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Recovery of ammonia from poultry litter using flat gas permeable membranes

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ABSTRACT

The use of flat gas-permeable membranes was investigated as components of a new process to capture and recover ammonia (NH_3) in poultry houses. This process includes the passage of gaseous NH_3 through a microporous hydrophobic membrane, capture with a circulating dilute acid on the other side of the membrane, and production of a concentrated ammonium (NH_4) salt. Bench- and pilot-scale prototype systems using flat expanded polytetrafluoroethylene (ePTFE) membranes and a sulfuric acid solution consistently reduced headspace NH_3 concentrations from 70% to 97% and recovered 88% to 100% of the NH_3 volatilized from poultry litter. The potential benefits of this technology include cleaner air inside poultry houses, reduced ventilation costs, and a concentrated liquid ammonium salt that can be used as a plant nutrient solution.

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1. Introduction

Volatilization of NH_3 gas from manure is one of the largest environmental concerns associated with confined poultry production. Excessive NH_3 accumulation in poultry housing air can adversely affect the health of both workers and birds (Kirychuk et al., 2006; Rylander and Carvalheiro, 2006). Several studies have shown the detrimental effect of high levels of NH_3 on bird performance (Dawkins et al., 2004; Ritz et al., 2004; Yahav, 2004). Increased ventilation can lower the NH_3 in poultry houses to safe levels, but energy costs during winter months represent a significant expense to the growers (Xin et al., 1996). Since NH_3 cannot be effectively contained within the house structure, NH_3 emissions may contribute to air pollution, atmospheric deposition, and health concerns for nearby residents (Wing and Wolf, 2000; Wheeler et al., 2006). Due to the high cost of commercial NH_3 fertilizers in today's marketplace, the conservation and recovery of nitrogen (N) are an important agricultural issue. Thus, there is major interest among producers and the public in implementing best control technologies that will abate NH_3 emissions from confined poultry operations by capturing and recovering N.

Technologies used by the poultry industry for NH_3 abatement can be classified into four broad categories based on their mode of action. The first and most widely used technology is simply increasing ventilation to keep NH_3 levels down inside the poultry houses (Xin et al., 1996). The second technology prevents the release of NH_3 into the environment by treating the exhaust air from the house ventilation system using scrubbing or filtration techniques. These techniques remove NH_3 from livestock houses by forcing the house air through an NH_3 trap, such as an acidic solution (scrubbers), or through a porous filter with nitrifying biofilms that oxidize NH_3 to nitrate – biotrickling or organic filters (Melse and Ogink, 2005; Pagans et al., 2005). The third technology uses dedicated ventilation systems independent of the house ventilation system to selectively pull and treat the air near the litter surface, where NH_3 levels are more concentrated (Lahav et al., 2008). The fourth approach involves directly mixing chemical amendments into the poultry litter to prevent NH_3 volatilization, without additional ventilation. These amendments act by either inhibiting microbial transformation of organic N into NH_3 or by conversion of volatile NH_3 into non-volatile NH_4 via acidification. Chemical amendments such as alum [$Al_2(SO_4)_3 \cdot 14\ H_2O$], sodium bisulfite ($NaHSO_4$), and acidified clays have been widely used to control or reduce NH_3 release from poultry litter and manure (Moore et al., 1995; Rothrock et al., 2008, 2010a). Using these amendments, N is conserved un-volatilized in the poultry litter but NH_3 is not recovered as a separate product as with the scrubbing methods. Recovery of NH_3 is a desirable feature because it can be exported off the farm, solving problems of N surpluses in concentrated poultry production regions.

This study describes a novel NH_3 removal approach using gas-permeable flat membranes placed inside the poultry house near the NH_3 source that combines some of the advantages and benefits of the technologies mentioned above. This new technology recovers N in a concentrated, purified form using the concept of integrated membrane separation and gas absorption also shared by hollow fiber membrane contactor techniques (Pabby and Sastre,

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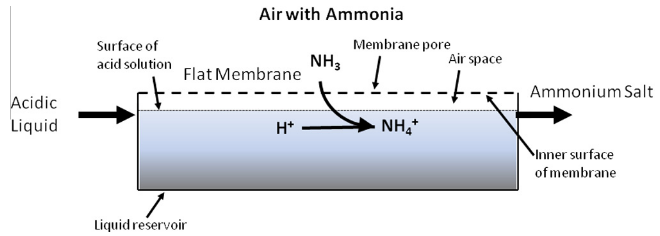

Fig. 1. Schematic diagram of NH_3 capture using flat, hydrophobic, gas-permeable membranes. The NH_3 gas permeates through hydrophobic membrane walls with micron-sized pores, where it combines with the free protons (H^+) in the acid solution to form non-volatile ammonium ions (NH_4^+).

2013). This concept includes the passage of gaseous NH_3 through microporous, hydrophobic, gas-permeable membranes and its capture in a circulated acidic solution with concomitant production of a concentrated NH_4 salt. Once NH_3 gas passes through the membrane and is in contact with the acidic solution, it reacts with free protons (H^+) to form non-volatile ammonium (NH_4^+) salt, which is retained and concentrated in the acidic solution (Fig. 1).

Hydrophobic gas-permeable membranes are made of different materials such as polypropylene (Shindo et al., 1981), polyethylene-polyurethane composites (Lee and Rittmann, 2000), and polytetrafluoroethylene (Blet et al., 1989). As a device that accomplishes mass transfer in gas–liquid systems, membrane contactor modules using a bundle of microporous tubular membranes have been studied in separation processes for gas stripping and absorption (Mansourizadeh and Ismail, 2009; Yang and Cussler, 1986). In these modules, gas flow or air movement accelerates the mass transfer between the gas and liquid phases. In fact, increased NH_3 gas flow in membrane contactors accelerates NH_3 removal rate (Klaassen et al., 2008). However, in a previous study, we found that tubular gas-permeable membranes made of expanded polytetrafluoroethylene (ePTFE) were effective for removing gaseous NH_3 from enclosures containing poultry litter in absence of air movement/ventilation (Rothrock et al., 2010b) and that they could be equally effective placed above or below the litter surface.

