

Ammonia Emissions from Anaerobically-digested Slurry and Chemical Fertilizer Applied to Flooded Forage Rice

Hong Hou · Sheng Zhou · Masaaki Hosomi ·
Koki Toyota · Kiori Yosimura · Yuuko Mutou ·
Taku Nisimura · Masao Takayanagi ·
Takashi Motobayashi

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Abstract Ammonia fluxes from application of anaerobically-digested slurry (ADS) and chemical fertilizer (CF) to flooded forage rice (*Oryza sativa L.*) in Japan were measured using a dynamic flow-through chamber method in lysimeters. The CF was applied at a rate of 300 N ha⁻¹ (three times) as ammoniacal-N fertilizer, and the ADS was applied to the lysimeters at total rates equivalent to 75, 100 and 150 kg N ha⁻¹, by broadcasting uniformly into the floodwater at three or six times (equal splits) between 17th June and 17th November, 2005. The emission fluxes for the first 2 days after application were very high from ADS, the

highest values being 679 compared with a maximum of 156 mg N m⁻² d⁻¹ from CF. Most (61–93%) of the ammonia loss occurred during the first 5 days after each application of fertilizer. The total N loss as ammonia from ADS (29.6–51.7%) was much higher than from CF (12.2%). The highest fluxes were observed in August (2005) when air temperature was highest. More ammonia was lost from the ADS applied at the early stages (i.e. root taking, tiller stages) than at later stages (i.e. elongation, fruiting stages) of rice growth.

H. Hou (✉) · M. Hosomi
Faculty of Engineering,
Tokyo University of Agriculture & Technology,
2-24-16 Naka-cho, Koganei, Tokyo 184-8588, Japan
e-mail: houghong@cc.tuat.ac.jp

S. Zhou
Graduate School of Engineering,
Tokyo University of Agriculture & Technology,
2-24-16 Naka-cho, Koganei, Tokyo 184-8588, Japan

K. Toyota
Graduate School of Bio-Applications and Systems
Engineering (BASE), Tokyo University of Agriculture
& Technology, 2-24-16 Naka-cho, Koganei,
Tokyo 184-8588, Japan

K. Yosimura · Y. Mutou · T. Nisimura ·
M. Takayanagi · T. Motobayashi
Faculty of Agriculture,
Tokyo University of Agriculture & Technology,
3-5-8 Saiwai-cho, Fuchui, Tokyo 183-8509, Japan

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1 Introduction

Release of ammonia from animal wastes following their field application has become one of the major air pollution problems in recent years stimulating worldwide concern. Combined ammonia emissions from fertilizer and livestock were reported to account for about 90 and 77% of anthropogenic ammonia release in Europe and in most of Asia, respectively (Ni, 1999). Ammonia emission from urban human activities and agriculture in the Tokyo metropolis is the highest in Japan (Kannari, Baba, & Hayami, 2001) and the wet deposition flux of NH₄⁺ observed in the

Tokyo urban area was the largest in Japan (Murano, 1998). The emission of ammonia from in the Kanto region by local emission sources had a significant influence on air quality and acidification of the environment in this area (Sakuraia, Fujitaa, Hayami, & Furuhashi, 2003).

Anaerobically-digested cattle slurry (ADS), which are waste products from power generation facilities, are a major concern from an environmental perspective, since they contain considerable amounts of organic matter. ADS is rich in nitrogen, an essential nutrient for plant growth. The use of ADS as organic manure may supplement the use of chemical fertilizers. These residues are available in large amounts for soil amendment and offer an opportunity to improve crop production, while at the same time avoiding adverse environmental impacts. ADS has been used as a liquid manure to be applied directly to land in the UK (Smith, Brewer, Crabb, & Dauven, 2000) and in some Asian countries such as China and India. However, in Japan, direct application of ADS to land is not widespread. In general, costly secondary processing of ADS is carried out before discharging to the environment. When ADS is applied to farmland, a large amount of NH_3 gas may be transferred to the atmosphere. This can lead to high local exacerbation of NH_4 and NH_3 loads. When deposited, such pollutants can cause undesirable changes in natural ecosystems (Jenkinson, 2001) and the field application of livestock slurry has been identified as a major source of atmospheric ammonia (Cai, 1997), especially when it is spread on the field surface. Therefore, before ADS can be effectively used in significant quantities without unacceptable environmental costs, there is a need to determine rates of ammonia emission from a plant-soil system in which ADS is applied. There are few studies of ammonia volatilization (N losses) from ADS applied to paddy field, and no national estimates of ammonia volatilization from paddy fields in Japan (Yang, Niimi, Kanda, & Suga, 2003). Moreover, comparisons of ammonia volatilization from ADS and chemical fertilizer have not been carried out at the same time and in the same environment. Although some experiments have measured ammonia emission from fields immediately after application of livestock slurry, very few have been conducted in paddy soil, with continuous measurement throughout the season. This study explored the potential use of anaerobically-digested slurry from

livestock waste, following fermentation to methane, as a nutrient source in the cultivation of the forage rice, which is an efficient use of farm waste as livestock feed (National Agriculture and Food Research Organization, Japan). In this paper, we compare the effect of different rates and frequency of ADS application and chemical fertilizer on ammonia emission from puddled transplanted rice grown with continuous ponding in lysimeters, throughout the growing season, in the vicinity of Tokyo. This study was part of the project to determine the N balance from forage rice cultivation fields using the ADS system. One of our objectives was to contribute to the development of reliable decision support systems to inform decision-making on rates and timings of ADS application in the cultivation of forage rice crops.

2 Materials and Methods

2.1 Site and Installation of Lysimeters

The field experiments were carried out at the farm (located at approximately $35^{\circ}72'N$ and $139^{\circ}4'W$) of the Field Science Center of Tokyo University of Agriculture and Technology, which is located in Fuchu, Tokyo. Soil was collected from a paddy field on this farm, and was air-dried, ground, and passed through a 2-cm screen to remove rocks, roots, and other large particles. The soil properties are shown in Table 1 (Kyaw, Toyota, Okazaki, Motobayashi, & Tanaka, 2005). An ammonia volatilization experiment was conducted using eight separate small-scale lysimeters, each made of steel with a height of 0.5 m and a diameter of 1 m. Gravel (about 10 cm in depth) was put into the bottom of the lysimeter, and soil (25 cm in depth) was placed above the gravel. There was a drainage outlet at the base of the each lysimeter; the mean drainage rate was $0.96 \pm 0.27 \text{ mm d}^{-1}$ among all the lysimeters during the study period. After packing the soil, the

Table 1 Properties of soil used in this study

Soil	Particle-size distribution			pH (H ₂ O)	Total N (g kg ⁻¹)	Total C (g kg ⁻¹)
	Sand (%)	Silt (%)	Clay (%)			
Clay loam	50	27	23	5.8	4.1	49

lysimeter was compressed by machine to achieve close packing. The packed lysimeters were plowed by rotary stirrer to 20 cm in depth to disperse the clay and reduce water infiltration. The soil surface was flooded a depth of 5 cm, and the water depth was maintained at this level as far as possible during the experimental period by regular additions of water through pipes connected to a pump. Finally, the paddy soil lysimeters were installed in a paddy field of the Field Science Center.

