

Effects of option mitigating ammonia volatilization on CH₄ and N₂O emissions from a paddy field fertilized with anaerobically digested cattle slurry

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Abstract A lysimeter experiment was carried out to evaluate the effects of the NH₃ volatilization mitigation by adding anaerobically digested cattle slurry (ADCS) alone, with wood vinegar (WV) or with a higher level of floodwater (HFW), on emissions of CH₄ and N₂O from a paddy soil planted with fodder rice. We have carried out the following treatments: (1) chemical fertilizer, (2) ADCS, (3) ADCS + WV, and (4) ADCS + HFW; the height of floodwater was 10 cm in the latter treatment, and it was 3 to 4 cm in the other treatments just before fertilizer applications. Nitrogen fertilizer rate added to soil in each treatment was 30 g NH₄⁺-N m⁻² (split in one basal and two top-dressing additions). Ammonia volatilization in the ADCS treatment was 2.7 g NH₃-N m⁻² throughout the growing season, and it was significantly reduced by 79% and 55% in the ADCS + WV and ADCS + HFW treatments, respectively. The total amount of CH₄ emitted in the ADCS treatment in the growing season was not significantly enhanced by the mitigation of NH₃ volatilization either by

adding wood vinegar or by increasing the height of the floodwater. Negligible N₂O emissions were observed in all treatments during the growing period.

Keywords Ammonia mitigations · Anaerobically digested cattle slurry · Wood vinegar · Floodwater management · Fodder rice

Introduction

During the period from 1960 to 2005, the rice consumption per capita is decreased from 115 to 61 kg year⁻¹ with an increase in livestock products from 32 to 137 kg year⁻¹ in Japan (Ministry of Agriculture, Forestry and Fisheries 2006). To increase the domestic production of fodder and stimulate the use of paddy fields, cultivation of whole crop rice varieties, in which the whole plant parts are used for fodder, is now increasing (Ministry of Agriculture, Forestry and Fisheries 2007).

The increased animal husbandry is associated with the management of waste, which can lead to environmental pollution. The anaerobic digestion of the animal waste draws a great attention since it can generate renewable energy and can compensate the CO₂ emissions from fossil energy sources. However, the management of the relative residue, the anaerobically digested slurry, can be a problem.

We have studied the use of anaerobically digested cattle slurry (ADCS) as a fertilizer in paddy fields (Hou et al. 2007; Sunaga et al. 2010). The ADCS fertilization of paddy fields planted with whole crop rice varieties can be advantageous for recycling the organic waste, reducing the use of chemical fertilizers, and increasing fodder production if negative impacts on the plant and the environment do not occur. However, ammonia volatiliza-

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tion can occur in paddy fields fertilized with ADCS because of the high pH and high ammonia concentration of ADCS (Hou et al. 2007). We have shown that ammonia volatilization was decreased by 63% to 82% when ADCS was applied with an acid residue, wood vinegar, or the floodwater level was increased (Win et al. 2009). However, the addition of wood vinegar may increase CH₄ emission because its major constituent acetic acid is a substrate for methanogens (Kyuma 2004) and the decomposition of ADCS can stimulate reduction processes. In addition keeping high floodwater level may also stimulate CH₄ emission due to the decrease in redox potential (Mosier et al. 2004). Therefore, it is needed to estimate the effect of these ammonia mitigation treatments on emissions of different greenhouse gases. The objective of this study was to evaluate the effects of two ammonia mitigation treatments (addition of wood vinegar and increase in the floodwater level at the application) on CH₄ and N₂O emissions from a paddy soil fertilized with ADCS.

Materials and methods

Experimental site and treatments

This experiment was conducted in 12 stainless steel lysimeters (1 m×1 m, 0.5 m depth), prepared in 2007 with a gravel layer at the lowest 10 cm, a compacted 7 cm subsoil layer, and a top 20 cm plowed layer. The soil was a gray lowland soil (Fluvisols: total C 35.0 gkg⁻¹; total N 5.0 gkg⁻¹; NH₄⁺-N 4.35 mg Nkg⁻¹; pH [H₂O] 6.0) collected from the Field Museum Hommachi, Field Science Centre, Tokyo University of Agriculture and Technology, Fuchu, Tokyo. The experiment is that already reported by Win et al. (2009) and including the following four treatments, each replicated three times: (1) chemical fertilizer (CF), (2) ADCS, (3) ADCS + wood vinegar (ADCS + WV), and (4) ADCS + higher floodwater level (ADCS + HFW).

