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# Effect of fertilizer application on methane emission/production in the paddy soils of Taiwan

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**Abstract** Methane production in three types of rice paddy soil was investigated under greenhouse conditions. The amount of methane produced during the first crop season (March to July) was 2–6 times higher than that in the second crop season (August to December). Application of organic fertilizer hastened the drop in redox potential and increased methane production and emission. Methane production also increased with the depth of soil with high values in soil samples from 18 to 30 cm depth. Methane production in the first crop season was 18.0, 54.3 and 49.4 mg $cm^{-3}$ for 6 t $ha^{-1}$ straw application for Linkou, Tzawchyau and Jiaushi soils, respectively. The value was 33.4 mg $cm^{-3}$ for the second crop season in Jiaushi soil. Methane emission was high during the flowering and maturity stages in the first crop season and the values were high during the tillering and flowering stages in the second crop season. Methane emission was high in Tzawchyau and Jiaushi soils in the first crop season. Methane emission rate reached a maximum from 12 noon to 3 p.m. due to high temperature and a minimum at 3 to 6 a.m. in both planted and unplanted soils.

**Key words** Methane production · Methane emission · Paddy soil · Redox potential · Organic fertilizer

## Introduction

The increase in methane concentration in the atmosphere from 1.50 to 1.72 µg $cm^{-3}$ during the past decade has recently received increasing attention. The major source sites of methane production are rice paddies, wetlands and sediments (Ehhalt 1985). The decrease in redox potential after flooding favors methane production in soils. Organic or chemical fertilizer application enhances methane production (Lindau and Bollich 1993; Banik et al. 1996). Methane produced in paddy soil is emitted directly through the soil column or transported by plants to the atmosphere (Cicerone and Shetter 1981).

Rice is the main crop in Taiwan. The cultivation area in 1995 was 197 571 ha in the first crop season and 165 908 ha in the second crop season. The methane flux fluctuates markedly with climate, soil characteristics, rice variety, fertilizer application, and agricultural practices (Masscheleyn et al. 1993; Sass et al. 1994; Yang et al. 1994; Chang and Yang 1997; Yang and Chang 1997). In this paper, the results on the effect of fertilizer application on methane production and emission in the paddy soils of Taiwan are reported.

## Materials and methods

Pot experiments

*Rice variety and soils*

Tainung No. 67, a Japonica rice variety, used in this study, was provided by Taiwan Provincial Taoyuan District Agricultural Improvement Station. Three soils [Linkou Landipreox clay (pH 4.9, organic matter 7.2 g $kg^{-1}$, total nitrogen 0.7 g $kg^{-1}$, Brag#1 extractable phosphorus 1.6 mg $kg^{-1}$, CEC 9.7 cmol $kg^{-1}$, and sand:clay:silt=8.1:64.4:27.5), Tzawchyau Hapludults sandy loam (pH 5.8, organic matter 13.6 g $kg^{-1}$, total nitrogen 0.8 g $kg^{-1}$, Brag#1 extractable phosphorus 13.6 mg $kg^{-1}$, CEC 7.8 cmol $kg^{-1}$, and sand:clay:silt=59.7:2.6:37.7), and Jiaushi Haplaquepts silty clay (pH 6.9, organic matter 20.7 g $kg^{-1}$, total nitrogen 1.5 g $kg^{-1}$, Brag#1 extractable phosphorus 8.8 mg $kg^{-1}$, CEC 7.6 cmol $kg^{-1}$, and sand:clay:silt=6.9:44.8:48.3)] were used in this study.

*Rice straw*

Rice straw, used in studies on the effect of organic amendments, contained cellulose 387 g $kg^{-1}$, hemicellulose 183 g $kg^{-1}$, lignin 150 g $kg^{-1}$, total nitrogen 6.6 g $kg^{-1}$, total phosphate 1.96 g $kg^{-1}$ and ash 122 g $kg^{-1}$.

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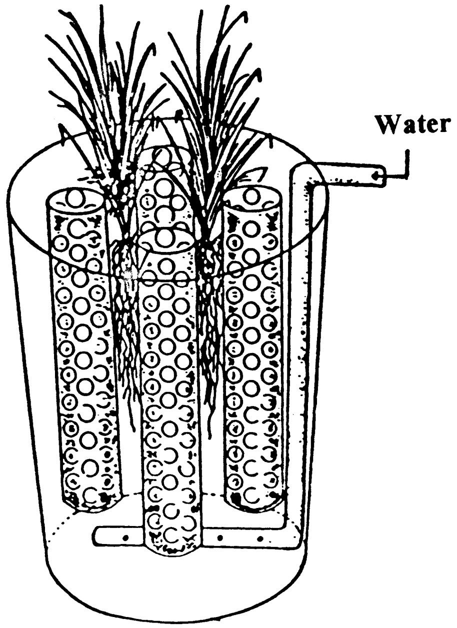