2.2 Experimental Materials

The ADS came from a dairy cattle thermophilic methane fermentation (Hokkiado, Japan) and had a low dry matter content (5.36%), a high ammonium N concentration ($\text{NH}_4\text{-N}$ 1320 mg l^{-1} , T-N 3850 mg l^{-1}), abundant K (water-soluble 2916 mg/l), constant amounts P (T-P 726 mg/l), and a high pH (8.9). Hukugou Rinkaan 42 (Kumiai Chemical Ind., Co., Ltd.) was used as the chemical fertilizer treatment; it consisted of 14%-N, 14%- P_2O_5 , and 14%- K_2O , and the major nitrogen source was ammonia (Ammonium phosphate). It is a highly soluble solid.

Forage rice seedlings (Kusahonami, *Oryza sativa L.*, which a new rice variety for whole-crop silage (WCS) was developed by Japan's National Institute of Crop Science in 2002. Sakai et al., 2003) were transplanted in three rows at a row spacing of 25 cm in each lysimeter on 20th June, 2005. The harvest was carried out on 17th November, 2005. During the forage rice growth period, plant height and total numbers of stems and SPAD value were measured fortnightly. Rice fresh weights, DM yield and N and C concentrations were determined at the time of harvest. During the growing season, there was frequent rainfall which amounted to approx. 810 mm and mean air temperature was 23.0°C.

2.3 Experimental Design

Four treatments were tested (Table 2) with two replicates per treatment. The ADS was applied to the lysimeters at total rates equivalent to 75, 100 and 150 kg N ha^{-1} , by broadcasting uniformly into the floodwater at three or six times (equal splits) between 17th June and 17th November, 2005. After the basal application onto the wet soil surface, the soil was immediately stirred throughout the profile (20 cm) using a soil mixer, while top-dressing, only the floodwater was mixed artificially.

In this study, the rate of fertilizer application was calculated according to N content in CF and ADS, leading to differences in the amounts of P and K in each treatment. However, on the basis of the amount of nitrogen included in the plant body after harvest, we observed that there were no significant differences in the N content, or N uptake above-ground among all the treatments (Table 3). This suggests that the difference of P and K in each treatment may not influence either nitrogen absorption by the rice plant or the plant growth.

2.4 Ammonia Volatilization

A dynamic flow-through chamber method (Kissel, Brewer, & Arkin, 1977) was used to measure NH_3 emissions. A simplified schematic diagram is shown in Fig. 1. It consists of a volatilization chamber, a chemical trap, and a vacuum pump. The chamber, which consisted of a stainless steel cylinder (0.16 m dia and 0.3 m deep), capped with a removable lid, was located above the floodwater surface, with no plants enclosed. One end of a teflon tube was used as an atmospheric inlet and was set up at a height of 2 m above the floodwater surface; the other end was

Table 2 Description of treatments

Sites		Basal dressing		Top-dressing				
		(Lysimeter)	17-Jun	20-Jul	19-Aug	14-Sep	14-Oct	19-Oct
CF 300 (chemical fertilizer)	(A, H)		100/44/83		100/44/83		100/44/83	
ADS 300 (anaerobically-digested slurry)	(B, G)		100/55/221		100/55/221		100/55/221	
ADS 450-1 (anaerobically-digested slurry)	(C, F)		150/83/332		150/83/332		150/83/332	
ADS 450-2 (anaerobically-digested slurry)	(D, E)		75/41/166	75/41/166	75/41/166	75/41/166	75/41/166	75/41/166

*Application rate of N/P/K: kg-N , P, K ha^{-1}

Table 3 Plant maximum height, mean total numbers of stem, mean SPAD value, and contents of N in the above-ground part of rice palnt

Site	CF 300	ADS 300	ADS 450-1	ADS 450-2	LSD
The maximum height (cm)	118	116	117	117	ns
Average number of stem	10	8.3	8.8	10	ns
SPAD value	41	40	41	41	ns
N content (%)	1.9	2.0	2.1	2.1	ns
N uptake above-ground (kg ha ⁻¹)	256	228	221	302	ns

ns, not significant at $P < 0.05$.

connected to the fresh air inlet on the chamber. The air inside the chamber was mixed by a single-speed vacuum pump. When the chambers were in place, the chamber penetrated the paddy soil to a depth of 8–14 cm forming a seal between the floodwater surface and the air within the chamber. The chambers were placed on the lysimeters at approximately 9:00 A.M. for least 1 h before the first sampling. Once the chamber reached steady state conditions, atmospheric ammonia concentration at the center of each lysimeter was determined by drawing air through a vacuum system across the soil surface and passing the air through