Tubular membrane performance can be affected by changes in the pressure of the liquid. Increasing pressure in the liquid side can lead to membrane pore wetting and drastically reduce gas absorption performance in long-term operation (Mansourizadeh and Ismail, 2009). As shown in Fig. 1, our design using flat membranes configured for removing NH_3 from poultry litter could be an advantage to tubular membranes because the liquid absorbent is not in direct contact with the membrane. The objective of this study was to test the feasibility of using flaePTFE, gas-permeable membrane as a new approach to remove and recover NH_3 from poultry litter. To achieve this, flat ePTFE membrane manifolds were developed and installed in bench- and pilot-scale chambers using a circulated acidic solution to evaluate their ability to recover and concentrate N from poultry litter under normal and enhanced NH_3 volatilization conditions. Results from this study and Rothrock et al. (2010b) have been used in the filing of a U.S. Patent on the process (Szögi et al., 2011).

2. Materials and methods

2.1. Gas-permeable membrane

The flat gas-permeable membrane was made of ePTFE with a thickness of 0.044 mm and a bubble point of 21 kPa (FL1001, Phillip Scientific, Rock Hill, SC). The ePTFE membrane was supported with a 0.229-mm thick spun-bond polypropylene fabric that faced the acid.

2.2. Process configurations

Two laboratory experiments and one pilot-scale prototype experiment were performed to test the feasibility of using flat membranes to capture and recover NH_3 volatilized from poultry litter. The basic process configuration used in both laboratory and pilot-scale experiments in this study is shown in Fig. 2. An acidic solution (1 *N* H_2SO_4) contained in an acid tank was continuously re-circulated using a peristaltic pump into an air-tight enclosure containing the poultry litter. Once inside the enclosure, the acid was contained and circulated though a plastic reservoir covered by the flat, gas-permeable membrane sheet, allowing for the passage of NH_3 gas emitted by the litter and subsequent recovery and concentration of the N as an NH_4 salt. For all bench- and pilot-scale experiments, NH_3 concentrations in the headspace of the chamber were measured using the Dräger Chip Management System (Dräeger Safety, Inc., Pittsburgh, PA). The NH_3 chips were in the measuring range of 100–2000 μg NH_3 L^{-1} (20 °C) with an accuracy of ±10% of the measured values according to the manufacturer. The instrument was further tested under laboratory conditions (21 °C) with a 9% accuracy of the measured values using a 500 μg L^{-1} certified NH_3 standard (National Specialty Gases, Durham, NC).

2.3. Laboratory experiments

Two bench-scale experiments were performed to test the feasibility of using flat ePTFE membranes in conjunction with an acidic solution to capture and recover NH_3 volatilized from poultry litter under natural and enhanced conditions using lime [Ca $(OH)_2$] to accelerate NH_3 volatilization. In both experiments, lime raised the pH of the litter to convert available non-volatile ammonium-N (NH_4–N) to volatile NH_3–N. The addition of lime was to test how quick NH_3 could be recovered from the litter but also because lime has been historically used for disinfection and NH_3 management of poultry litter (Yushok and Bear, 1948; Shah et al., 2012). The bench-scale experiments included two replicates. The acid solution and headspace air were sampled daily. The pH of the acidic solution was monitored using pHydrion Insta-Chek 0–13 litmus paper (Micro Essential Laboratory, Brooklyn, NY). In all experiments, the pH values of the liquid in the acid tank were always below pH 2, which is an indication of a 100% NH_3 absorption efficiency (Lahav et al., 2008; Rothrock et al., 2010b). Liquid acid samples from the acid tank (0.3 mL) were diluted into 2.7 mL ultrapure water and stored in capped vials at 4 °C until analysis. Final litter samples were taken once the enclosures were opened after the last air/acid sampling; all litter samples were stored at −20 °C until analysis. The experiments were performed in a laboratory at ambient pressure and temperature conditions (23–25 °C).

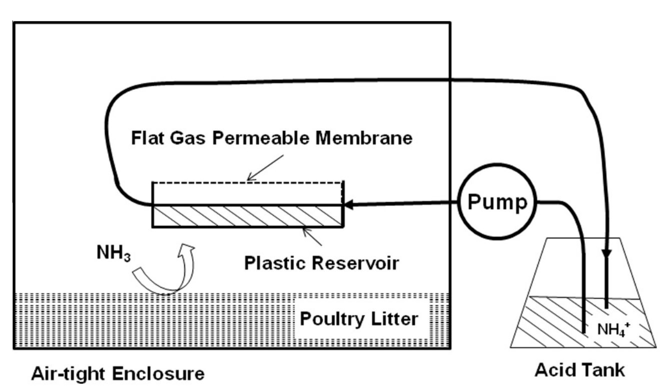

Fig. 2. Schematic diagram of the basic process for NH_3 recovery.

2.3.1. Experiment 1

The basic concept and general feasibility of a flat gas-permeable membrane for NH_3 recovery from poultry litter were tested using a flat membrane prototype shown in Fig. A.1a. The prototype consisted of a 200-mL polyvinyl chloride (PVC) plastic reservoir, a flat membrane with a surface area of 196 cm^2 (14.0 × 14.0 cm), and a PVC frame lid with uniformly spaced openings (3.9 × 1.0 cm) to fasten the membrane onto the reservoir.