Nitrogen application rate (30 g NH₄⁺-N m⁻²) was higher than that (10 g NH₄⁺-N m⁻²) normally used by farmers because we wanted to test the effect of the enriched waste treatments on crop production. The fertilizer was split into the basal application (10 g NH₄⁺-N m⁻²) on June 4, the first top dressing (10 g NH₄⁺-N m⁻²) at the maximum tillering stage (July 31) and the second top-dressing (10 g NH₄⁺-N m⁻²) at the flowering stage (September 20). ADCS had the following chemical properties: pH 7.5, NH₄⁺-N 1.94 g L⁻¹ and total N 3.49 g L⁻¹. Three 1-month-old rice seedlings (*Oryza sativa* L. var. Leaf star) were transplanted per hill with a spacing of 30 cm×15 cm on June 11; thus, these were 28 hills per lysimeter. There was no drainage, and thus, water was lost by evapotranspiration

from lysimeters. Irrigation was done every 3 to 4 days to keep the floodwater level at about 4 cm, except for the ADCS + HFW treatment, where the level was kept high at about 10 cm, just before each fertilizer application. Insecticides (Perdon, Cartap hydrochloride) and fungicides (Rinber, Furametpyr) were each applied at the rate of 30 kg ha⁻¹ (conventional dosage) on July 21 and August 26 for the suppression of rice stem borer and sheath blight, respectively. Rice plants were harvested on November 7, 2008, and no management was done after the harvest.

Analysis

Ammonia volatilization was monitored by the dynamic flow-through chamber method (Kissel et al. 1977), with measurement and calculation as mentioned by Hou et al. (2007) and Win et al. (2009). Gas samples were taken at weekly intervals during the growing period and every 2 months during the fallow period by using the closed chamber method. The gas sampling was done during the day time (from 10:00 to 16:00) using four chambers. A plexiglas chamber consisting of two parts was used: The lower part was 30 cm×30 cm and it was 50 cm high, whereas the upper one was 30 cm×30 cm and it was 100 cm high. The upper part was installed with an inner fan operated by a 6-V battery and a 1-l Tedlar[®] bag for regulating the inside pressure. At the time of sampling, the paddy soil surface with two rice hills was firstly covered by the lower part of the chamber. The physical disturbance brought by the insertion of chamber sometimes triggers accidental emissions of bubbles, and therefore, the upper part was connected 10 min after the insertion of the lower part. Gas samples (30 ml) were collected with a 50-ml syringe after 0, 10, and 20 min and immediately transferred into pre-evacuated 10-ml vials. Temperature in the chamber was recorded by a micro-temperature thermometer (PC-9125, AS ONE Co., Tokyo, Japan). Both CH₄ and N₂O concentrations were analyzed by a gas chromatograph with flame ionization detector (FID) (GC-14B, Shimadzu, Kyoto, Japan) and electron capture detector (GC-14A, Shimadzu, Kyoto, Japan), respectively. The FID-GC was equipped with a Porapak N (80/100 mesh) column; CH₄ analysis was done by using carrier gas (He) with column temperature at 80°C, injection, and detector temperatures at 180°C; N₂O analysis was done as reported by Yanai et al. (2007).

Based on the linear rate increase of the gas concentrations with time (min), the gas fluxes Q (mg m⁻² min⁻¹) were calculated by the following equation of Rolston (1986).

$$Q = (V/A) \times (\Delta C/\Delta T) \times (M/22.4) \times (273/K) \quad (1)$$

where V is the headspace volume (m³) of the chamber, A is the base chamber area (m²), $(\Delta C/\Delta T)$ is the change in the

gas concentration (mg m^{-3}) per time unit T (min), M is the molar weight of the gas, and K is Kelvin temperature of air inside the chamber.

Cumulative emission was obtained by multiplying the daily flux at each measurement for the time interval and sum up the values. Caution is required to consider total gas emissions because our sampling frequencies were sparse, especially during the fallow period, and day and night fluctuation was not considered in this study.

At each gas sampling, pH and temperature in the floodwater were measured by a portable pH meter (WM-22 EP, DKK-TOA Co., Tokyo, Japan), and the depth (cm) of the floodwater was measured by a ruler. Plant height, tiller numbers, and SPAD values (SPAD-502, Konica Minolta Censing Inc., Sakai, Japan) were periodically measured by selecting five plants per lysimeter.

At harvest (November 7, 2008), six plants per lysimeter were sampled, and the total dry matter of each plant was measured. Leaves and grains were separately measured after incubation at 80°C for 24 h. Three composite soil samples were collected after harvesting (November 14, 2008) from two soil layers (0 to 2 cm) and (2 to 20 cm) from each lysimeter and analyzed for the total C and N contents by a CN coder (MT-700, YANACO New Science Inc., Kyoto, Japan).

Statistical analysis

The gas emissions (g m^{-2}) during the growing season were analyzed by one-way ANOVA (a software package Excel statistics version 12, SPSS Inc., Tokyo, Japan), whereas the seasonal gas fluxes ($\text{mg m}^{-2}\text{day}^{-1}$) and floodwater properties throughout the growing season were analyzed by two-way ANOVA. Mean comparison was done by $\text{LSD}_{0.05}$ (Fisher).

