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Reduction of Ammonia Emission from Stored Poultry Manure Using Additives: Zeolite, Al+clear, Ferix-3 and PLT

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Abstract. *Manure storage can be a significant source of ammonia emission that impacts the environment. Ammonia emission from manure storage can be controlled by using physical, chemical and/or biological methods. Five treatment agents, including zeolite, liquid Al+Clear (aluminum sulfate), granular Al+Clear (aluminum sulfate), and granular Ferix-3 (ferric sulfate), and PLT (sodium hydrogen sulfate) were topically applied to stored fresh layer manure. Each agent was tested at three application rates, i.e., low, medium and high. Manure was stored in 19-litter Teflon-lined vessels under a constant ambient temperature of 23 °C with a constant airflow of 3 liter per minute. The ammonia concentrations and emissions from the vessels were measured and ammonia emission reductions by the treatment regimens were evaluated as compared to the control. Reduction of ammonia emission as a result of topical application of the tested manure treatment agents, when compared to the control, over a 7-day manure storage period was as following: A) 68%, 81% or 96%, respectively, for zeolite applied at 2.5%, 5% or 10% of the manure weight; B) 63%, 89%, or 94%, respectively, for liquid Al+Clear applied at 1, 2, or 4 kg m^{-2} of manure surface area; C) 81%, 93%, or 94%, respectively, for dry granular Al+Clear applied at 0.5, 1.0, or 1.5 kg m^{-2}; D) 82%, 86%, or 87%, respectively, for Ferix-3 applied at 0.5, 1.0, or 1.5 kg m^{-2}; and E) 74%, 90%, or 92%, respectively, for PLT applied at 0.5, 1.0, or 1.5 kg m^{-2}.*

Keywords. Laying hen, belt house, manure storage, ammonia emission, additives.

Introduction

Ammonia (NH_3) volatilization from intensive livestock operation not only reduces fertilizer nitrogen (N) value when manure is applied to agricultural land, but also contributes to environmental pollution. Effective technologies that reduce ammonia loss during animal housing, manure storage and land application would have positive economic and environmental benefits.

Laying hen manure is typically either stock-piled in the lower level of high-rise houses or removed from belt cage layer houses to manure storage facilities once to seven times a week. Various mechanisms are involved in conserving N in poultry manure during storage, including immobilization of ammonium through addition of easily decomposable, N-poor materials, adsorption of ammonium (NH_4^+) and NH_3 on suitable amendments, and pH regulation of the manure solution (Kirchmann and Witter, 1989).

Numerous additives have been investigated to reduce NH_3 volatilization from livestock manure. McCroy and Hobbs (2001) published a comprehensive review of a wide range of additives, i.e., acidifying agents, absorbing agents, and bacterial additives, for reducing ammonia from livestock wastes. Natural zeolite is a cation-exchange medium that has high affinity and selectivity for NH_4^+ ions due to its crystalline, hydrated properties resulted from its infinite, 3-dimentional structures (Mumpton and Fishman, 1977). It has been widely used as amendment to poultry litter (Maurice et al., 1998; Nakaue and Koelliker, 1981b), in anaerobic digesters treating cattle manure (Borja et al., 1996), during composting of pig slurry and poultry manure (Bernal et al., 1993; Kithome et al., 1999), air scrubber packing material to improve poultry house environment (Koelliker et al., 1980), and as a filtration agent in deep-bedded cattle housing (Milan et al., 1999). Kithome et al. (1998) investigated the kinetics of NH_4^+ adsorption and desorption by natural zeolite clinopilolite [(Na_4K_4) (Al_8Si_{40}) $O_{96}\cdot 24H_2O$] for its ability to adsorb N in its NH_4^+ form at various pH values and initial NH_4^+ concentrations.

The volatilization of ammonia has been attributed to microbial decomposition of nitrogenous compounds, principally uric acid, in poultry manure. Manure pH plays an important role in ammonia volatilization. Ammonia concentration tends to increase with increasing pH. Ammonia release remains small when pH is below 7.0, but can be substantial when pH is above 8.0. Uric acid decomposition is most favored under alkaline (pH>7) conditions. Uricase, the enzyme that catalyzes uric acid breakdown, has maximum activity at a pH of 9 with uric acid decreasing linearly for more acid or alkaline pH values. The NH_3 emission can be inhibited by acidulants, which can lower manure pH and reduce conversion of ammonium to ammonia. The acidulants also inhibit the activities of bacteria and enzymes that are involved in the formation of ammonia, reducing ammonia production. Liquid Al+Clear and dry granular Al^+Clear (aluminum sulfate), Ferix-3 (ferric sulfate) and PLT (sodium hydrogen sulfate) are acidulants that produce hydrogen ions (H^+) when they dissolve, and the hydrogen ions produced by this reaction will attach to ammonia to form ammonium. Because of these reactions, the amount of ammonia emitted from the manure will be reduced, which will increase the nitrogen (N) content of the manure. Al^+Clear and PLT had been applied to poultry litter control ammonia volatilization (Moore et al., 1995, 1996; Kithome et al., 1999; Lefcourt and Meisinger, 2001, Armstrong et al., 2003). Ferix-3 usually is used for industrial and municipal water and wastewater treatment over a wide pH range. Uses include color removal, organics removal, phosphorous removal, bacteria reduction, arsenic removal, sludge conditioning, turbidity reduction, COD/BOD reduction, enhanced coagulation, and heavy metals removal. It performs very well in soil remediation applications. However, information on the three acidulants efficacies on ammonia mitigation with laying hen manure is meager.