The depth of the acid solution flowing inside the plastic container was controlled by the height of the acid outflow port. This configuration of the acid solution outflow created an air space of about 1.0 cm between the inner surface of the membrane and the surface of the acidic liquid (Fig. A.1b). The membrane manifold was tested in an enclosure (45.7 × 45.7 × 21.6 cm) that possessed in-flow and out-flow ports for the acid solution, as well as an access port near the top of the enclosure to measure headspace NH_3. Tygon tubing (4.75 mm I.D, 6.35 mm O.D., 0.8 mm wall; Cole Parmer, Vernon Hills, IL) was used for the inflow and outflow lines outside of the chamber connected to the acid tank. The acid tank consisted of a 2-L glass flask containing 0.3 L 1.0-*N* H_2SO_4 which was re-circulated at a rate of approximately 3.0 L d^{-1} using a Manostat pump (Cole Parmer). The enclosure contained 200 g of poultry litter that was amended with 2% (w v^{-1}) lime [56.0 g Ca$(OH)_2$ kg^{-1}] to accelerate the NH_3 volatilization.

2.3.2. Experiment 2

A second bench-scale prototype evaluated the flat membrane assembled in a trough module (Fig. A.2). It was evaluated for its technical feasibility to recover NH_3 under a range of increasing NH_3 concentration levels in the headspace. Experiment 2 tested two identical prototypes contained in two separate enclosures (45.7 × 45.7 × 21.6 cm each) within a Plexiglas box (862-CGA, Plas Labs, Lansing, MI). As in experiment 1, each enclosure possessed in-flow and out-flow ports for the acid solution, as well as an access port near the top of the enclosure to measure headspace NH_3. Each enclosure contained 1000 g of poultry litter split within two plastic bins arranged on either side of the plastic membrane module. Each membrane module consisted of a PVC trough (Model No. 400, NDS, Inc., Fresno, CA) with an U-shape (29.0 cm × 11.0 cm (top) × 8.0 cm (interior depth); Fig. A.2a). A PVC frame cover (Model No. 241-1, NDS, Inc.) with large openings (7.0 × 1.0 cm) that were uniformly spaced (0.5 cm) was used to fasten the membrane assembly to the trough. The depth of the acid solution flowing inside the trough was about 2.5 cm (dictated by height of the acid outflow port), and the depth of the membrane air space between the surface of the acid and the inside of the membrane was about 4.0 cm (Fig. A.2). The flat ePTFE membrane on the trough module had a surface area of 0.0213 m^2. The acid tank consisted of a 2-L glass flask containing 1.0 L 1.0-*N* H_2SO_4 solution which was re-circulated at a rate of 10.0 L d^{-1} using a Manostat pump.

To increase NH_3 volatilization in the headspace, lime was applied at increasing rates [2.8, 5.6, 14, and 56 g of $Ca(OH)_2$] to 1000-g poultry litter samples to have lime treatments equivalent to 0.1, 0.2, 0.5, and 2% w v^{-1}, respectively. Each treatment was run in duplicate and a control treatment with no lime (0%) was included. After mixing the lime, the litter was evenly split between the two plastic bins and placed within the Plexiglas enclosure. Prior to sealing the enclosure, litter samples (10.0 g) were retrieved from each bin and pooled to form the initial litter sample. Triplicate acid samples and headspace air readings were taken daily for 4 d. Litter samples were taken once the enclosure was opened after the last air/acid sampling at the end of the experiment. In situations where gaseous NH_3 concentrations exceeded the upper detection limit of the Dräger Chip Management System (2000 μL L^{-1}), headspace NH_3 concentrations were estimated from the same time point measurements using the same chamber but with 200 g of amended poultry litter.

2.4. Pilot scale experiment

A pilot-scale prototype was tested in Florence, South Carolina. It consisted of a flat membrane trough manifold system (Fig. A.3) contained within a 2.51-m^3 enclosure (Deluxe Cold Frame G-80, The Greenhouse Catalog, Salem, OR). The enclosure had inflow and outflow ports for the acid solution as well as an access port at the top of the enclosure for headspace NH_3 measurements. Tygon tubing (4.75 mm I.D, 6.35 mm O.D., 0.8 mm wall; Cole-Parmer) was used for the inflow and outflow lines outside of the chamber. Inside the enclosure, 32.5 kg of poultry litter was contained within a large plastic bin (1.22 × 1.22 × 0.18 m). The flat membrane manifold system was placed on the perimeter of the plastic bin. The manifold system consisted of four troughs with the dimension 130 cm (length) × 11.0 cm (top) × 8.0 cm (interior depth), each connected in series with an acid flow pipe. The combined membrane surface area of the four troughs was 0.3716 m^2. The volume of acid solution was adjusted to maintain a fluid connection through the manifold system. The acid tank contained 13.5 L of diluted acid solution (1.0-*N* H_2SO_4) which was re-circulated at a rate of 27 L d^{-1} using a Manostat pump. To ensure acid flow through the system, the four troughs of the manifold were placed at successively lower heights to produce a gravity flow within the manifold system.

Lime was added to 32.5 kg of poultry litter at rates of 0.0 and 56.0 g kg^{-1} to achieve 0 and 2% w v^{-1} lime treatments, respectively. After homogenization, initial samples (50 g) were taken by pooling 10 randomly selected 5-g grab samples. The enclosure was closed and sealed using an air-tight foam sealant. Triplicate acid samples and headspace air readings were taken daily for 9 (0% lime) or 13 days (2% lime). The pH of the acid tank was monitored daily using a pH combination electrode (Denver Instruments, Bohemia, NY). The pH values of the acid tank during the pilot scale experiment were in the range of 0.7–0.9 units. Air temperature inside the enclosure was monitored using temperature data loggers (SI-23039-52; Cole Parmer). Mean and standard deviation of temperatures within the enclosure of the pilot experiment were 31.7 ± 7.0 °C with 0% lime and 26.2 ± 4.8 °C with 2% lime treatment.