Results

Ammonia, CH_4 , and N_2O emissions

Ammonia volatilization was significantly ($P < 0.05$) lower in the CF, ADCS + HFW, and ADCS + WV treatments than

in the ADCS treatment at all fertilization times (the basal, first top dressing and second top dressing) (Fig. 1). Total NH_3 volatilization from June 4 to September 27 as a percentage of the applied $\text{NH}_4^+\text{-N}$ amounted to 8.9% ($2.66 \text{ g NH}_4^+\text{-N m}^{-2}$), 4.0% ($1.20 \text{ g NH}_4^+\text{-N m}^{-2}$), and 1.9% ($0.564 \text{ g NH}_4^+\text{-N m}^{-2}$) in the ADCS, ADCS + HFW, and ADCS + WV treatments, respectively.

There were no significant differences among the treatment means of daily CH_4 and N_2O fluxes, whereas there were differences among the relative seasonal variations in fluxes. In the CF, ADCS, and ADCS + DFW treatments, CH_4 fluxes occurred about 1 month after submergence, and peak emissions were observed at the maturity stage (October 3, 9, and 16, respectively) (Fig. 2). Methane fluxes in the three treatments from October 3 to 16 were $0.802 \text{ g CH}_4 \text{ m}^{-2}\text{day}^{-1}$, 2.26 and $2.35 \text{ g CH}_4 \text{ m}^{-2}\text{day}^{-1}$, respectively. In the ADCS + WV treatment, the flux started to increase 1 month after submergence, and the maximum peak ($2.11 \text{ g CH}_4 \text{ m}^{-2}\text{day}^{-1}$) was observed on July 17 (Fig. 2). Mean CH_4 fluxes throughout the growing season were 0.408 ± 0.516 , 0.930 ± 1.27 , 1.03 ± 0.817 , and $1.07 \pm 1.37 \text{ g CH}_4 \text{ m}^{-2}\text{day}^{-1}$ in the CF, ADCS, ADCS + VW, and ADCS + HFW treatments, respectively, whereas those during the fallow period were 4.9 ± 4.8 , 8.3 ± 2.7 , 8.9 ± 5.0 , and $10.6 \pm 12.1 \text{ mg CH}_4 \text{ m}^{-2}\text{day}^{-1}$, respectively. Though annual CH_4 emission was not significantly different among the treatments, all ADCS treatments emitted about twice as CF did (59.5 , 146 , 147 , and $153 \text{ g CH}_4 \text{ m}^{-2} \text{ season}^{-1}$ in CF, ADCS, ADCS + WV, and ADCS + HFW treatments, respectively).

The maximum negative N_2O flux ($-7.1 \text{ mg N}_2\text{O m}^{-2}\text{day}^{-1}$) was observed in the ADCS + HFW treatment on August 19, whereas the maximum positive flux ($2.2 \text{ mg N}_2\text{O m}^{-2}\text{day}^{-1}$) occurred on August 7 (maximum tillering) in the ADCS + WV treatment, and mean fluxes were -0.76 ± 1.54 , -0.44 ± 1.07 , -0.05 ± 1.28 , and $-0.51 \pm 1.90 \text{ mg N}_2\text{O m}^{-2}\text{day}^{-1}$ in the CF, ADCS, ADCS + WV, and ADCS + HFW treatments, respectively (Fig. 3). The N_2O emission during the growing season (from June 30, 2008 to October 27, 2008) were negative in all the treatments, $-0.13 \text{ g N}_2\text{O m}^{-2}$, $-0.06 \text{ g N}_2\text{O m}^{-2}$, and $-0.01 \text{ g N}_2\text{O m}^{-2}$ and $-0.12 \text{ g N}_2\text{O m}^{-2}$, respectively, and those during the

Fig. 1 Ammonia volatilization after the application of fertilizers at basal and first and second top dressing. Bars represent standard deviation of the mean ($n=3$). CF chemical fertilizer, ADCS anaerobically digested cattle slurry, WV wood vinegar, HFW higher level of floodwater