Materials and Methods

Air Emission Vessels

Eight emission vessels were designed and built for the study (Fig. 1). The vessels were placed in an environment-controlled room with a constant temperature 23 °C at the Livestock Environment and Animal Physiology (LEAP) Lab II of Iowa State University. The vessels were made of 19-liter (5-gal) plastic containers. To prevent potential interference of the vessel material with ammonia emission measurement, each vessel was lined with Teflon FEP100 film (200A, DuPont Teflon ® Films, Wilmington, DE). Both air inlet and outlet were located in the air-tight lid. Teflon tubing (1/4″ diameter) and manifold, along with PVC compression fittings, were used in constructing the emission vessel system.

The vessels were operated under positive pressure. A diaphragm pump (Model DOA-P104-AA, Gast Manufacturing, Inc., Benton Harbor, MI) was used to supply fresh air to the emission vessels. Flow rate of the fresh supply air was controlled and measured with an air mass flow controller (0 to 30 LPM, stainless steel wetted part, AAlborg Instruments and Control Inc., Orangeburg, N.Y.). The supply air was connected to a distribution manifold where air was further divided via eight identical flowmeters (0.2 to 4 LPM, stainless steel valve, VFB-65-SSV, Dwyer Instruments, Inc., Michigan City, Indiana). A flow rate of 3 LPM was introduced into each vessel, resulting in an air exchange rate of 11 air changes per hour (ACH). Each vessel was equipped with a small stirring fan (12VDC, Radio Shack) located 6 cm below the lid for uniform mixing of the headspace. Gas exhausted from the vessels was connected to a common 5 cm PVC pipe that was routed to the building vent outlet. A photographical view of the experimental setup is shown in Figure 2.

Samples of the exhaust air from each of the eight vessels, the supply air, and the room air were sequentially taken at 6-min intervals, with the first 4 minutes for stabilization and the last 2 min for measurement. This yielded a measurement cycle of one hour for each vessel. The sequential sampling was achieved by controlled operation of eight solenoid valves (Type 6014, 24V, stainless steel valve body, Burkert Contromatic USA, Irvine, CA). A Teflon filter was placed in front of each solenoid valve. A photoacoustic infrared (IR) ammonia gas analyzer (Chillgard RT Refrigerant Monitor, MSA, Pittsburg, PA) was used to measure the NH_3 concentrations. The analyzer uses an internal pump to draw sample air at a flow rate of approximately 1.0 LPM. Manure temperature was measured with type T thermocouples (0.2 ºC resolution). Air temperature and relative humidity of the room were monitored with a temp/RH data logger (HOBO Pro RH/Temp, Onset Computer Corporation, Bourne, MA). Analog outputs from the thermocouples, NH_3 analyzer, and the mass flow meter were logged at 20-s intervals into a measurement and control module (Model CR10, Campbell Scientific, Inc., Logan, UT).

Laying Hen Manure and Mitigation Options Tested

Hen manure that accumulated on belt for less than a day in a commercial manure-belt layer house was used in the evaluation of the treatment agents. Manure samples with an initial weight of 2.5 kg were used as the experimental units. The 2.5 kg sample was placed either in a 3.8-liter (1-gal) container (surface area of 0.02 m^2) that was further placed inside the 19-liter (5-gal) emission vessel or directly into the emission vessel (surface area of 0.05 m^2).

Five treatment additives at various application rates were tested, including natural zeolite, two forms (liquid and dry) of Al^+Clear, Ferix-3, and PLT. The treatment agents were topically applied to the manure samples at 2.5%, 5% or 10% of the manure weight for zeolite; 1, 2, or 4 kg m^{-2} of manure surface area for liquid Al^+Clear; and 0.5, 1.0, or 1.5 kg m^{-2} for dry granular Al^+Clear, Ferix-3, and PLT. The application rates of Al^+Clear, Ferix-3, and PLT referred to the application rates of alum on the broiler litter (Armstrong et al., 2003). Properties of the four chemicals tested are listed in Table 1.

Each treatment regime had 4 to 6 replications. The trials with the four chemical agents' treatment lasted 7 days. In the case of zeolite treatment, three trials were conducted. The first two trials examined the effects of single application at one of the afore-mentioned three rates on ammonia emissions over a 14-day storage period, where the third trial examined the effect of multiple applications (every two days, coinciding with manure loading) at the 5% application rate on ammonia emission during a 14-day test. Manure samples were taken from the top 2.5 cm and their physical and chemical properties were analyzed by a certified commercial analytical laboratory.

Results and Discussion

Effect of Topical Application of Zeolite on NH_3 Emission from Hen Manure

Surface-applied zeolite on fresh manure substantially decreased NH_3 emission during 14-d storage period and the effect were generally proportional to the application rates. Daily NH_3 emissions of zeolite on manure in batch trials were illustrated in Figure 3. The adsorption of NH_3/NH_4^+ took effect right after its application at Day 0 and resulted in largest ER reduction on Day 1. Ammonia emissions were reduced by 66, 91 and 96% at the end of Day 1, with application rates of 2.5, 5 and 10%, respectively. Daily ammonia emission of the Ctrl vessels became stabilized after day 3, whereas emissions of the Trt vessels continued to increase with the Trt2.5 being most obvious. Ammonia emissions of Trt5 and Trt10 were significantly lower than that of the Ctrl ($P<0.01$) throughout the 14-d trial period, whereas this was true for the Trt2.5 regimen during the first 7 d ($P<0.01$). Addition of two or more layers of manure did not seem to increase NH_3 emission on a per vessel basis (g d^{-1} or g $m^{-2}d^{-1}$), largely due to the same emitting surface area in the vessel. However, on a per unit manure mass basis, daily ER decreased progressively with the addition of manure (Fig. 4).

Table 2 summarizes the effects of single or multiple topical applications of zeolite at the three dosages on NH_3 emission reduction. Cumulative NH_3 ER reductions at the end of Day 7 and Day 14 were 68% and 20% for Trt2.5, 81% and 50% for Trt5, and 96% and 77% for Trt10. Fourteen-day daily average NH_3 ERs were 0.231, 0.185, 0.116 and 0.053 g d^{-1} kg^{-1} initial manure for control, Trt2.5, Trt5 and Trt10, respectively.

