et al.*, 1996; Humer and Lechner, 1999a) or by sulfuric acid accumulation (Furusawa *et al.*, 1984; Yang and Allen, 1994) as the pH optimum of methanotrophs lies between 6 and 8 (Park *et al.*, 1992; Humer and Lechner, 1999a). However, at EBRT's that are common for biofiltration of methane containing air, *i.e.* several minutes to hours, no significant inhibition or acidification is expected due to the corresponding low loading rates of NH_3 and H_2S.

The aim of this study was to develop a pilot-scale biofilter for reduction of methane emission from livestock husbandry and to assess its feasibility for full-scale operation at a regular livestock production facility.

5.2 Materials and methods

5.2.1 Inoculum

Prior to the biofilter experiments, laboratory experiments were performed to determine the optimal culture of methanotrophic bacteria for inoculation of the biofilter. Four cultures were tested: two pure cultures, *Methylomonas methanica* (ATCC 51626) and a *Methylocystis* strain (Dunfield and Conrad, 2000), and two mixed cultures, one originating from activated sludge taken from the final settling tank of a local municipal wastewater treatment facility and one from a compost biofilter treating methane containing land-fill gas.

The methane degradation rate of each culture was determined at 20°C in a single batch experiment using a continuously shaken vessel by regularly measuring the decrease of the methane concentration in the headspace from 25 to 5 g m^{-3} with a gas chromatograph (Interscience 8000-series; column: Chromosorb 102; detector: FID). The maximum growth rate (μ_{max}) of each culture had been determined in duplicate at 20°C using optical density measurements (OD_{450}) in batch experiments. In previous experiments the relation between OD_{450} and dry weight was determined; an OD_{450} of 1 equals a dry weight of 180 mg L^{-1}.

For use of these cultures for inoculation of a biofilter, the effect of temperature, acidity (pH) and ammonia concentration on the biomass activity are important parameters. Therefore the effects on the maximum growth rate were determined in duplicate for different temperatures (10°C, 20°C, and 30°C) for all cultures. Based on the results of these experiments from both the pure cultures and from the mixed cultures the culture with the highest mean methane degradation was selected for further testing. For the *Methylocystis* strain and the mixed culture from the compost biofilter the effects on the maximum growth rate were determined in duplicate for low pH (pH 5) and for different ammonia concentrations (10, 100, and 1000 mg N L^{-1} H_2O) at a constant pH. For the latter experiment, ammonia, nitrite, and nitrate concentrations were measured in duplicate at the start and the end of the experiment spectrophotometrically (LANGE cuvette test; LCK 303, 305, 339-342).

All experiments were performed in a standard nutrient solution for methanotrophic bacteria (ATCC 1306 medium) containing: 1.0 g L^{-1} $MgSO_4.7H_2O$, 0.2 g L^{-1} $CaCl_2.6H_2O$, 4 mg L^{-1} Fe-EDTA, 1.0 g L^{-1} KNO_3, 0.272 g L^{-1} KH_2PO_4, and 0.717 g L^{-1} $Na_2HPO_4.12H_2O$; the phosphate acts as pH buffer. The initial pH-value of the medium was 6.4. For the growth experiment on pH 5, the pH of the medium was lowered using chloric acid.

Based on the results of the microbiological experiments a culture was selected for inoculation of the pilot biofilter. The selection was made based on the maximum growth rate, the methane degradation rate and the sensibility to changes in temperature, pH and ammonia concentrations of the different cultures.

5.2.2 Biofilter setup and operating conditions

The experiments are carried out with ventilation air of a covered liquid manure storage and not with exhaust air from an animal house, in order to limit the size and costs of the pilot-scale biofilter system.

The biofilter consisted of a plexiglass cylinder (height: 0.95 m; inner diameter: 0.49 m) which was packed with a mixture of expanded perlite (bulk density: 95 kg m^{-3}; particle size: 0.6 - 7.5 mm) and garden compost (bulk density: 450 kg m^{-3}; dry matter content: 348 g kg^{-1};) in a volume ratio of 40 : 60 which equals a mass ratio of about 75 : 25. The height of the filter packing was 86 cm and the bed volume equaled 160 L. The total nitrogen content of the compost was 3.2 g kg^{-1}. The total phosphorus content of the compost was raised from 0.3 to 0.5 g kg^{-1} by addition of 1.5 g bone meal (P content: 15% m/m) per kg of compost and the pH was raised from 5.3 to 7.0 by addition of lime fertilizer, as these conditions are generally considered as optimal for biofilter operation. Dry matter, nitrogen, phosphorus, and pH analyses were carried out in duplicate according to standard methods

(NEN, 1998). At day 0, the biofilter was inoculated with 35 L of activated sludge taken from the final settling tank of a local municipal wastewater treatment facility.

From underneath the cover of a 6 m^3 storage tank filled with liquid pig manure (dry matter content: 50 - 100 g m^{-3}), methane containing air was withdrawn with a membrane pump and, after passing through a washing bottle to remove excess water, entered the biofilter at the bottom. Both air inlet and outlet tubes were equipped with an air sampling port. A schematic of the setup is shown in Figure 5.1.

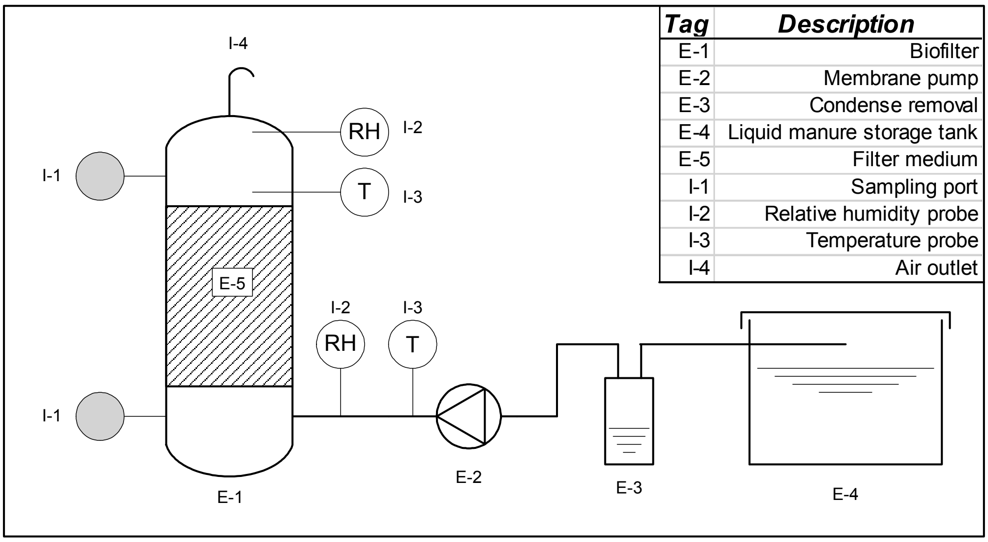


Figure 5.1 Schematic of the setup of the pilot-scale biofilter system.

In order to determine the performance of the biofilter under different methane loading rates, the air flow through the biofilter was gradually increased from 0.75 to 8.5 m^3 m^{-3} $hour^{-1}$ (EBRT = 7 - 80 minutes) using an adjustable valve in the air inlet tube. The experimental setup did not allow independent adjustment of methane concentration and air flow. The methane production rate was determined by the bacteriological processes in the manure storage tank and the actual methane concentration was the result of the set air flow rate and the autonomously occurring air-exchange with the outside.

The biofilter was continuously operated for two months (October - December 2002). At the end of the experiment the filter packing was analyzed and the pH, dry matter content, and the concentrations of ammonium (NH_4^+), nitrite (NO_2^-), and nitrate (NO_3^-) were measured.

5.2.3 Biofilter measurements

Because the ventilation air from liquid manure storages and animal houses contains some ammonia, some nitrification and denitrification is likely to occur in the biofilter. It is known that during incomplete nitrification and denitrification some nitrous oxide (N_2O) may be produced as a by-product (Rogers and Whitman, 1999). Because N_2O is a greenhouse gas, measurements on N_2O production were included in this study; the emission of 1 kg N_2O equals an emission of 296 kg CO_2 on a time horizon of 100 years (IPCC, 2001).

Two to three times a week an air sample was taken from the gas inlet and outlet of the biofilter. The concentrations of CH_4, CO_2, and N_2O were determined with a gas chromatograph (Carbo Erba Instruments, GC 8000 Top; column: Molsieve 5A (CH_4, CO_2), Haysep (N_2O); detector: FID/HWD (CH_4); ECD/HWD (N_2O), HWD (CO_2)); the within-repeatability of this method is 2.8%. NH_3 and H_2S concentrations were measured with gas detection tubes (Kitagawa; 105SD, 120SB). The system was allowed a minimum period of 24 hours after every change in air flow, before new air samples were taken.

The air flow was measured with a rotameter. Temperature of inlet and outlet air was measured continuously (Rotronic Hygromer Serie 1200). The pressure drop over the biofilter was measured with a digital pressure gauge (Special Instruments, Digima Premo B-026).

5.2.4 Economic evaluation

Finally, on basis of the experimental results an estimation of the costs of a full-scale biofilter system was made. Figures on the investment, maintenance, and operational costs of the designed biofilter system were provided by a biofilter manufacturer (Pure Air Solutions BV, Steenwijk, Netherlands) and a consulting firm (Stork Product Engineering BV, Amsterdam, Netherlands).

5.3 Results and discussion

5.3.1 Inoculum

The average methane degradation rate over a headspace concentration range from 25 to 5 mg CH_4 L^{-1} at 20°C was found to be 6.0 mg CH_4 g^{-1} dry weight $hour^{-1}$ for *Methylocystis*, 8.0 mg CH_4 g^{-1} dry weight $hour^{-1}$ for activated sludge inoculum, and 8.2 mg CH_4 g^{-1} dry weight $hour^{-1}$ for the compost biofilter inoculum, respectively. The degradation rates followed first-order kinetics for the whole concentration range. First-order methane oxidation kinetics will occur as long as the liquid phase methane concentration << half-saturation coefficient (K_m).

Assuming equilibrium gas and liquid phase concentrations, a Henry's law constant of 1.5 x 10^{-3} M atm^{-1}, and a K_m value of 15.1 μM CH_4 (Gebert *et al.*, 2003), first-order kinetics will occur as long as the gas phase concentration is far below 7 g CH_4 m^{-3} as a liquid phase concentration of 15.1 μM CH_4 equals a gas phase concentration of 7 g CH_4 m^{-3}. If mass transfer of methane from gas to liquid phase is rate limiting, however, first-order kinetics will also occur at higher methane concentrations. This explains why the removal of methane showed first-order kinetics for the whole concentration range (from 25 to 5 g CH_4 m^{-3}) in these experiments. Apparently the transport of methane from gas to liquid phase was limited despite the continuous shaking of the samples.

Because of first-order kinetics the maximum methane degradation rate (V_{max}) and the half-saturation constant (K_M) of the cultures could not be determined. For unknown reasons, no degradation of methane was observed for *Methylomonas*.

The maximum growth rate at 20°C was 0.039 $hour^{-1}$ (SD = 0.007) for *Methylomonas*, 0.054 $hour^{-1}$ (SD = 0.029) for activated sludge, and 0.042 $hour^{-1}$ (SD = 0.000) for compost biofilter culture. The maximum growth rate for *Methylocystis*, *viz* 0.012 $hour^{-1}$ (SD = 0.002) was significantly lower than for the other cultures, whereas the degradation rate is only little lower. Our hypothesis is that *Methylocystis* has a less effective metabolism in the degradation of methane than the other tested cultures. The low growth rate of *Methylocystis* may be explained by a high maintenance energy of this strain during growth on methane.

From 20 to 10°C, the maximum growth rate of the cultures decreased with on average 54% (46 - 61%). From 20° to 30°C, the maximum growth rate increased with 24% and 37% for the activated sludge and compost biofilter culture, respectively, and increased to a three- and five-fold value for *Methylomomas* and *Methylocystis*, respectively. The relation between temperature and methanotrophic activity that was found compares with literature (Gebert *et al.*, 2003; Humer and Lechner, 1999a; Streese *et al.*, 2001).

At pH 5 the maximum growth rate of the *Methylocystis* strain decreased by a factor 25, *viz* from 0.063 $hour^{-1}$ to 0.0024 $hour^{-1}$, whereas the maximum growth rate of the compost culture decreased by a factor 2.5, *viz* from 0.056 $hour^{-1}$ to 0.021 $hour^{-1}$, compared to neutral pH (pH 6.4). These results confirm that the biofilter should be operated around neutral pH for optimal methanotrophic growth (Humer and Lechner, 1999a; Park *et al.*, 1992).

Growth up to ammonia concentrations of 1,000 mg N L^{-1} occurred for both the *Methylocystis* strain and the culture from the compost biofilter. The addition of ammonia did not affect the maximum growth rate of the compost mixed culture; the growth of the pure *Methylocystis* culture was slightly inhibited. For

Methylocystis, the maximum growth rate was reduced with 40%, 62%, and 32%, respectively, at concentrations of 10, 100 and 1,000 mg ammonia-N L^{-1}. The pH-value was constant during all experiments (pH 6.4) due to the buffering effect of the medium. Since no clear relation was found between increase of the ammonia concentration and decrease of the maximum growth rate, probably no direct inhibition by ammonia took place. However, the decrease of the maximum growth rate may be caused by competition for certain enzymes by the nitrification process or by the directly inhibiting effect of nitrite that is formed by nitrification. These inhibition effects are known phenomena for methanotrophs (Bedard and Knowles, 1989; Bronson and Mosier, 1993; King and Schnell, 1994; Boeckx *et al.*, 1996; Humer and Lechner, 1999a).

The maximum growth rate of *Methylocystis* was significantly lower at both 10 and 20°C. Furthermore, the tested pure culture (*Methylocystis*) appeared to be more susceptible to variations in acidity and high ammonia concentration than the tested mixed culture (compost biofilter). This made the *Methylocystis* culture an unsuitable candidate for inoculation of the biofilter. As the ability of the *Methylomonas* culture to degrade methane was lost during the experiment for unknown reasons this culture was not selected either. Because of the lower susceptibility of the mixed culture and for economical reasons, it was decided to inoculate the pilot biofilter with activated sludge from a municipal wastewater treatment plant, which is a mixed culture that is readily available and can be obtained free of charge.

5.3.2 Methane removal

In Figure 5.2, the inlet and outlet concentration of methane and the removal efficiency of the pilot biofilter are shown. During the first two weeks, the air flow was set to 0.75 m^3 m^{-3} $hour^{-1}$. From day 0, the methane removal efficiency showed a sharp increase and after two weeks the removal efficiency stabilized at 80 - 85% during. This course indicates that, after an adaptation period, an equilibrium was established between the availability of methane substrate and the amount of methanotrophs; this is called the start-up period of the biofilter. From day 18, the air loading rate of the system was gradually increased (see Figure 5.3). At the moment that the air load was reduced again (day 48), the methane removal efficiency sharply increased.

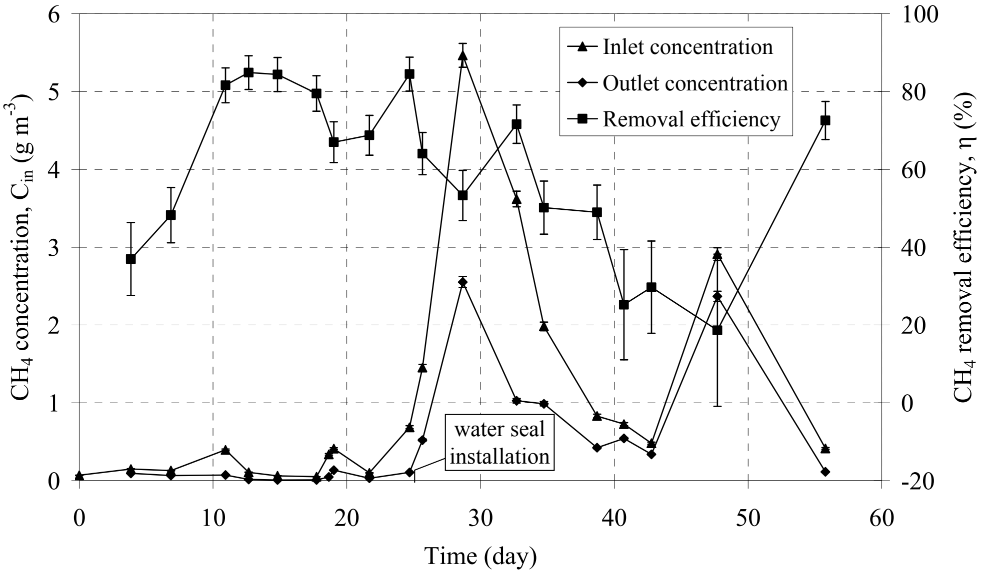


Figure 5.2 Methane removal by the biofilter system.

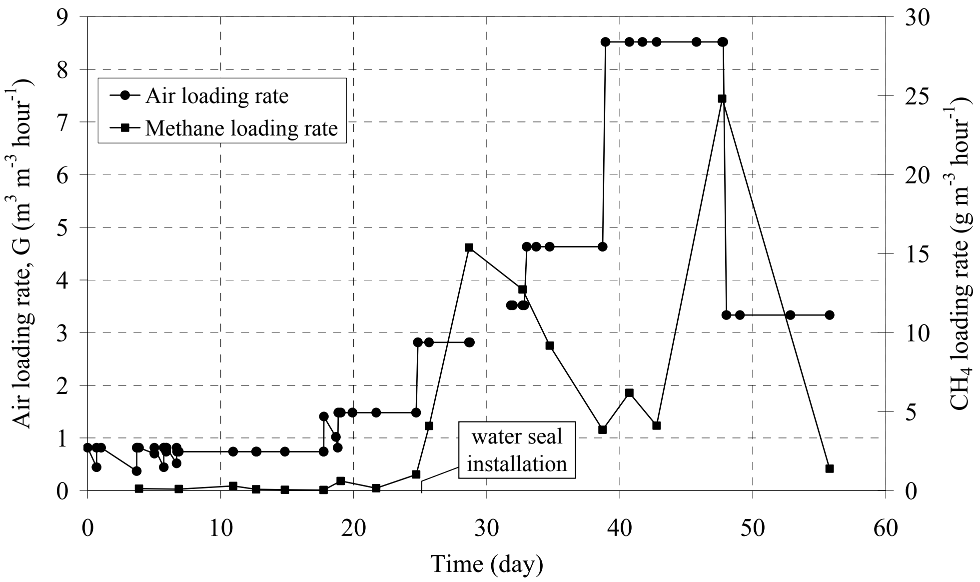


Figure 5.3 Loading rate of air and methane.

Furthermore, the quality of the cover of the liquid manure storage tank was significantly improved by installing a water seal on day 25, thus reducing the air-exchange with outside air. This immediately resulted in a sharp increase of the methane concentration from 700 mg m^{-3} on day 25 to 5,500 mg m^{-3} on day 29.

In the air flow range that was applied in our experiments, *i.e.* 0.75 to 8.5 m^3 m^{-3} $hour^{-1}$, the pressure drop turned out to be directly proportional to the air loading rate; at an air load of 8.5 m^3 m^{-3} $hour^{-1}$ the pressure drop was 20 Pa m^{-1}.

5.3.3 Emissions of N_2O, NH_3, H_2S, and odor

In the first week of the experiment the N_2O content of the outlet air increased from atmospheric concentration, *i.e.* 0.6 mg m^{-3}, to 10 mg m^{-3} and decreased to about 1.0 mg m^{-3} in another week. For the further duration of the experiment, the N_2O outlet concentration remained unchanged. The N_2O that was emitted in the first two weeks had possibly been accumulated during the airtight storage of the compost and resulting anaerobic conditions prior to the experiment. The emission of N_2O during the further duration of the experiment indicates that still some N_2O production occurred in the biofilter, despite the aeration that takes place. After the start-up period, the contribution of N_2O to the total GHG concentration in the outlet air, *i.e.* the sum of CH_4 and N_2O, expressed as CO_2 equiv, varied from 0.4 to 64%.

The NH_3 and H_2S air inlet concentrations of the biofilter varied from 3 to 15 mg m^{-3} and from 0.4 to 6 mg m^{-3}, respectively, which equals a biofilter loading rate of 3 to 83 mg m^{-3} $hour^{-1}$ for NH_3 and 0.6 to 16 mg m^{-3} $hour^{-1}$ for H_2S. At these low ammonia and dihydrogen sulfide loading rates no acidification or significant inhibition is expected; accordingly, removal efficiencies were high, *viz* 90 - 100% for NH_3 and 100% for H_2S. At the end of the experiment the pH of the packing material was 6.8 and the dry matter content 376 g kg^{-1}; the concentration of NO_3^--N, NO_2^--N, and NH_4^+-N was 140 mg m^{-3}, < 1 mg kg^{-1}, and 200 mg kg^{-1}, respectively, which indeed indicates that no acidification of the biofilter had taken place and no significant inhibition by ammonium or nitrite had occurred.

The odor characteristics of the air changed from 'manure' for the inlet air to the smell of 'woods' for the outlet air; the actual odor concentration (OU_E m^{-3}; *49*), however, was not determined.

5.3.4 Temperature and relative humidity

The temperature of the inlet air was on average 12.0°C (minimum: 4.7°C; maximum: 21.1°C) and reflected the outside temperature together with its day-night cycle. The temperature of the outlet air was on average 1.4°C higher than the inlet temperature; this temperature rise is probably due to the insulating effect of the shed the biofilter was located in. Both inlet and outlet air appeared to be close to water saturation as was indicated by condensation inside the tubing.

5.3.5 Influence of concentration and air flow on methane removal

After an adaptation period, in which accumulation of methanotrophic biomass occurred, the biofilter is considered in stable operation from day 25 to 60; the methane measurements that were done during this period are further analyzed.

The removal capacity for a component in a biofilter system is usually expressed as:

$$EC = (C_{in} - C_{out}) \,.\, Q / V \qquad \text{[Eq. 5.1]}$$

where EC is the elimination capacity for a component (g m^{-3} hour^{-1}), C_{in} is the inlet concentration (g m^{-3}), C_{out} is the outlet concentration (g m^{-3}), Q is the air flow (m^{3} hour^{-1}), and V is the biofilter volume (m^{3}).

Assuming plug-flow conditions and first-order kinetics for methane oxidation, it can be derived that:

$$Q / V = k / \ln(C_{in} / C_{out}) \qquad \text{[Eq. 5.2]}$$

where k is the first-order rate constant for methane oxidation (hour^{-1}).

When the term Q / V in Equation 5.1 is replaced by Equation 5.2, Equation 5.1 changes to:

$$EC = k \,.\, (C_{in} - C_{out}) / \ln(C_{in} / C_{out}) = k \,.\, C_{m,log} \qquad \text{[Eq. 5.3]}$$

where $C_{m,log}$ (g m^{-3}) is the logarithmic mean of the inlet and outlet concentration.

In Figure 5.4, the measured elimination capacity for methane is plotted against $C_{m,log}$. The linear relationship that is shown in Figure 5.4, with a k value of 2.5 hour^{-1}, indicates that indeed first-order kinetics occur. This k value corresponds to the range from 0.3 to 6.6 hour^{-1} that can be calculated from the data of other studies at logarithmic mean methane concentrations from 0.4 to 22 g m^{-3} (Sly *et al.*, 1993; Du Plessis *et al.*, 2003; Streese and Stegmann, 2003). Finally, the methane elimination capacity that was observed, at an average air inlet temperature of 12°C, may significantly increase if the biofilter is operated at a higher temperature. The methanotrophic activity increases with temperature as was found in the laboratory experiments and in literature (Humer and Lechner, 1999a; Streese *et al.*, 2001; Gebert *et al.*, 2003).

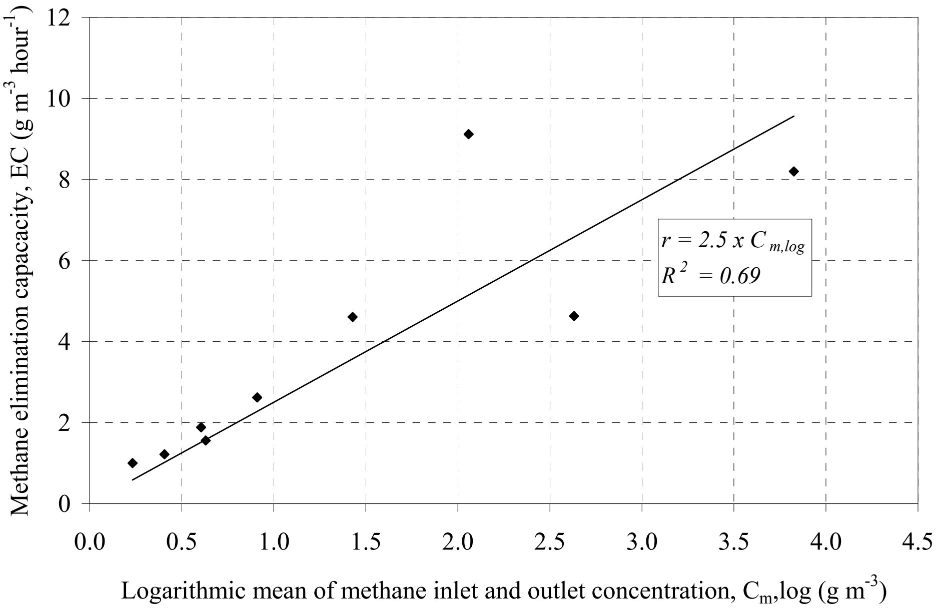


Figure 5.4 Dependence of methane elimination capacity on concentration.

