

Multi-pollutant Scrubbers for Removal of Ammonia, Odor, and Particulate Matter from Animal House Exhaust Air

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Description:

Intensive poultry and pig operations concentrated in the south and east of the Netherlands are major contributors to ammonia, odor and particulate matter (PM) emissions. In the Netherlands, livestock production is responsible for 95% of the national ammonia emission causing acidification and eutrophication of natural ecosystems, and 50% of the total emission of all acidifying compounds (Koch *et al.*, 2003; EDC, 2007). Furthermore, odor 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. Finally, a large number of premature deaths and health problems are associated with the emission of particulate matter (PM) (Mokdad *et al.*, 2004; WHO, 2006b), *i.e.* tiny solid or liquid particles that are suspended in the air (e.g. dust, dirt, soot, smoke, and liquid droplets). Approximately 20% of the primary PM10 production in the Netherlands is estimated to originate from poultry and pig operations (Chardon and Hoek, 2002)¹. Besides, ammonia is a known precursor of secondary particulates so by being a significant emitter of ammonia, livestock farming also contributes indirectly to PM emissions. New air quality regulations from the World Health Organization (WHO) and European Union (EU) impose limits on PM10 and PM2.5 emissions on all major sources, including farms, to keep PM concentration in ambient air below critical standards to protect human health (EC, 1999, 2005; WHO, 2006a).

In several European countries (Germany, Denmark, Netherlands), one-stage acid scrubbers and bio-scrubbers are considered off-the-shelf techniques for effective removal of ammonia from exhaust air from pig houses and, to less extent, for poultry houses and odor removal (Melse and Ogink, 2005). In Table 1 the market size of air scrubbers is shown for livestock operations in the Netherlands as per January 1st 2008.

Table 1. Scrubber application for ammonia removal in pig and poultry operations in the Netherlands (based on statements from manufacturers, as per January 1st, 2008).

	Installed capacity (m3/hour)	Number of farms
Acid scrubbers	64 million	790
Biotrickling filters	14 million	90
Total:	79 million	880
Pig	76 million (*)	850
Poultry	3 million (**)	30
Total:	79 million	880

(*) This equals 10% of the exhaust air of all pig farms nationwide.

(**) This equals 0.4% of all exhaust air of all poultry farms nationwide.

Currently, a new generation of scrubbers is being developed for livestock operations: "multi-pollutant air scrubbers". This type of scrubber should be able to drastically reduce the emission of not only ammonia but of three pollutants:

- Ammonia
- Odor
- Particulate matter (PM10 and PM2.5).

Multi-pollutant air scrubbers usually consist of two or more scrubbing stages, each stage aims for the removal of one type of compounds. The first prototypes of multi-pollutant scrubbers for pig farms, combining the concepts of acid scrubbing, bio-scrubbing, water-curtains, and biofiltration have recently been tested in Germany (Arends *et al.*, 2006) and the Netherlands, and are in operation now at a limited number of farms. Recent research on particulate matter removal by such multi-pollutant air scrubbers showed an average removal efficiency ranging from 62 to 93% for PM10

¹ PM10 (also called inhalable particles) represents the fraction of particles that have an aerodynamic diameter of 10 µm or less; PM2.5 (also called respirable 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/m³ that has the same terminal settling velocity in the gas as the particle of interest.

and from 47 to 90% for PM_{2.5} (Aarnink *et al.*, 2007, 2008a, 2008b; 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. Data on PM removal by one-stage ammonia scrubbers (acid scrubbers or biological air scrubbers) is currently not available.

Market conditions demand an increasing scale of operation for both pig and poultry farms whereas air quality regulations restrict their size, unless emissions can be drastically reduced. Multi-pollutant scrubbers may play an important role here. It is expected that within the next years the implementation of air scrubbers will expand in intensive livestock production areas in Europe to comply with European regulations for the protection of natural ecosystems and ambient air quality. However, considerable research and development efforts are needed to keep operational costs at acceptable levels.

Recently, an innovation and implementation program has been set up by the Dutch national government that aims to stimulate the development and introduction of multi-pollutant air scrubbers. The program includes farm-scale research on five pilot locations where experimental multi-pollutant scrubbers are tested during a three-year period.

The objectives of this paper are to:

- Give an overview of technical principles applied in scrubbers for livestock operations.
- Present the preliminary results of the Dutch research program on multi-pollutant scrubbers with regard to removal efficiencies and operational parameters of these scrubbers.

Mitigation Mechanism and Applicability:

Acid scrubbers and bio-scrubbers

Since the 1990's, single-stage air scrubbers have been implemented on intensive livestock operations mainly to minimize ammonia emissions for the protection of nearby located sensitive ecosystems. Two types of scrubbers have been generally applied: 1) acid scrubbers, 2) bio-scrubbers or biotrickling filters.