Kithome et al. (1999) reported that NH_3 loss was decreased by 44% when composting poultry manure over 56 days with a surface application of 38% zeolite. Bernal et al. (1993) also reported that more than 90% of N-loss was trapped by placing 12% (by weight) zeolite in air stream over 13-day composting of pig slurry and chopped straw mixture. Zeolite additions at 2.5% and 6.25% into dairy slurry reduced NH_3 emissions by 22% and 47%, respectively, over 4-d storage period (Lefcourt and Meisinger, 2001).

Effects of Al^+Clear, Ferix-3, and PLT Treatment on NH_3 Emission from Layer Manure

Surface-applied liquid and granular Al^+Clear, Ferix-3, and PLT on fresh manure substantially decreased NH_3 emission during 7-d storage period. Daily NH_3 emissions from all treatment and control were illustrated in Figure 3. Ammonia emissions for each regimen, emission reduction by the treatment as compared to the control, and manure properties are summarized in Table 3. Reduction of ammonia emission as a result of topical application of the tested manure treatment agents, when compared to the control, over a 7-day manure storage period was as following: A) 63%, 89%, or 94%, respectively, for liquid Al[+]Clear applied at 1, 2, or 4 kg m^{-2} of manure surface area; B) 81%, 93%, or 94%, respectively, for powder Al[+]Clear applied at 0.5, 1.0, or 1.5 kg m^{-2}; C) 82%, 86%, or 87%, respectively, for Ferix-3 applied at 0.5, 1.0, or 1.5 kg m^{-2}; and D) 74%, 90%, or 92%, respectively, for PLT applied at 0.5, 1.0, or 1.5 kg m^{-2}. Ammonia emission reduction from each of the three application rates (denoted as low, medium and high) was significantly lower than that of the control ($P<0.001$). After 7 days, the NH_3 emission reductions from all low application rates were lower than the higher application rates ($P<0.001$).

Daily NH_3 ER of control vessels became stabilized after Day 3, while those of medium and high application treatment vessels stayed with very low NH_3 ERs (Fig. 5). Ammonia ERs (<0.01 g NH_3 kg^{-1} initial manure) of medium and high application rate on every single day were not different ($P>0.70$) during the 7 days. Ammonia ERs of low application rate vessels started to increase from the 3rd, 5th, 6th and 7th day for liquid Al[+]Clear, dry granular Al[+]Clear, Ferix-3, and PLT, respectively.

Results of the manure properties in Table 2 showed that manure samples receiving the higher application rates had lower pH, lower TAN, and higher total N in the top 2.5 cm manure after the 7-day storage period. The average TAN from the controls, low, medium and high application rate vessel were 11.3, 9.9, 8.2, and 6.9 g kg^{-1} (as-is), respectively. The average pH values from the controls, low, medium, and high application rate vessel were 7.6, 7.4, 7.1 and 6.6 respectively. The average total N from the controls, low, medium, and high application rate vessel were 18.5, 18.6, 21.6, and 22.9 g kg^{-1} (as-is), respectively. The more nitrogen was conserved in the manure with higher application rate.

Conclusions

Surface-applying fresh layer manure with zeolite, Al[+]Clear, Ferix-3 and PLT is an effective means to reduce NH_3 emission during storage. Reduction of ammonia emission as a result of topical application of the tested manure treatment agents, when compared to the control, over a 7-day manure storage period was as following: A) 68%, 81% or 96%, respectively, for zeolite applied at 2.5%, 5% or 10% of the manure weight; B) 63%, 89%, or 94%, respectively, for liquid Al[+]Clear applied at 1, 2, or 4 kg m^{-2} of manure surface area; C) 81%, 93%, or 94%, respectively, for dry granular Al[+]Clear applied at 0.5, 1.0, or 1.5 kg m^{-2}; D) 82%, 86%, or 87%, respectively, for Ferix-3 applied at 0.5, 1.0, or 1.5 kg m^{-2}; and E) 74%, 90%, or 92%, respectively, for PLT applied at 0.5, 1.0, or 1.5 kg m^{-2}.

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Table 1.Physical and chemical properties of Al$^+$Clear, Ferix-3 and PLT

	Liquid Al$^+$Clear	Dry Al$^+$Clear	Ferrix-3	PLT
Molecular formula	$Al_2(SO_4)_3 \cdot 14H_2O$	$Al_2(SO_4)_3 \cdot 14H_2O$	$Fe_2(SO4)_3 \cdot 9H_2O$	$NaHSO_4$
Molecular weight	594	594	562	120
pH	2.0 (approx)	3.5 (1% solution)	1.02 (10% solution)	<1 (5% solution)
Appearance	Clear	White granules	Yellowish granules	Off-white granules
Physical state	48.5% in water	Dry solid	Dry solid	Dry solid
Odor	Odorless	Odorless	Slight	Odorless

Table 2. Effects of topical application of zeolite at various rates on reduction of ammonia emission from laying hen manure storage. The application rates, expressed in % of manure weight, were 0% (Ctrl), 2.5% (Trt2.5), 5% (Trt5), and 10% (Trt10), respectively.