Rewriting Equation 5.2 leads to a relationship between the methane removal efficiency and the air loading rate of the biofilter:

$$\eta = (C_{in} - C_{out}) / C_{in} = 1 - e^{-k(V/Q)} = 1 - e^{-k/G} \quad \text{[Eq. 5.4]}$$

where η is the removal efficiency (-) and G is the air loading rate (m^3 m^{-3} $hour^{-1}$). This relationship, which is plotted in Figure 5.5, describes that, as a result of first-order kinetics, any desired methane removal efficiency can be achieved by adjusting the air loading rate of the biofilter, independent of the actual methane inlet concentration.

5.3.6 Biofilter design calculations

In a liquid manure storage tank, the methane production rate is assumed to be constant at 40 g m^{-3} manure day^{-1}. For a storage tank with an amount of 1,000 m^3 of liquid manure, the necessary biofilter size can be calculated from Equation 5.4 for any methane inlet concentration and desired removal efficiency. In Figure 5.6, for a methane concentration range up to 75% of the Lower Explosion Limit (LEL methane = 30 g m^{-3}), the required biofilter volume is calculated from Equation 5.4 for several removal efficiencies. It is noted that, in contrast to most biofilter

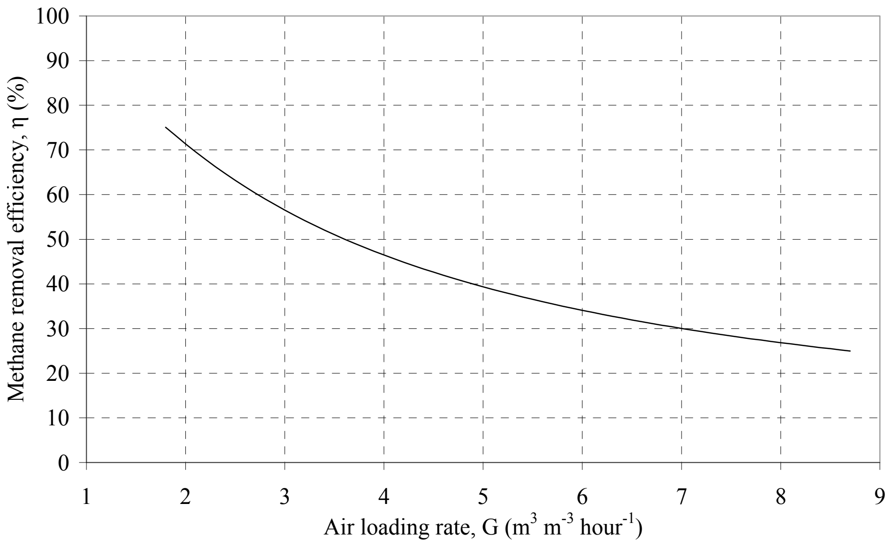


Figure 5.5 Dependence of methane removal efficiency on air loading rate.

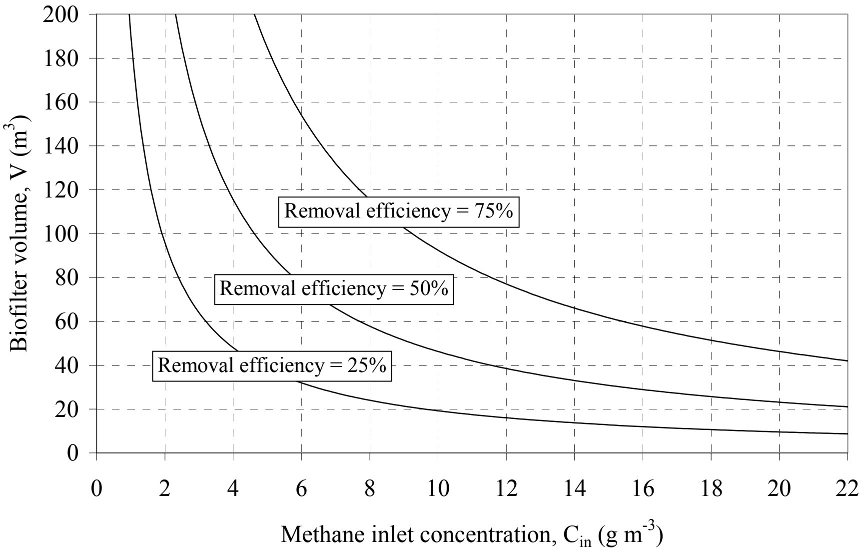


Figure 5.6 Biofilter design for treatment of exhaust air from a 1,000 m^3 liquid manure storage at constant methane production rate of 40 g m^3 manure.

applications, the air flow rate and the inlet concentration are inversely proportional (the methane production rate is constant) which results in a proportional relationship between the removal efficiency and the biofilter volume, as is shown in Figure 5.6. As our experiments were achieved only up to methane concentrations of 5.5 g CH_4 m^{-3}, the calculations for the range from 5.5 to 22 g CH_4 m^{-3} are extrapolated assuming that first-order methane degradation still occurs at these concentrations.

In previous experiments on methane degradation in biofilters, first-order kinetics were observed for methane concentrations up to 16 g m^{-3} due to mass transfer limitations (Streese and Stegmann, 2003). The batch experiments that are presented in this paper for determination of the methane degradation rate of several cultures will suffer less from mass transfer limitations than in a biofilter system due to the continuous shaking of the system. However, also in these well-shaken batch systems first-order kinetics were observed, up to headspace concentrations of 25 g m^{-3}. Therefore, it is assumed that first-order kinetics are indeed valid for biofilter methane inlet concentrations up to 22 g/m^3.

As was expected, Figure 5.6 clearly shows that in order to minimize the biofilter volume the methane inlet concentration should be maximized. For a liquid manure storage tank, this means that the cover must be further improved to reduce unwanted dilution with ambient air. The methane concentration can then be raised to the level that is still considered safe with regard to explosion hazard. Furthermore, from Figure 5.6 it can be concluded that biofiltration of exhaust air from animal houses, with a methane concentration of only 5 to 100 mg m^{-3}, is not a realistic option as very large biofilter volumes would be required. Therefore the economic evaluation only focuses on the treatment of air from a liquid manure staorage.

5.3.7 Economic evaluation

Based on the pilot-scale biofilter experiments, a design is made for three full-scale biofilter systems that treat exhaust air of a liquid manure storage (75 m^3 $hour^{-1}$) with a methane inlet concentration of 22 g m^{-3}. Figure 5.6 shows that at this methane inlet concentration a biofilter of 10 m^3, 20 m^3, and 40 m^3 is required to achieve a methane removal efficiency of 25%, 50%, and 75%, respectively, assuming that first-order methane degradation still occurs at this concentration. In Table 5.1 a cost calculation of these biofilters is given for application at a 1,000 m^3 liquid manure storage assuming a methane production rate of 40 g m^{-3} manure day^{-1}. The details on the cost calculation are mentioned in the footnotes of Table 5.1. The reduction costs of emission of GHG's are expressed as USD t^{-1} of CO_2 equiv where 1 kg CH_4 stands for 23 kg of CO_2 equiv. The production of N_2O in the biofilter can be neglected, as its contribution is very small in comparison with the high methane outlet concentrations in this calculation.

Table 5.1 Economic evaluation of a biofilter treating exhaust air from a 1,000 m^3 liquid manure storage at a methane inlet concentration of 22 g m^{-3}.[a]

	Methane removal efficiency		
	25%	50%	75%
Biofilter volume (m^3)	10	20	40
EBRT (minutes)[b]	7	17	33
Methane removal (t of CO_2 equiv $year^{-1}$)[c]	84	168	252
Investment costs (USD)[d]			
Rectangular concrete tank[e]	5,000 – 10,000	10,000 - 20,000	15,000 - 30,000
Packing[f]	600 - 1,100	1,200 - 2,200	2,400 - 4,400
Moistening system[g]	750 - 1,5000	1,500 - 3,000	3,000 - 6,000
Engineering and projectmanagement	5,000 - 10,000	7,500 - 12,500	10,000 - 15,000
Fan[h]	350 - 750	350 - 750	350 - 750
Total (average):	17,525	29,500	43,450
Operational costs (USD $year^{-1}$)[d]			
Depreciation[i]	1,753	2,950	4,345
Interest[j]	263	443	652
Electricity use[k]	44	44	44
Packing replacement[l]	536	924	1,700
Total:	2,595	4,361	6,741
Greenhouse gas emission reduction costs (USD t^{-1} CO_2 equiv)[c]	31	26	26

[a] Biofilter volumes, loading rates, and corresponding removal efficiencies are based on extrapolation of pilot-scale biofilter (160 L) experiments at maximum methane inlet concentrations of 5.5 g m^{-3} and a methane production rate of 40 g m^{-3} manure day^{-1}. [b] EBRT = empty bed air residence time; air flow is 75 m^3 $hour^{-1}$. [c] $GWP_{methane}$ is 23. [d] Investment and operational cost were provided by Pure Air Solutions BV, Steenwijk, Netherlands (biofilter manufacturer) and Stork Product Engineering BV, Amsterdam, Netherlands (consulting firm); all prices are excluding value added tax. [e] Including a synthetic roof to keep out rain. [f] Compost/perlite mixture in a 60 : 40 volume ratio; USD 60-110 m^{-3}; packing height is 1 m. [g] Moistening of the biofilter takes place by two meshes of drip irrigation tubes that are located at two depths. [h] 75 m^3 $hour^{-1}$, 50 W. [i] Linear depreciation of 10% per year, residual value is 0. [j] 6% of the net present worth, *i.e.* 3% of the initial investment. [k] 50 W, USD 0.10 kWh^{-1}. [l] Packing replacement once per 5 years; including labor (USD 35 $hour^{-1}$), crane rental (USD 80 $hour^{-1}$), packing disposal costs (USD 35 m^{-3}), and purchase of new packing (USD 60-110 m^{-3}).

From Table 5.1 it follows that the investment and operational costs per m^3 of biofilter decrease with increasing biofilter size, as is usually the case. The average investment costs of a biofilter of 10 m^3, 20 m^3, and 40 m^3 are USD 1800, USD 1500, and USD 1100 per m^{-3}, respectively, as can be derived from Table 5.1. The operational costs, including both fixed and variable costs, are USD 260, USD 220, USD 170 per m^3, respectively. The GHG emission reduction costs of biofiltration, as a measure for reduction of GHG emission from animal husbandry, are USD 31 t^{-1} CO_2 equiv for a biofilter of 10 m^3 and decrease to USD 26 t^{-1} CO_2 equiv with increasing biofilter size. Although no real stock exchange market price exists for emission reduction of CO_2 equiv so far, these costs are generally considered too high for commercial application, as cheaper techniques for GHG mitigation are available.

The investment costs of the biofiltration system, however, may be substantially decreased if the biofilter is integrated in the construction of the liquid manure storage tank. The biofilter probably could be built on top of the liquid manure storage tank using its roof as a basal area; the biofilter and the storage tank could also share the same wall construction if the storage tank was newly built. Furthermore, thermal insulation of the liquid manure storage tank might further decrease the costs of biofiltration, as raising the operating temperature of the biofilter will result in a reduction of the necessary biofilter size.

Alternative techniques for treatment of the exhaust air from a liquid manure storage might be flaring off or burning the methane in a combined heat and power (CHP) installation, the latter replacing some fossil fuel use. Further investigations should determine the cost effectiveness of such techniques in comparison with biofiltration. Finally, it is foreseen that an increasing demand for GHG mitigation techniques will lead to higher emission reduction costs per CO_2 equiv so that relative expensive mitigation techniques, such as biofiltration, may become economically feasible in future.

Acknowledgements

This work was supported by Novem (Nederlandse Organisatie voor Energie en Milieu) and the Dutch Ministry of Agriculture, Nature and Food Quality (LNV). Biofiltration cost figures were provided by Pure Air Solutions BV, Steenwijk, Netherlands and Stork Product Engineering BV, Amsterdam, Netherlands. We thank I.J.T. Dinkla, N.W.M Ogink and H.C. Willers for their suggestions on the manuscript, G. Kupers and J. Slangen for carrying out the laboratory analyses, and M.A. Hilhorst for the project idea.

References

Apel, W.A.; Dugan, P.R.; Wiebe, M.R. (1991). Use of methanotrophic bacteria in gas phase bioreactors to abate methane in coal mine atmospheres. *Fuel.*, *70*, 1001-1003.

Bedard, C.; Knowles, R. (1989).Physiology, biochemistry and specific inhibitors of CH_4, NH_4 and CO oxidation by methanotrophs and nitrifiers. *Microbiol Rev.*, *53*, 68–84.

Bender, M.; Conrad, R. (1992). Kinetics of CH_4 oxidation in oxic soils exposed to ambient air or high CH_4 mixing ratios. *FEMS Microbiol Ecol.*, *101*, 261-270.

Boeckx, P.; Van Cleemput, O.; Villaralvo, I. (1996). Methane emission from a landfill and the methane oxidising capacity of its covering soil. *Soil Biol Biochem.*, *28*, 1397-1405.

Bosse, U.; Frenzel, P. (1997). Activity and distribution of methane-oxidizing bacteria in flooded rice soil microcosms and in rice plants (*Oryza sativa*). *Appl Environ Microb.*, *63* (4), 1199-1207.

Bronson, K.F.; Mosier, A.R. (1993). Suppression of methane oxidation in aerobic soil by nitrogen fertilizers, nitrification inhibitors and urease inhibitors. *Biol Fert Soils.*, *17*, 263-268.

CEN (2003). *Air quality - Determination of odour concentration by dynamic olfactometry*. EN13725. European Committee for Standardization: Brussels, Belgium, 2003.

Czepiel, P.M.; Mosher, B.; Crill, P.M.; Harriss, R.C. (1996). Quantifying the effects of oxidation on landfill methane emissions. *J Geophys Res.*, *101*, 16721-16729.

Dammann, B.; Streese, J.; Stegmann, R. (1999). Microbial oxidation of methane from landfills in biofilters. In *Proceedings Sardinia '99, 7th International Waste Mangement and Landfill Symposium*; Cagliari, Italy, October 4-8, 1999; Christensen, T.H., *et al.*, Eds.; CISA: Cagliari, Italy; Vol. II, pp 517-524.

Dunfield, P.F.; Conrad, R. (2000). Starvation alters the apparent half-saturation constant for methane in the Type II methanotroph *Methylocystis* strain LR1. *Appl Environ Microb.*, *66* (9), 4136-4138.

Devinny, J.S.; Deshusses, M.A.; Webster, T.S. (1999). *Biofiltration for air pollution control.* Lewis Publishers: Boca Raton, 1999; ISBN 1-56670-289-5.

De Visscher, A.; Thomas, D.; Boeckx, P.; Van Cleemput, O. (1999). Methane oxidation in simulated landfill cover soil environments. *Environ Sci Technol.*, *33*, 1854-1859.

De Visscher, A.; Van Cleemput, O. (2003). Simulation model for gas diffusion and methane oxidation in landfill covers soils. *Waste Manage.*, *23*, 581-591.

Du Plessis, C.A.; Strauss, J.M.; Sebapalo, E.M.T.; Riedel, K.H.J. (2003). Empirical model for methane oxidation using a composted pine bark biofilter. *Fuel.*, *82*, 1359-1365.

EAA (2002). *Greenhouse gas emission trends in Europe, 1990 - 2000*. Topic report 7/2002. European Environment Agency: Copenhagen, Denmark, 2002; ISBN: 92-9167-516-4. http://reports.eea.eu.int/topic_report_2002_7/en/.

EPA (2004). *Inventory of U.S. greenhouse gas emissions and sinks: 1990-2002*. Report EPA 430-R-04-003. U.S. Environmental Protection Agency: Washington, DC, 2004. http://www.epa.gov/globalwarming/publications/emissions.

Figueroa, R.A. (1996). Landfill gas treatment by biofilters. In *Landfilling of waste: Biogas*; Christensen, T.H., et al., Eds.; E & FN Spon: London, 1996; ISBN 0-419-19400-2; pp 535-549.

Freibauer, A. (2002). *Biogenic Greenhouse Gas Emissions from Agriculture in Europe – Quantification and Mitigation*. PhD thesis. University of Hohenheim: Stuttgart, Germany, 2002. http://www.uni-hohenheim.de/ub/opus/volltexte/2002/22/.

Furusawa, N.; Togashi, I.; Hirai, M.; Shoda, M.; Kubota, H. (1984). Removal of hydrogen sulfide by a biofilter with fibrous peat. J Ferment Technol., *62* (6), 589-594.

Gebert, J.; Groengroeft, A.; Miehlich, G. (2003). Kinetics of microbial landfill methane oxidation in biofilters. *Waste Manage.*, *23*, 609-619.

Hahne, J.; Vorlop, K.-D. (2001). Treatment of waste gas from piggeries with nitrogen recovery. *Landbauforsch Volk.*, *3* (51), 121-130.

Hanson, R.S.; Hanson, T.E. (1996). Methanotrophic bacteria. *Microbiol Rev.*, *60* (2), 439-471.

Heipieper, H.J.; De Bont, J.A.M. (1997). Methane oxidation by Dutch grassland and peat soil microflora. *Chemosphere*, *35* (12), 3025-3037.

Humer, M.; Lechner, P. (1999a) Alternative approach to the elimination of greenhouse gases from old landfills. *Waste Manage Res.*, *17*, 443-452.

Humer, M.; Lechner, P. (1999b) Methane oxidation in compost cover layers on landfills. In *Proceedings Sardinia 1999, 7th International Waste Management and Landfill Symposium*; Cagliari, Italy, October 4-8, 1999; Christensen, T.H., *et al.*, Eds.; CISA: Cagliari, Italy; Vol. III, pp 403-410.

IPCC (2001). Climate change 2001: The scientific basis. Contribution of working group I to the third assessment report of the Intergovernmental Panel on Climate Change; Houghton, J.T., et al.; Eds.; Cambridge University Press: Cambridge, U.K. and New York, NY, USA, 2001; ISBN 0521-80767-0 and ISBN 0521-01495-6; 881 pp. http://www.grida.no/climate/ipcc_tar/.

Jones, H.A.; Nedwell, D.B. (1993). Methane emission and methane oxidation in land-fill cover soil. FEMS Microbiol Ecol., 102, 185-195.

Kennes, C.; Veiga, M.C.; Prado, O. (2001). Non-biological treatment technologies. In *Bioreactors for waste gas treatment*; Kennes, C.; Veiga M.C., Eds.; Kluwer Academic Publishers: Dordrecht, Netherlands, 2001; ISBN 0-7923-7190-9, pp 17-46.

Kightley, D.; Nedwell, D.B.; Cooper, M. (1995). Capacity for methane oxidation in landfill cover soils measured in laboratory-scale microcosms. *Appl Environ Microb.*, *61* (2), 592-601.

King, G. M.; Schnell, S. (1994). Ammonium and nitrite inhibition of methane oxidation by *Methylobacter albus* BG8 and *Methylosinus trichosporium* OB3b at low methane concentrations. *Appl Environ Microb.*, *60*, 3508–3513.

Krumholz, L.R.; Hollenback, J.L.; Roskes, S.J.; Ringelberg, D.B. (1995). Methanogenesis and methanotrophy within Sphagnum peatland. *FEMS Microbiol Ecol.*, *18*, 215-224.

Kussmaul, M.; Gebert, J. (1998). Ein neues Verfahren zum biologischen Methan- und Geruchsabbau von Gasen aus Abfalldeponien mit passiver Entgasung. *Müll und Abfall.*, *30* (8), 512-518.

Martinec, M.; Hartung, E.; Jungbluth, T.; Schneider, F.; Wieser, P.H. (2001). Reduction of gas, odor and dust emissions from swine operations with biofitlers. In *Proceedings of the 2001 ASAE Annual International Meeting*; Sacramento, USA, July 30-August 1, 2001; Paper 014079, 8 pp.

Melse, R.W. (2003). *Methane degradation in a pilot-scale biofilter for treatment of air from animal houses and manure storages* (in Dutch). Report 16. Agrotechnology & Food Innovations, Wageningen UR: Wageningen, Netherlands, 2003; ISBN 90-5406-240-1.

NEN (1998). *Overview of standards for analysis of water and sludges* (in Dutch). Nederlands Normalisatie Instituut: Delft, Netherlands, 1998.

Park, S.; Shah, N.N.; Taylor, R.T.; Droege, M.W. (1992). Batch cultivation of Methylosinus trichosporium OB3b: II. Production of particulate methane mon-oxygenase. *Biotechnol Bioeng.*, *40 (1)*, 151-157.

Park, S.; Brown, K.W.; Thomas, J.C. (2002). The effect of various environmental and design parameters on methane oxidation in a model biofilter. *Waste Manage Res.*, *20*, 434-444.

Park, S.Y.; Brown, K.W.; Thomas, J.C. (2004). The use of biofilters to reduce atmospheric methane emissions from landfills: Part I. Biofilter design. *Water Air Soil Poll.*, *155*, 63-85.

Petersen, S.O.; Amon, B.; Gattinger, A. (2005). Methane oxidation in slurry storage surface crusts. *J Environ Qual.*, *34 (2)*, 455-461.

Rogers, J.E.; Whitman, W.B. (1999). *Microbial production and consumption of greenhouse gases: methane, nitrogen oxides, and halomethanes*; American Society for Microbiology: Washington, 1999; ISBN 1-55581-035-7; 298 pp.

Scharff, H.; Oonk, H.; Vroon, R.; Van der Sloot, H.A.; Van Zomeren, A.; Hensen, A. (2001). Improved methane oxidation by means of forced aeration under a landfill cover. In *Proceedings Sardiania 2001, 8th International Waste Management and Landfill Symposium*; Cagliari, Italy, October 1-5, 2001; Christensen, T.H., *et al.*, Eds.; CISA: Cagliari, Italy; Vol. II, pp 555-564.

Sly, L.I.; Bryant, L.J.; Cox, J.M.; Anderson, J.M. (1993). Development of a biofilter for the removal of methane from coal mine ventilation atmospheres. *App Microbiol Biotechnol.*, *39*, 400-404.

Streese, J.; Stegmann, R. (2003). Microbial oxidation of methane from old landfills in biofilters. *Waste Manage.*, *23*, 573-580.

Streese, J.; Dammann, B.; Stegmann, R. (2001). Reduction of methane and trace gas emissions from former landfills in biofilters. In *Proceedings Sardinia 2001, 8th International Waste Management and Landfill Symposium*; Cagliari, Italy, October 1-5, 2001; Christensen, T.H., *et al.*, Eds.; CISA: Cagliari, Italy; Vol. II, pp 575-584.

Stein, V.B.; Hettiaratchi, J.P.A. (2001). Methane oxidation in three Alberta soils: influence of soil parameters and methane flux rates. *Environ Technol.*, *22*, 101-111.

Straka, F.; Crha, J.; Musilova, M.; Kuncarova, M. (1999). LFG-biofilters on old landfills. In *Proceedings Sardinia 1999, 7th International Waste Management and Landfill Symposium*; Cagliari, Italy, October 4-8, 1999; Christensen, T.H., *et al.*, Eds.; CISA: Cagliari, Italy; Vol. II, pp 507-516.

UNFCC (1997). *Kyoto Protocol to the United Nations Framework Convention on Climate Change*. Document FCCC/CP/1997/Add. 1, 1997. http://unfccc.int/resource/docs/convkp/kpeng.pdf.

Whalen, S.C.; Reeburch, W.S.; Sandbeck, K.A. (1990). Rapid methane oxidation in a landfill cover soil. *Appl Environ Microb.*, *56* (11), 3405-3411.

Yang, Y.; Allen, E.R. (1994). Biofiltration control of hydrogen sulfide. 1: Design and operational parameters. *J Air Waste Manage.*, *44* (7), 863-868.

Chapter 6

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

R.W. Melse; G. Mol

Wageningen University and Research centre, Agrotechnology & Food Innovations, Wageningen, Netherlands.