Acid scrubbers are based on the entrapment of ammonia in acid liquid that is recirculated over a packed bed and the frequent discharge of the resulting ammonium salt solution at a concentration of about 150 g/L. Usually sulfuric acid is applied and pH is kept between 2 and pH 4. Melse and Ogink (2005) reported average ammonia removal efficiencies of 96% for farm-scale operated acid scrubbers. Reported average removal efficiency for odor was only 31% and showed a large variation. Acid scrubbers are considered as a state-of-the-art technique in cases where very high reductions of ammonia emissions are required.

In bio-scrubbers, or biotrickling filters, bacteria convert ammonia into nitrite and nitrate. Nitrogen concentrations in the water are kept below inhibiting levels by regular discharge of the recirculation liquid. The biomass is partly attached to the packed bed and partly suspended in the recirculation liquid. As compared to chemical scrubbers the discharge volume of biotrickling filters is about 8 to 10 times higher. Average ammonia removal efficiency at farm operations amounted 70%, whereas for odor removal a large variation was found with an average removal efficiency of 44%. In general, biotrickling filters have a higher odor removal potential than acid scrubbers because a wide array of odor components dissolved in the circulation water are broken down by the biomass, whereas in chemical scrubbers only part of the odor components are kept in solution due to a low pH.

Scrubbers are mainly applied in pig housings with central ventilation ducts. Only a few examples are known where they are applied in poultry houses. The high dust content of ventilation air increases the risks of blockage of the packing bed causing high pressure drop and increased energy use.

Multi-pollutant scrubbers

In the late 1990's, multi-pollutant air scrubbers for livestock operations were initially developed in Germany to ensure a high and sustainable removal of livestock odor (Arends *et al.*, 2006; Hartung *et al.*, 2006). The basis of this development was the known high odor removal capacity of biofilters with an organic packing material (usually a mixture of materials like compost, wood bark, wood chips, peat, perlite, and organic fibers), which is related to the huge absorption capacity per unit of volume. The functioning and lifespan of the biofilter is improved by pre-treating incoming air, leading it first through an acid scrubber and mist eliminator before entering the biofilter. This approach addresses four general disadvantages associated with the use of biofilters with organic packing material in livestock operations:

- By removing the main part of the ammonia load before entering the biofilter the formation of nitrite/nitrate salts is minimized. High ammonia loads eventually result in excessive nitrite/nitrate concentrations that block a proper functioning of micro-organisms in the biobed and leads to acidification, thus undermining the removal capacity of the bed, often without being noticed by users. In practice this can be prevented by replacement of the biofilter packing at regular intervals. Pre-treating the air allows a much longer packing lifetime, and thus reduces refilling costs.
- Biofilters have to be kept moist for adequate microbial functioning. Especially at the air inlet side the filterbed may dry out because the inlet air is not water saturated. By scrubbing the incoming air first, the air will be water saturated and helps to moisten the filter bed.

- Biofilters are sensitive to dust loads that clog the packing, increase the pressure drop, and subsequently lead to increase of ventilation energy. Together with inadequate moistening, clogging may lead to preferential air flows which will decrease the removal performance of the filter bed. By scrubbing the air first, total dust load is drastically reduced.
- The total pressure drop over the filterbed can be very high in practice (>200-300 Pa) and requires a relatively high energy input per unit of air volume. By pre-treating the air by a scrubber the height of the filter bed can be reduced thus reducing the pressure drop.

In a subsequent development the concept of specialized treatment for different compounds in consecutive scrubbing units has been further improved. Instead of one pre-treating scrubbing step, two scrubbing steps are implemented followed by a vertically oriented biobed. The first step consists of packing material over which water is recirculated to remove dust, the second step operates as an acid scrubber to remove ammonia, and the third step is designed as a biobed wall to remove the remaining odor. The collection basins are separated, and a mist eliminator is placed between the acid step and biobed. By placing the treatment steps as three consecutive walls on short distance directly after each other, the air can pass straightforward through the system without extra turns that increase the pressure drop. A few Dutch and German companies produce installations for pig farms based on this three-step approach. Further modifications involve two-phase setups leaving out the biobed, where an acid-based treatment for ammonia removal is followed by a water-based treatment to remove remaining odors, and where discharge water of the water treatment is used as recirculation liquid in the first acid-based phase. Eventually, a water-based treatment step might turn into a biotrickling step. Recently, manufacturers have started to develop and test denitrification treatment in order to reduce the amount of water that might be discharged from the biotrickling stage.