		Single Application (in 1-gal emission vessels)				Four Layers (5-gal vessels)	
		Ctrl	Trt2.5	Trt5	Trt10	Ctrl	Trt5
Amount of manure, kg		2.5				2.5 kgx4 = 10	
Surface area of manure, m^2 (ft^2)		0.02 (0.22)				0.05 (0.54)	
Application rate	kg m^{-2}	0	3.125	6.25	12.5	0	2.55
	lb ft^{-2}	0	0.639	1.277	2.555	0	0.52
Number of zeolite application		Once - at the beginning				Four - once per layer	
Trial/treatment duration, day		14				14	
Avg. daily ER per unit of manure weight or surface area over trial period	g $kg^{-1}d^{-1}$	0.231	0.185	0.116	0.053	0.137	0.069
	g $m^{-2}d^{-1}$	29.9	24.0	15.0	6.9	16.1	9.7
7-d cumulative emission, g kg^{-1}		1.6	1.0	0.62	0.14	-	-
7-d emission reduction rate		-	68%	81%	96%	-	33% [b]
Total cumulative emission, g kg^{-1} [a]		3.0	2.5	1.4	0.7	1.7	1.0
Total cumulative emission reduction		-	20%	50%	77%	-	44%
8-d emission reduction rate [c]		-	-	-	-	-	54%

[a] comparison tests lasted 14 days for vessel trials

[b] represents cumulative emission reduction over 7 days following the last-layer addition of hen manure

[c] represents cumulative emission reduction during first 8 days of manure additions

$$\text{Emission Reduction Rate} = \frac{CumuEmission_{Treament}}{CumuEmission_{Control}} \times 100\%$$

Table 3. Effects of topical application of liquid Al+Clear, dry granular Al+Clear, Ferix-3 and PLT at different rates on reduction of ammonia emission from laying hen manure storage

		Liquid Al+Clear, kg m^{-2}				Dry Al+Clear, kg m^{-2}				Ferix-3, kg m^{-2}				PLT, kg m^{-2}			
		Ctrl	1	2	4	Ctrl	0.5	1.0	1.5	Ctrl	0.5	1.0	1.5	Ctrl	0.5	1.0	1.5
Amount of manure, kg		2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5
Surface area, m^2		0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02
Application rate	kg m^{-2}	0	1.0	2.0	4.0	0	0.5	1.0	1.5	0	0.5	1.0	1.5	0	0.5	1.0	1.5
	lb ft^{-2}	0	0.20	0.41	0.82	0	0.10	0.20	0.31	0	0.10	0.20	0.31	0	0.10	0.20	0.31
Avg. daily ER over trial period	g kg^{-1}d^{-1}	0.187	0.070	0.020	0.011	0.150	0.029	0.011	0.009	0.075	0.014	0.011	0.010	0.144	0.037	0.014	0.012
	g m^{-2}d^{-1}	21.1	7.87	2.30	1.27	17.0	3.23	1.23	1.07	8.41	1.56	1.19	1.09	16.3	4.18	1.57	1.38
Cumulative emission [€]	g kg^{-1}	1.31	0.49	0.14	0.08	1.05	0.20	0.08	0.07	0.52	0.10	0.07	0.07	1.01	0.26	0.10	0.09
	g m^2	148	55.1	16.1	8.90	119	22.6	8.62	7.48	58.8	10.9	8.33	7.60	114	29.2	11.0	9.64
Reduction Rate [ϕ]		-	63%[b]	89%[a]	94%[a]	-	81%[b]	93%[a]	94%[a]	-	82%[b]	86%[a]	87%[a]	-	74%[b]	90%[a]	92%[a]
Dry content		28.1	29.9	31.1	30.8	27.1	27.9	27.1	30.8	28.3	34.1	31.9	33.9	27.0	29.0	30.5	32.3
Total N, g kg^{-1} (as-is)		17.6	16.5	21.0	24.1	18.5	18.8	20.0	19.1	21.1	23.0	23.5	24.9	16.6	16.2	21.9	23.4
Total N, gkg^{-1} (dry base)		62.6	55.2	67.5	73.5	68.3	67.4	73.8	62.0	74.6	67.4	73.7	73.5	61.5	55.9	71.8	72.4
TAN, g kg^{-1} (as-is)		10.5	9.8	6.0	5.4	11.1	12.5	12.3	10.4	13.2	8.6	7.1	5.6	10.5	8.6	7.3	6.0
TAN, g kg^{-1} (dry base)		37.4	32.8	19.3	16.5	41.0	44.8	45.4	33.8	46.6	25.2	22.3	16.5	38.9	29.7	23.9	18.6
pH		7.6	7.53	7.01	6.42	7.68	7.65	7.65	6.82	7.37	7.2	6.92	6.55	7.6	7.3	6.8	6.7

[€] Comparison tests lasted 7 days for vessel trials

[ϕ] Represents cumulative emission reduction during 7 days

$$\text{Emission Reduction Rate} = \frac{CumuEmission_{Treament}}{CumuEmission_{Control}} \times 100\%$$

[ϕ] Values of emission reduction rate for each agent followed by the same superscript letters are not significantly different (P>0.05).