2.5. Poultry litter characteristics

The bedding material that constituted the base of the broiler litter in all bench- and pilot-scale experiments was wood chips. Broiler litter used for the experiments was collected from a 25,000-bird broiler house in Lee County, South Carolina. At the time of sampling, the house was empty and in between flocks during the middle of their annual production cycle (five flocks per year). Large composite litter samples were taken in two transects along the house and in its center section (between water lines). They were placed in 160-L containers. The containers were sealed and transported to the laboratory. For the bench-scale experiments, a 15-kg portion of the litter was passed through a 5.8-mm sieve and placed in cold storage (−65 °C) prior to laboratory experiments. For the pilot-scale experiment, the litter was placed in cold storage (−20 °C) until placement within the experimental enclosure. The properties of the litter are listed in Table 1.

2.6. Analytical methods

All liquid samples were analyzed for NH_4-N according to Standard Method 4500-NH_3 G (APHA, 1998). Total Kjeldahl N (TKN) in solid samples was determined in digestion extracts using H_2SO_4 (Gallaher et al., 1976). The NH_4–N and NO_3–N were extracted from the litter using a 60:1 2 M KCl:litter mixture that

Table 1
Poultry litter properties.

Parameter	Unit	Bench-scale	Pilot-scale 0% Lime	Pilot-scale 2% Lime
Moisture content	%	24.2	22.3	20.6
Volatile solids	%	73.7	76.1	80.0
pH	–	8.78	8.59	8.82[a]
TKN[a]	g kg^{-1}	33.8	46.8	34.3
NH_4–N[b]	g kg^{-1}	2.11	5.11	1.88
Bulk density	g cm^{-3}	0.36	0.39	0.38

[a] Initial pH value before lime addition.
[b] Dry weight basis.

was shaken (200 oscillations min^{-1}) for 30 min followed by gravity filtration through Whatman filter paper, size 42 (Whatman International, Maidstone, England) (Peters et al., 2003). All NH_4–N, NO_3–N, and TKN analyses in solid samples were determined by colorimetry using the AutoAnalyzer II (Technicon Instruments Corp., Tarrytown, NY). All litter analyses were reported on a dry weight basis. Moisture content of the poultry litter was determined by oven drying the litter at 105 °C to constant weight. The dried sample was ignited in a muffle furnace at 550 °C for 30 min to determine volatile solids (VS). Litter pH was measured electrometrically using a combination pH electrode at a 5:1 de-ionized water to litter ratio. Data were statistically analyzed by means and standard errors (proc MEANS), analysis of variance (proc ANOVA), and least significant difference at a 0.05 probability level ($LSD_{0.05}$) for multiple comparisons among means with SAS Version 9.2 (SAS Institute, Cary, NC).

3. Results and discussion

3.1. Feasibility of flat gas-permeable membrane for nitrogen recovery

In experiment 1, changes in gaseous NH_3 concentration levels within the enclosure headspace and the mass of NH_4–N recovered in the acid tank using the flat membrane tray system are shown in Fig. 3. Gaseous NH_3 concentrations increased rapidly to 1370 μL L^{-1} in the headspace of the enclosure within the first day as a result of liming. These NH_3 gas concentrations decreased quickly to a uniform level (36.5 μL L^{-1}) within the headspace enclosure in the next two days. The total reduction in NH_3 gas concentration was 97.3%. Simultaneously, the daily increase of NH_4–N mass recovered in the acid solution matched the decrease in NH_3 gas concentration (Fig. 3). By the third day of the experiment, 337.2 mg (94.5%) of the initial mass of NH_4–N (356.2 mg, dashed line in Fig. 3) in poultry litter was recovered in the acid solution.

From the results of this first prototype experiment with a flat, hydrophobic, gas-permeable membrane, it was concluded the following: (1) this systems was effective to both reduce NH_3 gas from poultry litter and recover the volatilized NH_3 in a non-volatile liquid NH_4 salt form; and (2) a prototype design containing an air space between the inner side of the gas-permeable membrane and the surface of the acid solution was efficient to remove and recover NH_3 from poultry litter (Fig. 1). The air space between membrane and absorbing liquid is an advantage of the prototype designed in this study over tubular membrane manifolds because the absorbing liquid is not in contact with the membrane. Thus, no pore wetting would occur, preventing frequent cleaning measures or membrane replacement. Such a design greatly simplified the construction of flat membrane modules. For this reason, plastic trough modules were constructed in subsequent laboratory and pilot-scale membrane system experiments using an air space in between the acidic liquid surface and the membrane.

3.2. Nitrogen recovery with increasing NH_3 gas concentration

In experiment 2, lime treatments were applied at different rates (0.1, 0.2, 0.5 and 2.0% w v^{-1}) to enhance NH_3 volatilization by increasing litter pH. The experiment included a control treatment with no lime application (0% w v^{-1}). Each treatment was run in duplicate. Changes in gaseous NH_3 levels within the enclosure headspace and the levels of NH_4–N in the acid solution were monitored over a 4-d period. As a result of liming, gaseous NH_3 concentrations increased rapidly in the headspace of the enclosures within 6 h for all lime treatments (Fig. 4); higher NH_3 headspace concentrations resulted with higher lime application rates. However, in all lime treatments, the NH_3 gas decreased to a uniform level within 4 d with the membrane system; the NH_3 concentration in the headspace were consistently reduced 73–96% with respect to the initial 6-h peak concentrations (Fig. 4).