**Fig. 1** Diagram of the experimental pot, the soil cylinders and irrigation system for rice cultivation

*Pot cultivation*

Rice was cultivated in all experiments in $4.5\times10^4$ $cm^3$ PVC pots (i.d. 38.0 cm, o.d. 38.4 cm, and height 48.0 cm with a soil depth of 40.0 cm) under greenhouse conditions. A subsurface irrigation system for initial internal flooding and gas displacement consisted of a perforated 2 cm (i.d.) PVC pipe on the bottom of a container connected to a vertical nonperforated 2 cm (i.d.) PVC pipe to the surface. Four sampling cylinders (i.d. 3.2 cm, o.d. 3.5 cm and height 40.0 cm) were placed vertically in the pot as illustrated in Fig. 1. The cylinder consisted of polycarbonate tubing with 180 holes (diameter 7.9 mm) drilled over the entire length and circumference. The tube was covered with nylon cloth (Nylon 200 HD, 0.48 mm) to prevent the penetration of rice roots into the soil column. The cylinder allowed sampling of a soil core with a home-made acylic tube (i.d. 1.12 cm, o.d. 1.20 cm and height 50.0 cm) during the growth of rice without damaging the roots. Subsequent irrigation consisted of the manual addition of deionized water from the surface to maintain a water depth of 5 cm. Four weeks after flooding, four hills (each hill containing three plants) of 21-day-old rice seedlings were transplanted into the experimental pots 10 cm apart (Yang et al. 1994). All experiments were triplicates.

*Fertilizer*

Each pot was treated with chemical fertilizer ($CaHPO_4$ 4.4 g, urea 5.2 g and KCl 3.0 g) or organic fertilizer (rice straw 70.0 or 140.0 g, equivalent to 6 or 12 t $ha^{-1}$) and thoroughly mixed with the top 20 cm of soil before irrigation. Nonfertilized and unplanted pots were used as the controls in studies on methane production or emission.

*Methane production and production rate*

A soil core (30.0 cm length) from the top to 35 cm depth was obtained from the experimental pots as described above. Each 6.0-cm length of soil (diameter 1.12 cm) was put into a 50-$cm^3$ serum bottle which had been previously flushed with oxygen-free nitrogen gas and sealed with a butyl rubber stopper. After shaking, the gas (1.0 $cm^3$) in the head space was withdrawn and 0.3 $cm^3$ was injected into a glass column (0.26 mm×2.0 m) packed with Porapak Q (80/100 mesh) in a Shimadzu 14A Gas Chromatograph (Shimadzu Co., Japan) equipped with flame ionization detector. Column temperature was set at 55 °C, and injection and detector temperatures were set at 80 °C. Methane production and production rate were measured 7 times during transplanting, tillering, heading, flowering, maturity, harvest and resting stages in each crop season. Methane production rate was measured at 0.5-h intervals for 1.5 h, by examining the change in methane concentration in the head space after the bottle was flushed with oxygen-free nitrogen gas.

*Methane emission rate*

A home-made PVC chamber (diameter 32 cm, and height 40–70 cm depending on the growth stage of rice) was placed 10 cm under the soil 0.5 h before measurement to reduce soil disturbance. The methane emission rate during seven different growth stages was measured at 0.5-h intervals for 4 h from 10 a.m. to 2 p.m. Gas samples were removed through the rubber septum with a gas-tight syringe and stainless steel hypodermic needle.

*Estimation of methane emission*

Methane emission from paddy soil was estimated by the following equation (Rolston 1986):

$$f = (V/A)\,(\Delta C/\Delta t)$$

where $f$=methane emission rate (mg $m^{-2}$ $h^{-1}$), $V$=volume of chamber above the soil ($m^3$), $A$=cross-section of chamber ($m^2$), $\Delta C$=concentration difference between zero and $t$ times (mg $m^{-3}$), and $\Delta t$=time duration between the two sampling periods (h). Methane emission from paddy soil was calculated from the summation of methane emission in seven growth stages of rice.

Laboratory experiments

*Straw application and methane production*

Jiaushi soil (20 g, 20 mesh) was mixed with 0–0.4 g (0–2%) rice straw powder (40 mesh), placed in a 125-$cm^3$ flask, and 40 $cm^3$ deionized water was added. After thorough mixing, the flask was sealed with a butyl rubber stopper, flushed with oxygen-free nitrogen gas and incubated at 25 °C for 28 days. Methane concentration in the air space was determined every 2 days.

