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Research article

Continuous measurements of ammonia, nitrous oxide and methane from air scrubbers at pig housing facilities

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ABSTRACT

Ammonia, largely emitted by agriculture, involves a great risk for eutrophication and acidification leading to biodiversity loss. Air scrubbers are widely applied to reduce ammonia emission from pig and poultry housing facilities, but it is not always clear whether their performance meets the requirements. Besides, there is a growing international concern for the livestock related greenhouse gases methane and nitrous oxide but hardly any data concerning their fate in air scrubbers are available. This contribution presents the results from measurement campaigns conducted at a chemical, a biological and a two-stage biological air scrubber installed at pig housing facilities in Flanders. Ammonia, nitrous oxide and methane at the inlet and outlet of the air scrubbers were monitored on-line during one week using a photoacoustic gas monitor, which allowed to investigate diurnal fluctuations in the removal performance of air scrubbers. Additionally, the homogeneity of the air scrubbers, normally checked by gas detection tubes, was investigated in more detail using the continuous data. The biological air scrubber with extra nitrification tank performed well in terms of ammonia removal (86 ± 6%), while the two-stage air scrubber suffered from nitrifying bacteria inhibition. In the chemical air scrubber the pH was not kept constant, lowering the ammonia removal efficiency. A lower ammonia removal efficiency was found during the day, when the ventilation rate was the highest. Nitrous oxide was produced inside the biological and two-stage scrubber, resulting in an increased outlet concentration of more than 200%. Methane could not be removed in the different air scrubbers because of its low water solubility.

1. Introduction

Ammonia can harm the environment as it involves a risk for eutrophication and acidification leading to biodiversity loss (Oenema et al., 2012). As 94% of the ammonia emissions originates from agricultural activities (UNECE/LRTAP, 2012), this sector has a great effort to make. Since 2004, newly built pig housing facilities in Flanders are legally obligated to implement low ammonia emission techniques. This can be achieved e.g. by applying air scrubbers that remove ammonia from the outgoing ventilation air through absorption in water, followed by chemical and/or biological conversions and removal of the end products.

Intense contact between the gas and liquid phase ensures mass transfer of water soluble pollutants (e.g. ammonia) from the air to the washing water, where a chemical equilibrium is established, e.g. with ammonium:

$$NH_{3(g)} + H_2O_{(l)} \leftrightarrow NH_{3(l)} + H_2O_{(l)} \leftrightarrow NH^+_{4(l)} + OH^-_{(l)} \quad (1)$$

The washing water is recirculated to reduce water consumption. Eventually, the concentration of accumulating contaminants in the washing liquid becomes too high and discharge of the washing water becomes necessary. Fresh water must be added to compensate the water loss due to discharge and to evaporation. In a chemical air scrubber an acid is added to the washing water to decrease the pH and thereby increasing the driving force as more ammonia is converted into ammonium. In biological air scrubbers, ammonium captured in the washing water is oxidized by nitrifying bacteria to nitrite (NO_2^-) and subsequently to nitrate (NO_3^-). In a combined air scrubber, two or more scrubbing stages are placed in series. These can involve a water stage, mostly followed by a chemical or a biological scrubbing stage. More details on the operating principles of biological, chemical and two-stage air

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scrubbers can be found in the review of Van der Heyden et al. (2015).

Measurements at animal housing facilities (Mosquera et al., 2011) show that the ammonia removal efficiency of 70%, which is legally required in Flanders (MB31/05/2011), is not always reached in practise. Furthermore, there is a growing international concern for the livestock related greenhouse gases methane and nitrous oxide (Gerber et al., 2013). Still little attention is being paid to the performance of air scrubbers in terms of the reduction or production of these greenhouse gases. Measurements with high time resolution are necessary to follow the dynamics and to relate the emission pattern with underlying processes affecting the emission (Mosquera et al., 2014). However, a lot of studies present only single measurements of concentrations and removal efficiencies averaged over 1 h, mostly only focusing on ammonia (Girard et al., 2012; Melse and Mol, 2004; Melse and Ogink, 2005; Mosquera et al., 2011; Zhao et al., 2011). Melse et al. (2012a) already recognized that continuous measurement are needed to investigate day-night variations and fluctuations in the removal performance and performed a study at a biological air scrubber. Furthermore, to our knowledge, no continuous measurements of nitrous oxide and methane at air scrubbers were performed until present.

The aim of this study is to investigate the fluctuations in removal efficiency and concentration for ammonia, nitrous oxide and methane from a chemical, a biological and a two-stage air scrubber placed at commercial pig houses. The incoming and outgoing concentrations of ammonia, nitrous oxide and methane were continuously measured during one week to get insight in the performances of the respective air scrubbers.