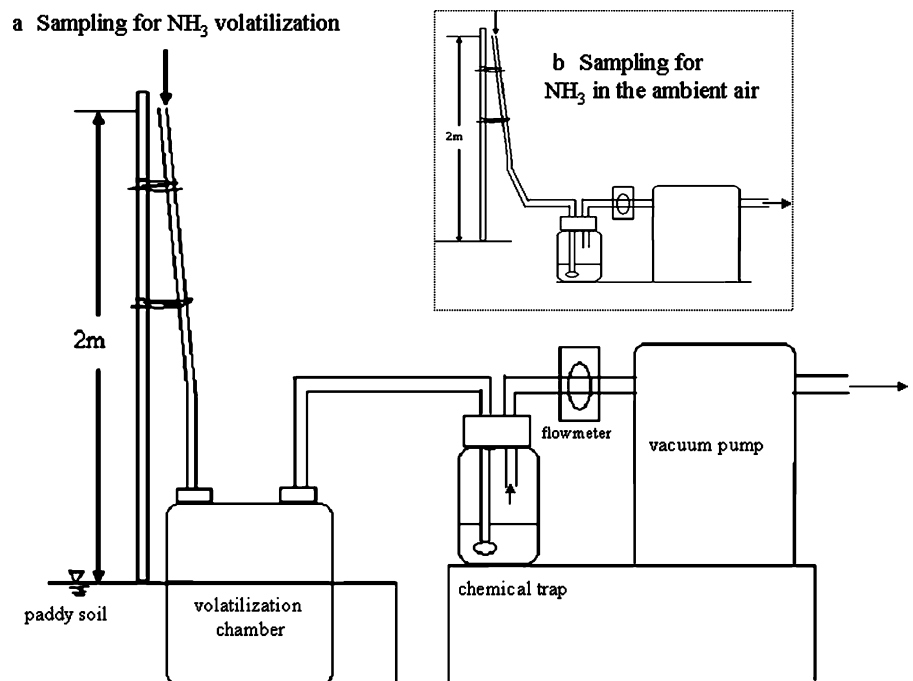
Teflon tubing (0.64 cm outside diameter, 0.40 cm inside diameter) and then through an acid trap containing 60 ml of 0.1 M H₂SO₄. The flow rate of air through the traps was 2.0 l min⁻¹ and the sampling period was 1 h. NH₃ volatilization flux was measured immediately after each application of fertilizer until volatilization was negligible. The measurement frequency was two times per day with 1 h of sampling time, and the times were, in the principle, 10:00–11:00 and 15:00–16:00 in the case of two samplings per day. A background ammonia sample was collected simultaneously from the ambient air at 2 m above the floodwater surface using the same method for each run (Fig. 1b). Ammonium nitrogen content of the traps was determined colorimetrically using the indophenol reaction method (U-2800A Spectrophotometer, HITACHI Japan).

Ammonia emission flux from the paddy was calculated using Eq. 1

$$F_a = [(C_1 - C_0) \cdot V] / (t \cdot A) \quad (1)$$

where F_a is the flux of ammonia gas, C_1 is the concentration of ammonium in the trap solution from the paddy soil, C_0 is the background ammonia concentration in the trap solution, V is the volume of the trap solution, t is duration of pump operation and A is the area of the base of the chamber.

Fig. 1 A sampling device for measurement of ammonia volatilization in a paddy field. The chemical trap bottle contained 60 ml of 0.1 M H₂SO₄ solution



The measurement period was from June 17 to November 2, 2005. Ammonium nitrogen concentration and pH of the floodwater were also measured three times a measurement day. Measurements of air temperature, relative humidity, radiation and rainfall were taken from an automatic measurement weather station less than 30 m away. No ADS was applied to the surrounding area.

The NH_3 volatilization was measured from the soil flooded 2 weeks after the installation of lysimeters, and prior to the addition of fertilizer. The result showed that ammonia fluxes from eight lysimeter soils were low, and average value was $2.34 \pm 1.3 \text{ mg N m}^{-2} \text{ d}^{-1}$. In this study, this value was assumed as background value of this measurement site. The emission factors were calculated according to the formula (2).

$$EF = (N_t - N_n) / N_a \quad (2)$$

Where EF is the emission factor, N_t is cumulative ammonia loss from the plot, N_n is cumulative ammonia loss from the control plot (the background value was used), and N_a is the amount of N applied.

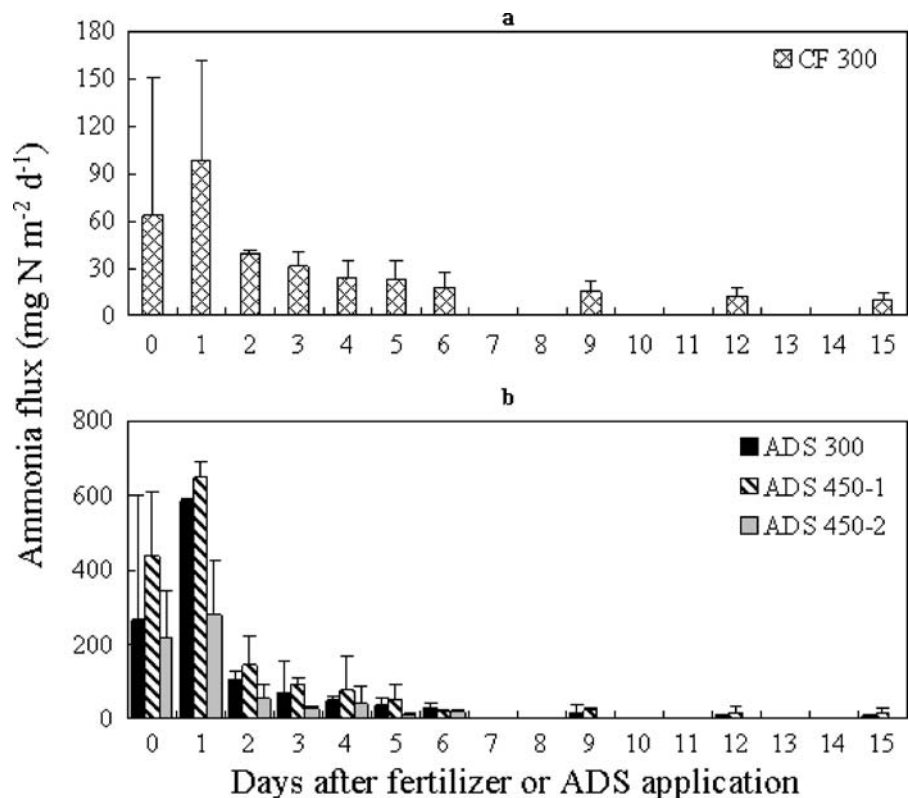
In this study, lysimeters were placed in the paddy field, and measurements of the ammonia volatilization were conducted under a natural situation. Efforts were made to ensure the same physical soil conditions in all lysimeters. Naturally, the artificial nature of the lysimeter may influence the objective evaluation of the ammonia volatilization, but it was considered that our results basically reflect an actual ammonia volatilization.