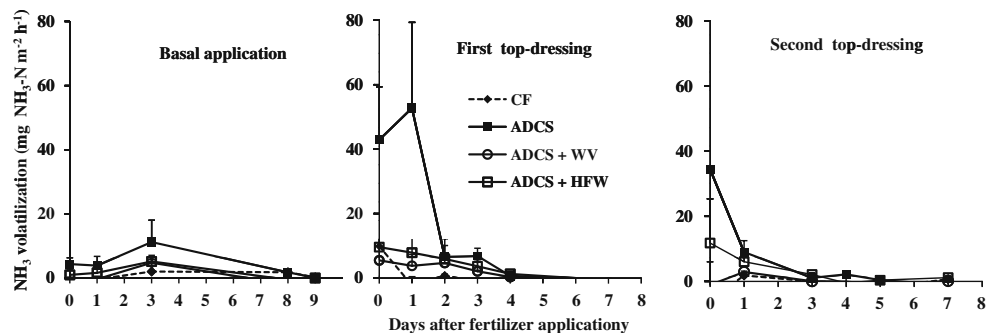
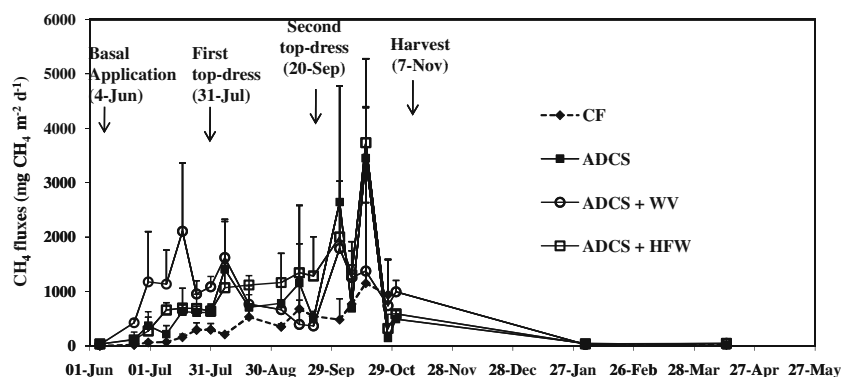


Fig. 2 Methane emissions throughout the whole year (rice growing season from June 4 to November 7 and fallow period). Bars represent standard deviation of the mean ($n=3$). CF chemical fertilizer, ADCS anaerobically digested cattle slurry, WV wood vinegar, HFW higher level of floodwater



fallow period (October 27, 2008 to April 13) were positive in all treatments ($0.17 \text{ g N}_2\text{O m}^{-2}$, $0.07 \text{ g N}_2\text{O m}^{-2}$, $0.24 \text{ g N}_2\text{O m}^{-2}$, and $0.14 \text{ g N}_2\text{O m}^{-2}$, respectively); thus, total amounts of N_2O fluxes in the whole year were 0.04 ± 0.33 , 0.01 ± 0.16 , 0.23 ± 0.86 , and $0.02 \pm 0.57 \text{ g N}_2\text{O m}^{-2}$, in the CF, ADCS, ADCS + WV, and ADCS + HFW, respectively.

Depth, temperature, and pH of floodwater

At the top-dressing days (July 31 and September 20), pH in the ADCS + WV treatment (5.9 ± 0.2) was significantly ($P < 0.05$) the lowest and followed by the CF (6.6 ± 0.2), ADCS (7.6 ± 0.2), and ADCS + HFW (7.7 ± 0.2) treatments. The paddy field was flooded from May 21 to October 31, and the floodwater height during the period ranged from 5.3 to 7.4 cm. Average height of the floodwater in the ADCS + HFW treatment ($8.8 \pm 1.4 \text{ cm}$), at the day of fertilizer application (basal application on June 4, first top dressing on July 31 and second top dressing on September 20) and first and second days after application, was significantly ($P < 0.05$) higher than that in the other treatments: CF ($5.8 \pm 3.1 \text{ cm}$), ADCS ($6.1 \pm 2.6 \text{ cm}$), and ADCS + WV ($5.3 \pm 2.5 \text{ cm}$).

The average seasonal temperature of floodwater was not significantly different among the treatments, ranging from 25.4°C to 25.7°C , whereas it was significantly ($P < 0.05$)

different among months, being the monthly means $27 \pm 3.0^\circ\text{C}$ in June, $28 \pm 3.3^\circ\text{C}$ in July, $31 \pm 1.1^\circ\text{C}$ in August, $24 \pm 2.7^\circ\text{C}$ in September, and $19 \pm 1.5^\circ\text{C}$ in October.

Soil C and N content

At harvest, total C contents in the 0 to 2 cm layer of CF soil were significantly ($P < 0.05$) lower than those in all the ADCS treatments (Table 1). Total C contents in the 2 to 20 cm layer were also higher in all the ADCS soils than in CF soil, although the difference was only significant for the ADCS + WV treatment. A similar tendency was observed in total N contents.

Growth and biomass production of rice

Rice plants in the CF treatment showed earlier tillering than those in the ADCS treatments during the early period (from June 16 to July 14) (Fig. 4). However, tiller number and SPAD value in the CF treatment decreased from July 14 to July 31. Average plant height in all treatments increased until October 3, when no significant differences were observed among all treatments ($141.4 \pm 1.8 \text{ cm}$). There were no significant differences in the tiller number among all treatments since July 31, and the maximum tiller number was observed on August 7 (12 hill^{-1}).