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 for a major part, about 80%, could be attributed to actual changes in the performance of the biotrickling filter. These changes were probably caused by variations in the composition of the air that were not completely reflected by the olfactometrically measured odour concentration, as 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 [OU_E/(m^3 filter)/s], which means that the absolute odour removal will probably rise at increasing load. 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 odour removal efficiency [%], the design of the filter probably needs to be optimized for both well and poorly water-soluble odour components.

Key words: *Air, Ammonia, Biotrickling filter, Odour, Olfactometry, Pig, Removal.*

6.1 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, ammonia (NH_3) emitted from livestock farming has become a major environmental concern in the Netherlands because it is one of the three main sources of soil acidification, together with nitrogen oxides (NO_x) and sulphur oxides (SO_x) (Heij and Erisman, 1995, 1997). In 2000, the ammonia emission from farming activities still accounted for 39% 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 mechanically ventilated 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.

6.2 Materials and methods

6.2.1 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. If the air contains water-soluble components, these components are partly transferred to the wet biofilm and are available for bacterial degradation. A schematic of a biotrickling filter is given in Figure 6.1.

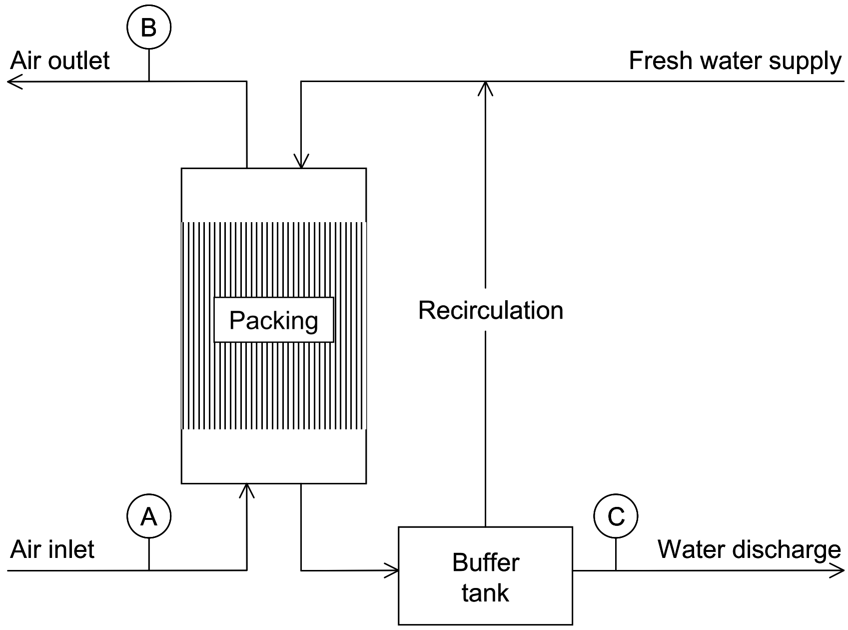


Figure 6.1 Schematic of biotrickling filter.

In the case of ammonia, the mass transfer largely depends on the pH of the water due to the equilibrium between NH_3 and NH_4^+ as is shown in Equation 6.1:

$$NH_3\ (g) + H_2O \text{ <---> } NH_3\ (aq) + H_2O \text{ <---> } NH_4^+ + OH^- \qquad \text{[Eq. 6.1]}$$

Subsequent bacterial oxidation from ammonium to nitrite (NO_2^-) and from nitrite to nitrate (NO_3^-) is called nitrification and mainly carried out by *Nitrosomonas* and *Nitrobacter* species respectively. In Equation 6.2 and 6.3 these processes are shown:

$$NH_4^+ + OH^- + 1.5\ O_2 \text{ ---> } NO_2^- + H^+\ 2\ H_2O \qquad \text{[Eq. 6.2]}$$

$$NO_2^- + H^+ + 2\ H_2O + 0.5\ O_2 \text{ ---> } NO_3^- + H^+ + 2\ H_2O \qquad \text{[Eq. 6.3]}$$

A stable operating biotrickling filter usually is in a steady-state condition which means there is an equilibrium between the processes shown in Equation 6.1 through 6.3 and the amount of nitrogen and H^+ that is removed from the system by water discharge. This normally results in the following conditions: 6.5 < pH < 7.5, 1 < [NH_4-N + NO_2-N + NO_3-N] (g /L) < 4, and 0.8 < [NH_4^+]/[NO_2^- + NO_3^-] < 1.2 on a molar basis (Scholtens, 1996; Den Brok *et al.*, 1997).

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 compounds are manifold (*e.g.* feed, animal, manure, bedding). Water solubility may vary from very low to very high. 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 a reduction of odour emission. It is known that many odorous compounds can be biologically degraded although biodegradability may vary from very low to very high.

The biotrickling filter that was studied is a commercially available system that had been installed at a regular pig production farm in the Netherlands in order to remove ammonia from part of the exhaust air from 650 fattening pigs. Based on an ammonia removal efficiency of 70% and an expected lifetime of 10 years, the sum of the capital and operational costs of a biotrickling filter for this application is about € 14 - € 17 per animal place per year or € 8 - € 10 per kg NH_3 abatement, excluding water discharge costs (Melse and Willers, 2003). 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^3 and a height of 1 m and had been designed for a maximum air flow of 20,000 m^3/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^3) and then partly discharged or recirculated. Both air inlet (Figure 6.1, point A) and air outlet (Figure 6.1, point B) tubes were equipped with an air sampling port and a sensor for measurement of temperature [°C] and relative humidity [%]. The air flow [m^3/s] was calculated from the frequency [Hz] of the fan and the technical specifications from the manufacturer. The discharge water was sampled at point C (Figure 6.1). The research took place between 8th October (day 0) and 19th December 2001 (day 72).

6.2.2 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 drawing air (120 L/hour) from the air sampling port 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 thus time-averaged over two hours. The 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 (NNI, 1998). Ammonia emission [g/s] was calculated by multiplying ammonia concentration [g/m^3] by air flow [m^3/s].

6.2.3 Odour measurements

Single 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. According to this method air samples were collected in Teflon odour bags (60 L) that were placed in airtight containers. The inlet of the initially evacuated odour bags was connected to the sampling port of the air inlet and air outlet respectively 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 were time-averaged over two hours. A filter (pore diameter: 1-2 μm) at the inlet of the sampling tube prevented the intake of dust that otherwise would 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 has 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 m^3 air [OU_E/m^3] (CEN, 1998). Odour emission [OU_E/s] was calculated by multiplying odour concentration [OU_E/m^3] by air flow [m^3/s].

6.2.4 Water analysis

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

6.3 Results and discussion

6.3.1 Ammonia removal

The results of the ammonia measurements are given in Figure 6.2. The ammonia concentration of the inlet air of the biotrickling filter varied from 14 to 20 mg N/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 m^3/hour.

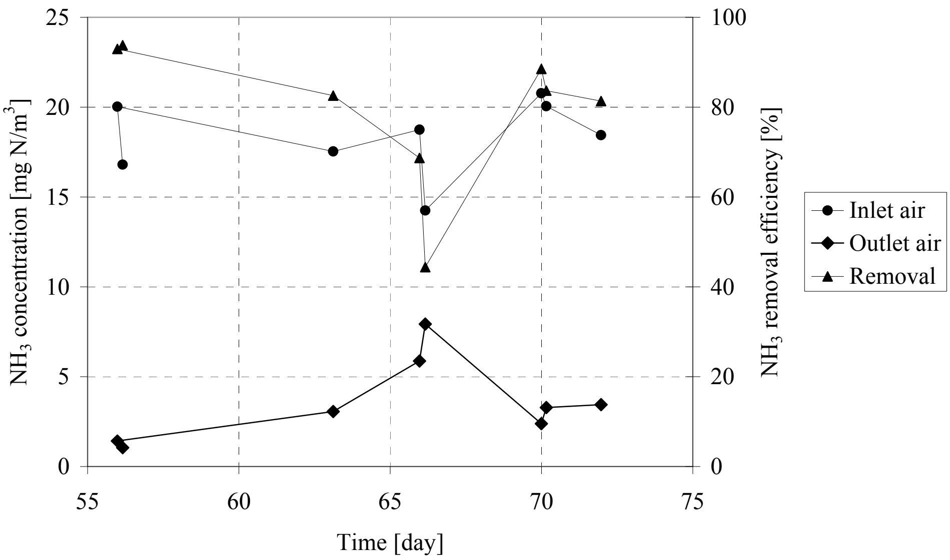


Figure 6.2 Ammonia removal by biotrickling filter treating exhaust air from a pig house.

The characteristics of the discharge water from the biotrickling filter (Table 6.1) 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 [NH_4^+] >> 0.4 g/L; *Nitrobacter* seems to be fully inhibited as [NO_3^-] ≈ 0. It is known that both ammonium and nitrite, in its undissociated forms of NH_3 (aq) and HNO_2 (aq), are inhibitive to nitrifying bacteria. Inhibition of *Nitrosomonas* by NH_3 (aq) starts at concentrations of 10 - 150 mg/L, whereas *Nitrobacter* is already inhibited by NH_3 (aq) at concentrations from 0.1 to 1 mg/L; moreover, *Nitrobacter* is inhibited by HNO_2 (aq) starting at concentrations of 0.2 - 2.8 mg/L (Anthonisen *et al.*, 1976). Assuming equilibrium, the NH_3 (aq) and HNO_2 (aq) concentrations in the discharge water are calculated to be 17 - 55 mg/L and 0.2 - 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 ammonium and nitrite concentrations that were found, resulting in accumulation of these compounds in the discharge water.

Table 6.1 Characteristics of discharge water of biotrickling filter treating exhaust air from a pig house.

Sample day	NH_4-N [g/l]	NO_2-N [g/l]	NO_3-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 filter nitrogen does not accumulate but leaves the system as nitrate with the discharge water. Measurements of the composition of the water that is sprayed over the bed (data not presented) show that the composition is equal to the composition of the discharge water (Table 6.1) indicating that hardly any fresh water is added to the system. Direct measurements of the fresh water intake are not available.

In the long run however, if no sufficient amount of fresh water is added biological activity will further decrease and accumulation of ammonium will proceed until hardly any ammonia will be removed from the air anymore because the equilibrium of Equation 6.1 will shift to the left more and more. In order to ensure successful ammonia removal and to stop the accumulation of nitrogen in the system, the fresh water intake must be drastically increased so that nitrogen will be removed from the system with the discharge water.

6.3.2 Odour removal

The results of the odour measurements are given in Figure 6.3. The odour concentration of the inlet air of the biotrickling filter varied from 1,000 to 2,400 OU_E/m^3 while the odour removal efficiency varied from -29 to 87% (49% on average).

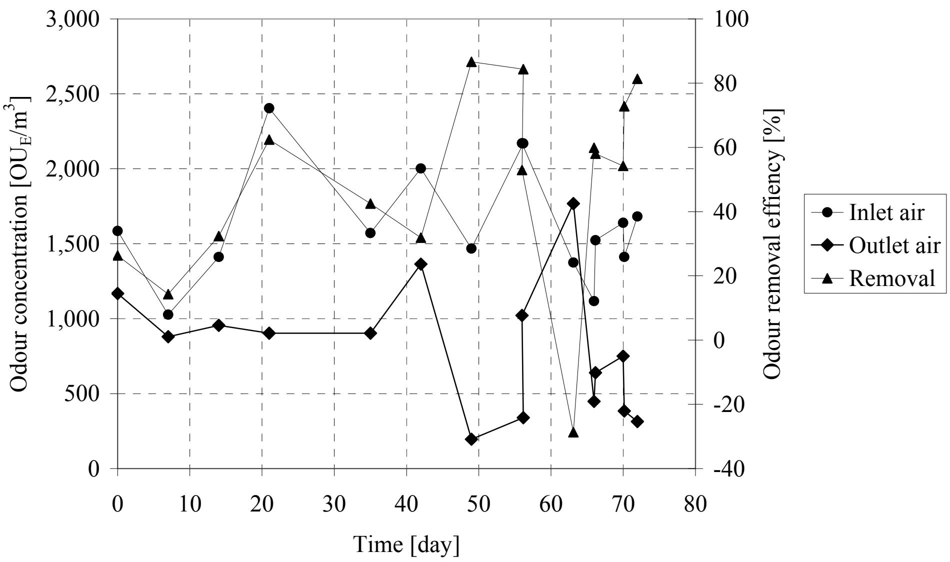


Figure 6.3 Odour removal by biotrickling filter treating exhaust air from a pig house.

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 variance 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 for about 20% to the total variance of the odour removal efficiency measurements, whereas the functioning of the biotrickling system contributes for about 80% to the total variance.

The reason for the variation of the odour removal efficiency being higher than for ammonia is possibly 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 in the air, 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 were not measured separately in this study, the phenomenon described here may explain the relatively large variation that was found for the odour removal efficiency.

In Figure 6.4 the odour removal [$OU_E/(m^3$ filter)/s] of the system is plotted against the odour load, *i.e.* the product of the inlet concentration and the air flow, showing a positive correlation. No data are shown for day 0-56 because no reliable air flow measurements are available for this period. It is often assumed that a biofiltration system has a maximum removal capacity for a polluting component (*e.g.* Deshusses and Johnson, 2000). This means that the absolute removal of the component [kg/(m^3 filter)/s] rises at increasing load until the maximum removal capacity is reached; if the load increases even further, the absolute removal will not further rise and the removal efficiency [%] will decrease. The reason for this phenomenon is that the removal process is usually limited by the degradation capacity of the bacterial community and not by the mass transfer from the air to the liquid phase. Therefore the absolute removal of a component is generally determined by the load of the component and not by the inlet concentration of the component. However, if a component is very poorly water-soluble, *i.e.* has a very high Henry's law constant, mass transfer may be the rate limiting step of the removal process.

As the odour removal in Figure 6.4 is still rising at increasing 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, in this study it is not possible to conclude that the odour removal is indeed determined by the odour load and not by the odour concentration because air flow and odour inlet concentration are positively correlated as is shown in Figure 6.5. As odorous air consists of both well and poorly water-soluble compounds, limitation of mass transfer may also play a part in this

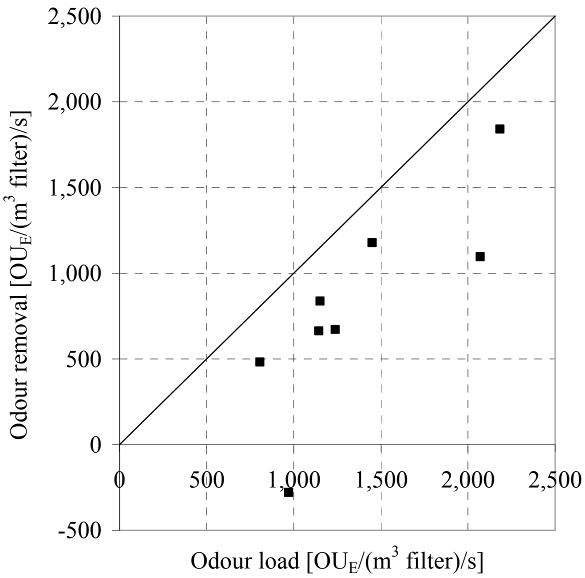


Figure 6.4 Odour removal versus odour load (day 56-72). The line indicates 100% removal efficiency.

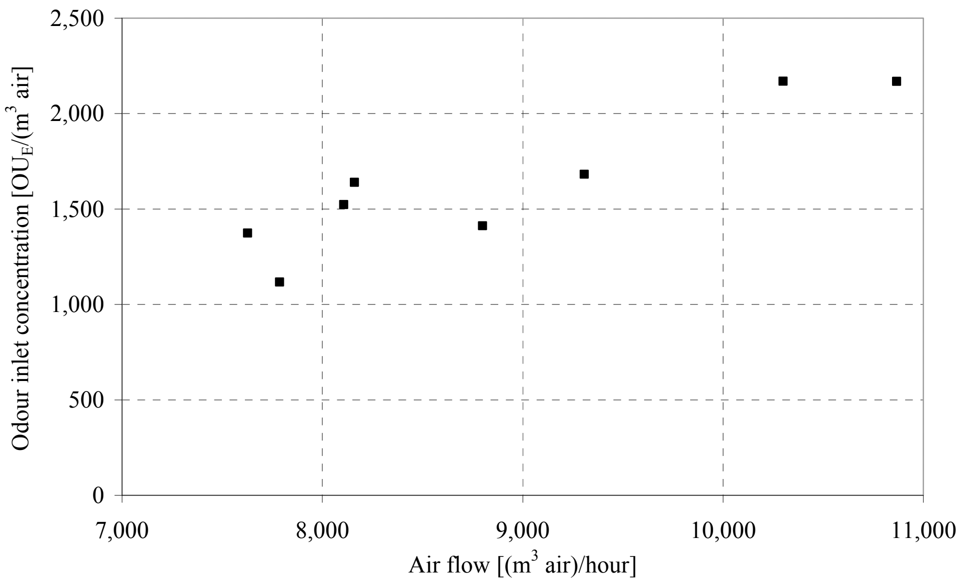


Figure 6.5 Odour concentration versus air flow (day 56-72).

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 odour that is still being emitted from the pig house. The removal efficiency now averages at about 50% of the incoming odour load which may be considered unsatisfactory with regards to nuisance. Because the odour removal is probably limited by the transfer of poorly water-soluble odour components from the air to the water phase, efforts to improve the odour removal should focus on changing system characteristics that influence the mass transfer process. Some options in this respect are the characteristics of the packing material, *e.g.* the specific surface area, the retention time, *e.g.* applying a lower air flow or a larger filter volume, and the design of a two- or multi-phase filter which is optimized for subsequent removal of poorly and well water-soluble odour components.

6.4 Conclusions

The removal of ammonia from the exhaust air of the mechanically ventilated 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 intake of the system needs to be increased to achieve a stable and reliable ammonia removal system.

The efficiency of the removal of odour from the exhaust air of the pig house showed a large variation that for a major part could be attributed to actual changes in the performance of the biotrickling filter. These changes in performance are probably caused by variations in the composition of the odorous air that are not completely reflected by the olfactometrically measured odour concentration, as 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 [OU_E/(m^3 filter)/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 odour removal efficiency [%], the design of the filter needs to be optimized for removal of both well and poorly water-soluble odour components.

Acknowledgements

This research was financed by the Dutch ministry of Agriculture, Nature and Food Quality (LNV).

References

Anthonisen A. C., Loehr R. C., Prakasam T. B. S. and Srinath E.G. (1976). Inhibition of nitrification by ammonia and nitrous acid. *Journal Water. Poll. Contr. Fed.*, 48, 835-852.

CBS (2002). *StatLine databank*, Statistics Netherlands website, *http://www.cbs.nl.*

CCDM (2002). *Emission Monitor for the Netherlands 2000 and estimates for 2001* [In Dutch], Report Series Environmental monitor 6. Coördinatiecommissie Doelgroepmonitoring, The Hague, Netherlands.

CEN (1998). *Odour concentration measurement by dynamic olfactometry*, Draft prEN13725, CEN/TC264/WG2, European Committee for Standardization, Brussels, Belgium.

Den Brok G. M., Verdoes N., Hoofs A. I. J. and Van Brakel C. E. P. (1997). *Varkensstallen met een lage ammoniakuitstoot* [Pig houses with a low ammonia emission], Report P2.32, Praktijkonderzoek Varkenshouderij, Rosmalen, Netherlands.

Deshusses M. A. and Johnson C. T. (2000). Development and validation of a simple protocol to rapidly determine the performance of biofilters for VOC treatment. *Environ. Sci. Technol.*, 34(3), 461-467; DOI: 10.1021/es9909172.

Heij G. J. and Erisman J. W. (eds.) (1995). *Acid Rain Research: Do We Have Enough Answers?* Proceedings of a Speciality Conference, 's-Hertogenbosch, Netherlands, 10-12 October 1994. Studies in Environmental Science, 64, 516 pp., Elsevier, Netherlands.

Heij G. J. and Erisman J. W. (1997). *Acid Atmospheric Deposition and its Effects on Terrestrial Ecosystems in the Netherlands. The Third and Final Phase (1991-1995).* Studies in Environmental Science, 69, 716 pp., Elsevier, Netherlands.

Melse R.W. and Willers H.C. (2003). *Toepassing van luchtbehandelingstechnieken binnen de intensieve veehouderij. Fase 1: Techniek en kosten* [Treatment of exhaust air from animal houses. Phase 1: Techniques and costs], Report 029, Agrotechnology & Food Innovations, Wageningen UR, Wageningen, Netherlands. ISBN 90-6754-739-5.

Mol G. and Ogink N. W. M. (2002). *Geuremissies uit de veehouderij II. Overzichtsrapportage 2000-2002* [Odour emissions from animal husbandry II. General report 2000-2002], Report 2002-09, IMAG, Wageningen, Netherlands.

NNI (1995). *NVN 2820 Luchtkwaliteit, sensorische geurmetingen met een olfactometer*, Nederlands Normalisatie Instituut, Delft, Netherlands.

NNI (1996). *NVN 2820/A1. Luchtkwaliteit, sensorische geurmetingen met behulp van een olfactometer*, Wijzigingsblad NVN 2820/A1. Nederlands Normalisatie-instituut, Delft, Netherlands.

NNI (1998). *Overview of standards for analysis of water and sludges* [In Dutch], Nederlands Normalisatie Instituut, Delft, Netherlands.

Ogink N. W. M. and Lens P. N. (2001). *Geuremissies uit de veehouderij I. Overzichtsrapportage 1996-1999* [Odour emissions from animal husbandry I. General report 1996-1999], Report 2001-14, IMAG, Wageningen, Netherlands.

O'Neill D. H. and Phillips V. R. (1992). Review of the control of odour nuisance from livestock buildings: Part 3: Properties of the odorous substances which have been identified in livestock wastes or in the air around them. *J. Agric. Engng Res.*, 53, 23-50.

Scholtens R. (1996). *Inspectie van luchtwassystemen voor mechanisch geventileerde varkensstallen* [Inspection of air treatment systems for mechanically ventilated pig houses.], Internal report, IMAG, Wageningen, Netherlands.

Van Ouwerkerk E. N. J. (1993). *Meetmethoden NH_3-emissie uit stallen. Werkgroep Meetmethoden NH_3-emissie uit stallen* [Methods to measure NH_3-emission from animal houses], Onderzoek inzake de mest- en ammoniakproblematiek in de veehouderij 16, DLO, Wageningen, Netherlands.

Chapter 7

Evaluation of four farm-scale systems for the treatment of liquid pig manure

R.W. Melse[1]; N. Verdoes[2]

[1]Wageningen University and Research centre, Agrotechnology & Food Innovations, Wageningen, Netherlands; [2]Wageningen University and Research centre, Animal Sciences Group, Lelystad, Netherlands.

ABSTRACT In some regions in the Netherlands, high pig concentrations and limited availability of arable land have led to a surplus of manure which results in high off-farm manure disposal costs. The aim of manure treatment is to lower manure transport costs by reducing the volume and to improve market prospects by changing the nutrient composition. The objective of this study was to promote the introduction of manure treatment in the Netherlands by giving research support to farmer initiatives and providing them with data on the actual performance of their system with regard to product composition, mass flows, gaseous emissions, and economic feasibility. Four farm-scale systems for treatment of liquid pig manure were studied: two systems for mechanical separation, one for nitrification / denitrification, and one for evaporation. The results showed that a wide range of manure products could be obtained that differ in dry matter, N, P, and K content. The emission of ammonia and odour varied from 1.8 to 55 g t^{-1} [manure] and 3.8×10^3 to 1.3×10^7 [European odour units] t^{-1} [manure] respectively. The nitrification / denitrification showed the highest emission of greenhouse gases (48 kg [carbon dioxide equivalents] t^{-1} [manure]), mainly nitrous oxide (N_2O), whereas the emission of the other systems was 12 to 17 kg [CO_2 equiv] t^{-1} [manure].

The critical success factor for operation of the manure treatment installations turned out to be not of technical but economical nature. The manure treatment costs, including variable and fixed cost, varied from 7 to 17 € t^{-1} (excluding value added tax). To be cost-effective in comparison with the disposal of untreated manure, these costs must be balanced out by the sale or the lower disposal costs of the manure products. As market prospects and disposal costs for manure and its products differ from case to case, no generally preferred manure treatment technique can be pointed out from this study, as local market circumstances must be taken into account.

7.1 Introduction

Pig production contributes substantially to the economy of many Western European countries in terms of employment and export of products. Production facilities are concentrated in several regions characterised by large-scale intensive farms. High pig concentrations and limited availability of nearby arable land led to a surplus of manure in some regions, which resulted in strict legislation (Burton & Turner, 2003).

In the Netherlands, this has resulted in high off-farm manure disposal costs for liquid manure which consist of sampling and analysis, transportation, land application, and, in some regions, an acceptance bonus for the landowner. The aim of manure treatment is two-fold. The first aim is to reduce transportation costs by volume reduction and the second aim is to make manure products that can compete with chemical fertilizer and untreated manure by adjusting the concentration and ratio of nitrogen (N), phosphorus (P), and potassium (K) according to the needs of the arable farmer.

