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## Enhancing recovery of ammonia from swine manure anaerobic digester effluent using gas-permeable membrane technology

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## ABSTRACT

Gas-permeable membrane technology is useful to recover ammonia from manure. In this study, the technology was enhanced using aeration instead of alkali chemicals to increase pH and the ammonium ( $\text{NH}_4^+$ ) recovery rate. Digested effluents from covered anaerobic swine lagoons containing 1465–2097 mg  $\text{NH}_4^+\text{-N L}^{-1}$  were treated using submerged membranes ( $0.13 \text{ cm}^2 \text{ cm}^{-3}$ ), low-rate aeration ( $120 \text{ mL air L-manure}^{-1} \text{ min}^{-1}$ ) and nitrification inhibitor ( $22 \text{ mg L}^{-1}$ ) to prevent nitrification. The experiment included a control without aeration. The pH of the manure with aeration rose from 8.6 to 9.2 while the manure without aeration decreased from 8.6 to 8.1. With aeration, 97–99% of the  $\text{NH}_4^+$  was removed in about 5 days of operation with 96–98% recovery efficiency. In contrast, without aeration it took 25 days to treat the  $\text{NH}_4^+$ . Therefore, the recovery of  $\text{NH}_4^+$  was five times faster with the low-rate aeration treatment. This enhancement could reduce costs by 70%.

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## 1. Introduction

Ammonia ( $\text{NH}_3$ ) emissions to the atmosphere are an environmental quality concern because they can contribute to eutrophication of surface waters, nitrate contamination of ground waters, and impair air quality (EPA, 2014). In the United States, the largest source of  $\text{NH}_3$  is livestock farming;  $\text{NH}_3$  emissions from animal husbandry operations (dairy, beef, poultry and swine) were estimated at 2.4 million tons/year in 2010 and 2.5 million tons/year in 2015 (EPA, 2014). In its volatile form,  $\text{NH}_3$  is a cause of air pollution and can create health problems for neighboring residents (Wing and Wolf, 2000). Ammonia runoff and subsequent accumulation in water sources leads to eutrophication and destruction of marine habitats (Paerl, 2006). On the other hand,  $\text{NH}_3$  is a valuable chemical for use in agricultural fertilizers and in the chemical industry. Current practices for  $\text{NH}_3$  production are energy intensive and contribute to global warming (Funderburg, 2013; IFA, 2009); manufacturing one metric tonne of anhydrous  $\text{NH}_3$  fertilizer requires  $1043 \text{ m}^3$  of natural gas. Therefore, developing new methods for removal and recovery of  $\text{NH}_3$  from swine manure is desirable for environmental and economical reasons.

Ammonia mitigation techniques for livestock farming typically focus on five areas: reduction of nitrogen (N) excretion through

dietary modifications, reduction of volatile N, building designs and manure managements, land application strategies, and emission capture and treatment (Ndegwa et al., 2008). Among technologies that focus on  $\text{NH}_3$  emission capture and treatment, some are focused on the recovery of the N for further use. These technologies include: (1) wet scrubber and stripping technologies (proposed for ammonia removal from swine manure wastewaters) (Bonmati and Flotats, 2003; Liao et al., 1995; Lin et al., 2014), (2) struvite precipitation with phosphate and magnesium (Nelson et al., 2000), (3) reverse osmosis using osmotic pressure (Masse et al., 2010), (4) ion exchange adsorption with zeolites (Milan et al., 1997), and (5) a gas-permeable membrane process at low pressure (Vanotti and Szogi, 2015).

The gas-permeable membrane process includes the passage of gaseous  $\text{NH}_3$  through a microporous hydrophobic membrane and subsequent capture and concentration in an acidic stripping solution on the other side of the membrane (Fig. 1). The membrane manifolds are submerged in the liquid manure and the  $\text{NH}_3$  is removed from the liquid before it escapes into the air (Vanotti and Szogi, 2011, 2015); the  $\text{NH}_3$  permeates through the membrane pores reaching the acidic solution located on the other side of the membrane. Once in the acidic solution,  $\text{NH}_3$  combines with free protons to form non-volatile ammonium ( $\text{NH}_4^+$ ) ions that are converted into a valuable  $\text{NH}_4^+$  salt fertilizer. The process is responsive to increased pH through addition of alkali chemicals (García-González and Vanotti, 2015), which leads to an increased release of  $\text{NH}_3$  from the manure and capture by the membrane (Fig. 1).