Table 2 shows both the increase in pH within the litter for each of the lime treatments and the recovery N balances and rates of removal by the membrane module. Initial litter pH in the 0.1%, 0.2%, 0.5%, and 2.0% lime treatments (9.25, 9.69, 10.79, and 12.75 pH units, respectively) was significantly higher ($p < 0.001$) than the pH with 0% lime control (8.78). As a consequence, a large percentage of the initial NH_4–N was likely in the volatile NH_3 form ($pK_a = 9.26$) and instantly available to be captured by the flat membrane system. The estimated percentages of NH_3 in volatile form

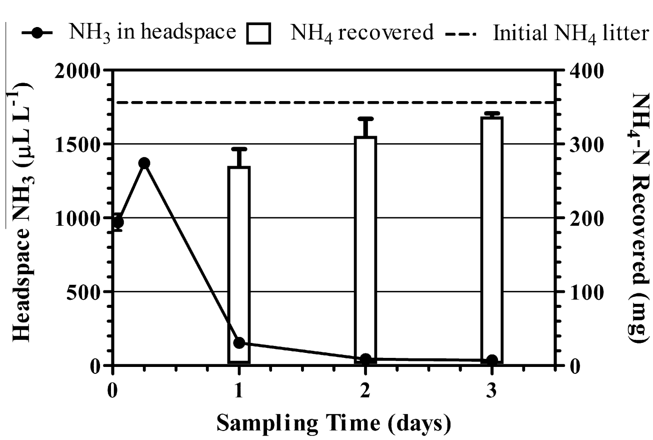

Fig. 3. Reduction of gaseous NH_3 from air in enclosure using the bench-scale flat membrane tray system and corresponding NH_4–N recovery in acid solution from poultry litter amended with 2% lime (w v^{-1}). The dashed line represents initial NH_4–N mass in the poultry litter (356.2 g); error bars are the standard deviation of duplicate measurements.

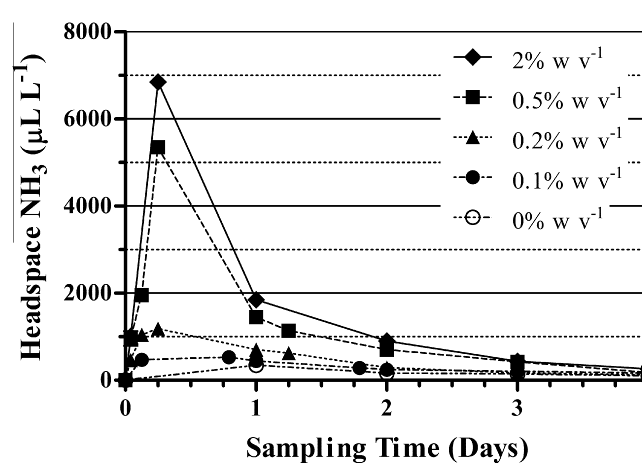

Fig. 4. Reduction of gaseous NH_3 from the headspace in enclosures using the bench-scale flat membrane trough system. Concentrations of NH_3 in the air were obtained using different rates of lime applied to the poultry litter at the beginning of the study (time = 0). Points represent the average of duplicate experiments.

Table 2
Recovery of nitrogen using the bench-scale flat membrane system under various lime application rates[a,b].

Lime, w v^{-1} (%)	pH Initial	NH_4 lost from litter[c] (mg N)	NH_4 recovered in the acid solution (mg N)	NH_4 recovery[d] (%)	Maximum NH_4 recovery rate[e] (g N m^{-2} d^{-1})	Average NH_4 recovery rate[f] (g N m^{-2} d^{-1})
0	8.78 ± 0.02	121 ± 15	107 ± 6	88.4	1.68	1.25
0.1	9.25 ± 0.01	383 ± 60	382 ± 14	99.7	8.65	4.49
0.2	9.69 ± 0.04	597 ± 79	598 ± 11	100.1	15.45	7.02
0.5	10.79 ± 0.04	1460 ± 108	1266 ± 44	86.7	28.62	14.86
2	12.75 ± 0.00	1603 ± 54	1515 ± 51	94.5	34.85	17.78

[a] 1000 g litter in a 0.045-m^3 enclosure, using 1.0 L 1 N H_2SO_4 (recirculation rate = 10 L d^{-1}).
[b] Average of duplicate 4-d experiment.
[c] NH_4 lost from litter = initial NH_4–N in litter − final NH_4–N in litter.
[d] NH_4 Recovery = (NH_4–N recovered in acid/NH_4–N lost from litter) × 100.
[e] Highest NH_4–N mass recovered in 1 day (Fig. 5); 0.0213 m^2 of flat membrane surface area.
[f] Average of NH_4–N mass recovered in 4-d experiment.

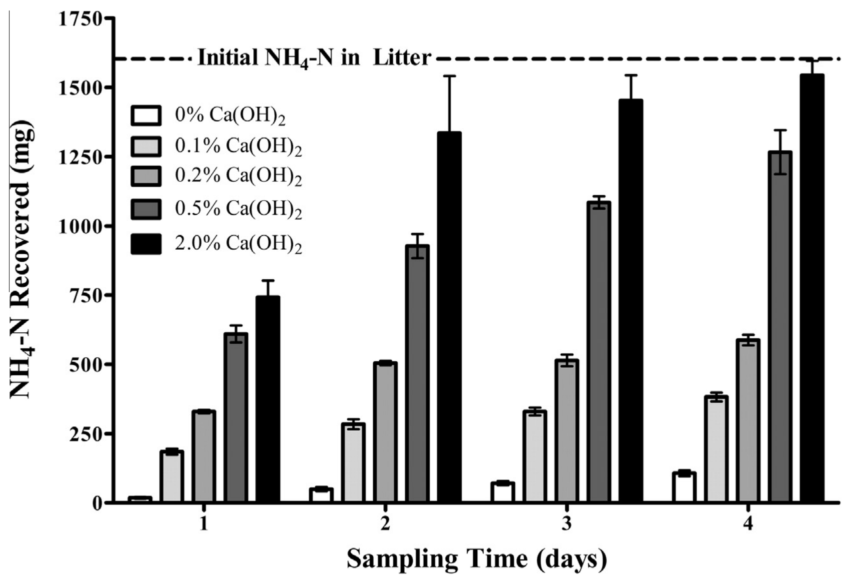

Fig. 5. Daily NH_4–N recovery from poultry litter under various lime application rates using the bench-scale flat membrane trough system. The dashed horizontal line represents the initial NH_4–N content of the poultry litter for all 5 treatments. The error bars are the standard deviation of duplicate experiments.

were 50%, 74%, 97% and 100% of the initial NH_4–N for the respective pH values 9.25, 9.69, 10.79, and 12.75 of the lime treatments.