This paper describes a study that was carried out on four farm-scale systems for treatment of liquid pig manure. The aim of the research was to promote the introduction of manure treatment in the Netherlands by giving research support to farmer initiatives in this field. The performance of each system was determined with regard to product composition, mass flows, gaseous emissions (ammonia, odour, and greenhouse gases), and costs, thus providing the farmers with a rational basis for making decisions with regard to the marketing of manure products and the economic feasibility of their system. Especially the data on gaseous emissions are relevant for getting a legal permit that allows building and operation of a manure treatment installation.

7.2 Materials and methods

7.2.1 General

All four installations were located at different pig farms in the Netherlands, which had the consequence that the composition of the manure to be treated varied from installation to installation. Each system had been in continuous operation without failures for at least two weeks prior to the measurements. For each system, product composition and flow were measured during four consecutive weeks in the period from November 2000 to April 2002. Also gaseous emissions were determined. Finally, the results obtained were used for an economic evaluation of each manure treatment system.

7.2.2 System 1: straw filtration inside a greenhouse

The first system that was studied was a straw filter placed in a greenhouse (500 m^3) made of 3-layered plastic foil (polyethylene - ethyl phenylacetate - polyethylene; total thickness: 0.2 mm) and is shown in Figure 7.1 (Melse & Gijsel, 2001).

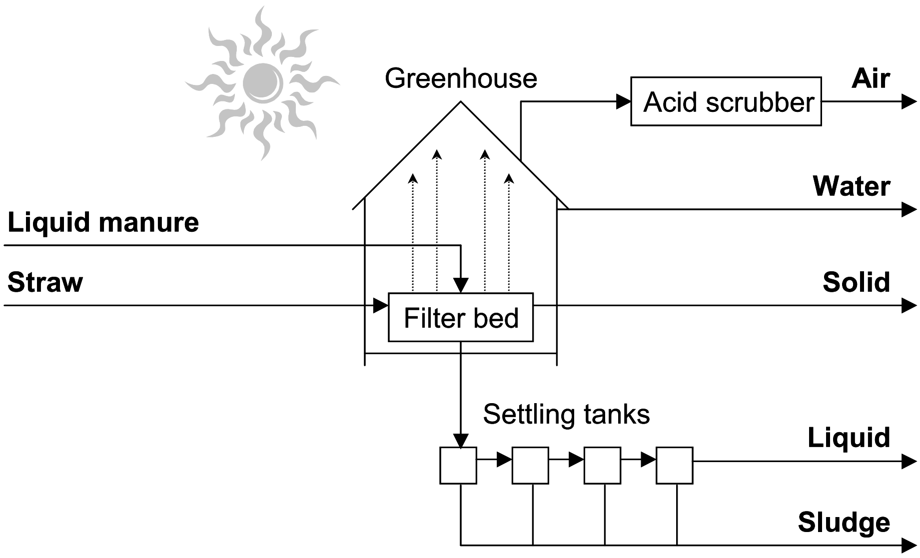


Figure 7.1 System 1: schematic of the straw filter system used for treatment of liquid pig manure; from Melse & Gijsel (2001).

The filter consisted of a layer of uncompressed straw (5 cm) on a trenched concrete floor (200 m^2); the use of straw is about 5 kg t^{-1} of treated manure. A mixture of liquid manure from sows and fattening pigs (1,600 t y^{-1}) was pumped to the greenhouse from a storage tank and periodically sprayed on the straw bed which operated as a cake filter. In a filtration system like this, directly after start-up only few solids are retained from the manure but as time passes the solids accumulate in the straw forming a cake which retains solid particles very well. The filtrate was processed by four cylindrically shaped settling tanks, connected in series (each tank: diameter, 2 m; height, 2 m; volume, 6 m^3). The settled filtrate was stored prior to land application whilst the settled sludge accumulated in the tanks during the evaluation period of four weeks. Inside the greenhouse part some of the liquid manure was evaporated by solar radiation and the water which condensed on the inner surface of the plastic foil was collected to be used as ammonium fertilizer. The greenhouse was mechanically ventilated (390 $m^3 h^{-1}$) and the exhaust air was scrubbed with sulphuric acid to remove ammonia. After four weeks of operation the filtration capacity of the straw bed had been drastically reduced by accumulation of solids. The addition of liquid manure was stopped and the straw bed was left for some days until the filtrate flow came to a complete stop. Then the straw bed, which can be considered as solid manure, was removed and stored on an open-air pile with a concrete floor. The drain water from this pile, including the collected rainwater, (about 0.3 t d^{-1}) was added to the settled filtrate.

Experience with the application of a straw bed filter for manure treatment has been gathered for some years now (Kalyuzhnyi *et al.*, 2000; PDV, 1994; Priem, 1980). New aspects in this study were the closed and mechanically ventilated environment in which the filter was operated, the evaporation of the liquid manure by the elevated temperature in the greenhouse, the condensation, and the subsequent collection of ammonia containing water.

7.2.3 System 2: mechanical separation and microfiltration

The second system that was studied was a mechanical separation and filtration system as shown in Figure 7.2. A flushing gutter system for manure removal had been installed in the pig house in order to reduce the ammonia emission (Satter *et al.*, 1997). A mixture of liquid manure from sows and fattening pigs (3,600 t y^{-1}) and flushing liquid was separated with a screw press (FAN Separator, Germany; pore diameter of 0.25 mm) into a filtrate and a solid fraction. Part of the filtrate was recirculated as flushing liquid, part of it was separated in a decanter centrifuge (Pieralisi, Italy, Baby II; capacity of 0.7 $m^3 h^{-1}$; 7.5 kW). The solid fractions from both the screw press and the centrifuge were stored; the ratio of the production rate of these fractions is 2 : 1 on a mass basis. The liquid fraction from the centrifuge was treated in a microfiltration unit (Zenon GmbH, Germany; pore diameter of 0.2 μm; membrane surface of 15 m^2) and a concentrate and a permeate were produced. Part of the permeate was recycled until desired concentrations in both liquids had been obtained. Every two weeks the membranes were cleaned. The entire system was placed in a closed shed which was mechanically ventilated at a rate of 3,000 $m^3 h^{-1}$.

7.2.4 System 3: nitrification and denitrification

The third system that was studied was a biological nitrification / denitrification system as shown in Figure 7.3. In such a system the main processes that take place are nitrification, *i.e.* bacterial conversion of ammonia (NH_4^+) to nitrite (NO_2^-) and nitrate (NO_3^-), denitrification, *i.e.* bacterial conversion of nitrate to dinitrogen gas (N_2), and oxidation of organic matter to carbondioxide (CO_2).

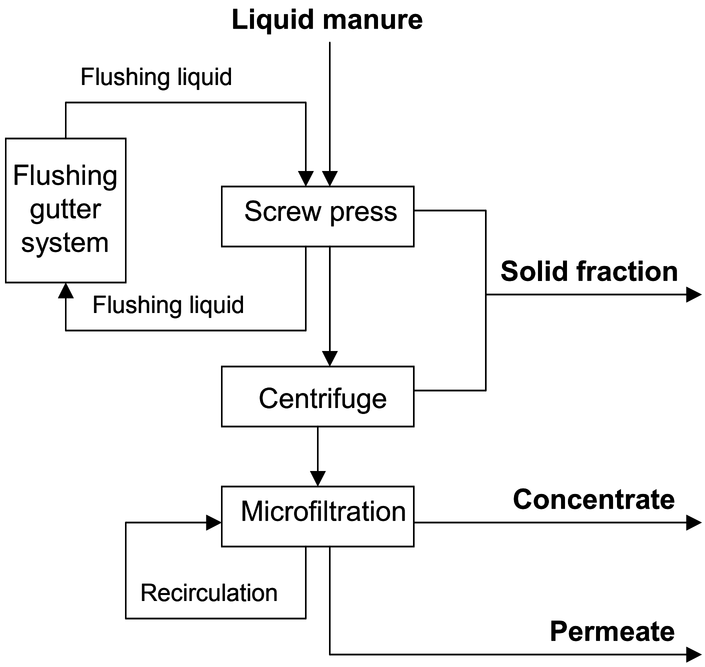


Figure 7.2 System 2: schematic of the mechanical separation and ultrafiltration system used for treatment of liquid pig manure.

With a peristaltic pump, liquid manure from sows (3,000 t y^{-1}) was pumped from a storage tank to the denitrification tank (polyester, 60 m^3) where it was mixed with the recirculating liquid from the nitrification tank (polyester, 60 m^3). The volume ratio of fresh manure input over recirculation flow was 1 : 10. After passing the denitrification tank the liquid entered the nitrification tank where membrane aerators provided continuous aeration of the liquid (100 - 120 m^3 [air] h^{-1}). Each tank has a hydraulic retention time of approximately one week. Finally the liquid entered a settling basin (600 m^3) where effluent and sludge were produced; the ratio of the production rate of these fractions is about 4 : 1 on a mass basis.

Hydrated lime [$Ca(OH)_2$] was added in order to precipitate phosphates and prevent acidification. Continuously, anti-foaming agent (1.3 *ml* t^{-1} [manure]) and molasses (0.1 *l* t^{-1} [manure]) was added, the latter to provide additional carbon for the denitrifying bacteria. This manure treatment system, excluding the centrifuge, was manufactured by Kamplan, the Netherlands.

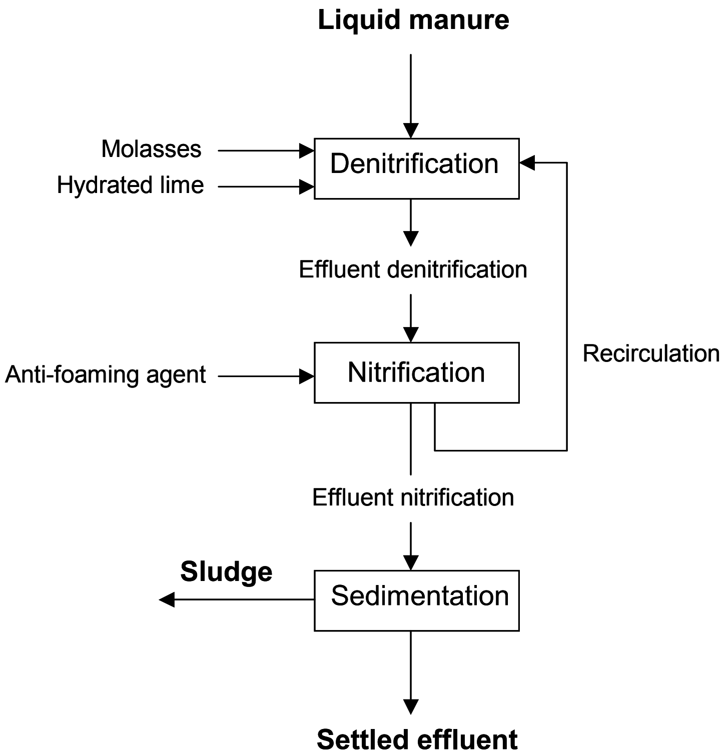


Figure 7.3 System 3: schematic of the biological nitrification / denitrification system used for treatment of liquid pig manure.

7.2.5 System 4: mechanical separation, evaporation, and condensation

The fourth system that was studied consisted of a mechanical separation step followed by evaporation and subsequent condensation in a mechanical vapour recompression (MVR) unit (Figure 7.4). A mixture of liquid manure from sows and fattening pigs (14,000 t y^{-1}) was pumped from a storage tank to a decanter centrifuge (Pieralisi; capacity: 5 - 6 m^3 h^{-1}; 11 kW). The solid fraction from the centrifuge was stored and might be subsequently composted. The temperature of the liquid fraction was raised in two subsequent heat exchangers with the heat that was generated in the MVR unit. The liquid then entered a degasser, in which ammonia and carbon dioxide were removed, and finally entered an evaporator. After cooling down the condensate from the MVR unit in heat exchanger 1, ammonia was removed by air stripping and scrubbing with sulphuric acid so that

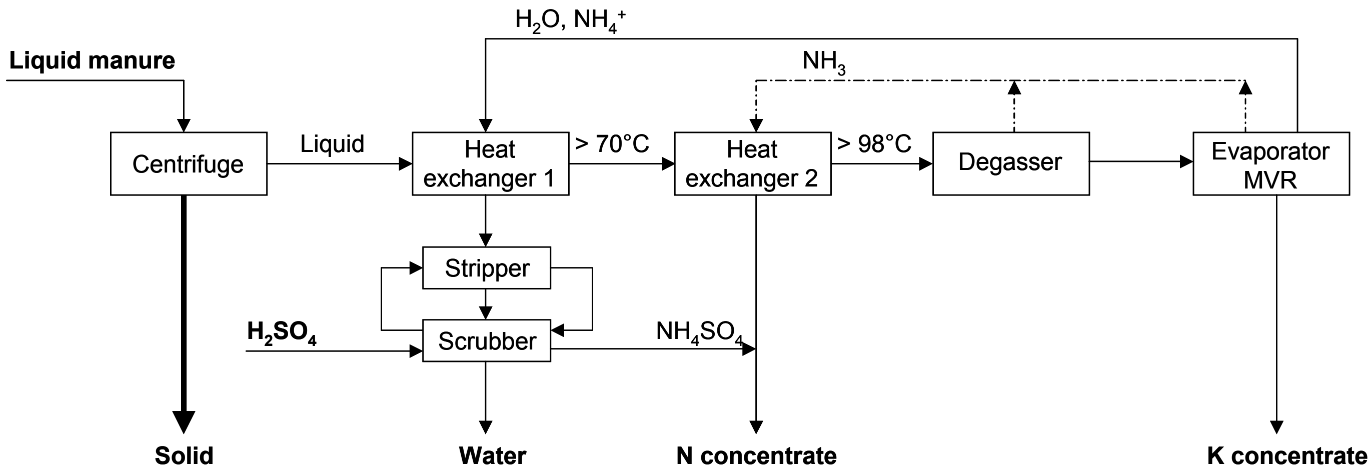


Figure 7.4 System 4: schematic of the system for mechanical separation, evaporation, and condensation of liquid pig manure; MVR, mechanical vapour recompression; – · – · –, vapour; ———, liquid; ▬▬▬, solid.

water and ammonium sulphate solution remained. The latter was mixed with the N concentrate that had been produced by condensation of ammonia vapour in heat exchanger 2. After evaporation a liquid NPK concentrate remained. The evaporation and condensation apparatus were placed in a closed unit in order to reduce emissions. The exhaust air from this unit (6 to 10 $m^3 h^{-1}$) was drawn through an active carbon filter to reduce the odour emission. This manure treatment system, excluding the centrifuge, was manufactured by Funki Manura, Denmark.

7.2.6 Measurements

Composition and flow of solids and liquids

Once a week a 1 *l* sample was taken from the input and output material of each system. The liquid manure was sampled directly from the pipe that supplied the manure to the treatment system and each product was sampled from a storage tank that contained the production of one week and was emptied afterwards. Sample analyses were performed according to standard methods (NNI, 1988). The following analyses were carried out: total solids (TS), total volatile solids (TVS), total phosphorus (TP), total Kjeldahl nitrogen (TKN), total ammoniacal nitrogen (TAN), nitrite (NO_2-N), nitrate (NO_3-N), total-potassium (TK), and pH. In order to be able to calculate discharge levies, for liquids concerned the chemical oxygen demand (COD), biological oxygen demand (BOD), and electrical conductivity (EC) was measured.

For system 1, the flow rate of the incoming liquid pig manure was determined by measuring the level of the manure storage tank at the start and the end of the experiments. For system 2, the flow rate of the incoming liquid pig manure could not be measured, as the liquid that enters the system already is a mixture of liquid manure and flushing liquid. Therefore this flow was calculated as the sum of the product flows of system 2. For system 3, the flow rate of the incoming liquid pig manure was measured continuously as the peristaltic pump was equipped with a pulse counter; every pulse equalled a known volume. For system 4, the flow rate of the incoming liquid pig manure was continuous measured with an electromagnetic flow meter. The flow rates of the liquid and solid products were determined by weighing the materials that had been collected; for system 3, however, the mass flows of the sludge and the settled effluent were not measured.

Gaseous emissions

The emission of ammonia (NH_3), carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O), and odour was measured twice. The first measurement was made during the evaluation period of four weeks; the second measurement was carried out within two months after the first. The sampling period was 2 to 4 hours for each single measurement thus time-averaged concentrations were determined. The measurements were corrected for the background concentrations measured upwind from the installation. The outside temperature and the temperature of the sampled air were measured.

The ammonia concentration in the air was determined by drawing air (120 *l* h^{-1}) through two gas washing bottles filled with nitric acid, in series (Van Ouwerkerk, 1993). Ammonia is trapped by the acid and accumulates in the bottles. Fluctuations in the ammonia 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 gas washing bottle, which serves as a control, does not contain more than 5% of the amount of ammonia trapped in the first gas washing bottle. By determining the ammonia content in the acid solution spectrophotometrically (NNI, 1988) the average ammonia concentration in the air was calculated.

Odour samples were collected in Teflon odour bags (60 *l*) that were initially evacuated. Each bag was placed in an airtight container and filled in 2 hours by creating an underpressure in the surrounding container. The sampling system was equipped with a heating system to prevent condensation and a filter to prevent the intake of dust. Odour concentrations were determined in compliance with the European olfactometric standard method EN13725 and expressed in European odour units per m^3 air (OU_E m^{-3}) (CEN, 2003).

The emission of gases such as CH_4, N_2O, and CO_2 contributes to the enhanced greenhouse effect. Air samples for the determination of CH_4, N_2O, and CO_2, were taken using an initially evacuated canister which was filled through a critical orifice. Concentrations were determined using a gas chromatograph (Carbo Erba Instruments, GC 8000 Top; column: Molsieve 5A (CH_4, CO_2), Haysep (N_2O); detector: FID/HWD (CH_4); ECD/HWD (N_2O), HWD (CO_2)). The global warming potential, expressed as carbon dioxide equivalents (CO_2 equiv), was calculated by multiplying the emission of CH_4 and N_2O with their respective global warming potential (GWP) values, *viz* 23 and 296 (on mass basis; time horizon of 100 years), and adding them (IPCC, 2001). Although CO_2 is a greenhouse gas as well, its emission from manure was neglected. The reason for neglecting CO_2 in this case is that this CO_2 does not originate from combustion of fossil fuels but from bacterial conversion of organic matter, which in turn has recently been assimilated from atmospheric CO_2. As such, this CO_2 is part of the short-term organic carbon cycle and does not contribute to the enhanced greenhouse effect.

7.3 Results and discussion

7.3.1 Composition and application of manure products

The different treatment techniques result in many products with widely varying composition. The average composition of the untreated manure and the products are listed in Table 7.1 for each manure treatment system. The products can be divided into several groups:

(1) solid manures - high TS and TP content,
(2) concentrates - high TKN, TAN or TK content,
(3) water - low TKN, TP and TK content, and
(4) miscellaneous compositions.

Every manure treatment system produces one solid product, which can be classified under solid manures. Products from this group can be applied in autumn for soil improvement because of the high TS content and for raising the phosphorus content of the soil.

Group 2 consists of three concentrates. The concentrate produced by system 2 is a TKN concentrate that can be applied in autumn when the need for a high nitrogen availability is limited. The N concentrate produced by system 4, with a high TAN content, can be applied during or just before the growing season when the crop needs a high nitrogen availability. The K concentrate produced by system 4, which also contains some P and N, can be used for moderate N and P fertilization on soils where high K doses are no problem.

Group 3 consist of the water that is produced by system 1 and 4. These liquids have low mineral contents and can be used for irrigation or may be discharged to the public sewer system.

The products in group 4 have various compositions. The possibilities for using the filtrate from system 1 and the permeate from system 2 depend on specific crop and soil needs. The composition of the sludge from system 1 is almost equal to the composition of the fresh manure, so this sludge can easily be recirculated by re-entering the system along with the fresh liquid manure. The settled effluent from system 3 was discharged to the public sewer system.

Alternatively, the compositions of the liquid fraction from system 1 and the permeate from system 2 could have been obtained by the use of a mechanical separator with flocculent addition (Van der Kaa & Den Brok, 1997).

In Table 7.2 the mass flows of the manure products and the distribution of nutrients (N, P, and K) are given. Due to errors in flow measurements, sampling, and analyses, the amount of nutrients that was found in the products never equalled 100%. In most cases, however, the error in the mass balance is below 10%; for system 2 and system 4 the error is even below 5%.

Although system 1 is designed to realise a substantial reduction of the manure volume, the amount of condensate produced is only 0.4% (Table 7.2). In summertime, however, the evaporation and condensate production rate can be eight to ten times higher (Huijben & Van Wagenberg, 1998). It is unclear why the error in the P balance of system 1 is relatively high, *viz* 16%.

For system 3, the nitrogen balance of Table 7.2 shows that 41% of the nitrogen was removed by the nitrification / denitrification process. The process appears to be hampered, however, as for comparable systems effluent TKN values were found which are five times lower than in this study (Feyaerts *et al.*, 2002). It is known that low nitrogen removal can be caused by insufficient aeration, as oxygen serves as electron donor for nitrification, or by low temperature, which decreases microbial activity. At the time of the research the average outside temperature was 5 to 10°C (winter time). It is necessary to do further experiments in order to find out which effect prevailed in this study. An increase of the nitrogen removal efficiency has a significant economic effect if the effluent is discharged to the public sewer system on a levy basis. Biological treatment of liquid waste streams and its underlying processes are described extensively in literature (Burton & Turner, 2003; Tchobanoglous, 1991). Furthermore, the phosphorus balance of system 3 shows a large error. In contrast with potassium, which is well soluble, the phosphorus is present in an insoluble form, either as precipitated calcium phosphate or bound to the organic matrix of the solid particles in the manure. Settlement of these phosphates in the denitrification tank will probably have led to unrepresentative sampling of the liquid in the tank. It is unclear what causes the large error in the K balance of the effluent of the denitrification tank. As the recirculation flow between the nitrification and denitrification tank is high in comparison to the fresh manure input (10 : 1), and no K is removed by the bacterial processes, the K content of the effluent of the nitrification and denitrifiction tank are expected to be equal.

7.3.2 Gaseous emissions

The gaseous emissions that were measured for each manure treatment system are presented in Figure 7.5. The emission of each component shows a large variation between the different systems.