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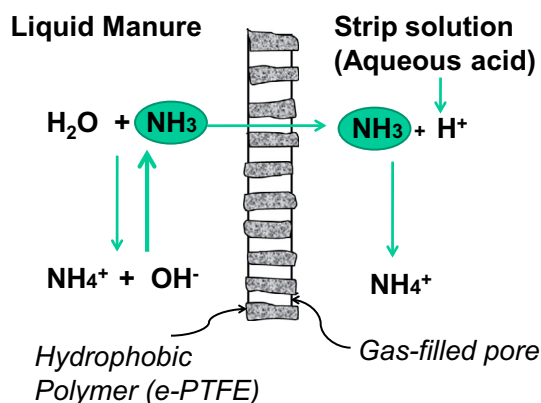


Fig. 1. Experimental device for  $\text{NH}_4^+$  capture from manure using gas-permeable membranes and low-level aeration to increase manure pH.

Gas-permeable membranes have been shown to effectively recover more than 98% of  $\text{NH}_4^+$  from liquid swine manure (Garcia-Gonzalez et al., 2015; Garcia-Gonzalez and Vanotti, 2015; Vanotti and Szogi, 2015). Zarebska et al. (2015) reviewed the pros and cons of six ammonia recovery methods including nanofiltration, reverse osmosis, gas-permeable membrane process (membrane distillation), air stripping, zeolite ion exchange, and struvite chemical precipitation and indicated the energy consumption for the gas-permeable membrane process was among the lowest ( $0.18 \text{ kW h kg NH}_3^{-1}$ ). For example, comparing gas-permeable membranes with air stripping, which both produce liquid ammonium sulfate, the energy consumption for the gas-permeable membrane process is 18 times lower than for air stripping. The main drawback from gas-permeable membrane systems is the cost of alkali chemicals to increase manure pH (Zarebska et al., 2015). Therefore, a strategy to reduce costs of the gas-permeable membrane process and improve farmer's adoption is to seek a simple and inexpensive alternative for raising the pH of the manure in a farm setting.

Vanotti and Szogi (2015) proposed the use of gas-permeable membranes with aeration instead of alkali chemicals to enhance the removal and recovery of  $\text{NH}_4^+$  from livestock effluents. Such conditions applied to stored livestock effluents results in a pH increase of about 1 unit and increased  $\text{NH}_3$  release. This effect has been demonstrated in experimentation involving the aeration of swine manure. In one study, passing air, 0.5%  $\text{O}_2$  or 4.9%  $\text{O}_2$  gas mixtures through slurry caused an increase in pH of about 1 unit in 1–2 days and about 2 units in 10 days (from 7 to between 8.5 and 9) (Stevens and Cornforth, 1974). Another study showed that aeration of swine lagoon wastewater without nitrification increased the pH of wastewater 1.5 units, from 7.5 to 9, in the first 18 h (Vanotti and Hunt, 2000). Others showed continuous aeration of manure increased pH almost 2 units (Zhu et al., 2001). In order to recover  $\text{NH}_3$  using gas-permeable membranes with aeration, nitrification must be inhibited or else it will oxidize  $\text{NH}_3$ , decrease pH, and affect overall  $\text{NH}_4^+$  recovery efficiency (Vanotti and Szogi, 2015). Nitrification inhibition can be achieved in various effective ways, for example: reducing aeration rates, reducing nitrifying biomass, increasing temperatures, or adding a commercial nitrification inhibitor (Vanotti and Szogi, 2015).