3 Results and Discussion

3.1 Ammonia Volatilization Following Application of Chemical Fertilizer or ADS

Ammonia fluxes from the floodwater following top-dressing of CF and ADS on Aug. 18, 2005 are shown in Fig. 2. The highest NH_3 fluxes occurred immediately after and on day 1 after fertilizer application, the highest values being 156, 590, 679 and 381 $\text{mg N m}^{-2} \text{ d}^{-1}$ for CF 300, ADS 300, ADS 450-1 and ADS 450-2, respectively. Fluxes declined sharply on the

Fig. 2 Ammonia fluxes following the application of chemical fertilizer **a** or anaerobically-digested slurries **b** into flooded forage rice from Aug. 18 to Sept. 2, 2005



third day. NH_3 fluxes had fallen to low levels by the seventh day after both CF and ADS application. At each date tested, losses of NH_3 were detected and most (61–93%) of the ammonia loss occurred during the first 5 days after each application of fertilizer. This was consistent with previous results (Yang et al., 2003; Smith et al., 2000). Ammonia volatilization shows that the pattern of decline was similar; the only difference was that the flux was significantly lower with CF than with ADS treatments on day 0. This may be due to the $\text{NH}_4^+ - \text{N}$ concentrations in the floodwater on day 0 after application for CF lower than ones for ADS (Fig. 3). After the high initial NH_3 emission, the rate generally fell rapidly as the concentration of $\text{NH}_4 - \text{N}$ in the floodwater decreased, perhaps due to emission, infiltration, nitrification, denitrification, soil immobilization, and plant uptake (van der Molen, Beljaars, Chardon, Jury, & Vanfaassen, 1990). Several investigators have reported that high NH_4^+ concentration in floodwater enhance NH_3 volatilization (e.g. Hayashi, Hishimura, & Yagi, 2006).

The relationships between NH_3 flux and time during the first 16 days after fertilizer application on 18th August were derived (Fig. 4). The data fitted an exponential relationship ($Y = a X^{-b}$: where Y is NH_3 flux and X is time), with the constants a and b varying depending on the application rate and the nature of the fertilizer. The constants were lowest with the CF 300 (85.7, 0.166 respectively) and highest with the ADS 450-2 (403, 0.486, respectively). All ADS treatments had similar a or b values, indicating that a and b may relate to the nature of the fertilizer.

3.2 Seasonal Variation in NH_3 Fluxes

Seasonal variation in ammonia flux from flooded forage rice for the four different treatments are presented in Fig. 5. The seasonal variation in NH_3 flux followed a similar pattern for all treatments. At each application time, ammonia fluxes were very high for the first 2 days after application, decreased rapidly by about 10-fold by the third day and continued to decrease by 20-fold from starting values by the seventh day. After 2 weeks, NH_3 fluxes approached background values. It appeared that ammonia emission showed no obvious seasonal variation and was affected principally by fertilizer application. This suggests that fertilizer application, especially ADS, is the main cause of large-scale NH_3 volatilization into the atmosphere. Misselbrook et al. (2002) stated that the spreading of animal manures on land accounts for approximately one third of the total NH_3 emissions from agriculture to the atmosphere. A similar conclusion was reported by other researchers (i.e. Cai, 1997; Pain, Van der Weerden, Chambers, Phillips, & Jarvis, 1998).

Rates of NH_3 emission from surface-applied ADS are markedly affected by current weather conditions, especially air temperature, wind-speed and solar radiation; the transport of NH_3 away from the surface to the atmosphere is therefore primarily a function of local meteorological conditions (Sommer & Hutchings, 2001). NH_3 fluxes during the middle of July to the beginning of September were higher than at other times (Fig. 5). This tendency corresponded to increases in air temperature suggesting a causative link. This

Fig. 3 Changes of $\text{NH}_4 - \text{H}$ concentration in the floodwater after application of chemical fertilizer or anaerobically-digested slurries from Aug. 18 to Sept. 2, 2005

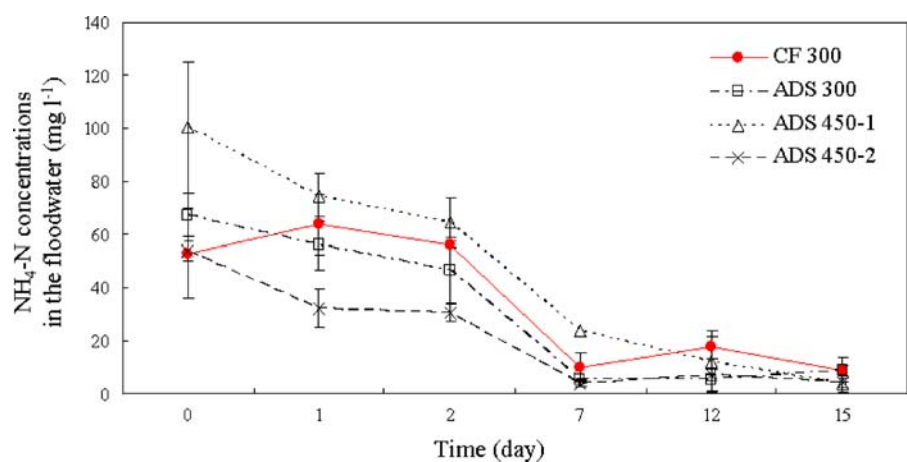


Fig. 4 Relationships between NH_3 flux from the paddy soil and time from the paddy soil after application of chemical fertilizer and anaerobically-digested slurries from 18th August to 2nd September, 2005