Ammonia emission is mainly determined by the TAN content and pH of the material, the size of the air / liquid contact area, the ventilation rate, and the temperature (Aarnink & Elzing, 1997; Monteny & Erisman, 1998). Although an acid

Table 7.1 Composition of liquid manure and of products after treatment for four treatment systems for liquid pig manure.[a]

	Total solids (TS) g kg^{-1}	Total volatile solids (TVS) g kg^{-1}	Total phosphorus (TP) g kg^{-1}	Total Kjeldahl nitrogen (TKN) g kg^{-1}	Total ammoniacal nitrogen (TAN) g kg^{-1}	Total potassium (TK) g kg^{-1}	pH	NO_2-N mg l^{-1}	NO_3-N mg l^{-1}
System 1, straw filtration									
Liquid manure	61 ± 11	42 ± 11	1.8 ± 0.2	7.0 ± 0.5	3.5 ± 0.5	5.5 ± 0.5	8.5 ± 0.2		
Straw[b]	637	602	1.5	9.8		18	6.7		
Water	<0.1	< 0.1	<0.001	0.56 ± 0.04	0.46 ± 0.05	<0.001	8.6 ± 0.3		
Solid[c]	181 ± 12	143 ± 1.3	5.0 ± 0.2	8.9 ± 1.6	3.2 ± 0.02	4.6 ± 0.4	8.3 ± 0.3		
Liquid[d]	19 ± 0	8.0 ± 2.6	0.4 ± 0.3	4.7 ± 0.8	2.7 ± 0.4	4.9 ± 0.5	8.4 ± 0.2		
Sludge[e]	53 ± 21	33 ± 19	1.7 ± 0.6	6.5 ± 0.8	3.8 ± 0.2	4.8 ± 0.2	8.3 ± 0.05		
System 2, mechanical separation									
Liquid manure	51 ± 22	34 ± 18	1.2 ± 0.6	5.1 ± 1.2	3.5 ± 0.6	4.7 ± 0.7	7.4 ± 0.2		
Solid fraction	283 ± 7	223 ± 23	5.8 ± 3.1	9.9 ± 3.4	4.0 ± 0.2	3.6 ± 1.6	7.9 ± 1.0		
Concentrate	81 ± 10	60 ± 7	1.2 ± 0.2	7.8 ± 0.6	4.0 ± 0.2	4.9 ± 0.2	7.7 ± 0.2		
Permeate[f]	18 ± 3	8.4 ± 1.8	<0.2	3.4 ± 0.4	3.1 ± 0.3	4.6 ± 0.5	7.8 ± 0.2	<0.3	<0.5
System 3, nitrification / denitrification									
Liquid manure[g]	53 ± 29	36 ± 21	1.5 ± 1.0	4.4 ± 0.9	2.9 ± 0.4	3.1 ± 0.2	7.9 ± 0.05	0.32 ± 0.15	1.4 ± 0.5
Effluent denitrification	25 ± 4	15 ± 3	0.5 ± 0.1	2.6 ± 0.4	1.8 ± 0.3	2.4 ± 1.3	8.6 ± 0.05	0.15 ± 0.10	1.1 ± 0.2
Effluent nitrification[h]	25 ± 3	14 ± 2	0.5 ± 0.1	2.6 ± 0.4	2.0 ± 0.6	2.8 ± 0.3	8.5 ± 0.1	0.13 ± 0.06	1.1 ± 0.1
Settled effluent[i]	9.1 ± 0.4	3.2 ± 0.3	0.028 ± 0.003	0.41 ± 0.07	0.22 ± 0.15	2.5 ± 0.3	8.2 ± 0.1	52 ± 15	135 ± 34
Sludge[b]	245 ± 41	126 ± 13	7.1 ± 5.5	7.2 ± 1.8	4.1 ± 2.4	3.0 ± 0.2	8.5 ± 0.2	0.25 ± 0.12	1.8 ± 0.18

(table continues on next page)

(Table 7.1 continued)

	Total solids (TS) g kg^{-1}	Total volatile solids (TVS) g kg^{-1}	Total phosphorus (TP) g kg^{-1}	Total Kjeldahl nitrogen (TKN) g kg^{-1}	Total ammoniacal nitrogen (TAN) g kg^{-1}	Total potassium (TK) g kg^{-1}	pH	NO_2-N mg l^{-1}	NO_3-N mg l^{-1}
System 4, evaporation									
Liquid manure	50 ± 2.9	28 ± 2.4	1.1 ± 0.1	5.2 ± 0.2	3.9 ± 0.08	5.1 ± 0.08	8.2 ± 0.05		
Solid	356 ± 13	218 ± 7.8	11.7 ± 0.3	9.2 ± 0.6	3.9 ± 1.0	4.4 ± 0.12	6.1 ± 0.3		
Water[j]	0.3 ± 0.2	0.2 ± 0.2	<0.03	0.34 ± 0.17	<1	<0.2	9.4 ± 0.3	<3	<20
N concentrate[k]	60 ± 18[d]	57 ± 16[d]	<0.03	130 ± 6.2	118 ± 3.4	<0.02	9.9 ± 0.1	<3	<20
K concentrate[l]	103 ± 7.2	51 ± 3.9	0.88 ± 0.06	3.9 ± 0.4	<1	20.3 ± 2.1	9.8 ± 0.1	9.8 ± 6.8	<20

[a] Values represent averages, followed by the standard deviation; n = 4, unless otherwise indicated.
[b] n = 1.
[c] n = 2.
[d] n = 3.
[e] From each settling tank, one sample was taken and analysed.
[f] chemical oxygen demand (COD) = 48 ± 57 g l^{-1}, n = 3; biological oxygen demand (BOD) = 22 ± 57 g l^{-1}, n = 2; electrical conductivity (EC) = 49 ± 16 mS cm^{-1}, n = 3.
[g] COD = 45 ± 27 g l^{-1}.
[h] Unsettled; COD = 16 ± 2 g l^{-1}.
[i] COD = 3.1 ± 0.1 g l^{-1}; EC = 10 ± 0.3 mS cm^{-1}.
[j] COD = 92 ± 15 mg l^{-1}.
[k] EC = 552 ± 10 mS cm^{-1}.
[l] EC = 68 ± 10 mS cm^{-1}.

Table 7.2 Mass balance and distribution of nutrients over the products of four treatment systems for liquid pig manure (including ammonia emission).

	Mass (% of input)[a]	N (% of input)[b] [c]	P (% of input)[b]	K (% of input)[b]
System 1, straw filtration; 16,000 t [manure] y^{-1}				
Water	0.4	<0.1	<0.1	<0.1
Solid	17	15	50	15
Liquid	65	61	16	61
Sludge	18	16	18	16
Ammonia emission		0.8		
Total of system 1	100	93	84	95
System 2, mechanical separation; 36,000 t [manure] y^{-1}				
Solid fraction	15	29	74	11
Concentrate	13	20	14	13
Permeate	72	48	9.5	70
Ammonia emission		0.3		
Total of system 2	100	97	97	95
System 3, nitrification / denitrification; 3,000 t [manure] y^{-1}				
Effluent denitrification	100	59	31	78
Effluent nitrification[d]	100	59	32	93
Ammonia emission		0.1		
Total of system 3	100	59	32	93
System 4, evaporation ; 14,000 t [manure] y^{-1}				
Solid	7.5	13	81	6.5
Water	67	4.4	2.1	0.9
N concentrate	2.7	67	0.1	<0.1
K concentrate	23	17	21	98
Ammonia emission		<0.1		
Total of system 4	100	103	104	105

[a] The mass flows of total input and output were set to 100%. [b] the nutrient composition of the starting material was set to 100%. [c] if N-NO_2^- and N-NO_3^- values were known, the N balance was calculated as the sum of TKN and N-NO_2^- and N-NO_3^-; otherwise only TKN values were used. [d] Before entering the settling basin; because the mass flow ratio of sludge over settled effluent was not measured, the settling basin is not included in the balance.

scrubber reduces the ammonia emission by 77%, system 1 still has the highest ammonia emission of all four systems (0.8% of the TKN content of the treated manure, Table 7.2). This can be explained by the manure spraying process that enhances contact between manure and the air, the large air / liquid contact area that is provided by the straw filter bed, and the relatively high temperature in the greenhouse which is about 30°C while the outside temperature is only 15°C. Laboratory experiments showed that in addition some ammonia diffusion and leakage through the foil occurred. Calculations showed, however, that this ammonia transport is negligible in comparison with the air that leaves the greenhouse with the mechanical ventilation system. In each process step of

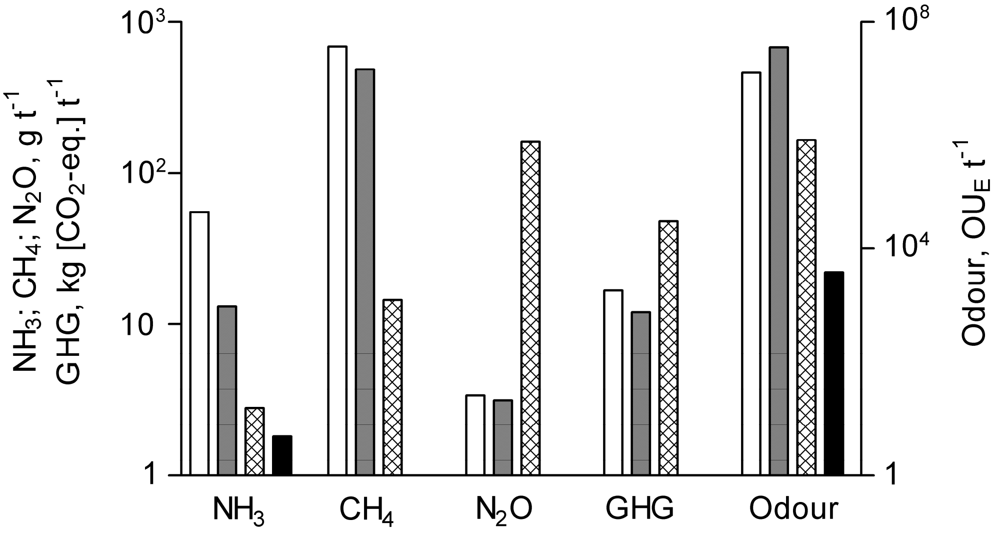


Figure 7.5 Average emission of NH_3, CH_4, N_2O, greenhouse gases (GHG; sum of CH_4 and N_2O), and odour from four treatment systems for liquid pig manure; emissions are expressed per tonne of manure input; for system 4, only odour and NH_3 emissions were measured: ▭, system 1, straw filtration; ▒, system 2, mechanical separation; ⊠, system 3, nitrification / denitrification; ■, system 4, evaporation.

system 2, there was direct contact between the ventilation air and the manure and its products, resulting in emission of ammonia. For system 3, the emission of ammonia is much lower as the concentration of ammonia in the liquid is kept on a low level as a result of the nitrification process. The ammonia emission from system 4 is the lowest from all four systems. This is because the system had been designed as a closed system with limited air ventilation and because of the efficient way in which the ammonia is captured by the sulphuric acid in the process.

The emission of methane and nitrous oxide from liquid manure, on the other hand, is hardly influenced by conditions such as ventilation rate and size of emitting surface because the water solubility of these compounds is very low. Methane is produced by conversion of organic matter by anaerobic bacteria that are present in manure. The mechanical processes of system 1 and 2 do not influence the methane production rate and a comparable emission is expected from a liquid manure storage without any treatment. System 3, however, has a much lower methane emission because the aerobic conditions in the nitrification tank inhibit methanogenesis. The relatively high emission of nitrous oxide for system 3, which equals 2.3% of TKN, can be explained by the fact that nitrous oxide is a by-product of bacterial nitrification and denitrification. Other researchers found emissions from biological treatment of animal slurry that are even four to fifteen times higher (Burton *et al.*, 1993; Osada *et al.*, 1995; Willers, 1996). The emission of nitrous oxide from incomplete denitrification may be decreased by increasing the residence time of the denitrification reactor (Béline & Martinez, 2002) or by a better control of the molasses addition by measurement of the redox potential.

Odour emissions are influenced by the size of the air / liquid contact area, ventilation rate, temperature, manure composition, occurrence of bacterial processes, and the use of air treatment techniques. In comparison with system 1 and 2, the emission from system 3 is low, which can be explained by the fact that the microbial consortium in the nitrification reactor degrades odorous compounds at aerobic conditions. The emission of system 4 is even lower because the exhaust air is treated with an activated carbon filter.

The data presented in Figure 7.5 can be of help when trying to obtain a legal permit for the building and operation of a manure treatment installation similar to the systems presented, as governmental officials often desire an estimation of the expected environmental impact with regard to odour (nuisance) and other emissions beforehand.

7.3.3 Economic evaluation

In Table 7.3 the investment and operational costs of each manure treatment system are presented. All costs presented in this study are excluding value added tax. The investment costs include additional costs such as necessary infrastructure, road paving, and extra storage facilities. The operational costs of the manure treatment systems, thus including variable and fixed costs, vary from 7.3 to 17.2 € t^{-1} of treated manure. These costs should be covered by the economical benefits of manure treatment.

For the studied manure treatment systems, the following economical benefits can be distinguished:

(a) The combined use of solid manures with a high P content (group 1, system 1 to 4) and of concentrates with a high N content (group 2, system 2 and 4), enables landowners to apply N and P at a ratio of their choice instead of using liquid manure with a fixed N to P ratio. In this way the fertilizing program can be adjusted to the crop needs so that overfertilization is prevented and plant production increases.

Table 7.3 Investment and operational costs of four manure treatment systems (excluding value added tax).[a]

	Costs, € t^{-1} [manure][b]			
	System 1 straw filtration 16,000 t y^{-1}	*System 2* mechanical separation 36,000 t y^{-1}	*System 3* nitrification / denitrification 3,000 t y^{-1}	*System 4* evaporation 14,000 t y^{-1}
Investment costs	25.0	42.3	39.8	71.9
Operational costs:				
Depreciation[c]	2.50	5.34	5.12	8.62
Maintenance[d]	0.75	1.27	1.20	2.16
Interest[e]	0.69	1.16	1.10	1.98
Electricity[f]	0.22	1.67	1.67	3.35
Diesel oil[g]	0.09	n/a	n/a	n/a
Chemicals	0.15	n/a	0.64	0.74
Miscellaneous	0.52[h]	0.56[i]	n/a	0.03[j]
Labour[k]	2.34	1.83	0.55	0.35
Total operational costs	*7.3*	*11.8*	*10.3*	*17.2*

[a] The presented costs are based on actually built and operated manure treatment faculties. More details can be found in Melse *et al.*, 2002a - 2002e. [b] based on 8,000 running hours y^{-1}. [c] depreciation time (linear; residual value is 0): 7.5 years for machinery; 10 years for buildings, tanks, infrastructure *etc.* [d] 3% of investment. [e] 5.5% of net present worth. [f] 0.06 € kWh^{-1}. [g] 0.68 € l^{-1}. [h] straw, 110 € t^{-1}. [i] membrane replacement. [j] active carbon filter replacement. [k] 18 € h^{-1}.

(b) In the Netherlands, liquid pig manure is often transported to other regions of the country which means expensive transport over a distance of 200 km. The benefit of system 1, 3, and 4 is that the manure volume is reduced by production of a water fraction with a low N, P or K content. This water fraction is applied close to the farm so only a small volume of the processed manure has to be transported to other regions.

However, for the farmers involved in this study it appeared to be hard to reduce the disposal costs of manure products in comparison with their previous disposal of untreated manure. The reasons for this are as follows:

(a) Many landowners in the Netherlands are used to receive a bonus when they use untreated manure for land application, so it is difficult to convince them to pay for manure products as long as they can alternatively use untreated manure.

(b) Many landowners in the Netherlands are accustomed to liquid manure application; if they start to apply solid manure products they have to invest in new manure spreading equipment.

(c) Due to the small scale of manure treatment, each systems is applied on one farm only, the supply of manure products is relatively small as compared to the supply and demand on the market of untreated liquid manure and chemical fertilizer. It is difficult to operate on such a market as traders are used to large product amounts. This is especially the case for concentrates (product group 2), which are produced in relatively small volumes, although the composition of *e.g.* a nitrogen concentrate can very well compete with chemical nitrogen fertilizer.

Furthermore, the costs of off-farm disposal of untreated manure have varied from 8 to 18 € t^{-1} in recent years. Banks and farmers do not consider this fluctuation to be a solid base for investment in manure treatment, as manure treatment systems have to be competitive with off-farm disposal costs.

In 2006, new regulations on the production and use of manure and minerals will be introduced in the Netherlands in order to comply with the Nitrates Directive (EEC, 1991) and standards for land application of nitrogen and phosphorus will become stricter. It is expected that the costs of manure disposal as a result of this will stabilize at a range from 13 to 15 € t^{-1} (De Hoop *et al.*, 2004). This will increase the economic feasibility of the manure treatment systems described in this study.

7.4 Conclusions

Using four different farm-scale treatment systems, liquid pig manure was processed into a wide range of products that had different dry matter and nutrient (P, N, K) compositions. The products were divided into four groups: solid manures, concentrates, water and miscellaneous products. The emission of ammonia varied from 1.8 to 55 g t^{-1} [manure] and the emission of odour from 3.8×10^3 to 1.3×10^7 [European odour units] t^{-1} [manure]. The nitrification / denitrification system showed the highest emission of greenhouse gases (48 kg [carbon dioxide equivalents] t^{-1} [manure]), mainly nitrous oxide (N_2O), whereas the emission of the other systems was 12 to 17 kg [CO_2-equiv] t^{-1} [manure]. The operational costs for manure treatment, including variable and fixed costs, varied from 7 to 17 € t^{-1} (excluding value added tax).

The critical success factor for the operation of these four manure treatment installations turned out to be not of technical but of economical nature. To have a treatment system that is cost-effective in comparison with the disposal of untreated manure, manure treatment costs must be covered by the sale or the lower disposal costs of the manure products. It is therefore concluded that a sound assessment of the market prospects of the products is the main criterion for choosing a specific manure treatment system with its specific product compositions. As market prospects differ from region to region and from country to country, no generally preferred manure treatment technique can be pointed out from this study, as local market circumstances must be taken into account. After such an assessment has been carried out for a specific farm situation, however, one of the available manure treatment system can be chosen for application.

Acknowledgements

The authors would like to thank all farmers that cooperated in the research: P. De Swart, B. Dirven, E. Evink, and J. Lavrijsen. Also we like to thank P. De Gijsel, G. Kupers, and J. Slangen for carrying out most measurements and analyses. The work was funded by the Dutch Product Boards for Livestock, Meat and Eggs.

References

Aarnink, A.J.A.; Elzing, A. (1997). Dynamic model for ammonia volatilization in housing with partially slatted floors, for fattening pigs. Livestock Production Science, 53, 153-169.

Béline, F.; Martinez, J. (2002) Nitrogen transformations during biological aerobic treatment of pig slurry: effect of intermittent aeration on nitrous oxide emissions. Bioresource Technology, 83, 225-228.

Burton, C.H.; Sneath, R.W.; Farrent, J.W. (1993). Emissions of nitrogen oxide gases during aerobic treatment of animal slurries. Bioresource Technology, 45, 233-235.

Burton, C.H.; Turner, C. (2003). Manure management. Treatment strategies for sustainable agriculture. Silsoe Research Institute, Silsoe, UK, ISBN 0 9531282 6 1.

CEN (2003). Air quality - Determination of odour concentration by dynamic olfactometry, EN13725, European Committee for Standardization, Brussels, Belgium.

De Hoop, D.W.; Luesink, H.H.; Prins, H.; Daatselaar, C.H.G.; Van Bommel, K.H.M.; Mokveld, L.J. (2004). Effecten in 2006 en 2009 van Mestakkoord en nieuw EU Landbouwbeleid [Effects in 2006 and 2009 of the Manure Agreement and new EU agricultural policy] Report 6.04.23, LEI, The Hague, Netherlands, ISBN 90 5242 963-4.

EEC (1991). Council Directive of 12 December 1991 concerning the protection of waters against pollution caused by nitrates from agricultural sources (91/676/EEC), European Council, Brussels, Belgium.

Feyaerts, F.; Huybrechts, D.; Dijkmans, R. (2002) Beste Beschikbare Technieken voor mestverwerking [Best Available Techniques for manure treatment], 2nd ed., Academia Press, Gent, Belgium, XXVI + 350 pp, ISBN 90 382 0434 5.

Huijben, J.J.H.; Van Wagenberg, A.V. (1998). Verdamping van water uit dierlijke mest met behulp van zonne-energie [Evaporation of water from slurry using solar radiation.] Report P1.213, Praktijkonderzoek Varkenshouderij, Rosmalen, Netherlands.

IPCC (2001). Climate Change 2001: The scientific basis. Contribution of working group I to the third assessment report of the Intergovernmental Panel on Climate Change (Houghton J T; Ding Y; Griggs D J; Noguer M; Van der Linden P J; Dai X; Maskell K; Johnson C A eds) Cambridge University Press. Cambridge, U.K., 881 pp, ISBN 0521 80767 0 and ISBN 0521 01495 6.

Kalyuzhnyi, S.; Sklyar, V.; Rodriguez-Martinez, J.; Archipchenko, I.; Barboulina, I.; Orlova, O.; Epov, A.; Nekrasova, V.; Nozhevnikova, A.; Kovalev, A.; Derikx, P.; Klapwijk, A. (2000). Integrated mechanical, biological and physico-chemical treatment of liquid manure streams. Water Science and Technology, 41 (12), 175-182.

Melse, R.W.; Gijsel, de, P. (2001). Systems for pig manure treatment in the Netherlands. ASAE Annual International Meeting, July 30-August 1, 2001, Sacramento, California, USA, paper 01-2239.

Melse, R.W.; Starmans, D.A.J.; Verdoes, N. (2002a). Mestverwerking varkenshouderij. Manura® 2000, Hollvoet te Reusel. [Pig manure treatment. Manura® 2000, Hollvoet in Reusel, Netherlands.] Praktijkboek, 4, Praktijkonderzoek Veehouderij, Lelystad, Netherlands.

Melse, R.W.; Starmans, D.A.J.; Verdoes, N. (2002b). Mestverwerking varkenshouderij. Manura® 2000, Houbensteyn te Ysselsteyn. [Pig manure treatment. Manura® 2000, Houbensteyn in Ysselsteyn, Netherlands.] Praktijkboek, 5, Praktijkonderzoek Veehouderij, Lelystad, Netherlands.

Melse, R.W.; Starmans, D.A.J.; Verdoes, N. (2002c). Mestverwerking varkenshouderij. Systeem Biovink, Evink te Oosterwolde (Gld). [Pig manure treatment. Biovink system, Evink in Oosterwolde (Gld), Netherlands.] Praktijkboek, 6, Praktijkonderzoek Veehouderij, Lelystad, Netherlands.

Melse, R.W.; Starmans, D.A.J.; Verdoes, N. (2002d). Mestverwerking varkenshouderij. Mestscheiding en microfiltratie, Dirven te Someren. [Pig manure treatment. Separation and microfiltration, Dirven in Someren, Netherlands.] Praktijkboek 7, Praktijkonderzoek Veehouderij, Lelystad, Netherlands.

Melse, R.W.; Starmans, D.A.J.; Verdoes, N. (2002e). Mestverwerking varkenshouderij. Strofilter in foliekas, De Swart te Alphen (NB). [Pig manure treatment. Straw filtration in a greenhouse, De Swart te Alphen (NB), Netherlands.] Praktijkboek, 8, Praktijkonderzoek Veehouderij, Lelystad, Netherlands.

Monteny, G.J.; Erisman, J.W. (1998). Ammonia emission from dairy cow buildings: A review of measurement techniques, influencing factors and possibilities for reduction. Netherlands Journal of Agricultural Science, 46(3/4), 225-248.

NNI (1988). Overview of standards for analysis of water and sludges (In Dutch). Nederlands Normalisatie Instituut, Delft, Netherlands, 26 pp.

NNI (1996). NVN 2820/A1. Luchtkwaliteit, sensorische geurmetingen met behulp van een olfactometer, Wijzigingsblad NVN 2820/A1. Nederlands Normalisatie-instituut, Delft, Netherlands

Osada, T.; Kuroda, K.; Yonaga, M. (1995) Reducing nitrous oxide gas emissions from fill-and-draw type activated sludge process. Water Research, 29(6), 1607-1608.

PDV (1994). Het HELERBO strofilter. [The HELERBO straw bed filter.] Final report SPOM project 94.094, Product Board Animal Feed (PDV), The Hague, Netherlands.

Priem, R. (1980). De strofiltratie van mengmest, onder dak uitgevoerd. Resultaten van het onderzoek en economische beschouwingen. [Straw filtration of liquid manure under a roof. Results and economic evaluation.] Landbouwtijdschrift, 33(4), 737-751.

Satter, I.H.G.; Hol, J.M.G.; Huis in 't Veld, J.H.W.; Groenestein, C.M. (1997) Vleesvarkensstal met mestverwijdering door spoelen met dunne mestfractie via spoelgoten. Report 97-1004. Praktijkonderzoek naar de ammoniakemissie van stallen XXXIV. IMAG-DLO, Wageningen, Netherlands.

Tchobanoglous, G. (1991). Wastewater engineering: treatment, Disposal, and Reuse / Metcalf & Eddy Inc.. Third edition, revised by G. Tchobanoglous and F.L. Burton. McGraw-Hill Book Co, 1334 pp, ISBN 0 07 100824 1.

Van der Kaa, C.C.R.; Den Brok, G.M. (1997) Concentrering fosfaat uit varkensmest met een decanteer-centrifuge. [Concentrating phospate from pig manure with a decanting centrifuge]. Report P 3.143. Praktijkonderzoek Varkenshouderij, Rosmalen, Netherlands.

Van Ouwerkerk, E.N.J. (1993). Meetmethoden NH_3-emissie uit stallen. Werkgroep Meetmethoden NH_3-emissie uit stallen. [Methods to measure NH_3-emission from animal houses.] Onderzoek inzake de mest- en ammoniakproblematiek in de veehouderij 16, DLO, Wageningen, Netherlands.

Willers, H.C.; Derikx, P.J.L.; Ten Have, P.J.W.; Vijn, T.K. (1996). Emission of ammonia and nitrous oxide from aerobic treatment of veal calf slurry. Journal of Agricultural Engineering Research, 63, 345-352.

Chapter 8

General discussion

Contents

8.1 Introduction

As described in chapter 1, the aim of this thesis is to better understand and improve the performance of air treatment techniques in livestock operations where these systems are applied at mechanically ventilated animal houses. The first three objectives, as defined in section 1.3, have been addressed in the previous chapters that are summarized in section 8.2

The fourth objective, an analysis of the problems and shortcomings related to current air scrubbing practices, will be addressed now. The topic is discussed in an integrated manner and recommendations are made for possible process improvement and future research. Section 8.3 discusses in more detail the performance and control of air scrubbers and section 8.4 comments upon various system concepts and their implications. Finally, in section 8.5 current and possible future developments are discussed.

8.2 Summary of previous chapters

In chapter 1 and 2 ("Introduction" and "Policy on emission control related to livestock production") it has been discussed that the increasingly stringent emission standards that animal production has to comply with require drastic emission reductions. Target pollutants include ammonia, odour, particulate matter, and greenhouse gases (methane and nitrous oxide). It is therefore important that suitable abatement technologies are available.