Fig. 3 Nitrous oxide emissions throughout the whole year (rice growing season from June 4 to November 7 and fallow period). Bars represent standard deviation of the mean ($n=3$). CF chemical fertilizer, ADCS anaerobically digested cattle slurry, WV wood vinegar, HFW higher level of floodwater

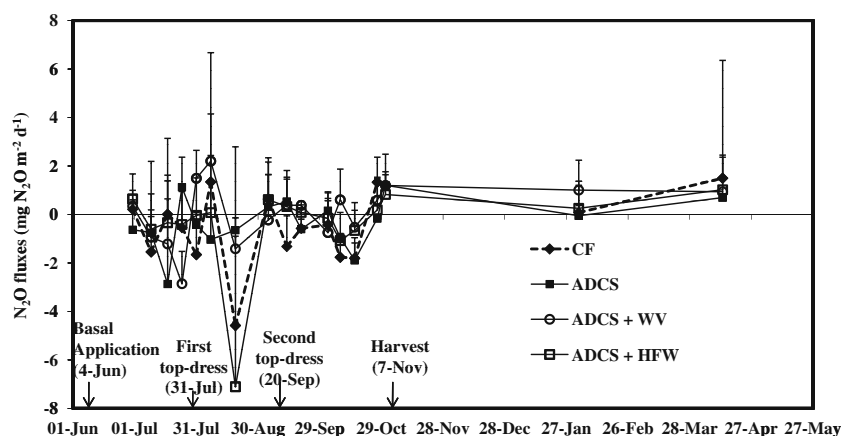


Table 1 Total plant biomass production of forage rice variety (*Oryza sativa* L. Leaf star) and soil C and N contents at harvest as affected by the treatments

Treatments	Grains	Leaves	Total	Soil C (g kg ⁻¹ soil)		Soil N (g kg ⁻¹ soil)	
				g (dry matter) m ⁻²		0–2 cm	2–20 cm
CF	727±8 a	2,550±115 a	3,280±18 a	39.7±2.6 b	36.6±0.4 b	3.7±0.4 b	3.3±0.1 a
ADCS	783±57 a	2,010±86 b	2,800±143 b	62.5±15 a	38.3±1.0 ab	4.6±0.7 ab	3.4±0.1 a
ADCS+WV	714±74 a	2,160±91 b	2,870±135 b	60.6±3.7 a	39.4±1.4 a	5.1±0.2 a	3.6±0.1 a
ADCS+HFW	753±32 a	2,040±81 b	2,800±103 b	65.0±14.9 a	37.8±0.9 ab	5.5±1.0 a	3.5±0.1 a

Different letters show a significant difference among the treatments ($P < 0.05$). Mean value ± standard deviation ($n = 3$)

CF chemical fertilizer, ADCS anaerobically digested cattle slurry, WV wood vinegar, HFW higher level of floodwater

Total rice yield at harvest ranged from 2.8 to 3.3 kg (dry matter) m⁻², and it was significantly ($P < 0.05$) higher in the CF treatment than in all ADCS treatments, whereas there was no significant difference among the treatments in the grain yield (Table 1). The C contents of leaves and panicle were 373 g C and 389 g C kg⁻¹ dry matter, respectively. Consequently, total C of the rice plant at harvest ranged from 1,060 to 1,230 g C m⁻² (Table 2).

Carbon balance

Soil C contents, which were enhanced in all the ADCS treatments, were much higher than amount of C fixed by rice plants that was enhanced in the CF treatment (Table 2). Global warming potential calculated on CH₄ and N₂O emissions was more than twice in the ADCS treatments than in the CF treatment. Methane emissions largely contributed to this result, while the contribution of N₂O was negligible. There was no significant difference in the net C balance among the treatments.

Discussion

Ammonia volatilization losses observed in this study (2008 growing year) were comparable to those previously observed in the 2007 (Win et al. 2009) and confirm that the NH₃

mitigation treatments, addition of wood vinegar (ADCS + WV), and increasing height of floodwater (ADCS + HFW) can decrease NH₃ volatilization.

The CH₄ emissions observed in this study (60 to 150 g CH₄ m⁻²) were comparable to those reported by Furukawa and Inubushi (2002) (80 to 113 g CH₄ m⁻², pot experiment) and Ali et al. 2008 (236 g CH₄ m⁻², pot experiment), but much higher than the average values of Japanese paddy fields (12 g CH₄ m⁻² year⁻¹, $n = 7$, field experiment) or of organic amended soils (17 to 46 g CH₄ m⁻² year⁻¹, $n = 11$, field experiment) (Kanno et al. 1997). Experimental conditions of our lysimeters were more similar to pot than field conditions since root mats were observed in the surface soil layer of the lysimeter, like it occurs in pot. The presence of root in the surface layer that is in a restricted soil volume can increase CH₄ emission due to higher amount of labile soil organic C available to methanogens than in the field (Zhang et al. 2007). It is well known that C inputs can stimulate CH₄ emission in flooded paddy field (Rath et al. 2005; Ma et al. 2008), as it does the increase in soil organic C content (Xu et al. 2003). The C contents of soil were not measured at the beginning of the 2008 growing season, but they were probably higher in the ADCS treated soils than in the CF treated soil. Thus, in addition to the stimulation by ADCS applications, probably there was a stimulation of CH₄ emissions due to the increased soil C contents of the ADCS treated soil.