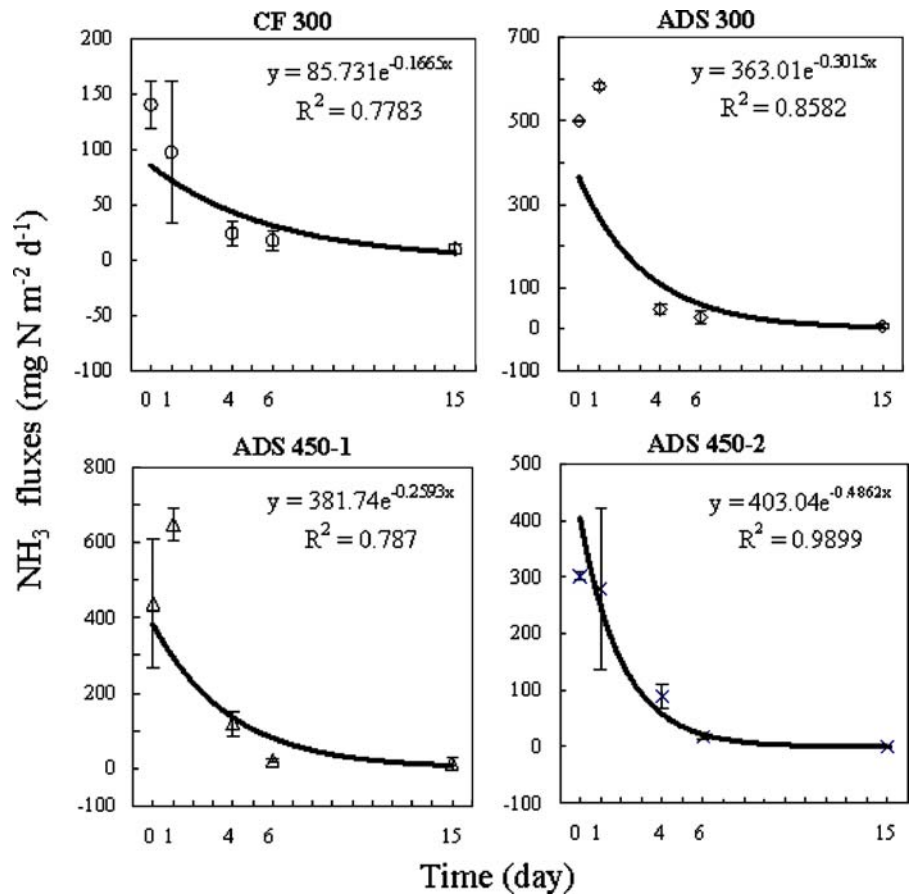


Fig. 5 Seasonal variations in temperature and ammonia flux from the flooded forage rice fields during a 151-day of rice cultivation period. The arrows showed times of fertilizers or anaerobically-digested slurry application. The solid ones showed that fertilizers were applied in the all treatments, and the hollow ones showed slurry application only to be conducted in the ADS 450-2. (open square) CF 300, (open triangle) ADS 300, (open circle) ADS 450-1, (filled circle) ADS 450-2, and (X symbol) temperature. Vertical bars represent one deviation of the ammonia flux

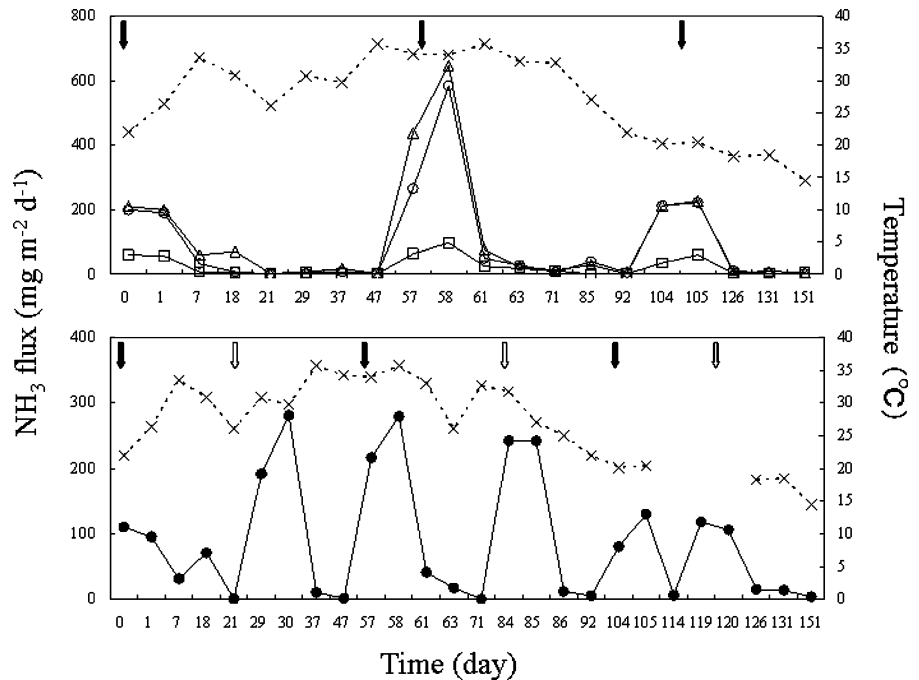


Table 4 Average values of ammonia flux, pH, cumulative emission and emission factor from the four treatments during the forage rice cultivation period

Site		CF 300	ADS 300	ADS 450-1	ADS 450-2
NH ₃ flux (mg N m ⁻² d ⁻¹)	Average	27.3	105	117	90.6
	Range	0–143	0–590	0–679	0–381
Cumulative NH ₃ emission (Kg N ha ⁻¹)	Average	36.7	155	170	133
	Range	0–216	0–890	0–1025	0–575
Emission factor* (%)	Average	12.2	51.7	37.7	29.6
Increase in NH ₃ loss, % relative to CF	Average	0.0	324	209	143
pH	Average	6.89	7.32	7.43	7.42
	Range	5.76–8.08	6.72–8.36	6.76–8.27	6.70–9.06

*: Emission factor, ammonia loss, % of applied N.

increasing air temperature presumably increases floodwater temperature and drives the evaporation of water thus effectively increasing the NH₃ concentration of the floodwater. A significant relationship between NH₃ flux and air temperature could not be derived directly in this study, but a positive correlation between has been verified by a number of researchers (Sommer, Friis, Bake, & Schjørring, 1997; Smith et al., 2000).

3.3 Effect of N Source on NH₃ Volatilization

Table 4 summarizes ammonia fluxes, pH values, cumulative emissions and emission factors from the four treatments during 151 days from basal application to 5 days after harvest. Variation in rates of emission was large reflecting the influence of a wide range of factors. The mean NH₃ losses (denoted here as emission factor %) were 12.2% of the application N for CF 300, 51.7% for ADS 300, 37.7% for ADS 450-1 and 29.6% for ADS 450-2 treatments. Thus, compared to the conventional chemical fertilizer, overall increases in NH₃ emissions following application were 324, 209, and 143% for ADS 300, ADS 450-1 and ADS 450-2, respectively. In the case of the same application rate, ammonia loss as well as emission factor from the ADS 300 was about four times higher than from CF 300. It is clear that emissions were consistently higher with ADS application compared to chemical fertilizer application. Comparison of the ammonia emission between ADS 450-1 and ADS 300 indicates that increasing application rate from 300 to 450 kg N ha⁻¹ decreased ammonia losses from 52 to 38% of the applied N. The ammonia loss decreased with increasing application rate was consistent with the result of Klarenbeek and Bruins (1991). From our results it appears that ammonia emission from the rice field was affected significantly by different N levels

and the nature of the fertilizer applied. Although both of ADS 450-2 and ADS 450-1 had the same application rate, the total NH₃ emission was much lower the more frequent but lower application rate of ADS 450-2. These effects may be attributed to a different level of initial build up of ammoniacal-N concentration in the floodwater, and partly due to different pH at the surface. A significant relationship ($r=0.656$, $p<0.01$) between the NH₄⁺ concentration in the floodwater and NH₃ flux is apparent in Fig. 6, indicating a greater dependence of NH₃ emission rate on NH₄⁺ content in the floodwater. Ammonia losses occur primarily from the surface of ammoniacal solutions in water and such liquid surfaces are found in association with fertilizers and slurries (Sommer & Hutchings 2001). Ammonia concentrations in air close to the slurry surface is in equilibrium with the dissolved NH₃ (Génermont & Cellier, 1997). This