In chapter 3 ("Air scrubbing techniques for ammonia and odor reduction at livestock operations: Review of on-farm research in the Netherland") the performance of on-farm operated air scrubbers has been reviewed and discussed. The main conclusions from this review are that bioscrubbers often experience operational problems (poor process control), that ammonia removal efficiency by scrubbers is relatively high (on average 96% for acid scrubbers and 70% for bioscrubbers), that odour removal is relatively low (on average 31% for acid scrubbers and 44% for bioscrubbers), and that bioscrubbers often show a large variation in odour removal efficiency.

In chapter 4 ("Size reduction of ammonia scrubbers for pig and poultry houses: Use of conditional bypass vent at high air loading rates") a novel scrubber design has been presented which comprises a partial air cleaning control system. Currently, scrubbers are designed and built to be able to treat the maximum airflow rate that may occur throughout the year. As a result, for most of the time air scrubbers are overdimensioned and underloaded. The presented partial air cleaning strategy, which uses an air flow controlled bypass vent that opens at high air loading rates, increases the efficiency of scrubber utilization and thus reduces mitigation costs, if removal efficiencies remains unaffected. Model calculations showed that a reduction of scrubber volume by *e.g.* 50% still enables treatment of 80 - 90% of the ammonia load throughout the year, which means that cost-optimization, *i.e.* reduction of operational cost per kg NH_3 load treated, is possible.

In chapter 5 ("Biofiltration for mitigation of methane emission from animal husbandry") it was shown in an experimental biofilter setup that biological oxidation of methane is possible, although the low water solubility of methane limits the degradation capacity[1]. Based on the found first-order rate constant a model was presented which shows the influence of concentration and air residence time on removal efficiency. Either relatively high methane concentrations (several grammes per m^3) or high air residence times (several minutes) are necessary for high removal efficiency at biofilter sizes that are considered achievable. This is why methane removal in common scrubber systems (air residence time in the order of seconds) that treat methane containing ventilation air from animal houses (methane concentration in the order of milligrammes per m^3) is expected to be negligible.

Finally, in chapter 6 and 7 ("Odour and ammonia removal from pig house exhaust air using a biotrickling filter" and "Evaluation of four farm-scale systems for the treatment of liquid pig manure") two case-studies were presented. In the first case-study the field performance of a specific biotrickling filter operated on a pig farm was described and discussed. It was found that the bioscrubber was not well operated, while no one was aware of that, and implies that poor performance of bioscrubbers might be a common problem which corresponds to some findings from chapter 3. This stresses the need for improvement of process control and monitoring by concerning parties (manufacturers, end-users, authorities). The second case-study is an evaluation of the technical performance and economics of several liquid manure treatment systems and includes an assessment of the gaseous emissions from these treatment systems. This study illustrates that within livestock farming scrubbers can also be used in application areas other than treatment of animal house ventilation air, by presenting several liquid manure treatment system that comprise an acid ammonia scrubber. Especially acid scrubbers might be used for effective abatement of ammonia emissions from such batch processes as this type of scrubber can be easily switched on and off without

[1] Whereas most work presented in this thesis deals with scrubbers, either acid scrubbers or bioscrubbers, in this chapter a biofilter was tested. Please see the definition in section 1.2.3 where this distinction is explained.

efficiency loss, in contrast to bioscrubber systems. Furthermore, the evaluation showed that emissions from manure treatment processes can be significant.

8.3 Performance and control

8.3.1 Pollutant removal

Ammonia

The ammonia removal of acid scrubbers is on average about 95% (chapter 3).[1] Apparently, current process control at acid scrubbers is sufficient to guarantee high and stable NH_3 removal efficiencies; the produced ammonium sulphate solution might be used as liquid nitrogen fertilizer.

The ammonia removal efficiency for bioscrubbers is significantly lower, on average 70% and shows a large variation (chapter 3). Although an average removal of 70% is considered normal for biotrickling filters treating animal house exhaust air at the applied air residence times and ammonia concentrations, the large variation that is found is not yet fully understood and needs further investigation. The broad range of ammonia removal efficiencies that is found, which includes occasionally measured high removal efficiencies, indicates that (average) ammonia removal efficiencies might be increased by better process control and by further optimization of the design. As adaptation of process control parameters does not affect scrubber dimensions it might be relatively easy to increase the cost efficiency of bioscrubbing for ammonia removal.

Also other techniques for ammonia removal from animal house exhaust air have been tested. An example is catalytic oxidation at high temperature (Monteny *et al.*, 1998); however, this technique was discarded because of the high costs. Recently oxidative treatment of ammonia inside an animal house was reported after UV radiation of inner walls coated with TiO_2 catalyst containing paint (Guarino *et al.*, 2008); a limitation of the system might be fouling of the reactive surface and UV absorption by the dust. However, indoor treatment techniques, such as the latter, offer some advantages over end-of-pipe treatment as no adaptations of the ventilation system are necessary (no increase of pressure drop, such as is the case for scrubbers) and no scrubber needs to be erected.

---

[1] Currently some acid scrubbers are operated with an aimed ammonia removal efficiency of just 70% as 95% removal is not always necessary. This is achieved by operation at a higher pH level, so the amount of acid that is added to the recirculating water is lower.

Odour

The acid scrubbers and biotrickling filters that are currently applied in livestock operations have been designed and optimized for the removal of NH_3 only; up to a few years ago the removal of odour by scrubbers has been considered as an unintentional, but welcome, circumstance in the Netherlands. However, currently there is a need for much higher odour removal potential than in the past as livestock facilities are more and more affected by odour regulations (see section 2.2). The design and operational strategy of air scrubber still need to be optimized for combined removal of more compounds than just ammonia, *e.g.* for odour. For example the effect of basic design parameters for ammonia removal (*e.g.* pH or air residence time) on odour removal is currently insufficiently known.

Another approach to increase odour removal is to make a multi-stage scrubbing system where each consecutive stage aims to remove one type of compounds, *e.g.* an acid scrubber for ammonia removal followed by a biotrickling filter or biofilter for further odour removal (see section 8.4.2 on multi-stage scrubbers). Other techniques that could be incorporated in a multi-stage approach include oxidative treatment with ozone, hydrogen peroxide, peracetic acid and ultraviolet radiation (Aarnink *et al.*, 2005; Jungbluth and Hartung, 2003; Koziel *et al.*, 2008) and catalytic oxidation at high temperature (Monteny *et al.*, 1998); some of these techniques might not only affect the odour emission but to some extent also the ammonia emission. However, due to the large exhaust airflows of animal houses many of these techniques are considered economically unfeasible.

For biotrickling filters it has been suggested that removal of poorly water-soluble odour components can be improved by addition of an organic solvent to the water phase, which might increase the availability of the odour component to the bacteria by enhancing mass transfer and thus the biodegradation rate (*e.g.* Césario, 1997; Van Groenestijn and Lake, 1999; Davidson and Daugulis, 2003). However, this principle has not been tested yet for odour emissions from livestock facilities.

It has been discussed (chapter 3 and 6) that the average odour removal efficiency is generally much lower than the average ammonia removal efficiency, both for bioscrubbers and acid scrubbers[1]. Furthermore, it was found that individual odour removal efficiency measurements strongly vary. The reason for the variation of the odour removal efficiency being higher than for ammonia is

[1] In this study odour concentrations have been measured by olfactometry which means that a dilution series of an air sample is tested by a human sensory panel. This results in an odour concentration which is the net result of the response of all individual odour compounds by olfactory receptors in the nasal cavity .

possibly that changes in the odour *composition* (the specific compounds that are present and their concentrations) are not fully reflected in the sensorially determined odour *concentration* levels (one value, OU_E/m^3, which reflects the olfactometric response to the sum of all compounds). In contrast with the removal of ammonia, which is easily transferred to the liquid phase and easily biodegraded, the sensorially measured removal of odour is net result of the removal of many separate odour components that each have different characteristics with regard to mass transfer and, in case of a bioscrubber, biodegradability. If, at a constant odour load, the concentration of an easily removable odour component increases in comparison with the other odour components in the air, 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 load in this study was determined olfactometrically and no odour components were measured separately, the phenomenon described here may explain the relatively large variation that was found for the odour removal efficiency. Finally, some odour compounds might be produced inside a scrubber and added to the air and thus affect the sensorially measured odour concentration level of the outlet air.

Further research into odour characteristics and the processes involved in odour removal by air scrubbers is necessary to understand the large variations and relatively low removal efficiency. A useful approach would be to combine olfactometric methods using a human panel with advanced analyses of individual compounds by gas chromatography - mass spectrometry (GC-MS). A better understanding of the odour characteristics, the identification and presence of critical odour components in exhaust air, and the principles of odour removal hopefully lead to improvement of current and development of new odour removal techniques.

Greenhouse gases

The main greenhouse gases associated with intensive livestock production are methane (CH_4) and nitrous oxide (N_2O).

Ventilation air from animal houses contains some methane, but because of the low water solubility of methane usually scrubber systems do not affect the methane concentration of the exhaust air. In chapter 5 it was shown that methane in ventilation air can be biologically degraded in a biofilter. However, high air residence times, minutes to hours, are needed to achieve successful oxidation at the relatively low methane concentration levels that are common in animal houses, whereas scrubbers for ammonia removal usually have air residence times in the order of seconds. This means that bioscrubber and biofilter systems suitable for methane removal will require very high investment costs; therefore bioscrubber and biofilter systems are not considered economically feasible for methane removal from animal house ventilation air. No low-cost treatment techniques are currently available that can be operated cost-effectively at these low methane concentrations. When scrubbers are applied at relatively low air flow rates, such as is the case in several manure treatment systems that are described in chapter 7, biofiltration for methane removal might be an interesting option, however.

Nitrous oxide might be formed as a by-product of the nitrification and denitrification processes that take place in biological water (see chapter 7) or air treatment systems (Trimborn, 2006). This means that application of bioscrubbers might increase the level of greenhouse emissions from animal houses. Especially under unfavourable or unstable process conditions there is a risk of considerable N_2O formation (Burgess *et al.*, 2002; Colliver and Stephenson, 2000; Hong *et al.*, 1993). Further research is necessary to find ways to prevent and control formation of gaseous N_2O in bioscrubbers.

Particulate matter

Although the experimental research described in this thesis did not deal with particulate matter removal by scrubbers, its potential for removal is discussed here briefly because particulate matter emission has become an important issue over the last few years (see chapter 2). Currently, PM10 and PM2.5 levels are exceeded in many places in the EU, whereas in regions with intensive agricultural activities the emission of PM from animal houses may account for a large part of the total PM emission. As mentioned in section 1.2.3, recent measurements of particulate matter removal by multi-stage scrubbers (see section 8.4.2) treating animal house exhaust air showed an average removal efficiency ranging from 62 to 93% for PM10 and from 47 to 90% for PM2.5 (Aarnink *et al.*, 2007, 2008a, 2008b; Zhao *et al.*, 2008; Ogink and Hahne, 2007). These data suggest that end-of-pipe air treatment may be of major importance for compliance with current and future PM10 and PM 2.5 standards. Therefore it is important to include the understanding and improvement of particulate matter removal by scrubbers as a research goal in current and future research. Scrubber technology may in future play an important role in reducing the particulate matter emission from animal houses, as alternative mitigation options are scarce.

8.3.2 Process control

Since scrubber technology for livestock facilities was introduced in the Netherlands about 25 years ago, the manufacturing costs of scrubbers have been reduced as manufacturers have optimized and upscaled their production processes.

Also the choice of construction materials and equipment (pumps, spray nozzles, measuring devices etc.) has been optimised to some extent. However, the pollutant removal process and control strategy itself have virtually remained unchanged, except for the fact that some treatment processes are now operated in series (see section 8.4.2. on multi-stage scrubbers).

Especially bioscrubbers experience quite often operational problems in day-to-day practice, which results in decreased ammonia removal efficiencies. Often this is caused by setting the water discharge flow rate to a (fixed) level that is lower than the minimum discharge rate that is necessary for successful ammonia removal. As a discharge is followed by addition of fresh water until the original water level is reached again, a too low discharge rate results in accumulation of inhibiting compounds (ammonium and/or nitrite), especially at high ammonia loading rates.

The operational problem of implementing a correct water discharge rate at bioscrubbers, which is illustrated by the findings in chapters 3 and 6, might be solved by introduction of a robust and cheap measure for the salt accumulation in the water, such as EC measurement (EC = Electrical Conductivity). Subsequently, the EC measurement can be used as a setpoint for automatic control of the water discharge rate. In such a system the water discharge rate is not fixed anymore but depends on the actual ammonia loading rate, as the accumulation of salts is linked to the ammonia load.

A drawback of EC measurement is that it can not distinguish between ammonium, nitrite, and nitrate. Whereas high EC values related to the build of nitrite indicate poor bioscrubber performance, high EC values related to nitrate build up do not necessarily indicate performance problems. However, if a stable EC level is achieved by controlled water discharge this would normally mean that production and discharge of these compounds is in balance. Still, it is necessary to carry out research in order to determine what EC level is desired for satisfactory process performance and how EC values are related to inhibition of ammonia removal and removal efficiencies.

In order to further reduce the amount of discharge water from bioscrubbers the water might be treated in a denitrification reactor and reused in the scrubber. During denitrification nitrate is converted to dinitrogen gas thus reducing the nitrogen content of the water. In this way the accumulation of salts is reduced which will decrease the need for discharge of the recirculation water and replacement by fresh water. Currently, systems where a scrubber for treatment of animal house exhaust air is combined with a denitrification system for the discharge water are in an experimental stage and are being tested on several farms.

Another possible technique to reduce the amount of discharge water is to concentrate the discharge water, *e.g.* by using membrane technology. The generated concentrate has a relatively small volume and might be removed from the farm as a liquid nitrogen fertilizer. The generated permeate has a relatively large volume and can be reused in the scrubber or used for irrigation.

By reducing the discharge water amount also the fresh water use is reduced.

8.3.3 Performance monitoring for regulatory purposes

Proper process control is considered a key element for reliable scrubber performance, especially for bioscrubbers. However, manufacturers of biological air treatment systems for livestock farming quite often seem to lack sufficient knowledge of the biological conversion processes that take place in a bioscrubber. As a result, process control parameters are often not properly set. Furthermore, on some farms the discharge water rate seems to be intentionally set to a too low level in order to reduce the operational costs of the system, as the disposal costs of discharge water can be significant (see footnote 1 on page 16). Inspection and control practices by local regulators and authorities are not that intense usually that these situations are quickly recognized and corrected.

In order to guarantee successful NH_3 removal by bioscrubbers process control and monitoring needs to be improved. One of the measures might be the installation of an electrical conductivity (EC) meter that controls the water discharge flow rate, as discussed in section 8.3.2.

Regulations in the Netherlands (VROM, 2002) currently lay down the amount of discharge water that must be produced in a bioscrubber, based on the amount of air treated, the average ammonia concentration and the expected removal efficiency. To be sure that the microbiological population is not inhibited and process performance is not at risk even at high ammonia loading rates, a safety margin is used when prescribing the discharge water flow rate. In other words: to be on the safe side regulations prescribe an average discharge flow rate that is (much) higher than would be necessary in a well controlled system where discharge is based on the actually monitored accumulation of salts (see section 8.3.2). An accurately controlled water discharge system (*e.g.* based on EC), which will reduce the amount of discharge water and thus disposal costs, might be incorporated in future regulations and replace the relatively high discharge water rate that is laid down in current regulations.

Another way of monitoring the performance of any type of scrubber, either acid scrubber or bioscrubbers, would be to directly measure the ammonia removal efficiency by using simple and cheap sensors for measuring ammonia concentrations in the inlet and outlet air. This information could be made available for regulatory inspections. However, the sensors that are currently available for ammonia measurement do not have the robustness and stability that is necessary for continuous gas measurement in this environment (high humidity, high dust concentration) and require cleaning, calibration and maintenance on a regular basis which makes them unsuitable for routine low-cost performance monitoring on site. Further research and development is necessary to develop sensors that can measure during long periods with a minimum maintenance requirement. This would facilitate both process control and regulatory inspection practices.

It is important that (national) authorities and manufacturers collaborate in order to set up proper monitoring and controlling systems. Such systems could be combined with electronic logging and made available for interested parties. If the need for proper monitoring an controlling is not addressed adequately, acceptance by society of large-scale intensive livestock production facilities equipped with an air scrubber might be at risk.

8.4 System concepts

8.4.1 End-of-pipe treatment

In comparison with other emission reduction principles, such as feed management and adaptation of housing system design, an advantage of end-of-pipe air treatment techniques such as air scrubbers is that very high reductions of ammonia emission can be achieved, up to nearly 100% for acid scrubbers. This aspect makes application of air scrubbing systems suitable for situations where other emission reduction options are inadequate.

Furthermore, air scrubbing techniques have the potential to remove not just one compound but to remove a variety of pollutants (ammonia, odour and particulate matter) in one system. In section 8.4.2 a system concept is discussed that aims to increase the removal of these pollutants by means of a multi-stage system.

The application of end-of pipe air treatment does not imply, however, that source measures, such as feed management, adaptation of the housing system or ventilation system, are irrelevant. In fact, changes of the housing system and its management might in some cases be combined with end-of-pipe measures so that the total emission reduction and/or the cost-efficiency of the abatement system can be increased. In sections 8.4.3 and 8.4.4 concepts are discussed that combine scrubber application with other housing management aspects.

8.4.2 Multi-stage scrubbers

In order to meet the increasingly stringent emission levels that livestock operations have to comply with, a new development is the application of multi-stage scrubbers (or combi scrubber). Whereas single-stage acid and biological air scrubbers have been designed for ammonia removal multi-stage air scrubbers also aim to achieve significant emission reduction of odour and particulate matter (PM 2.5 and PM 10). Usually multi-stage scrubbers are multi-stage systems where each stage aims for the removal of one type of compounds. The first prototypes of multi-stage scrubbers for pig and poultry farms, combining the concepts of acid scrubbing, bio-scrubbing, water-curtains, and biofiltration, are in operation now on a limited number of farms; research and development in this field has started recently (Ogink and Bosma, 2007; Arends *et al.*, 2008; Melse *et al.*, 2008). Multi-stage scrubbers may become of major importance for compliance with current and future emission standards. However, further research and development will be necessary to keep investment and operational costs at an acceptable level.

8.4.3 Partial air cleaning

In chapter 4 a novel scrubber design is presented, which aims to reduce both investment and operational costs. Currently, regulations in the Netherlands require that scrubbers are dimensioned for treating the maximum airflow rate that may occur throughout the year, so on average these systems are overdimensioned and underloaded. The new scrubber design focuses on maximizing the cost-effectiveness of ammonia scrubbers by looking into the emission pattern (air flow rate and concentration) throughout the year for different central ventilation configurations and animal categories. An analysis of ammonia emission datasets shows that scrubber size can be significantly reduced with a relatively small effect on the ammonia emission by introduction of a partial air cleaning strategy, which uses a bypass vent at high air loading rates.

In this partial air cleaning system, which assumes a central ventilation system, all air is treated in a scrubber until the maximum capacity (m^3 air/hour) of the scrubber is reached. All air exceeding this setpoint air flow rate is not treated but bypassed and directly emitted into the atmosphere. The bypass is only active during days when high ventilation rates are necessary. The calculations of this treatment strategy show that for example a reduction of scrubber size by 50% still enables treatment of 80 - 90% of the ammonia load. In this way both investment and operational costs (energy use of pumps) can be reduced. Recently this approach has been successfully tested on an acid scrubber on an experimental pig farm (Ellen *et al.*, 2007), although the technical design of the control system still needs to be improved.

The approach of partial air cleaning focuses aims for high *average* ammonia removal efficiencies whereas *occasional* low emission removal efficiencies are accepted. Scrubbers that are designed in this way have a large market potential because investment and operational costs seem to be much lower than for scrubbers that are designed for treatment of the maximum air flow rate. It is advisable that future legislation and regulations allow the use of this type of partial air cleaning systems, taking into account that the system must be monitored well to prevent unwanted emissions if the partial air cleaning control systems might fail. In this respect it is important that regulations clearly describe and define performance and removal efficiency requirements as efficiencies can be expressed in different ways (average efficiency, minimum efficiency, maximum number of times and duration that emission limit may be exceeded etc.).

For ammonia it is reasonable to focus on required *average* emission levels as the environmental impact is mainly related to the cumulative emission and resulting cumulative nitrogen deposition (kg/year) with long term effects on vegetation, and not the actual emission during a short period of time. For odour and particulate matter emissions, however, not the average but the *actual* emission level (peak level) and the duration of the peak emission might be relevant. The reason for this is that odour nuisance is affected by the peak level and duration of the emission, even if average emission levels remain unchanged. For particulate matter concentrations 24-hour limit values have been defined besides yearly average values, as the health effect of PM inhalation is related to the concentration level and exposure time. The effect of partial air cleaning on particulate matter and odour emission, and the connected nuisance and health effect, is a subject that needs to be further investigated.

Also partial air cleaning strategies are possible other than the bypass system that is described in chapter 4 for a central ventilation system. The first example is a system that is applied at an animal house equipped with a room-based ventilation system, where the fans of some rooms are connected to a scrubber and the fans of some rooms exhaust directly into the atmosphere. In contrast with the system described in chapter 4, all the time part of the air is emitted untreated, regardless of the total ventilation rate; this type of partial air cleaning system for ammonia removal is applied at several pig houses in Denmark (DAAS, 2004).

The second example is a ventilation system that is equipped with a number of fans that are sequentially switched on when the ventilation need increases. Such a system can be applied for instance at a broiler houses, which is usually one large room. The fans that are switched on first are connected to the scrubber system, the fans that are only used at maximum ventilation rate are not connected to the scrubber. Most of the time only the first fans are used and all ventilation air is treated by the scrubber. However, when maximum ventilation rates are applied, usually only during a few days every year, the fans that are not connected to the scrubber system are switched on as well and the air through these fans is exhausted into the atmosphere directly. In this way the investment and operational costs of air cleaning can be drastically reduced compared to an air scrubber system that has been designed for the maximum ventilation capacity.

The third example is an animal house where forced pit ventilation is applied (Sapounas *et al.*, 2008; Smits *et al.*, 2009). The ventilation air from the slurry pit is treated in a scrubber while the other ventilation air is left untreated. As the pit air has a relatively high ammonia concentration and low air flow rate, emission reduction can be realized at relatively low costs. This partial air cleaning technique can be applied both in mechanically and naturally ventilated houses.

The last example of a partial air cleaning system is a system where exhaust air is treated in a biofilter only when odour nuisance is expected on basis of atmospheric conditions. In this strategy the air is only treated in the biofilter when the atmosphere is stable, usually early in the morning and during cooler days, when mixing and dilution at the source is limited. When the atmosphere is less stable the air is emitted into the atmosphere directly without any treatment and odour nuisance is supposed to be reduced by mixing and dilution in the atmosphere (Hoff *et al.*, 2009).

8.4.4 Further integration of air scrubbing and housing management

Whereas the multi-stage scrubber system and, to a lesser extent, the partial air cleaning systems that are described in the previous sections can still be considered as "stand-alone" treatment systems, it is also possible to combine end-of-pipe air treatment with other housing management aspects. Air treatment might be applied as part of an integrated housing management system that takes into account pen design, manure collection, manure treatment, and ventilation control.

For example adaptations of the ventilation system of the animal house can directly affect the scrubber design parameters, if ammonia concentrations and/or air flow rates are changed. At several farms the application of an acid scrubber is now combined with a system for cooling the ventilation air that enters the building in summertime. This is done by leading the inlet ventilation air through a heat exchanger (*e.g.* a subsoil heat exchanger) prior to reaching the animals. As a result, both the average and maximum air flow rates of the house are decreased.

Consequently, a scrubber of a smaller size will suffice, assuming that the minimum empty bed air residence time is not changed, and treatment costs will be reduced.

Furthermore, concepts are possible that combine air conditioning with recirculation of ventilation air. An example is an experimental pig housing system where the exhaust air first goes through a scrubber for ammonia removal, then through a conditioning unit and finally is partly led back to the animals and partly replaced by fresh air (Mouwen and Plagge, 1995; Tolsma, 2000). In the conditioning unit the air is cooled down, condensate is removed, and treated by UV for desinfection. By recycling the air the inlet and outlet air flow of the house (m^3/hour) is much lower than for conventional ventilation systems. The inventors claim that this results in a better animal welfare status and a significant increase of growth performance.

Finally, an example of an approach which combines air treatment and manure treatment is provided by the Hercules pig fattening system (Ogink *et al.*, 2000). In this system the pig faeces and urine are collected separately using a conveyor belt beneath a slatted floor. The urine is acidified with nitric acid and used as scrubbing liquid in a packed bed where the exhaust ventilation air from the house is treated. As a result, ammonia is removed from the air, part of the urine is evaporated and a mineral concentrate remains.