As liming rate increased, more NH_3 was released into the air; and consequently more N was recovered (Fig. 5). Therefore, the amount of N removed by the membrane from the air was responsive to the increased concentration of NH_3 in air. For example, after one day 17.1, 184, 330, 610, and 742 mg NH_4–N were recovered in the 0, 0.1%, 0.2%, 0.5%, and 2.0% lime treatments, respectively (Fig. 5). In subsequent days, any lime application resulted in a significant increase ($p \leqslant 0.001$) in the mass of NH_4–N recovered with respect to the 0% treatment (control). By the end of the study, 107, 382, 598, 1266, and 1515 mg NH_4–N were recovered in the 0%, 0.1%, 0.2%, 0.5%, and 2.0% lime treatments, respectively (Table 3, Fig. 5). Most of the NH_4–N (>73%) was recovered within the first 2 days, from which the maximum recovery rates of the membrane in terms of surface area (1.68–34.85 g N m^{-2} d^{-1}) were estimated (Table 2). By day four, >86% of the NH_4–N lost from the litter was recovered in the acid solution for all treatments (Table 3 and Fig. 5). The average NH_4–N recovery rate by the flat membrane system increased with lime application in the range of 1.25 to 17.78 g N m^{-2} d^{-1} (Table 2). This range was similar to average recoveries obtained by Rothrock et al. (2010b) using tubular ePTFE gas-permeable membranes (1.29–16.52 g N m^{-2} d^{-1}) at increasing lime application rates (0–4% w/v). These results demonstrate that flat gas-permeable membrane systems provide an additional, effective configuration to significantly reduce gaseous NH_3 contamination of air and recover the volatilized NH_3 from poultry litter in a liquid form.

Table 3
Recovery of nitrogen using a pilot-scale flat membrane manifold system under normal and enhanced volatilization conditions[a].

Lime[b], w v^{-1} (%)	Initial NH_4 in litter (mg N)	Final NH_4 in litter (mg N)	NH_4 lost from litter[c] (mg N)	NH_4 recovered in the acid solution (mg N)	NH_4 recovery[d] (%)	Maximum NH_4 recovery rate[e] (g N m^{-2} d^{-1})	Average NH_4 recovery rate (g N m^{-2} d^{-1})
0	139,236 ± 2081	108,590 ± 757	30,646 ± 2355	29,942 ± 611	97.7	10.42	9.20
2	48,427 ± 1275	0	48,427 ± 1275	48,830 ± 387	100.8	28.63	10.32

[a] 32.5 kg litter in a 2.51 m^3 enclosure using 13.5 L 1*N* H_2SO_4 (recirculation rate = 27 L h^{-1}).
[b] Duration of experiment: 0% = 8.76 days; 2% = 12.73 days.
[c] NH_4 lost from litter = initial NH_4–N in litter − final NH_4–N in litter.
[d] NH_4 recovery = (NH_4–N recovered in acid/NH_4 lost from litter) × 100.
[e] Highest NH_4–N mass recovered in 1 day (Fig. 6); 0.3716 m^2 of flat membrane surface area.

3.3. Pilot-scale flat membrane manifold system

Considering the success of the bench-scale flat membrane system at recovering NH_4–N under enhanced NH_3 volatilization conditions, the pilot-scale flat membrane manifold system shown in Fig. A.3 was used to further test the efficacy of this technology. The pilot experiment was done twice: under normal conditions (0% lime), and under enhanced volatilization conditions using lime (2% w/v).

The effects of both normal and enhanced volatilization conditions on recovery of NH_4 from poultry litter are summarized in Table 3. Overall, the liming increased volatilization of NH_4 from the litter (22.0% without lime and 100% with 2% lime) and resulted in higher mass of N recovered (from 29,942 to 48,830 mg NH_4–N in the 0% and 2% treatments, respectively). The headspace peak NH_3 concentration of the pilot enclosure was 915 and >2000 $\mu L\ L^{-1}$ within the first day of the experiment in the normal and enhanced volatilization treatments, respectively. The recovery of NH_4–N in the acid solution of the pilot-scale manifold system was effective for both treatments. The NH_4–N concentrations in the acid tank were 2218 and 3617 mg L^{-1} at the end of the experiments with 0% and 2% lime treatment, respectively. These final concentration levels were the result of treating only one batch of poultry litter. The NH_4 concentration in the acid tank can be greatly increased by treating consecutive batches of waste with the same stripping solution. For instance, Vanotti and Szogi (2011) recovered 53,000 mg/L NH_4–N in a clear solution using tubular gas-permeable membranes for the removal of NH_4 from high strength swine wastewater. The same stripping solution was used in 10 consecutive batches treating raw swine manure. Concentrated acid was added to the acidic solution as needed to maintain a pH < 2 and high NH_3 absorption efficiency. Therefore, it is feasible to recover N as ammonium sulfate in a concentrated form that can be used as plant nutrient solution.