Running Research Program on Multi-pollutant Scrubbers:

Description of pilot program

In 2007, a three-year research program, funded by the Netherlands Ministry of Housing, Spatial Planning and the Environment (VROM), started that aims to promote successful application of multi-pollutant scrubbers at livestock operations. The goal of the program is to gain knowledge that will help scrubber manufacturers with the development of a new generation of scrubbers that not only remove ammonia but also achieve high removal efficiencies (at least 70%) for odor and particulate matter (PM10 and PM2.5). During a period of three years the operational parameters and removal efficiencies of ammonia, odor and particulate matter are followed on five farm locations where multi-pollutant scrubbers have been installed by four Dutch manufacturers. Based on these long-term experiences modifications and improvements will be proposed and realized. In Table 2 some characteristics of the pilot-locations are summarized. Empty bed residence times at maximum ventilation rates are between 0.1 and 1.6 s.

Table 2. Description of experimental multi-pollutant scrubbers at pilot locations in the Netherlands that are included in research program.

Pilot location	Animal category	Installed ventilation capacity (m ³ /hour)	EBRT (s)	Description ^[a]
1	30,000 broilers	75,000 ^[b]	0.43	Biotrickling filter (cross-flow) + denitrification unit
2	182 farrowing sows + 2,640 piglets	81,000	0.29	Acid scrubber + water scrubber (cross-flow)
3	400 dry and pregnant sows	60,000	1.0	Acid scrubber + water scrubber (cross-flow)
4	2,600 fattening pigs	160,000	1.6	Biotrickling filter (counter-current flow) + denitrification unit
5	21,000 broilers	180,000	0.12	Acid scrubber + water scrubber(counter-current flow)

^[a] EBRT = Empty Bed Residence Time (s), calculated as the volume of packing material (m³) divided by the air flow rate (m³/s).

^[a] A water scrubbers can be considered as a biotrickling filter on the long term as usually biomass develops.

^[b] The inlet air is treated in a heat exchanger connected to groundwater which results in lower ventilation rates.

Methods

General

The ammonia, odor and particulate matter measurements are carried out according to three protocols that are being developed by us on request by the Dutch government; the protocols will become mandatory in the near future.

Ammonia measurements

The NH₃ concentration in the inlet and outlet of the scrubber system is measured using an impinger ("bubble flask") method. A fraction of the air to be sampled 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). NH₃ is trapped by the acid and accumulates in the bottles; after 24 hours the measurement is stopped and the bottles are disconnected. Fluctuations in the NH₃ concentration of the sampled air are thus time-averaged over 24 hours. 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 NH₃ 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 NH₃. Finally, the NH₃ concentration of the air is calculated from the nitrogen content of the acid solution in the bottles, which is determined spectrophotometrically (NNI, 1998a), and the given air sampling flow rate.

The ammonia removal efficiency is expressed as $([\text{NH}_3\text{-in}] - [\text{NH}_3\text{-out}]) / ([\text{NH}_3\text{-in}]) \times 100\%$.

Odor measurements

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, from 10:00 AM to 12:00 AM. 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 m³ air (OUE m⁻³) (CEN, 2003). The accuracy of the sensory-based odor measurements is lower than the accuracy of common analytical 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%.

The odor removal efficiency is expressed as $([\text{Odor-in}] - [\text{Odor-out}]) / ([\text{Odor-in}]) \times 100\%$.

Particulate matter (PM) measurements

The dust concentrations (PM10 and PM2.5) of the incoming and outgoing air of the scrubbers were sampled for 24 hours. PM10 and PM2.5 were simultaneously measured. Dust concentrations were measured by drawing a known amount of air at a fixed air speed through a sampling head. Cyclone pre-separators for PM10 and PM2.5 within the sampling head separated the larger dust particles from the fractions that had to be measured (PM10 or PM 2.5) as described by Aarnink *et al.* (2008). PM10 and PM2.5 samples were collected on a glass fiber filter. The filters were weighed before and after sampling under standard conditions (NNI, 1998b; NNI, 2005).

The PM removal efficiency is expressed as $([\text{PM-in}] - [\text{PM-out}]) / ([\text{PM-in}]) \times 100\%$.

Preliminary results

In Table 3 the first results of the ammonia, odor and particulate matter removal efficiency measurements are shown for the different multi-pollutant scrubbers. All measurements were carried out during normal scrubber operation; when it was clear that a scrubber system was malfunctioning or had been malfunctioning recently (*e.g.* the system had ran out of acid, control parameters of the system were out of normal ranges, fresh water supply was halted) measurements were postponed for a week.

Table 3. Preliminary results of the measured removal efficiencies for ammonia, odor, and particulate matter (PM) by the farm-scale multi-pollutant scrubbers.

Ammonia removal	Odor removal	PM10 removal	PM2.5 removal
63 - 98%	0 - 83% ^[a]	41 - 46%	23 - 61%
average: 83%	average: 40%	average: 43%	average: 42%
n = 7	n = 8	n = 2	n = 2

^[a] Actually, in two cases an increase of the odor concentration was found.