8.5 Future outlook

8.5.1 General

From the previous sections it can be concluded that air scrubbing is a suitable technique for treatment of exhaust from mechanically ventilated houses and can still be improved and optimized. The operational strategy of air scrubbers has not been optimized yet for removal of other compounds than just ammonia. Furthermore, new system concepts are possible that aim to increase scrubber efficiency and/or cost-effectiveness. Finally, process control of bioscrubbers might be enhanced and result in improved robustness and performance of these systems.

In the next sections a future outlook is given on scrubber application with regard to IPPC regulations, market size, and research approach.

8.5.2 Best Available Techniques

In chapter 2 it has been mentioned that end-of-pipe emission reduction techniques such as air scrubbers were not considered as Best Available Techniques (BATs) for intensive livestock production in the legislatorial framework of the IPPC directive, as described in the first technical reference guide for intensive livestock facilities (EC,2003). The reason for this exclusion appeared to be of economic (high running costs), ecologic (high energy consumption, chemical use of acid scrubbers, waste water production), and technical nature (unstable performance of bioscrubbers) (VROM, 2007a; Hendriks, 2008).

However, successful improvements have been achieved in practice and research that should be taken into account. In the following section the raised objections are discussed:

(1) Economic: high running costs.

Scrubbers have been applied for 25 years now in intensive livestock farming in the Netherlands. The number of scrubbers that is operated nowadays in the Netherlands and other intensive livestock production areas in Europe is increasing (see section 8.5.3). This proves that, even when running costs are considered high, this technique is economically viable, at least in intensive livestock production areas where high emission standards have to be met.

(2) Ecologic: high energy consumption, chemical use, waste water production.

Manufacturers claim to have reduced energy use for about 50% since scrubbers were first introduced in livestock farming. This is achieved by more efficient pumps and low-pressure and/or discontinuous water distribution systems for wetting of the packing. Also energy use has been reduced by changes in scrubber dimensions: the air inlet surface areas has been increased and the thickness of the packing has been decreased, thus decreasing the pressure drop energy use at equal air flow rates and residence times. Furthermore, application of fans that have been chosen to work efficiently at higher backpressure levels (instead of the "normal" fans that are used for animal houses without a scrubber) has reduced energy use. However, the claimed decrease of energy use by scrubbers has currently not been sufficiently verified by independent research at the moment. Finally, introduction of partial air cleaning systems and inlet air cooling (see section 8.4.4) have further reduced energy use.

In an acid scrubber a chemical is used to capture ammonia, usually sulphuric acid. Although the use of chemicals has been stated as a drawback, the advantage of this is that liquid nitrogen fertilizer is produced as discharge water. In other words: nitrogen that would otherwise be emitted to the atmosphere is now being made available for use as fertilizer. The discharge water of acid scrubbers is increasingly being used for replacement of artificial fertilizer in the Netherlands, as facilitated by recent regulations (VROM, 2009). For bioscrubbers the amount of discharge water is larger and the nitrogen concentration is lower which makes competition with artificial fertilizer more difficult (see section 8.3.2 for possible techniques for discharge water volume reduction).

(3) Technical: unstable performance of bioscrubbers

It is true that bioscrubbers often experience operational problems (poor process control), as was also concluded in the review described in chapter 3. Although possibilities exist for improvement of process control and monitoring (see section 8.3.2 and 8.3.3), currently this objection has not been addressed adequately. Acid scrubbing, however, is generally considered as a stable and reliable technique as the process control for an acid scrubber is much more robust.

When considering the status of air scrubbing techniques in BAT regulations, a distinction should be made between acid scrubbers and bioscrubbers, as some objections specifically apply to acid scrubbers and some to bioscrubbers. Taking into consideration the objections that are discussed above, it might be advisable to reconsider the status of acid scrubbers in EU regulations when the list of BATs for intensive livestock farming is next reviewed. Acid air scrubbing has proven to be an effective and viable ammonia abatement technology in intensive livestock areas in Germany and the Netherlands, although independent research into the energy use of these systems is still necessary.

For bioscrubbers, however, the problem of unstable performance has not been addressed adequately until now. Therefore it is questionable to consider bioscrubbing as BAT for intensive livestock farming currently.

8.5.3 Scrubber market growth

For about 25 years air scrubbers have been applied in the Netherlands in intensive livestock farming, in particular for the emission reduction of ammonia. In 2004 biotrickling filters were operated on about 45 farms and acid scrubbers on about 160 farms, in total just over 200 locations (Melse and Ogink, 2004). However, during the last five years the application of air scrubbers has been multiplied; recent data show that in early 2008 scrubbers were operated on almost 900 farms in The Netherlands (Table 8.2). This market growth coincides with a general trend of increasing the scale of livestock operations in order to reduce costs. This increase of scale probably facilitates parties to invest in air-scrubber systems. In Germany a similar market growth can be noticed (Hahne, 2007). It is expected that in the coming years the implementation of air scrubbers will further expand in the Netherlands and in other intensive livestock production areas in Europe in order to comply with European regulations for protection of natural ecosystems and ambient air quality.

Table 8.2 Scrubber application for ammonia removal in pig and poultry operations in the Netherlands, based on statements from manufacturers, as per January 1st, 2008) (Melse et al., 2008).

	Installed ventilation capacity (m^3 $hour^{-1}$)	Number of farms (-)
Acid scrubbers	64 million	790
Biotrickling filters	14 million	90
Total:	79 million	880
Pig	76 million[a]	850
Poultry	3 million[b]	30
Total:	79 million	880

[a] This equals 10% of the exhaust air of all pig farms nationwide.
[b] This equals 0.4% of all exhaust air of all poultry farms nationwide.

8.5.4 Research approach

Currently, most scrubber related research in the field of livestock operations in the Netherlands deals with pilot- or full-scale testing of scrubber systems as they are provided by their manufacturers. More fundamental research, which tries to understand the processes that take place inside these systems, however, is scarce. It is necessary to profound the knowledge of the processes that take place inside the scrubber systems by means of laboratory research and standardized testing of (bench-scale) air scrubbers for this field of application. Further insight in these processes might lead to better insight in the boundaries and limitations of air scrubbers for treatment of animal house exhaust air and also give directions for improvement of the performance and the design of these systems. Possible subjects for future research include choice of packing material, ratio of liquid to gas flow rate, control of gaseous N_2O formation during ammonia conversion in bioscrubbers, and increase of odour removal efficiency.

Another interesting approach is to further investigate how the scrubber design characteristics (the dimensions of the packing material (length, width, height, and volume), system choice (cross- or counter-current), air flow direction (horizontal or vertical), pressure chamber dimensions etc.) affect the energy use for humidification, backpressure and energy use of the fans, investment costs, and removal performance of pollutants. Up till now (the combination of) these basic principles have not been investigated and reviewed thoroughly with regard to removal of ammonia, odour and particulate matter from animal house exhaust air. Further research into these issues might lead to improvement of process performance and cost-effectiveness, however.

References

Aarnink, A.J.A.; W.J.M. Landman; R.W. Melse; T. T. T. Huynh (2005). Systems for eliminating pathogens from exhaust air of animal houses. Proceedings of the 7th International Livestock Environment Symposium (ILES VII), May 18 - 20, 2005, Beijing, China; pp 239-244; ASAE, St. Joseph, MI, USA, ASAE Publication 701P0205, ISBN 1-892769-48-4.

Aarnink, A.; Y. Zhao; T. van Hattum; A. Hol (2007). Reductie fijn stof emissie uit een varkensstal door de combi-wasser van Big Dutchman. Animal Sciences Group van Wageningen UR, Lelystad, Netherlands; ISSN 1570 8616.

Aarnink, A.; Y. Zhao; T. van Hattum; A. Hol (2008a). Reductie fijn stof emissie uit een varkensstal door de combi-wasser van Uniqfill. Confidential report. Animal Sciences Group van Wageningen UR, Lelystad, Netherlands.

Aarnink, A.; Y. Zhao; T. van Hattum; A. Hol (2008b). Reductie fijn stof emissie uit een varkensstal door de combi-wasser van Inno+/Siemers. Confidential report. Animal Sciences Group van Wageningen UR, Lelystad, Netherlands.

Arends, F.; G. Franke; E. Grimm; W. Gramatte; S. Häuser; J. Hahne (2008). Exhaust Air Treatment Systems for Animal Housing Facilities, Techniques - Performance - Costs. KTBL-Schrift 464, KTBL, Darmstadt, Deutschland; ISBN 978-3-939371-60-1.

Burgess, J.E.; R.M. Stuetz; S. Morton; T. Stephenson (2002). Dinitrogen oxide detection for process failure early warning systems. Water Science and Technology, Vol 45, No 4-5, pp 247-254.

Césario, M.T. (1997). Water-immiscible solvents for the biological treatment of waste gases. PhD diss, Landbouwuniversiteit Wageningen, Wageningen, Netherlands; ISBN 90-5485-685-8.

Colliver, B.B.; T. Stephenson (2000). Production of nitrogen oxide and dinitrogen oxide by autotrophic nitrifiers. Biotechnology Advances, Vol 18, pp 219-232.

DAAS (2004). Acid scrubber, cleaning of 60 % of the exhaust air. BAT Data Sheet no. 106.04-58. Danish Agricultural Advisory Service, Natonal Centre, Aarhus, Denmark.

Davidson, C.T.; A.J. Daugulis (2003). Addressing biofilter limitations: A two-phase partitioning bioreactor process for the treatment of benzene and toluene contaminated gas streams. Biodegradation, Vol 14, pp 415-421.

EC (2003). Integrated Pollution Prevention and Control (IPPC). Reference Document on Best Available Techniques for Intensive Rearing of Poultry and Pigs; European Commission; http://ec.europa.eu/comm/environment/ippc/brefs/ilf_bref_0703.pdf.

Ellen, H.H.; J.M.G. Hol; A.I.J. Hoofs; J. Mosquera (2007). Vergelijking theorie en praktijk emissiereductie van chemische luchtwasser met bypassventilatoren; Rapport 74, Animal Sciences Group van Wageningen UR, Lelystad, Netherlands; ISSN 1570-8616.

Guarino, M.; A. Costa; M. Porro (2008). Photocatalytic TiO_2 coating - to reduce ammonia and greenhouse gases concentration and emission from animal husbandries. Bioresource Technology, Vol 99, pp 2650–2658.

Hahne. J. (2007). Actual development of waste air treatment in animal husbandry. Landtechnik, Vol 62, No 3, pp 178-179.

Hendriks, H.J.M. (2008). Personal communication. Member of the workgroup on the definition of the first BREF for Intensive Rearing of Poultry and Pigs.

Hoff, S.J.; J.D. Harmon; L. Chen; K.A. Janni; D.R. Schmidt; R.E. Nicolai; L.D. Jacobson (2009). Partial biofiltration of exhaust air from a hybrid ventilated deep-pit swine finisher barn. Applied Engineering in Agriculture, Vol 25, No 2, pp 269-280.

Hong, Z.; K. Hanaki; T. Matsuo (1993). Greenhouse gas - N2O production during denitrification in wastewater treatment. Water Science and Technology, Vol 28, No 7, pp 203-207.

Jungbluth, T.; E. Hartung (2003). Abschlußbericht zum Untersuchungsprojekt Ermittlung der Geruchsminderung der Abluft eines Mastschweinestalles durch eine Abluftreinigung mittels UV-Oxidation. Staatshaushaltsplan 2002 Kap. 0803 Tit. 686 83, Az. 22-8215.25, Universität Hohenheim, Stuutgart, Germany.

Koziel, J.A.; X. Yang; T. Cutler; S. Zhang; J. Zimmerman; S. J. Hoff; W. Jenks; Y. Laor; U. Ravid; R. Armon; J.H. van Leeuwen (2008). Mitigation of odor and pathogens from CAFAs with UV/TiO2: exploring cost effectiveness. Proceedings of the Mitigating Air Emissions From Animal Feeding Operations Conference, May 19 - 21, 2008, Des Moines, Iowa (IA), USA.

Melse, R.W.; N.W.M. Ogink (2004). Toepassing van luchtbehandelingstechnieken binnen de intensieve veehouderij, Fase 2: Mogelijkheden tot kostenverlaging van wassers; Rapport 271; Agrotechnology & Food Innovations, Wageningen UR, Wageningen, Netherlands; ISBN 90-6754-835-9.

Melse, R.W.; N.W.M. Ogink; B.J.J. Bosma (2008). Multi-pollutant scrubbers for removal of ammonia, odor, and particulate matter from animal house exhaust air. Proceedings of the Mitigating Air Emissions from Animal Feeding Operations Conference, May 19 - 21, 2008, Des Moines, Iowa (IA), USA.

Monteny, G.J.; M. Verhaak; E. Hartung (1998). Technical and economical feasibility of integration of a catalytic reactor system for removal of aerial pollutants from livestock operations. Framework BRITE-EURAM project, BEST-CT97-0556. Nota V 98-45, IMAG-DLO, Wageningen, Netherlands.

Mouwen, I.A.A.C.; J.G. Plagge (1995). Studie naar klimatisering van de dekstal in relatie tot emissie en energie (with English summary). Proefverslag P 1.125. Praktijkonderzoek Varkenshouderij, Raalte, Netherlands; ISSN 0922-8586.

Ogink, N.W.M.; H.C.Willers; A.H.M.Veeken; H.V.M.Hamelers (2000). The Hercules pig production system: a new farm system for sustainable production of pigs and fertilisers. Proceedings of the AgEng2000 conference, University of Warwick, 2-7 July 2000, United Kingdom. Paper 00-AP-006.

Ogink, N.W.M.; B.J.J. Bosma (2007). Multi-phase air scrubbers for the combined abatement of ammonia, odor and particulate matter emissions. International Symposium on Air Quality and Waste Management for Agriculture, Broomfield, Colorado, USA. CD-Rom Proceedings of the 16-19 September 2007. ASABE Publication Number 701P0907cd.

Ogink, N.W.M.; J. Hahne (2007). Removal of dust fractions by air scrubbers in livestock operations. Proceedings of DustConf 2007, International Conference in Maastricht, Netherlands, 23-24 April 2007; http://www.dustconf.com.

Ogink, N.W.M.; R.W. Melse ; J. Mosquera (2008). Multi-pollutant and one-stage scrubbers for removal of ammonia, odor, and particulate matter from animal house exhaust air. Proceedings of the 8th International Livestock Symposium (ILES VIII), August 31 - September 4, 2008, Iguassu Falls, Brazil.

Sapounas, A.A.; J.B. Campen; M.C.J. Smits; H.J.C. Dooren (2009). Simulating the effect of forced pit ventilation on ammonia emission from a naturally ventilated cow house with CFD. Proceedings of the Joint International Agricultural Conference, July 6 - 8, 2009, Wageningen, Netherlands; pp 81-89; Wageningen Academic Publishers, Wageningen, Netherlands, ISBN 978-90-8686-112-5.

Smits, M.C.J.; J.B. Campen; J.W.H. Huis in't Veld (2008). Emissiereductie door kelderluchtbehandeling in een vleeskalverstal; proof of principle. CFD modelberekeningen en pilot in een afdeling van een kalverhouderij. Report 179. Animal Sciences Group van Wageningen UR, Lelystad, Netherlands; ISSN 1570-8618.

Tolsma, A. (2000). BB Air system. Biological and bacterial air recycling system. Proceedings of the symposium "De varkensstal van de toekomst", 8 June 2000, WICC, Wageningen, Netherlands.

Trimborn, M. (2006). Biofilter/Biowäscher an Tierhaltungslagen als relevante Quelle von Lachgas durch Ammoniakabscheidung?; Schriftenreihe des Lehr- und Forschungsschwerpunktes USL, No 138; 59 pp; Landwirtschaftliche Fakultät der Universität Bonn, Bonn, Deutschland.

Van Groenestijn, J.W.; M.E. Lake (1999). Elimination of alkanes from off-gases using biotrickling filters containing two liquid phases. Environmental Progress, Vol 18, No 3, pp 151-155.

VROM (2002). Regeling Ammoniak en Veehouderij (RAV); 1 mei 2002; Staatsblad No 82. Ministry of Housing, Spatial Planning and the Environment, The Hague, Netherlands.

VROM (2007). Oplegnotitie bij de BREF voor de intensieve pluimvee- en varkenshouderij. SenterNovem/InfoMil. Ministry of Housing, Spatial Planning and the Environment, The Hague, Netherlands.

VROM (2009). Gebruikershandleiding over spuiwater uit chemische luchtwassers. SenterNovem/InfoMil. Ministry of Housing, Spatial Planning and the Environment, The Hague, Netherlands.

Zhao, Y.; A.J.A. Aarnink; P.W.G.G. Koerkamp; N.W.M. Ogink; M.C.M. de Jong (2008). Removal efficiency of dust and bacteria by multi-stage air scrubbers in pig houses Proceedings of the 8th International Livestock Symposium (ILES VIII), August 31 - September 4, 2008, Iguassu Falls, Brazil.

Summary

Intensive livestock production contributes substantially to the economies of many European countries in terms of employment and export of products. However, intensive livestock production is also connected with a number of environmental effects, which include emissions of pollutants to soils and surface waters (*e.g.* nitrogen, phosphorus, and heavy metals) and to the air, *e.g.* ammonia (NH_3), odour, non-CO_2 greenhouse gases (methane (CH_4) and nitrous oxide (N_2O)) and particulate matter (PM10 and PM2.5). In order to reduce the environmental impacts of livestock production both national and international regulations have come into effect. This thesis focuses on one of the emission sources: the abatement of gaseous emissions from animal houses.

Possible strategies for emission reduction from animal houses include adaptation of housing design and manure storage, feed management, and the application of end-of-pipe air treatment, *viz* acid scrubbers and bioscrubbers. In a scrubber contaminated air is brought in intensive contact with a scrubbing liquid; as a result the contaminant (*e.g.* ammonia) is removed from the air and transferred to the liquid. This is done by forcing the air through a porous bed that is continuously wetted. In an acid scrubber ammonia is captured in the liquid by acid; in a bioscrubber the ammonia is converted by bacteria, mainly to nitrite and nitrate. End-of pipe air treatment, which can only be applied for mechanically ventilated animal houses, can achieve very high emission reductions (up to 100% ammonia removal for acid scrubbers), whereas other emission reduction options might be insufficient to meet the emission standards in some cases (vicinity of vulnerable ecosystems or residential housings). Furthermore, air treatment techniques have the potential to remove not just one compound (ammonia) but to remove a variety of pollutants (also odour and particulate matter) in one system. In order to meet emission regulations and targets the successful application of scrubbers is of increasing importance. However, current applications of scrubber technology in livestock operations seem to be unsatisfactory with regard to long-term process robustness, removal efficiency, and cost-efficiency.

The aim of this thesis is to better understand and improve the performance of air scrubber techniques in livestock operations where they are applied at mechanically ventilated animal houses. The research focuses on the development of more efficient and reliable scrubber systems that are at the same time economically viable. The following objectives are defined:

(1) to determine how air scrubbers are performing at livestock farms, with regard to ammonia and odour removal (chapters 3, 6, and 7);

(2) to determine how the cost-efficiency of air scrubbers at livestock farms can be increased by applying a different treatment strategy (chapter 4);

(3) to determine if, in addition to ammonia and odour, methane can be removed by biological air treatment systems (chapter 5);

(4) to discuss the problems and shortcomings related to current air scrubbing practices and make recommendations for possible improvement and future research (chapter 8).

First of all the current performance of on-farm operated air scrubbers was reviewed and discussed with regard to ammonia and odour removal (chapter 3). It was concluded that bioscrubbers often experience operational problems (probably caused by poor process control), that ammonia removal efficiency by scrubbers is relatively high (on average 96% for acid scrubbers and 70% for bioscrubbers), that odour removal is relatively low (on average 31% for acid scrubbers and 44% for bioscrubbers), and that bioscrubbers often show a large variation in odour removal efficiency.

Next a novel scrubber design was presented which comprises a partial air cleaning control system (chapter 4). In this approach, which assumes a central ventilation system, the exhaust air is treated in a scrubber up to the point that the maximum capacity (m^3 air/hour) of the scrubber is reached. The surplus air exceeding this capacity is not treated but bypassed and directly emitted into the atmosphere. The bypass is only active during days when high ventilation rates are necessary. The presented partial air cleaning strategy, which uses an air flow controlled bypass vent that opens at high air loading rates, increases the efficiency of scrubber utilization (kg NH_3 treated/m^3 scrubber volume) and thus reduces mitigation costs per kg NH_3 if the removal efficiency remain unaffected. Model calculations showed that a reduction of scrubber volume by *e.g.* 50% still enables treatment of 80 - 90% of the ammonia load throughout the year, which means that cost-optimization, *i.e.* reduction of operational cost per kg NH_3 load treated, is possible.

In chapter 5 an experiment is described which shows that biological oxidation of methane is possible in a biofilter[1], although the low water solubility of methane limits the degradation capacity. Based on the found first-order rate constant a model was presented which shows the influence of methane concentration and air residence time on methane removal efficiency. Either relatively high methane concentrations (several grammes per m^3) or high air residence times (several

[1] In contrast with a scrubber, which is a packed tower filter with an inert packing material that is usually continuously wetted, the wording "biofilter" is used to describe a system with an organic-based packing material and a low water flow.

minutes) are necessary for high removal efficiency. This is why methane removal in common scrubber systems (air residence time in the order of seconds) that treat methane containing ventilation air from animal houses (methane concentration in the order of milligrammes per m^3) is expected to be negligible. Therefore bioscrubber and biofilter systems are not considered economically feasible for methane removal from animal house ventilation air. No low-cost treatment techniques are currently available that can be operated cost-effectively at these low methane concentrations.

Furthermore two case-studies were presented (chapter 6 and 7) that illustrate the practical application of air scrubbers in livestock farming. In the first case-study the field performance of a specific bioscrubber operated on a pig farm was described and discussed. It was found that the bioscrubber was not well operated, while no one was aware of that, and implies that poor performance of bioscrubbers might be a common problem which corresponds to some findings from chapter 3. The second case-study is an evaluation of the technical performance and economics of several liquid manure treatment systems and includes an assessment of the gaseous emissions from these treatment systems. This study illustrates that within livestock farming scrubbers can also be used in application areas other than treatment of animal house ventilation air, by presenting several liquid manure treatment system that comprise an acid ammonia scrubber. Especially acid scrubbers might be used for effective abatement of ammonia emissions from such batch processes as this type of scrubber can be easily switched on and off without removal efficiency loss, in contrast to bioscrubber systems.

Finally, in chapter 8 the problems and shortcomings related to current air scrubbing practices are discussed and recommendations are made for possible improvement and future research. It is concluded that air scrubbing is a suitable technique for treatment of exhaust of from mechanically ventilated houses but can still be improved and optimized.

First of all, design and operational strategy of air scrubbers have not been optimized yet for removal of other compounds than just ammonia. For example the effect of basic design parameters for ammonia removal, such as pH or air residence time, on odour removal is insufficiently known. Further research into odour characteristics and the processes involved in odour removal by air scrubbers is necessary to understand the large variations and improve the relatively low removal efficiency for odour.

Furthermore, new system concepts are possible that aim to increase scrubber efficiency and/or cost-effectiveness. An example is an approach in which several scrubber concepts are combined in a multi-stage system, where each stage aims to remove one type of compounds, *e.g.* a combination of an acid scrubber with a bioscrubber or biofilter. Another example is the application of a partial air cleaning strategy (chapter 4). This approach focuses on maximizing the cost-effectiveness of ammonia scrubbers by looking into the emission pattern (air flow rate and concentration) throughout the year for different central ventilation configurations and animal categories. In this way both investment and operational costs (energy use, water discharge costs) can be reduced. Current legislation and regulations in the Netherlands need to be adapted for application of this type of partial air cleaning systems. The effect of partial air cleaning on particulate matter and odour emission, and the connected nuisance and health effect, is a subject that needs to be further investigated.

Finally, the parameters that determine the actual value of the ammonia removal efficiency of bioscrubbers are not yet fully understood. At the applied residence times and concentrations an average removal efficiency of 70% is considered normal for ammonia removal; sometimes much higher or lower efficiencies are measured. The measurements showing a higher removal efficiency indicate that average ammonia removal efficiencies might be increased by better process control and further optimization of the design. Process control of bioscrubbers might be enhanced by measurement of electrical conductivity (EC) as a control parameter for water discharge. In this way the water discharge rate depends on the actual ammonia loading rate and the accumulation of inhibiting compounds (nitrite) can be prevented. Introduction of this control mechanism in future legislation might facilitate regulatory inspection and practices and improve process performance.