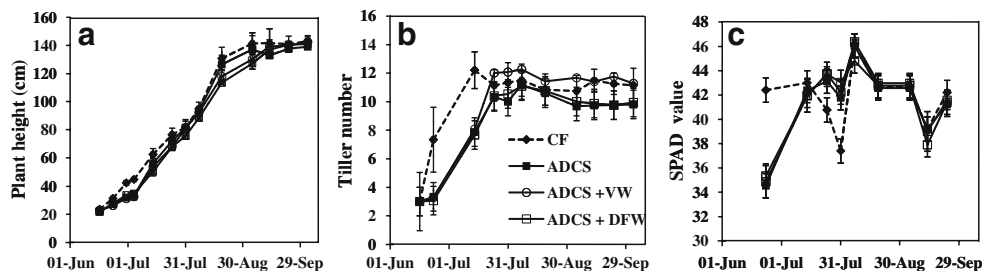


Fig. 4 Growth characteristic of forage rice variety (*Oryza sativa* L. var. Leaf star). **a** Plant height, **b** tiller number, and **c** soil plant analytical development (SPAD) leaf chlorophyll measurement value as

affected by the treatments. Bars represent standard deviation of the mean ($n = 3$). CF chemical fertilizer, ADCS anaerobically digested cattle slurry, WV wood vinegar, HFW higher level of floodwater

Table 2 Carbon balance ($\text{g C m}^{-2} \text{ season}^{-1}$) of the forage rice variety (*Oryza sativa* L. var. Leaf star)

Parameters		CF	ADCS		
			–	+WV	+HFW
(A) Carbon fixed by plant or soil (g C m^{-2})	Plant biomass C	1,230±72	1,060±54	1,080±51	1,060±39
	Soil C	–	739±281	898±174	701±349
	Subtotal	1,230±72	1,790±245	1,980±220	1,760±311
(B) Greenhouse gases ($\text{GWP CO}_2\text{-C m}^{-2}$)	CH ₄ emission	406±212	993±548	1,000±84	1,050±354
	N ₂ O emission	–10±8.5	–5±2.3	–1±0.1	–10±1.0
	Subtotal	396±205	998±550	1,000±84	1,040±355
(A–B) Net C balance ($\text{g C m}^{-2} \text{ season}^{-1}$)		838±244	806±531	980±166	720±364

Mean value ± standard deviation ($n=3$). Plant biomass C is total dry matter × C content. Soil C at 0–20 cm is calculated by subtracting the C content of the CF soil from that of the ADCS treatment. $\text{CO}_2 - \text{C}$ for CH₄ = CH₄ × 25 × 12/44 (IPCC 2007). $\text{CO}_2 - \text{C}$ for N₂O = N₂O × 298 × 12/44 (IPCC 2007)

CF chemical fertilizer, ADCS anaerobically digested cattle slurry, WV wood vinegar, HFW higher level of floodwater, GWP global warming potential

Wassmann et al. (2000) and Ali et al. (2008) have reported that CH₄ flux was the lowest immediately after planting and gradually increased with crop growth, reaching the maximum around the reproductive phase. This was the case in the CF, ADCS, and ADCS + HFW treated soils. In contrast, the CH₄ flux was the highest in mid-July in the ADCS + WV treated soil (Fig. 2); probably, the reason of the earlier peak might be due to the fact that the major constituent of wood vinegar, acetic acid, is a good substrate for methanogens (Kyuma 2004). Total CH₄ emission was not significantly higher in the ADCS + HFW than in the other ADCS treated soils. Water management is the main factor mitigating CH₄ emissions since intermittent irrigation and/or midseason drainage reduces CH₄ emissions compared to continuous flooding (Mosier et al. 2004; Minamikawa et al. 2005). The effect of different floodwater depths on CH₄ emission under continuous flooding are not known; increase in the floodwater depth from 5.4±2.4 cm in the ADCS to 6.5±2.6 cm in ADCS + HFW did not affect significantly CH₄ emissions.