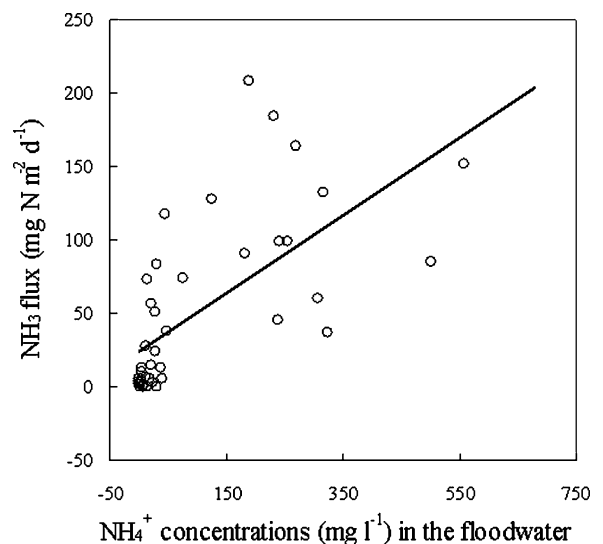


Fig. 6 Plot of NH₃ fluxes versus NH₄⁺ – N concentrations in the floodwater. $Y = 0.2645x + 24.277$, $r=0.656$, $p<0.01$

suggests that the concentration of NH_3 in the floodwater is a dominant factor among all the chemical and physical conditions controlling ammonia emissions. Fillery and Vlek (1986) also point out that the quantity of ammoniacal N in floodwater is an index of the potential for NH_3 volatilization. However, NH_3 emission is also relatively sensitive to pH over the range commonly found in ammonia solutions; a change of pH from 6.3 to 7.2 would increase emissions five times while a change from pH 7.2 to 8.4 would increase emissions by 10 times according to our preliminary indoors examination. NH_3 losses on the CF 300 treatment were lower than any of the ADS treatments perhaps because the mean pH ($n=27$) over time associated with CF300 (6.89 ± 0.37) was about 0.5 units lower than for all the ADS treatments (7.32 ± 0.38 for ADS300; 7.43 ± 0.39 for ADS450-1; 7.42 ± 0.42 for ADS450-2). Previous studies have shown pH to affect

NH_3 emissions (Aneja et al., 2001; Singh & Nye, 1988). The results above suggest that the three most important parameters among the biological and chemical processes which determine the NH_3 equilibrium and production rate are nitrogen content, pH and temperature.

Although information for ADS being applied to paddy soil was not available, ammonia losses (emission factor %) from the field or paddy, cited from literature, were compared with our data (Table 5). Our emission factor for CF 300 in Tokyo was comparable to all the values cited in other areas, except for one in Fengqiu, China. Our data for ADS also fell within the cited ranges for ADS applications to non-paddy systems, and was near the median. However, NH_3 losses from the paddy treated with ADS were lower than that in Kyusyu, Japan (Yang et al., 2003). This may be because their experiment was conducted on

Table 5 Averages or ranges of ammonia emission from paddy or field as reported in the literature

Sites	Fertilizer types	Application rate	Land use	Sampling method	Emission factor % N applied	References
Fengqiu, China	Ammonium bicarbonate	90 kg N ha ⁻¹	Rice field	Micrometeorological method	39.1	Zhu et al. (1989)
Indian	Urea	90 kg N ha ⁻¹	Rice field	Open acid-trap method	30.1	Dhyani and Mishra (1992)
	Chemical fertilizer	60–240 kg N ha ⁻¹	Rice field	Open acid-trap method	4.58–14.7	
Dan Yang, China	Urea	90 kg N ha ⁻¹	Rice field	Micrometeorological method	8.8	Cai, Zhu, Trevitt, Freney, and Simpson (1986)
	Ammonium bicarbonate				18.2	
Thailand	Ammonium sulfate	50, 100 kg N ha ⁻¹	Rice field	Dynamic flow-through chamber method	12.4, 9.2	Wetselaar, Shaw, Firth, Oupatham, Thitiopoca (1977)
UK	Livestock slurry	30–40 kg N ha ⁻¹	Grassland and Arable	Micrometeorological method	5.3–63.8	Smith et al. (2000)
UK	Livestock slurry	21–51 m ³ ha ⁻¹	Grassland	Micrometeorological method	12–48	Misselbrook et al. (2002)
		25–54 m ³ ha ⁻¹	Arable land		20–83	
kyusyu, Japan	Cattle slurry	109 kg N ha ⁻¹	Grassland	Micrometeorological method	60	Yang et al. (2003)
Swiss	Liquid cattle manure	30 t ha ⁻¹	Grassland	Passive samplers method	27–94	Henzi, Katz, Fahrni, Neftel, and Frick (1998)
Tokyo, Japan	Chemical fertilizer	300 kg N ha ⁻¹	Rice field (forage rice)	Dynamic flow-through chamber method	12.2±2.4	This study
	Anaerobically-digested slurry (orginate in cattle slurry)	300 kg N ha ⁻¹			51.7±2.9	
		450 kg N ha ⁻¹			33.7±6.5	