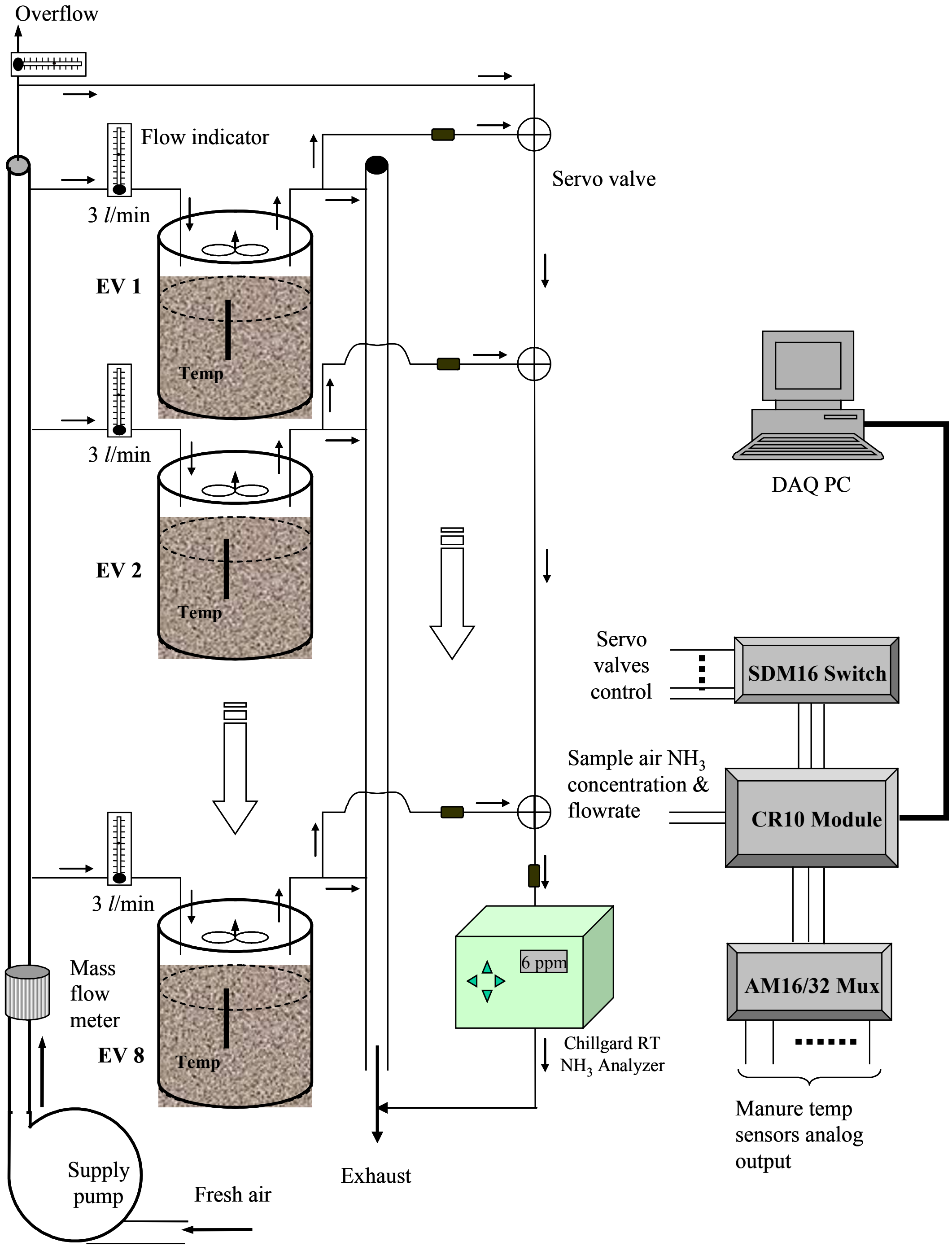


Figure 1. Schematic representation of the experimental setup for evaluating efficacy of treatment agents on ammonia emission reduction from laying hen manure (EV = emission vessel). (Courtesy of Xin, 2005)

Figure 2. Photographs of the laboratory setup for evaluating efficacy of air emission mitigation strategies. Pictured to the right is topical application of zeolite on laying hen manure at various dosages.

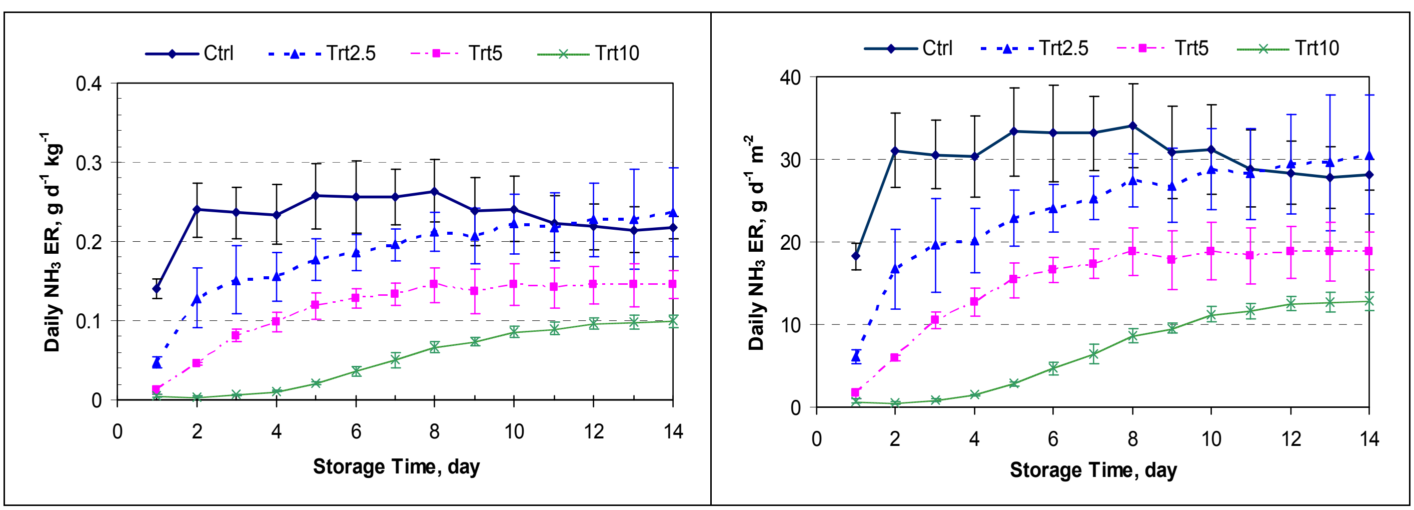


Figure 3. Daily ammonia emissions of ventilated laying hen manure storage with various rates of single surface application of zeolite (Ctrl: no zeolite; Trt2.5: 2.5% zeolite by weight; Trt5: zeolite 5% by weight; Trt10: 10% zeolite by weight).