The recovery of the NH_4–N lost from the litter was 97.7% for the normal conditions (0% lime), and 100% for the enhanced volatilization (2% lime) treatment. However, maximum recovery rates of the membrane system estimated from daily mass NH_4–N recovery data (Fig. 6) were almost threefold larger with lime application (28.63 g N $m^{-2}\ d^{-1}$) than without lime application (10.42 g N m^{-2} d^{-1}, Table 3). According to these results, the limiting factor for high mass recovery was the NH_3 concentration in the headspace rather than the membrane surface area. Even though the NH_3 was recovered efficiently in both situations (Table 3), the average rate of recovery through the membrane (per unit surface area) was about 12% higher with the increased NH_3 availability due to liming. It was concluded that the performance efficiency of NH_3 removal with the gas-permeable membrane system is consistently high (>97%) across a wide range of NH_3 volatilization conditions (normal or enhanced) in poultry litter.

3.4. Economic considerations

The annualized cost of NH_3 recovery with the gas-permeable membrane system was calculated for a typical poultry house size (1800 m^2) growing 20,000 broilers (42 days per flock, 6 flocks per year). Assuming an average production of 0.77 g N $bird^{-1}\ d^{-1}$ (Tao and Mancl, 2008) and a manure N loss of 38% as NH_3 gas (Moore et al., 1995), the daily loss of N from this type of operation is about 5.9 kg N d^{-1}. With an average recovery efficiency of 96%, the annual N recovery with the membrane system is 1427 kg N per year (5.9 kg N $d^{-1} \times 0.96 \times 42$ d $\times$ 6 flocks). On the other hand, the membrane system has a capacity to recover N at a rate of 31.74 g NH_4–N $m^{-2}\ d^{-1}$ (pooled maximum N removal rates obtained in this study, Tables 2 and 3). Thus, a membrane surface of 186 m^2 is required to recover the daily NH_3 losses in the typical poultry house described above. Equipment cost estimates include the flat membrane at \$26.90 m^{-2} and additional manifold components at \$10.00 m^{-1} (includes the trough, connectors, pump, and acid tank). Taking into account 8% interest and 10-year useful life, annualized cost of equipment is \$4385 (flat membrane plus plastic channel). The amount of acid needed for 1 year operation was estimated as the mass of acid needed per mole of $(NH_4)_2\ SO_4$ (98 g of sulfuric acid per 28 g of N), which is equivalent to 3.5 kg of acid per kg of N recovered. Thus, 4995 kg of sulfuric acid (98% w/v) are needed to recover 1427 kg of N per year with the gas permeable system. Since the cost for the sulfuric acid is \$0.329 kg^{-1}, the annual cost for the acid is \$1643. The estimated total annual cost for a flat membrane system (20,000 birds, 6 flocks per year) is \$6028 (\$4385 + \$1643).

The concentrated ammonium sulfate produced (1427 kg N) during production of six flocks (1 year) has an equivalent fertilizer value of \$3339 assuming a value of \$2.34 per kg N as ammonium

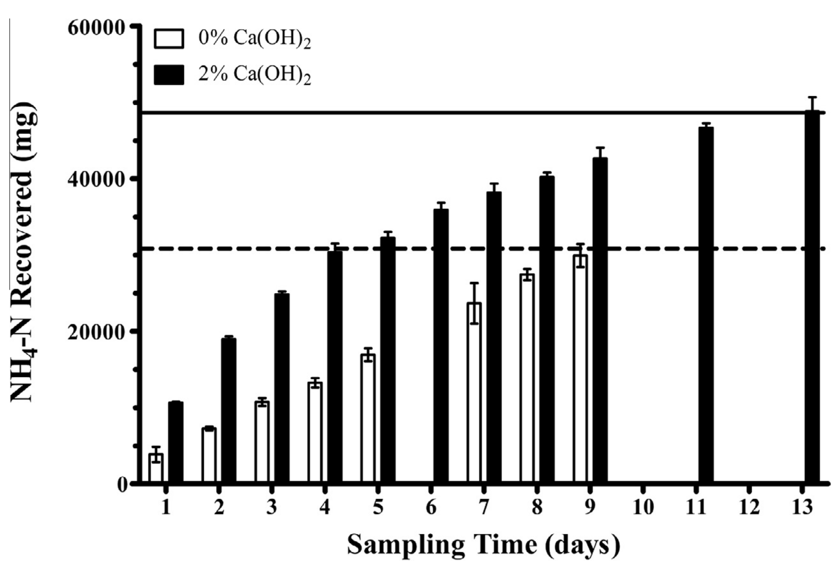

Fig. 6. Daily NH_4–N recovery from poultry litter using the pilot-scale flat membrane manifold system. The 0% and 2% lime treatments lasted 9 and 13 days, respectively. The solid line represents the cumulative NH_4–N loss from the 2% treatment, and the dashed line represents the cumulative NH_4–N loss from the 0% treatment. The error bars are the standard deviation of duplicate experiments.