2. Materials and methods

2.1. Description of the air scrubbers

The study was conducted on three air scrubbers placed at different pig housing facilities, located in Flanders, Belgium. The main design characteristics can be found in Table 1. The scrubber packings are made from inert plastic.

The chemical air scrubber is installed at a commercial fattening pig barn with 1344 animal places (Asse, Belgium). The pH of the washing water is controlled below 4, until the maximum electrical conductivity setpoint of 180 mS cm^{-1} is reached, to stay below the maximum allowed ammonium sulfate concentration in the washing water set in Flanders of 2.1 mol L^{-1} or 58.6 g N L^{-1} (MB31/05/2011). At that point, sulphuric acid is no longer dosed until the pH reaches 7 and the washing water is discharged to a separate storage tank. This is to obtain a neutral ammonium sulfate discharge product which can be used for field applications.

The biological air scrubber was located in Linter (Belgium), treating the exhaust air of a barn housing 144 sows and 3120 piglets. The installation was extended with an additional nitrification tank with bacteria grown in granular sludge to treat the washing water before reuse. In Flanders, the maximum allowed concentration of nitrogen in the biological washing water is set to 3.2 g N L^{-1} (MB31/05/2011), which corresponds to an EC setpoint of approximately 18 mS cm^{-1}. When this value is reached in the biological scrubbing section, part of the washing water is discharged towards to manure pit of the housing facility and fresh water is added.

The two-stage air scrubber (Mollem, Belgium) consisted of a water stage to remove dust, followed by a larger biological stage of 1.80 m packing material with a lower specific surface area. When an EC setpoint of 15 mS cm^{-1} in the biological scrubber section (stage 2) is reached, the washing water from the water scrubbing section (stage 1) is discharged to the manure pit of the animal housing facility, the washing water from stage 2 flows to stage 1 and fresh water is added to stage 2. In this way, the washing water of section 1 can have a higher nitrogen content without inhibiting the nitrifying bacteria in stage 2. This scrubber was installed at a fattening pig housing facility of 1515 animal places.

2.2. Measuring equipment

Gas concentration measurements of NH_3, CH_4 and N_2O were performed using an Innova photoacoustic gas monitor 1314 (Table 2) connected to a CBISS multipoint sampler (LumaSense Technologies, Denmark). The analyzer was calibrated by the manufacturer and validated with 30 ppm ammonia from a gas bottle before the measurements started. This equipment is listed as one of the prescribed measuring techniques in the VERA test protocol for air cleaning technologies (VERA, 2010), which provides a standardized protocol in Europe to test the efficiency of air scrubbers. Using a photo-acoustic monitor not only allows to measure ammonia at a high time resolution but also allows the measurement of nitrous oxide and methane at the same time and is therefore chosen in this study. However, caution must be paid when measuring low concentrations ($<$2 ppm NH_3) as they could be overestimated when using this system (Mosquera et al., 2014; Osada et al., 1998). Measuring multiple gases with an infrared photoacoustic monitor can also have an effect of non-compensated interferences which could lead to under- or overestimations (Hassouna et al., 2013; Zhao et al., 2012). This is especially important when concentrations are measured to determine emission factors (absolute values) but is of minor importance when the removal efficiency is calculated as the difference between incoming and outgoing concentrations is used (relative values). After calibration, cross-interferences are normally compensated, allowing proper operation of the photoacoustic gas monitor.

The gas flow rate through the air scrubber, or thus the ventilation rate of the housing facility, was estimated based on continuous pressure difference measurements over the ventilation fans with a P26 differential pressure transducer (halstrup-walcher, Germany) which were linked at certain points with the estimated ventilation rate given on the climate control computer inside the housing facility. Ventilation rates are expressed as a percentage of the maximum ventilation rate. Temperature and relative humidity of the incoming and outgoing air were measured using thermocouples (EE071 probe, E+E Elektronik, Germany). The washing water of each scrubber was sampled at least three times during each measuring campaign and analysed in the lab for pH and electrical conductivity (EC) according to EN 13037 and EN 13038. For the chemical air scrubber, the washing water was further analysed for NH_4^+ and SO_4^{2-}, for the biological and two-stage air scrubber for NH_4^+, NO_2^- and NO_3^-, using extraction and ion chromatography according to EN 13652.

2.3. Measuring strategy

Continuous measurements were performed on each air scrubber during one week in June 2014. Gas samples were taken sequentially at five sampling points; one before and four behind the respective air scrubber. Each sampling consisted of at least 6 consecutive measurements with a 3 min time interval. Only the last measurement was taken into account to overcome a possible measuring delay of the gas monitor. The sampling point before the scrubber in the pressure chamber included a dust filter to prevent dust to enter the measurement device. The sampling tubes behind the scrubber were heated to 110 °C to prevent condensation. Dilution by wind was avoided by using protective ducts around the sampling points. It was chosen to sample a number of four points behind the scrubbers to compromise minimal time resolution and