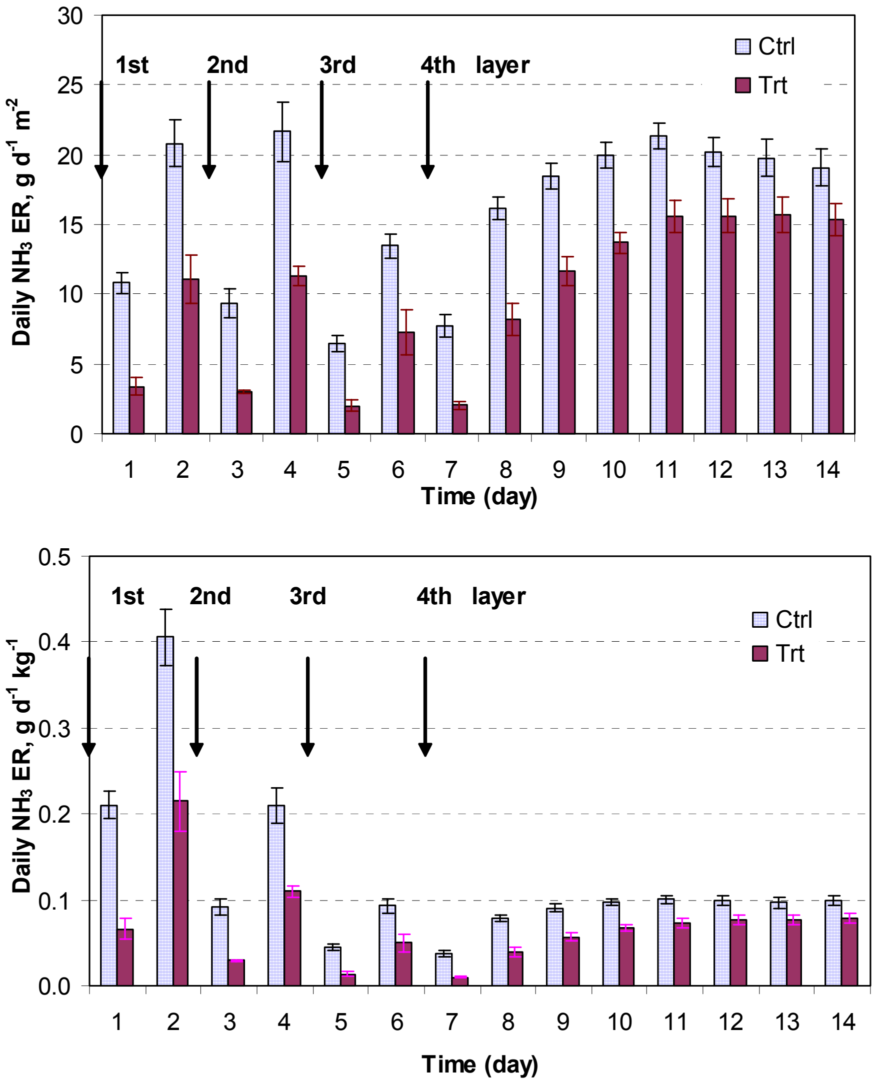
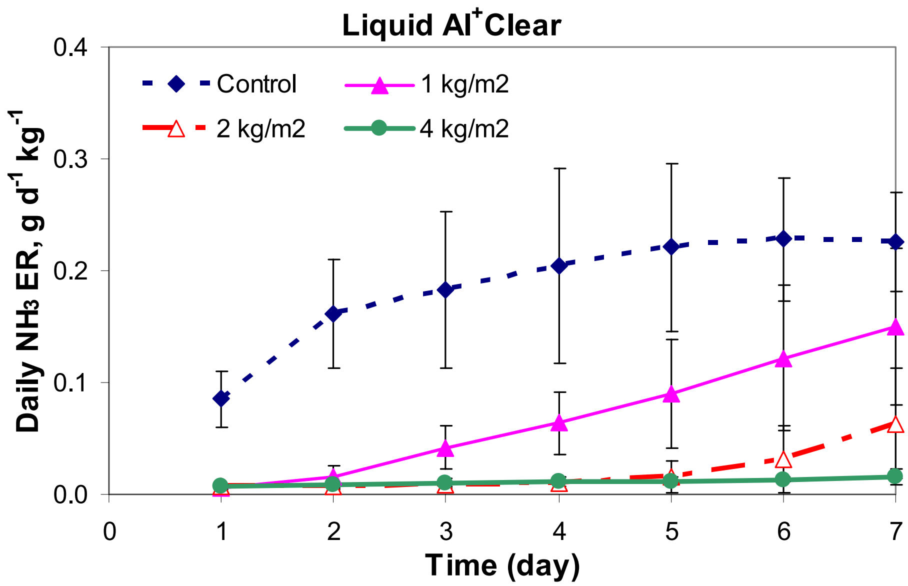


Figure 4. Daily ammonia emissions of ventilated hen manure storage. Fresh manure was added and zeolite topically applied on days 0, 2, 4, and 6 (Ctrl – no zeolite; Trt – 5% zeolite by weight).