In our study, N₂O emissions were not important because CO₂-equivalent CH₄ emissions ranged from 439 to 1,120 g C m⁻²year⁻¹, while CO₂-equivalent N₂O emissions ranged from 0.6 to 18.7 g C m⁻²year⁻¹. The mean N₂O fluxes during the growing season were negative (–0.05 to –0.76 mg m⁻²day⁻¹), suggesting the absorption of N₂O by the paddy soils. Chapuis-Lardy et al. (2007) found a maximum negative N₂O emission of –2.3 mg N₂O m⁻²day⁻¹, in waterlogged rice in China. Negative emissions (–2.4 mg N₂O m⁻²day⁻¹) were also reported by Majumdar (2005), whereas Xiong et al. (2007) reported low N₂O emission (0.05 g N₂O m⁻² season⁻¹) in a continuously flooded paddy field and Tsuruta et al. (1997) reported no

emission or uptake of N₂O during the rice growing period. Akiyama et al. (2005) reported an average N₂O emission of 1.07±1.49 mg N₂O m⁻² season⁻¹ ($n=17$) in fertilized paddy soils. Therefore, the importance of N₂O emissions as greenhouse gas emission is low in continuously flooded paddy soils compared to those of upland soils (Luo et al. 2008; Ram et al. 2009).

At harvest, soil carbon contents (0 to 2 cm) in the lysimeter soils were significantly ($P<0.05$) higher in the ADCS treatments than in the CF treatment. The use of ADCS in forage rice under flooded condition can enhance soil C sequestration (Table 2). Triberti et al. (2008) reported that soil C content was not increased by the application of chemical fertilizer, but it was increased at rates of 0.18 and 0.26 t ha⁻¹year⁻¹ by applying slurry or manure in an upland maze–wheat rotation system.

The CF treatments showed a higher value in C fixed by rice plants than the ADCS treatment did, while the ADCS treatments showed much higher values in the soil C accumulation (Table 2). However, this benefit in the ADCS treatments was offset by their enhanced CH₄ emission. Consequently, there was no significant difference in the net C balance between the CF and ADCS treatments. The present study did not show clear benefit in the ADCS application to paddy field. Of course, nutrients of ADCS can be taken up by plants, thus reducing the risk of being leached. In addition, the net C balance demonstrated that the options used in this study for mitigating ammonia volatilization did not increase CH₄ emission and thereby did not affect the C balance in the paddy field fertilized with anaerobically digested cattle slurry.

According to Nishimura et al. (2008), soil C budgets of single cropping of paddy rice plots were positive (79 to 137 g C m⁻²year⁻¹), while those of the single cropping of

upland rice and soybean-wheat plots were negative (-343 to -275 g C m⁻² year⁻¹ and -361 to -256 g C m⁻² season⁻¹, respectively). These findings indicated that paddy rice field may contribute to the mitigation of global warming potential.

This study revealed that the application of ADCS into paddy field enhances soil C accumulation, but it enhances CH₄ emission more than the C accumulation in terms of global warming potential, while N₂O emission was negligible. Therefore, CH₄ emissions should be mitigated so as to utilize ADCS in forage rice cultivation as an environmentally friendly system.