Using raw swine manure that contained high  $\text{NH}_4^+$  concentration and high carbon (chemical oxygen demand  $17 \text{ g L}^{-1}$ ), Garcia-Gonzalez et al., 2015 showed that 98% recovery of  $\text{NH}_4^+$  can be obtained with gas-permeable membranes using low-rate aeration for increasing pH while reducing operational costs by 57% when compared to alkali chemical addition. The objective of this research was to determine if the aeration approach – with nitrification inhi-

bition – is also effective to increase pH and recover  $\text{NH}_4^+$  from anaerobically digested effluents containing high  $\text{NH}_4^+$  concentration and low organic carbon (chemical oxygen demand  $< 2.5 \text{ g L}^{-1}$ ). We used anaerobically digested manure effluent from two swine farms with covered anaerobic lagoons in North Carolina, USA.

## 2. Methods

### 2.1. Experimental procedure

Batch experiments were conducted in 2-L wastewater vessels made of polyethylene terephthalate (PET) with an effective volume of 1.5 L (Fig. 2). The acid tank used to concentrate the  $\text{NH}_4^+$  consisted of 500-mL Erlenmeyer flasks with 250 mL of a 1 N sulfuric acid ( $\text{H}_2\text{SO}_4$ ) stripping solution. This stripping solution was continuously recirculated using a peristaltic pump (Cole-Parmer, Masterflex L/S Digital Drive, Illinois, USA) at  $4 \text{ mL min}^{-1}$  through a tubular gas-permeable membrane submerged in the reactor. In the aerated treatments, air was delivered to the bottom of the manure vessel at a low-rate of  $0.18 \text{ L-air min}^{-1}$  ( $0.12 \text{ L-air L-manure}^{-1} \text{ min}^{-1}$ ) using an aquarium pump, a shielded air flow meter with a precision valve (GF-9260, Gilmont Instruments, Illinois, USA) and an aquarium diffuser stone that provided fine bubbles. This low airflow rate was selected to effectively increase the pH of manure based on preliminary aeration tests and at the same time avoid nitrification of the  $\text{NH}_4^+$  (that reduces pH in manure). Aeration rate was half the aeration rate used in the experiments of Garcia-Gonzalez et al. (2015) with raw swine manure ( $0.24 \text{ L-air L-manure}^{-1} \text{ min}^{-1}$ ), and about 8 times lower than aeration rates used by Magri et al. (2012), that greatly inhibited nitrite production activity in experiments of partial nitrification of swine wastewater ( $0.9 \text{ L-air L-liquid}^{-1} \text{ min}^{-1}$ ). Another strategy to avoid nitrification was the addition of a commercial nitrification inhibitor (Vanotti and Szogi, 2015). In this study we used both low-aeration and a nitrification inhibitor (nitrapyrin) to stop  $\text{NH}_4^+$  oxidation in the aerated treatments. The vessels were not sealed and had 5 ports in the lid: two ports for acid recirculation, one sampling port, one port for aeration and one port that remained open to allow air to escape.

Gas-permeable membrane made of expanded polytetrafluoroethylene (ePTFE) (Phillips Scientific Inc., Rock Hill, SC) with a length of 60 cm, outer diameter of 10.25 mm and wall thickness

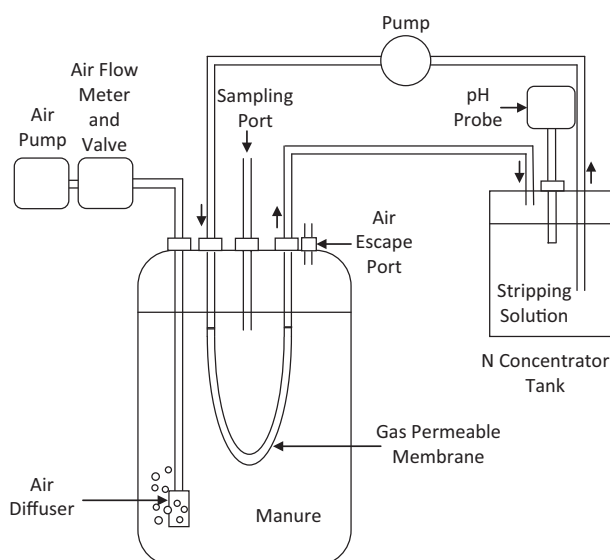


Fig. 2. Process diagram of gas-permeable membrane system for removal and recovery of anaerobically digested swine effluent.

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