For about 25 years air scrubbers have been applied in the Netherlands in intensive livestock farming now, in particular for the emission reduction of ammonia. However, during the last five years the application of air scrubbers has been multiplied and nationwide already 10% of the exhaust air of pig farms is treated by air scrubbing. Besides ammonia the reduction of odour and particulate matter emissions has become of increasing importance. The growth of the number of air scrubbers coincides with a general trend of increasing the scale of livestock operations. It is expected that the coming years the implementation of air scrubbers will further expand in the Netherlands and in other intensive livestock production areas in Europe in order to comply with European regulations for protection of natural ecosystems and ambient air quality.

Samenvatting

De titel van dit proefschrift is: "Luchtbehandelingstechnieken voor beperking van emissies uit de intensieve veehouderij".

De intensieve veehouderij is een omvangrijke economische activiteit in veel Europese landen en draagt zo bij aan werkgelegenheid en export. Daarnaast is deze sector verantwoordelijk voor het optreden van emissies naar bodem en oppervlakte water (bijv. stikstof, fosfor en zware metalen) en emissies naar de lucht, zoals ammoniak (NH_3), geur, broeikasgassen (voornamelijk methaan (CH_4) en lachgas (N_2O)), en fijnstof (PM10 and PM2.5). Om deze emissies te verminderen is zowel nationale als internationale wet- en regelgeving van kracht. Dit proefschrift richt zich op een van de emissiebronnen uit de veehouderij: de gasvormige emissie uit stallen.

Er bestaan verscheidene methoden om emissies uit stallen tegen te gaan, zoals aanpassing van stalontwerp en mestopslagsysteem, aanpassing van de voersamenstelling en toepassing van een nageschakeld luchtbehandelingssysteem. Dit laatste is alleen toepasbaar in het geval van mechanisch geventileerde stallen. De meest gangbare luchtbehandelingssystemen voor stallen zijn zure wassers ("chemische wassers) en biowassers. In een wasser wordt de te behandelen lucht in intensief contact gebracht met een wasvloeistof, zodat de verontreiniging (o.a. ammoniak) uit de lucht wordt gewassen. Dit wordt gedaan door de lucht te leiden door een poreus, meestal kunststof pakkingsmateriaal dat continu bevochtigd wordt. In een zure wasser wordt ammoniak in de wasvloeistof gebonden met zuur en in een biowasser wordt ammoniak door bacteriën omgezet in hoofdzakelijk nitriet en nitraat. Luchtbehandelingssystemen zijn in staat om een hoge emissiereductie te realiseren (tot 100% ammoniakverwijdering bij een zure wasser) terwijl andere emissiebeperkende maatregelen soms ontoereikend zijn om te voldoen aan geldende emissienormen (bijv. in de nabijheid van kwetsbare natuurgebieden of bebouwing). Bovendien scheppen nageschakelde luchtbehandelingssystemen de mogelijkheid om niet slechts één (ammoniak) maar meerdere componenten (ook geur en fijnstof) uit de lucht te verwijderen. Om te kunnen voldoen aan steeds strenger wordende emissie-eisen is een succesvolle toepassing van luchtwassers op het boerenbedrijf van toenemend belang. In de praktijk blijkt echter dat de toepassing van wassers niet altijd even succesvol is en dat de processtabiliteit en verwijderingsefficiëntie op de lange termijn soms te wensen overlaten.

Het doel van dit proefschrift is het beter begrijpen en verbeteren van het functioneren van luchtwassers zoals deze worden toegepast voor de behandeling

van ventilatielucht van mechanisch geventileerde stallen. Het onderzoek richt zich zowel op het verhogen van de efficiëntie en betrouwbaarheid als op het verlagen van de kosten van luchtwassers voor stallen. De doelstellingen zijn als volgt:

(1) het bepalen hoe luchtwassers voor stalluchtbehandeling in de praktijk functioneren met betrekking tot ammoniak- en geurverwijdering (hoofdstuk 3, 6 en 7);

(2) het bepalen hoe de kostenefficiëntie van luchtwassers kan worden verbeterd door het toepassen van een alternatieve luchtbehandelingsstrategie (hoofdstuk 4);

(3) het bepalen of, naast ammoniak en geur, methaan kan verwijderd worden met behulp van een biologisch luchtbehandelingssyteem (hoofdstuk 5);

(4) het analyseren van de problemen en tekortkomingen van de huidige luchtbehandelingssystemen voor stallucht en het formuleren van aanbevelingen voor verbetering en nader onderzoek (hoofdstuk 8).

Allereerst is in kaart gebracht hoe de huidige generatie luchtwassers voor stallen in de praktijk functioneert wat betreft ammoniak- en geurverwijdering (hoofdstuk 3). Geconcludeerd wordt dat biowassers vaak te maken hebben met operationele problemen en storingen (waarschijnlijk veroorzaakt door gebrekkige procesregeling), dat de ammoniakverwijdering door wassers relatief hoog is (gemiddeld 96% voor zure wassers en 70% voor biowassers), dat de geurverwijdering relatief laag is (gemiddeld 31% voor zure wassers en 44% voor biowassers) en dat biowassers vaak een grote variatie van het geurverwijderingsrendement vertonen.

Vervolgens is een nieuw wasserontwerp gepresenteerd waarbij in het geval van hoge ventilatiebehoefte niet de volledige luchthoeveelheid wordt behandeld (zoals in de huidige praktijk het geval is) maar slechts een deelstroom; een deel van de ventilatielucht ontwijkt dan rechtstreeks naar de atmosfeer zonder dat deze behandeld is (hoofdstuk 4). De ventilatiebehoefte hangt enerzijds af van het weer (warm/vochtig) en anderzijds van het aantal en gewicht van de dieren in de stal; bij lage luchtdebieten wordt wel de volledige luchthoeveelheid door de wasser geleid. Dit wasserontwerp leidt er toe dat de gemiddelde ammoniakbelasting (kg NH_3/m^3 wasservolume) toeneemt waardoor de emissiereductiekosten per kg NH_3 afnemen bij ongewijzigde verwijderingsrendementen. Modelberekeningen wijzen uit dat wanneer een wasser tweemaal zo klein is nog steeds 80 - 90% van de totale hoeveelheid ammoniak door de wasser wordt geleid gemiddeld over een jaar. Een verlaging van de emissiereductiekosten is dus mogelijk met een dergelijk systeem.

In hoofdstuk 5 wordt een experiment beschreven waarin aangetoond wordt dat biologische oxidatie van methaan mogelijk is in een biofilter[1], hoewel de lage wateroplosbaarheid van methaan de afbraakcapaciteit van het biofilter beperkt. Op basis van de bepaalde eerste-orde reactie constante wordt berekend op welke wijze de methaan afbraakcapaciteit van het biofilter afhangt van de methaanconcentratie van de aangeboden lucht en van de luchtverblijftijd in het filter. Om een hoge verwijderingsefficiëntie te bereiken is ofwel een hoge methaanconcentratie noodzakelijk (enige grammen per m^3), ofwel een hoge luchtverblijftijd (enige minuten). Aangezien de gangbare luchtwassystemen voor stallucht (methaanconcentratie in de ordegrootte van milligrammen per m^3) een lage luchtverblijftijd hebben (in de ordegrootte van seconden) volgt hieruit dat de methaanverwijdering door deze luchtwassers verwaarloosbaar zal zijn. Daarom worden biowasser en biofilter systemen ongeschikt geacht voor kostenefficiënte verwijdering van methaan uit stallucht. Op dit moment zijn geen verwijderingstechnieken bekend die bij dergelijke lage methaanconcentraties economische haalbaar zijn.

Daarna worden twee case-studies gepresenteerd (hoofdstuk 6 en 7). In de eerste case study wordt beschreven hoe een specifieke biowasser in de praktijk draait op een varkensbedrijf. De biowasser bleek niet goed te functioneren terwijl niemand zich daarvan bewust was. Dit impliceert dat het slecht functioneren van biowassers mogelijk een regelmatig optredend probleem is, hetgeen bevestigd wordt door de bevindingen uit hoofdstuk 3. De tweede case study is een technische en economische evaluatie van een aantal mestbewerkingssystemen voor drijfmest; ook de gasvormige emissies worden meegenomen. In een aantal gevallen maakt een zure wasser deel uit van het mestbewerkingssysteem waarmee geïllustreerd wordt dat luchtwassers ook op andere gebieden binnen de veehouderij kunnen worden ingezet dan voor stalluchtbehandeling alleen. In het bijzonder zure wassers zijn geschikt om ingezet te worden voor het terugdringen van de ammoniakemissie van dergelijke batch processen, aangezien dit type wasser eenvoudig aan en uit kan worden geschakeld zonder verlies van ammoniakverwijderingsefficiëntie, in tegenstelling tot biowassers.

Tenslotte worden in hoofdstuk 8 de problemen en tekortkomingen bediscussieerd zoals we deze aantreffen bij de huidige luchtbehandelingssystemen en wordt een aantal aanbevelingen gedaan voor procesverbetering en nader

[1] In tegenstelling tot een wasser, waarmee een systeem wordt bedoeld met een continu bevochtigd inert (kunststof) pakkingsmateriaal, wordt met "biofilter" een systeem bedoeld waarbij sprake is van een organisch pakkingsmateriaal en een relatief lage bevochtigingssnelheid.

onderzoek. Geconcludeerd wordt dat luchtwassing een geschikte techniek is voor de behandeling van ventilatielucht van stallen maar nog steeds verbeterd en geoptimaliseerd kan worden.

In de eerste plaats is de dimensionering en procesregeling van luchtwassers voor stallucht nog niet geoptimaliseerd voor de verwijdering van andere componenten dan ammoniak. Zo is het effect van pH en luchtverblijftijd, wat basale ontwerpparameters zijn voor ammoniakverwijdering, op geurverwijdering onvoldoende bekend. Nader onderzoek naar geursamenstelling en de processen die een rol spelen bij geurverwijdering door luchtwassers is nodig om de grote variatie in geurverwijdering te begrijpen en de relatief lage geurverwijderingsefficiëntie te verhogen.

In de tweede plaats zijn nieuwe systeemconcepten mogelijk die als doel hebben de verwijderingsefficiëntie en/of kosteneffectiviteit te verhogen. Een voorbeeld hiervan is het combineren van verscheidene wasserconcepten in een meertraps-wassersysteem, waarbij elke wastrap als doel heeft een specifiek type verbindingen te verwijderen, bijvoorbeeld een combinatie van een zure wasser met een biowasser of een biofilter. Een ander voorbeeld is het toepassen van systemen die (onder bepaalde condities) een deelstroom van de lucht behandelen en een deel van de lucht onbehandeld laten ontwijken (hoofdstuk 4). Deze benadering richt zich op het maximaliseren van de kosteneffectiviteit van ammoniakwassing door te kijken naar het emissiepatroon (luchtdebiet en concentratie) zoals dat jaarrond wordt waargenomen bij verschillende centrale ventilatiesystemen en diercategorieën. Op deze manier kunnen zowel investeringskosten als exploitatiekosten (energiegebruik, spuiwaterafzet) worden verlaagd. Om de toepassing van dergelijke deelstroom-luchtbehandelingssystemen mogelijk te maken is aanpassing van de Nederlandse wet- en regelgeving nodig. Het effect van dergelijke systemen op de emissie van fijnstof en geur, en de gevolgen hiervan voor de omgeving (hinder, gezondheidseffecten), dient nader onderzocht te worden.

Tenslotte wordt nog niet volledig begrepen welke factoren de hoogte van het ammoniakverwijderingrendement bij biowassers bepalen. Bij luchtverblijftijden en concentraties zoals die in de praktijk gevonden worden, wordt meestal een rendement in de buurt van 70% ammoniakverwijdering gevonden; soms worden beduidend hogere of lagere waarden gevonden. De metingen waarbij hogere rendementen worden gevonden wijzen er op dat de gemiddelde ammoniakverwijdering wellicht kan verhoogd worden door betere procesregeling en optimalisatie van het ontwerp. De procesregeling van biowassers zou kunnen verbeterd worden door sturing van de waswaterhoeveelheid op basis van meting van de elektrische geleidbaarheid (EC) van het waswater. Op deze manier hangt de spuiwaterhoeveelheid af van de werkelijke ammoniakbelasting van het systeem en kan ophoping van remmende verbindingen (nitriet) worden voorkomen. Het opnemen van een dergelijk regelmechanisme in toekomstige wet- en regelgeving zou handhaving en inspectie kunnen vergemakkelijken en kunnen leiden tot een verbetering van de werking van het proces.

In Nederland worden wassers voor behandeling van ventilatielucht van stallen reeds een jaar of 25 toegepast, in het bijzonder om de emissie van ammoniak tegen te gaan. In de laatste 5 jaar is de toepassing van wassers echter verveelvoudigd en wordt reeds 10% van alle ventilatielucht van varkensstallen in Nederland behandeld in een luchtwasser. Naast ammoniak is de emissiereductie van geur en fijnstof steeds belangrijker geworden. De groei van het aantal luchtwassers valt samen met een algemene tendens van schaalvergroting binnen de veehouderij. De verwachting is dat de komende jaren steeds meer luchtwassers zullen toegepast worden in Nederland en in andere gebieden met intensieve veehouderij in Europa om te kunnen voldoen aan Europese regelgeving op het gebied van natuurbescherming en luchtkwaliteit.

Dankwoord

In de eerste plaats wil ik promotor Wim Rulkens en co-promotor Nico Ogink van harte danken voor hun inzet en begeleiding, in het bijzonder bij het schrijven van de algemene discussie en het publiceren van een aantal artikelen. Daarnaast wil ik mijn dank uitspreken aan alle andere (oud)collega's die hebben meegewerkt aan de artikelen in dit proefschrift, dan wel als co-auteur, dan wel door de uitvoering van metingen en lab-analyses, dan wel door het becommentarieren van de manuscripten: Cor ter Beek, Peter de Gijsel, Max Hilhorst, Annemieke Hol, Geert Kupers, Gerben Mol, Julio Mosquera, Johan Ploegaert, Jean Slangen, Nico Verdoes, Victor van Wagenberg, Arjan van der Werf en Hans Willers. Verder wil ik mijn collega's André Aarnink, Hilko Ellen, Karin Groenestein, Jan van Harn, Michel Smits en Dick Starmans bedanken bij wie ik af en toe kon aankloppen wanneer ik advies behoefde. Iedereen, ook de niet bij name genoemde collega's: bedankt voor de prettige samenwerking door de jaren heen! Tenslotte wil ik Alida bedanken voor haar onmisbare hulp bij de opmaak van het proefschrift.

Curriculum vitae

The author, Roland Willem Melse was born on December 11, 1971 in Middelburg, the Netherlands. After finishing his secondary education (VWO) in 1990 in the same city, he spent an orientation year at the Evangelical Academy in Amersfoort. Next he moved to the Wageningen Agricultural University where he qualified cum laude with an MSc in environmental sciences in 1996. Since then, he has been working at several companies (Bioway BV, Ede; Pure Air Solutions BV, Steenwijk) and research institutes (Research Institute for Animal Husbandry (PV), Rosmalen/Lelystad; Institute of Agricultural and Environmental Engineering (IMAG), Wageningen; Animal Sciences Group, Lelystad). In 2005 he moved to South Africa with his family and spent a year and a half at the University of Potchefstroom. During his professional career, Roland's work has focused on solid waste management (*e.g.* manure treatment) and abatement of airborne emissions, both in agricultural and industrial setting. As from June 2007 he is working as a researcher at the Animal Sciences Group, a contract research group within Wageningen University and Research Centre. Here he is working on a research program funded by the Dutch Ministry of Agriculture, Nature and Food Quality (LNV) and the Ministry of Housing, Spatial Planning and the Environment (VROM), which aims to support and stimulate the use of air cleaning techniques within intensive livestock farming for emission reduction of ammonia, odour, and particulate matter. Roland is married to Alida Boonstra and they have three children; the family lives in Wageningen.

List of Publications

Refereed journal papers:

Melse, R.W.; Ogink, N.W.M.; Rulkens, W.H. (2009) Overview of European and Netherlands' regulations on airborne emissions from intensive livestock production with a focus on the application of air scrubbers. Biosyst. Eng. Vol 104 No 3 pp 289-298. doi:10.1016/j.biosystemseng.2009.07.009.

Melse, R.W.; Timmerman, M. (2009) Sustainable intensive livestock production demands manure and exhaust air treatment technologies. Bioresour. Technol. Vol 100 No 22 pp 5506-5511. doi:10.1016/j.biortech.2009.03.003.

Melse, R.W.; Ogink, N.W.M.; Rulkens, W.H. (2009) Air treatment techniques for abatement of emissions from intensive livestock production. Open Agr. J. Vol 3 pp 6-12. doi:10.2174/1874331500903010006.

Melse, R.W.; Wagenberg, A.V. van; Mosquera, J. (2006) Size reduction of ammonia scrubbers for pig and poultry houses: Use of conditional bypass vent at high air loading rates. Biosyst. Eng. Vol 95 No 1 pp 69-82. doi:10.1016/j.biosystemseng.2006.05.006.

Melse, R.W.; Ogink, N.W.M. (2005) Air scrubbing techniques for ammonia and odor reduction at livestock operations: Review of on-farm research in the Netherlands. T. ASAE. Vol 48 No 6 pp 2303-2313.

Melse, R.W.; Van der Werf, A.W. (2005) Biofiltration for mitigation of methane emission from animal husbandry. Environ. Sci. Technol. Vol 39 No 14 pp 5460-5468. doi:10.1021/es048048q.

Melse, R.W.; Verdoes, N. (2005) Evaluation of four farm-scale systems for the treatment of liquid pig manure. Biosyst. Eng. Vol 92 No 1 pp 47-57. doi:10.1016/j.biosystemseng.2005.05.004.

Melse, R.W.; Mol, G. (2004) Odour and ammonia removal from pig house exhaust air using a biotrickling filter. Water Sci. Tech. Vol 50 No 4 pp 275 - 282.

Conference papers:

Ogink, N.W.M.; **Melse, R.W.**; Mosquera, J. (2009) Development of new methods and strategies for monitoring operational performance of emission mitigation technology at livestock operations. Proceedings of the Joint International Agricultural Conference, July 6 - 8, 2009, Wageningen, Netherlands.

Ogink, N.W.M.; **Melse, R.W.**; Mosquera, J. (2008). Multi-pollutant and one-stage scrubbers for removal of ammonia, odor, and particulate matter from animal house exhaust air. Proceedings of the 8th International Livestock Symposium (ILES VIII), August 31 - September 4, 2008, Iguassu Falls, Brazil.

Melse, R.W.; Ogink, N.W.M.; Bosma, B.J.J. (2008). Multi-pollutant scrubbers for removal of ammonia, odor, and particulate matter from animal house exhaust air. Proceedings of the Mitigating Air Emissions from Animal Feeding Operations Conference, May 19 - 21, 2008, Des Moines, Iowa (IA), USA.

Ogink, N.W.M.; Mosquera, J.; **Melse, R.W.** (2008). Standardized testing procedures for assessing ammonia and odor emissions from animal housing systems in The Netherlands. Proceedings of the Mitigating Air Emissions from Animal Feeding Operations Conference, May 19 - 21, 2008, Des Moines, Iowa (IA), USA.

Aarnink, A.J.A.; Landman, W.J.M.; **Melse, R.W**.; Huynh, T.T.T. (2005) Systems for eliminating pathogens from exhaust air of animal houses. Proceedings of the 7th International Livestock Environment Symposium, May 18 - 20, 2005, Beijing, China.

Melse, R.W.; Willers, H.C. (2005) Theoretical evaluation of ammonia scrubbers for governmental approval. Proceedings of the 7th International Conference on Construction, Technology and Environment in Livestock Farming, March 2-3, 2005, Braunschweig, Germany; pp 295-300, Kuratorium für Technik und Bauwesen in der Landwirtschaft e.V. (KTBL), Darmstadt, Germany. ISBN 3-7843-2185-2.

Aarnink, A.J.A.; Landman, W.J.M.; Huynh, T.T.T.; **Melse, R.W.**; De Gijsel, P.; Fabri, T. (2004) Cleaning exhaust air from pathogens and environmental pollutants in animal houses. Proceedings of the AgEng 2004 Conference Engineering the future, September 12-16, 2004, Leuven, Belgium.

Willers, H.C.; **Melse, R.W.**; Aarnink, A.J.A.; Hoeksma, P. Technology for use of animal wastes in socially responsible agricultural practice. Proceedings of the AgEng 2004 Conference Engineering the future, September 12-16, 2004, Leuven, Belgium.

Melse, R.W.; Hilhorst, M.A. (2004) Reduction of methane emission from liquid manure storage using biofiltration. Proceedings of the International Conference on Greenhouse Gas Emissions from Agriculture - Mitigation Options and Strategies; A. Weiske (Ed.). February 10 - 12, 2004. Leipzig, Germany; pp 288-289.

Willers, H.C.; **Melse, R.W.**; Ogink N.W.M. (2003) Concentration of urine from fatteners combined with ammonia removal by scrubbing exhaust air of a pig house. Proceedings of the Ninth International Symposium on Animal, Agricultural and Food Processing Wastes, 12-15 October 2003, Durham, North Carolina, USA, pp. 584-589 (ASAE Publication Number 701P1203).

Melse, R.W.; Mol, G. (2003) Odour and ammonia removal from pig house exhaust air using a biotrickling filter. Second IWA International Workshop and Conference on Odour and VOCs. Measurement, Regulation and Control Techniques. 14-17 September 2003, Singapore.

Melse, R.W. (2002) Animal production: a challenge for environmental technology; experiences from the Netherlands. Conference on Integrated Solutions to Manure Management: Working Together on Challenges and Opportunities. Canadian Environment Industry Association. London, Ontario, Canada. September 11 - 13, 2002.

Melse, R.W.; Starmans, D.A.J.; Verdoes, N. (2002) Gaseous emissions from systems for pig slurry treatment. Proceedings of the 10th International Conference of the FAO European Co-operative Research Network on Recycling of Agricultural, Municipal and Industrial Residues in Agriculture (RAMIRAN). Strbske Pleso, Slovak Republic. May 14 - 18, 2002.

Melse, R.W.; Starmans, D.A.J.; Verdoes, N. (2002) Gaseous emissions from systems for pig slurry treatment. Proceedings of Third Symposium on Non-CO_2

Greenhouse Gases, Maastricht, Netherlands, 21-23 January 2002, pp. 51-52. ISBN 90-77017-70-4.

Hilhorst, M.A.; **Melse, R.W.**; Willers, H.C.; Groenestein, C.M.; Monteny, G.J. (2002) Reduction of methane emissions from manure. Proceedings of Third Symposium on Non-CO_2 Greenhouse Gases, Maastricht, Netherlands, 21-23 January 2002, pp. 435-440. ISBN 90-77017-70-4.

Melse, R.W.; De Gijsel, P. (2001) Systems for pig manure treatment in the Netherlands. 2001 ASAE Annual International Meeting. July 30-August 1, 2001. Sacramento, California, USA. Paper number 01-2239.

Van Wagenberg, A.V.; **Melse, R.W.** (2000) Evaporation of water from solid pig manure. AgEng 2000 International Conference on Agricultural Engineering in the Third Milennium, University of Warwick, England, 2-7 July 2000. Paper number: 00-AP-040.

Kraakman, N.J.R.; **Melse, R.W.**; Koers, B.; Van Dijk, J. (1998). Biological treatment of waste gases containing H_2S in combination with either odor or CS_2. Proceedings of the 1998 USC-TRG Conference on Biofiltration, October 22-23, 1998. Los Angeles, California, USA, p. 91-98.

Melse, R.W.; Kraakman, N.J.R. (1998). Biological treatment of waste gas containing H_2S and CS_2 combined with production of concentrated NaOH and H_2SO_4. Proceedings of Symposium of the Forum Of Applied Biotechnology, September 24-25, 1998. Gent, Belgium.

Kraakman, N.J.R.; Constanje, R.; Hamelers, H.V.M.; **Melse, R.W.** (1997) Characterisation of air flow in biofilters. Proceedings of the Fourth International Symposium of Biological Waste Gas Cleaning, Maastricht, Netherlands, April 28-29, 1997 / Eds: Prins, W. L; Ham, van, J. Düsseldorf : VDI Verlag, 1997.

The research described in this thesis was carried out at the Animal Sciences Group (ASG) which is part of the Wageningen University and Research centre (WUR, Wageningen, Netherlands), the former Research Institute for Animal Husbandry (PV), the former Institute of Agricultural and Environmental Engineering (IMAG), and the Agrotechnology and Food Innovations group (A&F).
Parts of the work presented in this thesis were financially supported by the Netherlands Ministry of Agriculture, Nature Management and Food Quality (LNV), the Netherlands Ministry of Housing, Spatial Planning and the Environment (VROM), SenterNovem (agency of the Dutch Ministry of Economic Affairs), and the Dutch Product Boards for Livestock, Meat and Eggs (PVE). All support to carry out this work is gratefully acknowledged.