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References

- Akiyama H, Yagi K, Yan X (2005) Direct N₂O emissions from rice paddy fields: summary of available data. *Glob Biogeochem Cycles* 19:1–10
- Ali MA, Lee CH, Kim PJ (2008) Effect of silicate fertilizer on reducing methane emission during rice cultivation. *Biol Fertil Soils* 44:597–604
- Chapuis-Lardy L, Wrage N, Metay A, Chotte JL, Bermoux M (2007) Soils, a sink for N₂O? A review. *Glob Chang Biol* 13:1–17
- Furukawa Y, Inubushi K (2002) Feasible suppression technique of methane emission from paddy soil by iron amendment. *Nutr Cycl Agroecosyst* 64:193–201
- Hou H, Zhou S, Hosomi M, Toyota K, Yosimura K, Mutou Y, Nisimura T, Takayanagi M, Motobayashi T (2007) Ammonia emissions from anaerobically digested slurry and chemical fertilizer applied to flooded forage rice. *Water Air Soil Pollut* 183:37–48
- Intergovernmental Panel on Climate Change (2007) Climate change 2007: the physical science basis. IPCC Fourth Assessment Report (AR4), Contribution of working group 1 to the fourth assessment report of the intergovernmental panel on climate change. <http://www.ipcc.ch>
- Kanno T, Miura Y, Tsuruta H, Minami K (1997) Methane emission from rice paddy fields in all of Japanese prefecture: relationship between emission rates and soil characteristics, water treatment and organic matter application. *Nutr Cycl Agroecosyst* 49:147–151
- Kissel DE, Brewer HL, Arkin GF (1977) Design and test of a field sampler for ammonia volatilization. *Soil Sci Soc Am J* 40:1133–1138
- Kyuma K (2004) Fertility considerations for paddy soils (V)—fundamental biological and biochemical reactions in submerged paddy soils. In: Kyuma K (ed) *Paddy soil science*. Kyoto University Press, Kyoto, pp 82–95
- Luo J, Lindsey SB, Ledgard SF (2008) Nitrous oxide emissions from animal urine application on a New Zealand pasture. *Biol Fertil Soils* 44:463–470
- Ma J, Xu H, Yagi K, Cai Z (2008) Methane emission from paddy soils as affected by wheat straw returning mode. *Plant Soil* 313:167–174
- Majumdar D (2005) Past, present and future of N₂O emissions from rice field: a treatise. In: Livingston JV (ed) *Trends in air pollution research*. Nova Science, New York, pp 53–130
- Minamikawa K, Sakai N, Hayashi H (2005) The effects of ammonium sulfate application on methane emission and soil carbon content of a paddy field in Japan. *Agric Ecosyst Environ* 107:371–379
- Ministry of Agriculture, Forestry and Fisheries (2006) Annual report on food, agriculture and rural areas in Japan FY 2006. Ministry of Agriculture, Forestry and Fisheries Web. http://www.maff.go.jp/e/pdf/fy2006_rep.pdf
- Ministry of Agriculture, Forestry and Fisheries (2007) Annual report on food, agriculture and rural areas in Japan FY 2007. Ministry of Agriculture, Forestry and Fisheries Web. http://www.maff.go.jp/e/annual_report/2007/pdf/e_index.pdf
- Mosier A, Wassmann R, Verchot L, Khing J, Palm C (2004) Methane and nitrogen oxide fluxes in tropical agricultural soils. *Environ Dev Sustain* 6:11–49
- Nishimura S, Yonemura S, Sawamoto T, Shirato Y, Akiyama H, Sudo S, Yagi K (2008) Effect of land use change from paddy rice cultivation to upland crop cultivation on soil carbon budget of a cropland in Japan. *Agric Ecosyst Environ* 152:9–20
- Ram CD, Iain RG, Neal WM (2009) Nitrous oxide emission from feedlot manure and green waste compost applied to Vertisols. *Biol Fertil Soils* 45:809–819
- Rath AK, Ramakrishnan B, Rao VR, Sethunathan N (2005) Effects of rice-straw and phosphorus application on production and emission of methane from tropical rice soil. *J Plant Nutr Soil Sci* 168:248–254
- Rolston DE (1986) Gas flux. In: Klute A (ed) *Methods of soil analysis*, part 1, 2nd ed. Agronomy monograph. Soil Science Society of America and American Society of Agronomy, Madison, pp. 1103–1119
- Sunaga K, Yoshimura N, Hou H, Win KT, Tanaka H, Yoshikawa M, Watanabe H, Motobayashi T, Kato M, Nishimura T, Toyota K, Hosomi M (2010) Impacts of heavy application of anaerobically digested slurry to whole crop rice cultivation in paddy environment on water, air and soil qualities. *Jpn J Soil Sci Plant Nutr* 80:596–605 (in Japanese with English summary)
- Triberti L, Nastri A, Giordani G, Comellini G, Baldoni G, Toderi G (2008) Can mineral and organic fertilization help sequester carbon dioxide in cropland? *Eur J Agron* 29:13–20
- Tsuruta H, Kanda K, Hirose T (1997) Nitrous oxide emission from a rice paddy field in Japan. *Nutr Cycl Agroecosyst* 49:51–58
- Wassmann R, Neue U, Lantin RS (2000) Characterization of methane emissions from rice fields in Asia. I. Comparison among field sites in five countries. *Nutr Cycl Agroecosyst* 58:1–12
- Win KT, Toyota K, Motobayashi T, Hosomi M (2009) Suppression of ammonia volatilization from a paddy soil fertilized with anaerobically digested cattle slurry by wood vinegar application and floodwater management. *Soil Sci Plant Nutr* 55:190–202
- Xiong ZQJ, Xing GX, Zhu ZL (2007) Nitrous oxide and methane emissions as affected by water, soil and nitrogen. *Pedosphere* 17:146–155
- Xu H, Cai ZC, Tsuruta H (2003) Soil moisture between rice-growing seasons affects methane emission, production, and oxidation. *Soil Sci Soc Am J* 67:1147–1157
- Yanai Y, Toyota K, Okazaki M (2007) Effects of charcoal addition on N₂O emissions from soil resulting from rewetting air-dried soil in short-term laboratory experiments. *Soil Sci Plant Nutr* 53:181–188
- Zhang P, Zheng J, Pan G, Zhang X, Li L, Tippkötter R (2007) Changes in microbial community structure and function within particle size fractions of a paddy soil under different long-term fertilization treatments from the Tai Lake region, China. *Colloids Surf, B* 58:264–270