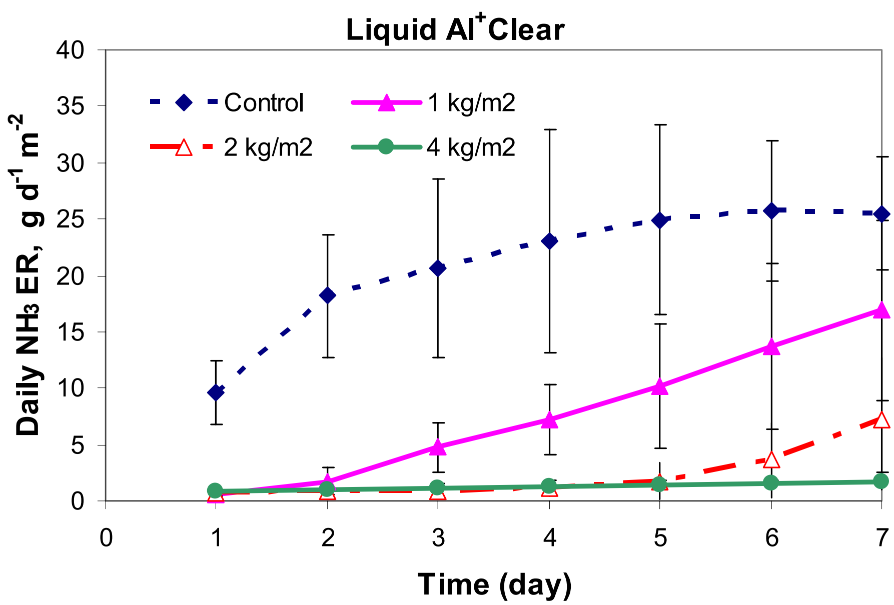
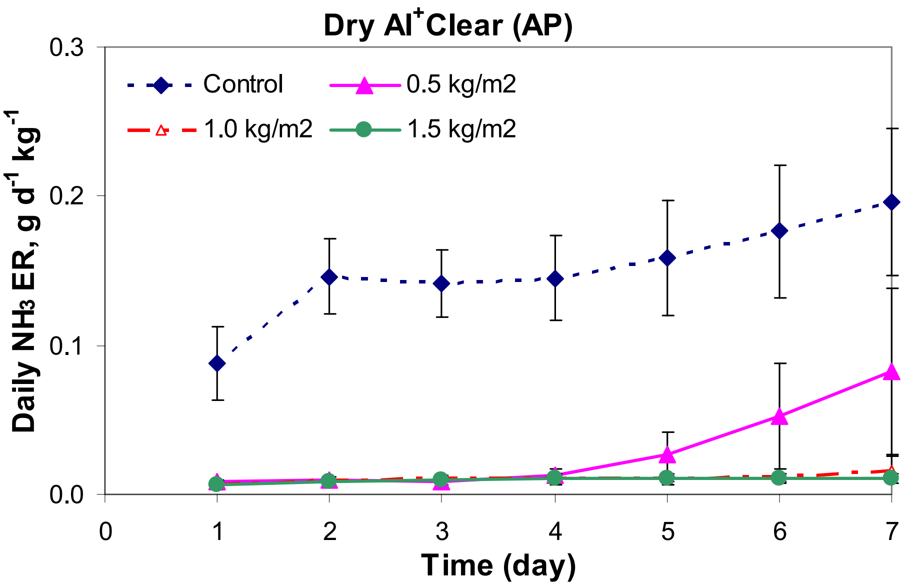


Figure 5. Daily ammonia emission rate (mean and standard error, n=6) of ventilated storage of laying hen manure with different rates of topical application of liquid Al^{+}Clear.



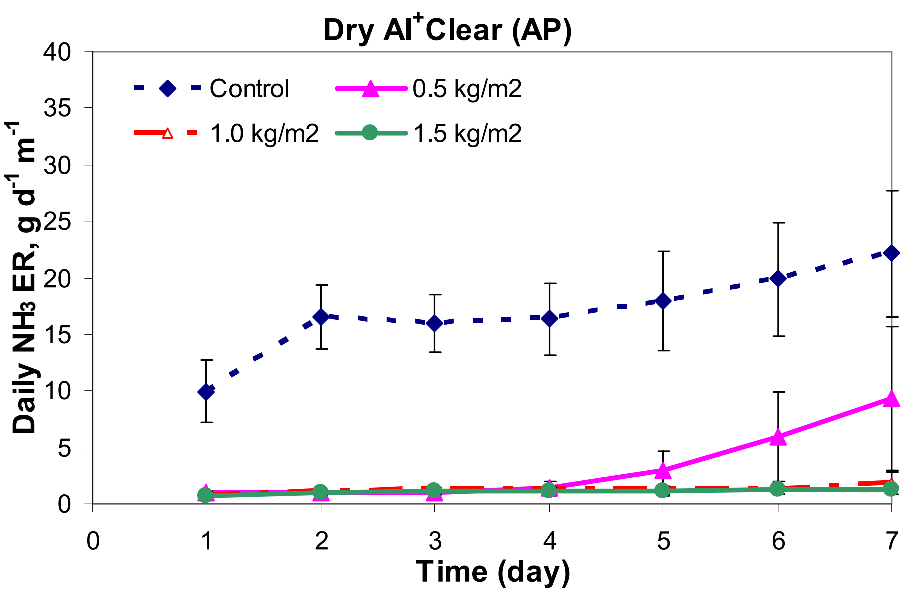
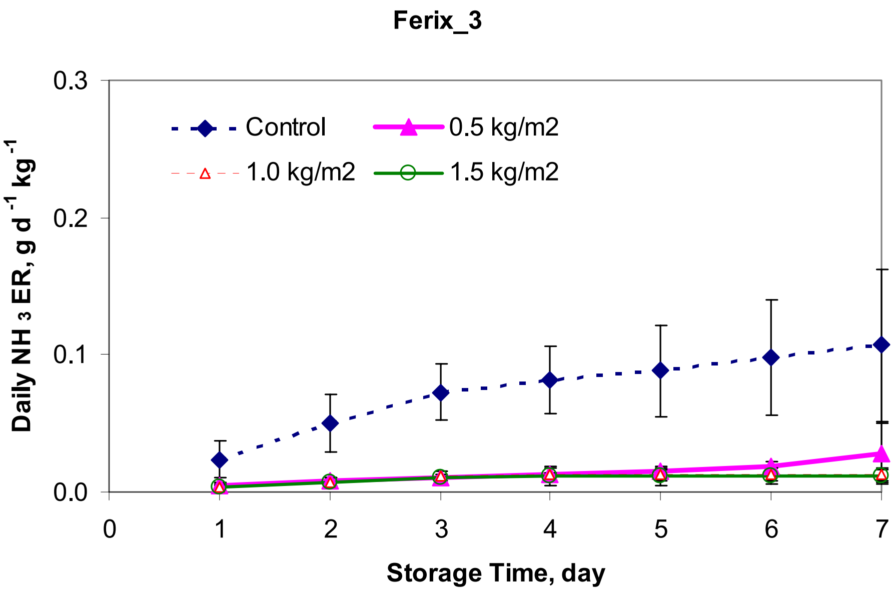