Table 3 shows that the aimed ammonia removal of at least 70% can usually be achieved. The odor and PM removal, however, is much lower and needs to be increased in order to meet the requirement of 70% removal. It must be noted that the number of PM measurements is low at present. An analysis of known PM removal efficiencies by scrubbers reveals that the removal efficiency is probably proportional to the air residence time in the packing (Aarnink *et al.*, 2007, 2008a, 2008b).

Future measurements will be carried out in the coming two years in order to get reliable data on long-term performance of the multi-pollutant scrubber systems. Based on the results additional research will be carried out in order to improve performance, reliability, and stable operation of the systems. Although the multi-pollutant scrubbers are running at farm-scale size, they are still being considered as experimental systems at present.

Cost:

In Table 4 a cost-calculation is given both for acid scrubbers, biotrickling filters and multi-pollutant scrubbers, based on a newly built production facility. In case of modification of a pig house, investment costs will be higher because the ventilation system has to be modified from (usually) separately ventilated compartments to a central ventilation duct. Multi-pollutant scrubbers require a higher investment as compared to the other scrubber systems. These extra costs are related to the extra scrubbing phases that are included. Investments in extra ventilation capacity are the same for both scrubber types. It is expected that with the introduction of more multi-pollutant scrubber systems and larger series volumes, investment differences between conventional and multi-pollutant scrubbers will get smaller.

The total operational costs of multi-pollutant scrubbers are expected to be about 5% lower than for biotrickling filters. This is due to the reuse of discharge water from the biotrickling step in the acid scrubbing step, thus reducing the amount of discharge water and partly replacing the fresh water intake. As compared to acid scrubbers, the total operational costs of multi-pollutant are expected to be about 25% higher, mainly due to fixed costs of the investment.

Table 4. Investment and operational cost of scrubbers for newly built production facilities in €/ animal space (based on Arends *et al.*, 2006; Melse and Ogink, 2005; Melse and Willers, 2004; Ogink and Bosma, 2007)^[a].

	Acid scrubber	Biotrickling filter	Multi-pollutant scrubber (3-stage water/acid/biotrickling)
Investment costs	32.8	43.5	50.3
<i>Operational costs (year⁻¹)</i>			
Depreciation (10%)	2.6	3.4	4.2
Maintenance (3%)	1.5	1.8	2.0
Interest (6%)	0.8	1.0	1.2
Electricity use (€ 0.11 kWh ⁻¹)	3.3	3.8	3.7
Water use (€ 1.0 m ⁻³)	0.6	1.7	0.6
Chemical use (€ 0.6 L ⁻¹ H ₂ SO ₄ , 98%)	1.4	n/a ^[c]	0.7
Water discharge ^[b]	0.6	2.5	1.0
Total operational costs (year ⁻¹)	10.8	14.3	13.5

^[a] The investment costs are based on a maximum ventilation capacity of 60 m³ animal place⁻¹ h⁻¹.

^[b] Water disposal costs are assumed of € 10/m³ for discharge from acid scrubbing and € 2/m³ for discharge from biotrickling or water scrubbing. For the multi-pollutant scrubber, discharge water from the biotrickling or water scrubbing step is reused in the acid scrubbing step. The systems do not include a denitrification unit which might significantly decrease water discharge costs.

^[c] n/a = not applicable.

Important aspects for possible cost reduction are reduction of energy and alternative use or treatment of discharge water (e.g. denitrification, optimization of discharge control). Furthermore, decreasing scrubber size, which can e.g. be made possible by using bypass options at maximum ventilation (Melse *et al.*, 2006) or by cooling of incoming ventilation air, will reduce costs.

Technology Summary:

In The Netherlands, Germany and Denmark packed-bed biotrickling filters and acid scrubbers for removal of ammonia from exhaust air of animal houses are off-the-shelf techniques. These scrubbers are mainly applied in pig housings with central ventilation ducts; only a few are applied in poultry housings because of the relatively high dust concentrations in this air. At the moment a new generation of so-called "multi-pollutant scrubbers" is being developed and tested that not only remove ammonia but also aim for significant removal (at least 70%) of odor and particulate matter (PM10 and PM2.5) from the air. This combination provides an attractive option for large scale livestock operations to remain in operation in areas with nearby located residential areas and sensitive ecosystems. Multi-pollutant air scrubbers usually consist of two or more scrubbing stages where each stage aims for the removal of one type of compounds. Recently a 3-year research program has started that monitors and aims to improve the performance of five farm-scale multi-pollutant scrubber from different manufacturers. The preliminary results show that ammonia removal is relatively high but that the removal of odor and particulate matter needs to be improved further. Finally a detailed cost calculation is presented.

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