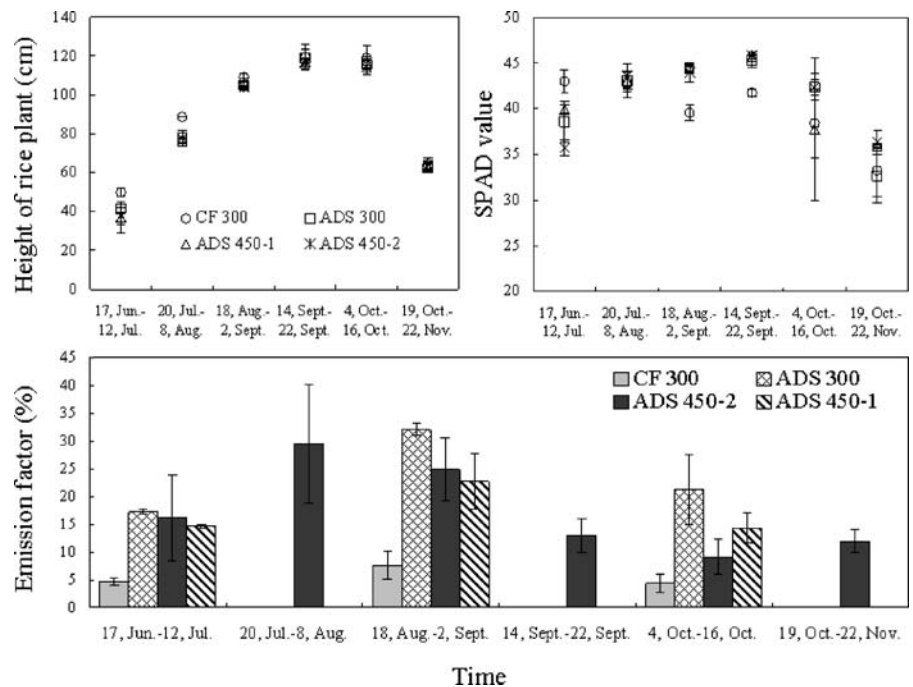
fields ploughed after harvesting corn without any growing plants and the different sampling methods used. The lower ammonia volatilization in our study may be due to the fertilizer or slurry applied being diluted with floodwater, lowering ammonia concentrations in the floodwater. Other researchers have also reported that as the water content in the soil increases, NH_3 emission tends to decrease (Roelle & Aneja, 2002). In addition, a low air-exchange was used in our study and this may also have led to lower estimates of NH_3 emissions. Moreover, the areas for both studies were completely different with different environmental conditions, thus leading to distinct NH_3 loss rates since NH_3 volatilization depends partially on environmental conditions (Zhu et al., 1989).

3.4 Time of ADS Application

Ammonia emission from ADS applied to a flooded rice field varied markedly depending on the time of application and stage of plant growth (Fig. 7). The ammonia loss expressed as percentage of applied N decreased with delay in slurry application. When slurry was broadcast into the floodwater from July to August, and at early stages of plant growth, ammonia loss was highest (above 30% of the applied N). The rice plant is smaller at this stage, resulting in less

shading to restrict photosynthetic activity of micro-organisms. In addition, rice plants have an inefficient root system for absorption of applied N in the early stages (Dhyani & Mishra, 1992). Furthermore, this period coincided with the highest average temperature in Japan, leading to highest floodwater temperature. These factors resulted in an increasing potential for NH_3 volatilization. Ammonia losses were lower (10–15% of the applied N) when ADS were applied at panicle initiation and maturation periods. At these stages of growth, the plant roots rapidly absorbed applied N, and the plant canopy may have shaded the floodwater reducing floodwater temperatures and restricting air movement at the water surface. Zhou, Nakai, and Hosomi (2006) used the same rice variety and observed that the nitrogen absorption rate of the forage rice increased sharply about seven times 2 months after transplanting. The net result was low NH_3 gas concentrations in the floodwater, low transfer coefficients and low rates of volatilization. Previous researchers (Craswell & Vlek, 1982 and Fillery, Simpson, & De Datta, 1984) stated that timing of N application to the crop may affect ammonia volatilization with the maximum loss of N occurring at the early growth stage of the crop. However, this result is not consistent with findings of Dhyani and Mishra (1992), in which the maximum ammonia loss

Fig. 7 Ammonia emission factors in the four treatments from flooded rice fields after application of fertilizers as affected by plant height, numbers of stem, and SPAD value



occurred at the time of basal dressing application. This may be attributed to different fertilizer practices. In this study, anaerobically-digested slurries were mixed immediately with floodwater and the plough layer using a soil mixer after basal dressing application. This may have caused rapid reduction of ammoniacal-N concentration in the floodwater, resulting in less ammonia loss in our study than in the study of Dhyani and Mishra. Lorenz and Steffens (1997) also reported a negative relationship between sward height and NH_3 emission for trailing shoe applications to grassland.

4 Conclusions

In this experiment which investigated the effects of fertilizer type, application rate and amount of application on measured NH_3 fluxes, we found all treatments to yield similar temporal patterns of NH_3 emission. However, NH_3 emission factors among these treatments were very different showing a ranking of $\text{CF } 300 < \text{ADS } 450\text{-}2 < \text{ADS } 450\text{-}1 < \text{ADS } 300$, indicating that ammonia emission is directly affected by the amount of N application, application rate, and fertilizer type. Experiments in which chemical fertilizer and ADS were broadcast into floodwater indicate that the important variables controlling ammonia emissions are the $\text{NH}_4^+ - \text{N}$ concentration, pH and air temperature; other factors exerted their influences through effects on those primary variables. Therefore, further work should focus on field and laboratory studies to further investigate these relationships to help refine the NH_3 budget for managing ADS application to paddy soil. Moreover, further work is certainly warranted to determine whether these results for ADS apply to different soils and floodwater in other environments. The various results from our study indicate that in this particular Japanese environment, application of ADS increased ammonia emission more than chemical fertilizer. Thus, a new method of ADS application is required to decrease ammonia emission to the atmosphere.