Figure 5 (continued). Daily ammonia emission rate (mean and standard error, n=6) of ventilated storage of laying hen manure with different rates of topical application of dry granular Al^{+}Clear



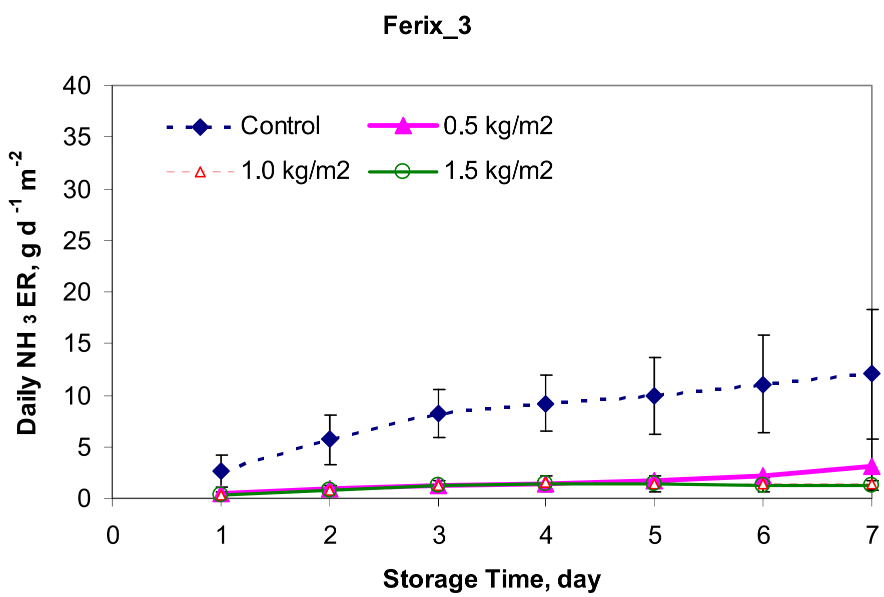
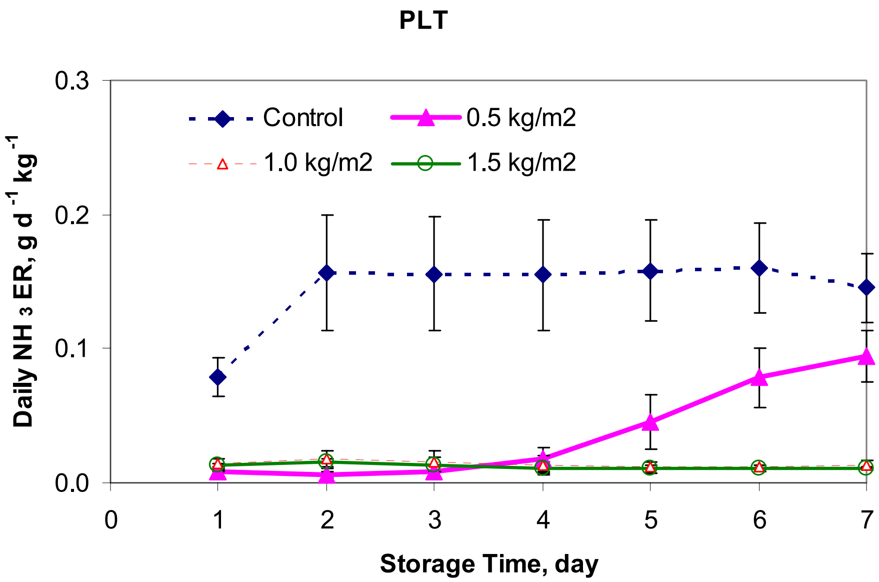


Figure 5 (continued). Daily ammonia emission rate (mean and standard error, n=4) of ventilated storage of laying hen manure with different rates of topical application of Ferix-3.



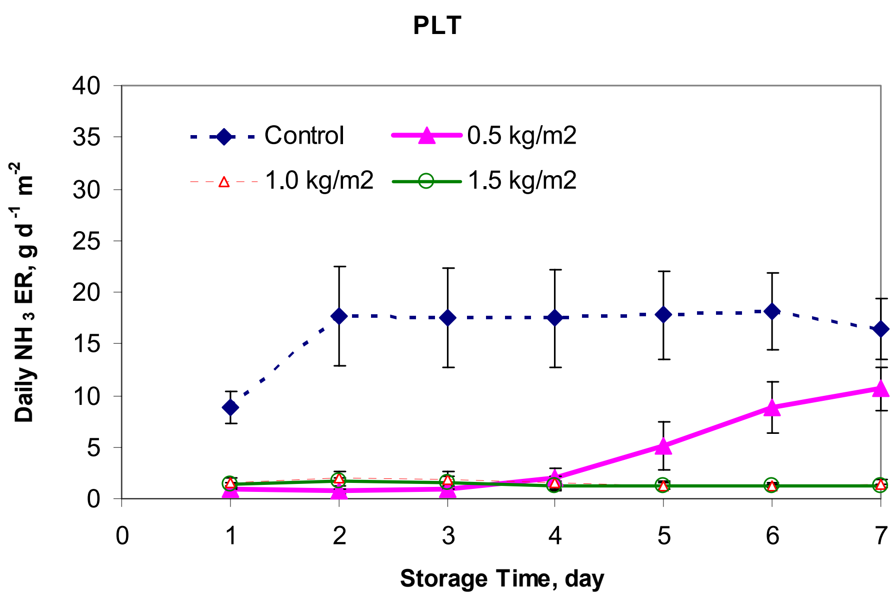


Figure 5 (continued). Daily ammonia emission rate (mean and standard error, n=4) of ventilated storage of laying hen manure with different rates of topical application of PLT.