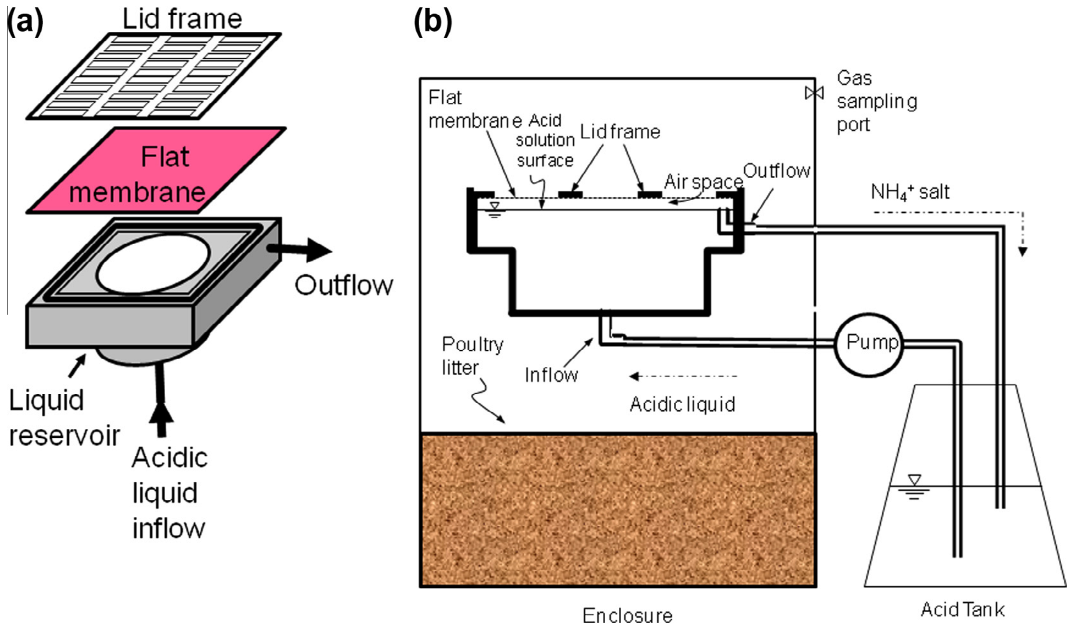

Fig. A.1. Flat membrane system used in laboratory experiment 1: (a) diagram of the bench-scale membrane tray manifold for NH_3 recovery and (b) cross-section diagram of the bench-scale system (diagrams are not to scale).

sulfate (USDA-ERS, 2012). Considering the value of recovered N, the net cost is \$2689 per year (\$6028–\$3339). A complete economic analysis of this membrane system would have to consider unexpected expenses (recovered NH_3 storage reservoir, operator training, lab analysis, etc.) as well as additional economic benefits expected from energy savings (reduction on ventilation rate and fuel consumption during winter) and improved productivity (reduced bird mortality). In view of Attar and Brake (1988), they modeled the economic benefit of NH_3 control in poultry houses as relate to reduced ventilation rate and fuel consumption. With their model they calculated that if outside temperature was 7 °C, then the cost of producing broilers in a similar house (19,000 birds) was reduced \$3800 per flock. For example, the net cost turns into a net benefit when reduced fuel consumption is considered. So one flock produced during cold conditions could make the membrane system profitable (\$2870 net cost – \$3800 fuel savings = \$930 profit). In addition, the bird mortality is reduced with the improved air quality, which should be an important additional economic benefit from implementation of this invention. For example, Moore et al. (1995) used alum applied to the litter to reduce atmospheric ammonia inside the poultry houses and reduced the bird mortality rates about half, from 8.3% in a control to 3.6% in the treatment that reduced atmospheric ammonia.

4. Conclusions

Results of this study support the concept and technical feasibility of using flat gas-permeable membrane systems to recover NH_3 from poultry waste. Flat membrane systems reduced headspace NH_3 concentrations from about 70% to 97% while recovering 88% to 100% of the volatilized NH_3 within an acidic liquid. The potential benefits of this technology include improved bird productivity and health, reduction in NH_3 emissions, and lower energy requirements for ventilation since NH_3 is being passively removed. In addition this technology produces cleaner air and recovers NH_3 into a concentrated ammonium salt, a valuable plant nutrient product.

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Appendix A.

See Figs. A.1–A.3.

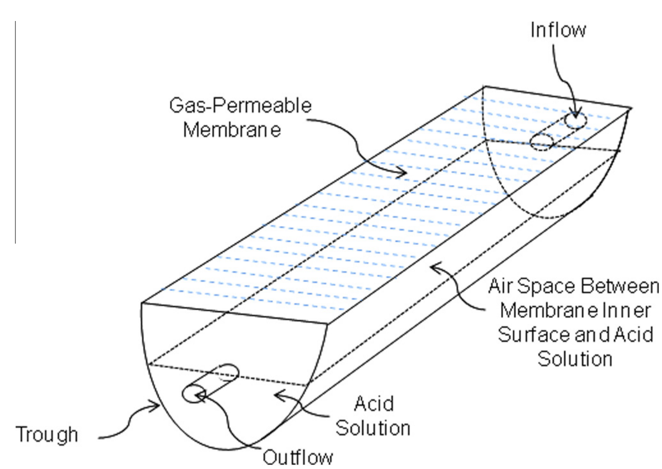

Fig. A.2. Diagram of the bench-scale membrane trough module (diagram is not to scale).

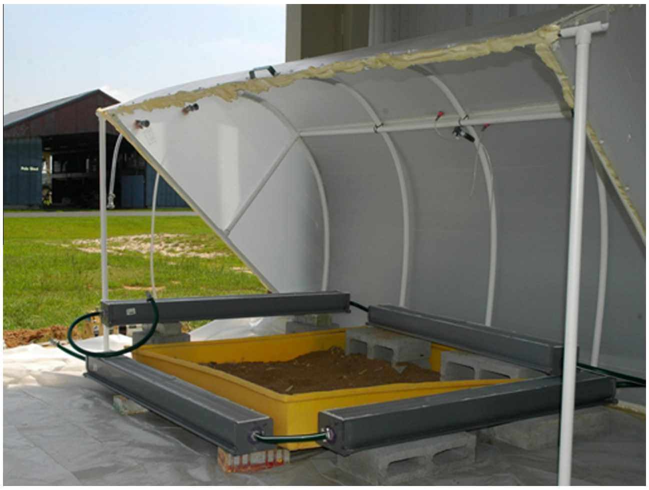

Fig. A.3. Pilot-scale experiment with the flat membrane manifold system connected in series and placed on the perimeter of the NH_3 source (poultry litter) prior to closing the air-tight enclosure.

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