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References

- Aneja, V. P., Roelle, P. A., Murray, G. C., Southerland, J., Erisman, J. W., Fowler, D., et al. (2001). Atmospheric nitrogen compounds: Emissions, transport, transformation, deposition and assessment. *Atmospheric Environment*, *35*, 1903–1911.
- Cai, G. X. (1997). Ammonia volatilization. In Z. L. Zhu, Q. X. Wen, & J. R. Freney (Eds.), *Nitrogen in Soils of China* (pp. 193–213). Dordrecht, The Netherlands: Kluwer.
- Cai, G. X., Zhu, Z. L., Trevitt, A. C. F., Freney, J. R., & Simpson, J. R. (1986). Nitrogen loss from ammonium bicarbonate and urea fertilizers applied to flooded rice. *Fertilizer Research*, *10*, 203–215.
- Craswell, E. T., & Vlek, P. L. G. (1982). Nitrogen management for submerged rice soils. In *Vertisols and Rice Soil of the Topics* (pp 158–81). Proceedings of 12th International Congress of Soil Science, held during 8–14 February 1982 at New Delhi.
- Dhyani, B. P., & Mishra, B. (1992). Effect of nitrogen-application schedule on ammonia volatilization from field of rice (*Oryza sativa*). *Indian Journal of Agricultural Science*, *62*, 73–74.
- Fillery, I. R. P., Simpson, J. R., & De Datta, S. K. (1984). Influence of field environment and fertilizer management on ammonia loss from flooded rice. *Soil Science Society of America Journal*, *48*, 914–920.
- Fillery, I. R. P., & Vlek, P. L. G. (1986). Reappraisal of the significance of ammonia volatilization as an N loss mechanism in flooded rice fields. *Fertilizer Research*, *9*, 79–98.
- Génermont, S., & Cellier, P. (1997). A mechanistic model for estimating ammonia emission from slurry applied to bare soil. *Agricultural and Forest Meteorology*, *88*, 145–167.
- Hayashi, K., Nishimura, S., & Yagi, K. (2006). Ammonia volatilization from the surface of a Japanese paddy field during rice cultivation. *Soil Science and Plant Nutrition*, *52*, 545–555.
- Henzi, H., Katz, P. E., Fahmi, M., Nefel, A., & Frick, R. (1998). A simple empirical model based on regression analysis to estimate ammonia emissions after manure application. *Atmospheric Environment*, *32*, 301–307.
- Jenkinson, D. S. (2001). The impact of humans on the nitrogen cycle, with focus on temperate arable agriculture. *Plant and Soil*, *228*, 3–15.
- Kannari, A., Baba, T., & Hayami, H. (2001). Estimation of ammonia emission in Japan. *Journal of Japan Society for Atmospheric Environment*, *36*, 29–38 (in Japanese).
- Kissel, D. E., Brewer, H. L., & Arkin, G. F. (1977). Design and test of a field sampler for ammonia volatilization. *Soil Science Society of America Journal*, *40*, 1133–1138.
- Kyaw, K. M., Toyota, K., Okazaki, M., Motobayashi, T., & Tanaka, H. (2005). Nitrogen balance in a paddy field planted with crop rice (*Oryza sativa* cv. Kusahonami) during two rice growing season. *Biology and Fertility of Soils*, *42*, 72–82.
- Klarenbeek, J. V., & Bruins, M. A. (1991). NH_3 emission after land spreading of animal slurries: NH_3 and odour emissions from livestock production. In V. C. Nielsen, J. H. Voorburg, & P. L'Hermité (Eds.), *Elsevier Applied Science* (pp. 105–115). London and New York.

- Lorenz, F., & Steffens, G. (1997). Effect of application techniques on ammonia losses and herbage yield following slurry application to grassland: Nitrogen emissions from grasslands. In S. C. Jarvis & B. F. Pain (Eds), (pp. 287–192). UK: CAB International.
- Misselbrook, T. H., VanderWeerden, T. J., Pain, B. F., Jarvis, S. C., Chambers, B. J., Smith, K. A., et al. (2002). Ammonia emission factors for UK agriculture. *Atmospheric Environment*, *34*, 871–880.
- Murano, K. (1998). Wet deposition of ammonium and atmospheric distribution of ammonia and particulate ammonium in Japan. *Environmental Pollution*, *102*(S1), 321–326.
- Ni, J. Q. (1999). Mechanistic models of Ammonia release from liquid manure: A review. *Journal of Agricultural Engineering Research*, *72*, 1–17.
- Pain, B. F., van der Weerden, T. J., Chambers, B. J., Phillips, V. R., & Jarvis, S. C. (1998). A new inventory for ammonia emissions from UK agriculture. *Atmospheric Environment*, *32*, 309–313.
- Roelle, P. A., & Aneja, V. P. (2002). Characterization of ammonia emission from soils in the upper coastal plain, North Carolina. *Atmospheric Environment*, *36*, 1087–1097.
- Sakai, M., Iida, S., Maeda, H., Sunohara, Y., Nemoto, H., & Imbe, T. (2003). New rice varieties for whole crop silage use in Japan. *Breeding Science*, *53*, 271–275.
- Sakuraia, T., Fujitaa, S. I., Hayami, H., & Furuhashi, N. (2003). A case study of high ammonia concentration in the nighttime by means of modeling analysis in the Kanto region of Japan. *Atmospheric Environment*, *37*, 4461–4465.
- Singh, R., & Nye, P. H. (1988). A model of ammonia volatilization from applied urea. Development of the model. *Journal of Soil Science*, *37*, 9–20.
- Smith, K. A., Brewer, A. J., Crabb, J., & Dauven, A. (2000). A survey of the production and use of animal manures in England and Wales; III. Cattle manure from dairy and beef production. Draft paper accepted for publication, Soil Use and Management (2000). (Also Parts I and II relating to pig and poultry manures, respectively).
- Sommer, S. G., Friis, E., Bake, A. B., & Schjørring, J. K. (1997). Ammonia emission from pig slurry applied with trail hoses or broadcast to winter wheat: Effects of crop developmental stage, microclimate, and leaf ammonia absorption. *Journal of Environmental Quality*, *26*, 1153–1160.
- Sommer, S. G., & Hutchings, N. J. (2001). Ammonia emission from field applied manure and its reduction – invited paper. *European Journal of Agronomy*, *15*, 1–15.
- Van Der Molen, J. A., Beljaars, C. M., Chardon, W. J., Jury, W. A., & Vanfaassen, H. G. (1990). Ammonia volatilization from arable land after application of cattle slurry. 2. Derivation of a transfer model. *Netherlands Journal of Agricultural Science*, *38*, 239–254.
- Wetselaar, R. T., Shaw, T., Firth, P., Oupatham, J., & Thitiopoca, H. (1977). Ammonia volatilization from variously placed ammonium sulphate under lowland rice field conditions in central Thailand. Proc. Int. Seminar SEFMIA. October 10–14, 1977. Tokyo, Japan. Soc of Sci of Soil and Manure, Japan.
- Yang, Z., Niimi, H., Kanda, K. I., & Suga, Y. (2003). Measurement of ammonia volatilization from a field, in upland Japan, spread with cattle slurry. *Environmental Pollution*, *121*, 463–467.
- Zhou, S., Nakai, S., & Hosomi, M. (2006). Nitrogen transformation in surface flow wetland planted forage rice receiving river water. *Proceedings of 10th International Conference on Wetland Systems for Water Pollution Control*, 219–227.
- Zhu, Z., Cai, G., Simpson, J. R., Zhang, S. L., Chen, D. L., Jackson, A. V., et al. (1989). Processes of nitrogen loss from fertilizers applied to flooded rice fields on a calcareous soil in north-central China. *Fertilizer Research*, *18*, 101–115.