*Incubation temperature and methane production*

Jiaushi soil (20 g, 20 mesh) was mixed with 0.2 g rice straw powder (40 mesh), placed in a 125-$cm^3$ flask, and 40 $cm^3$ deionized water was added. The flask flushed with oxygen-free nitrogen gas and incubated at 12, 15, 20, 25, 30, 35 and 40 °C for 8 days. Methane concentration was measured every 2 days.

*Methane oxidation rate*

Twenty-gram portions of Jiaushi soil (20 mesh) were placed in a 125-$cm^3$ serum bottle, sealed with a butyl rubber stopper under atmospheric pressure, injected with 0.2 $cm^3$ pure methane gas (China Petroleum Corporation, Taiwan), and incubated at 25 °C for 130 h. The methane oxidation rate was measured at 6-h intervals, in terms of the decrease in the concentration of methane in the air space. Soil sterilized at 121 °C for 40 min served as control.

Chemical analyses

Soil pH was determined directly or on a 1:1 (w/w) soil to water suspension with a pH meter. Total nitrogen was determined by the modified Kjeldahl method (Yang et al. 1991). Organic carbon content was determined by wet oxidation by the Walkley-Black method and the organic matter content was calculated as the value of 1.724 times of organic carbon (Nelson and Sommers 1982). *Eh* was measured with a Hanna No. 081-854 potential meter (Code HI 3131 B) under 5–20 cm depth of topsoil using the Pt electrode.

## Results and discussion

### Growth of rice plants

Linkou soil contained less organic matter, total nitrogen and extractable phosphorus than the other soils. Rice growth was poor in Linkou soil even with fertilizer application. The time for heading in Linkou soil was about 10 days later than in Tzawchyau and Jiaushi soils. The growth period in the first crop season was 127 days, while it was 117 days in the second crop season. The daily average soil temperature in 1993 was 27.7 °C in the first crop season, and 27.1 °C in the second crop season. The daily soil temperature increased during the cultivation in the first crop season, whereas it fell in the second crop season. Fertilizer application stimulated the growth and the height of rice plants.

### pH change during rice cultivation

The pH values of all test soils gradually decreased during rice cultivation, while they almost kept a constant value in unplanted soils (Fig. 2). The soil pH decrease during rice cultivation might have been due to the accumulation of root secretions and microbial metabolites.

### Redox potential change during rice cultivation

The *Eh* value decreased very significantly in the first 10 days after flooding treatment and with the application of chemical and organic fertilizer. Tzawchyau and Jiaushi soils had lower *Eh* values than the Linkou soil. The difference in *Eh* values between planted and unplanted soils in all tests was non-significant. The *Eh* of soil at a depth of 20 cm was lower than at a depth of 10 cm in all test soils (Fig. 3). Yagi and Minami (1990) also reported that application of organic fertilizer retarded the *Eh* value, and enhanced methane production.

### Methane production and production rate

In the first crop season, methane production in core samples of Linkou soil (0–30 cm depth) was detected first at 30 days after transplantation with 6 t $ha^{-1}$ straw application. The maximum methane production of soil core from 24–30 cm depth was before the heading stage in Linkou soil, and during the flowering stage in the Tzawchyau and Jiaushi soils. Methane production during the rice cultivation and unplanted soil was presented in Table 1. Chemical and organic fertilizer application increased methane production. Methane production was higher in the Tzawchyau and Jiaushi soils than in the Linkou soil. The C/N ratios of Linkou, Tzawchyau and Jiaushi soils were 6.0, 9.9 and 8.0, respectively. The soils with higher C/N ratios had higher methane production. A similar phenomenon was also described by Bouwman (1991).

The methane production in Jiaushi soil during rice cultivation in the second crop season is shown in Fig. 4.

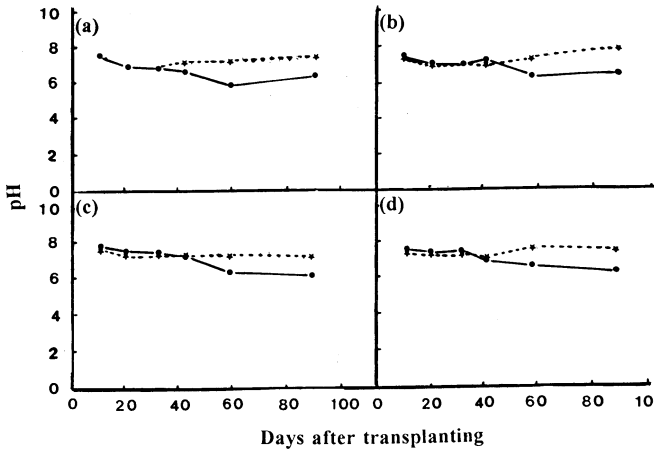


**Fig. 2 a–d** Soil pH of Jiaushi soil in the second crop season of 1993. **a** Non-fertilized soil, **b** chemically fertilized soil, **c** 6 ton $ha^{-1}$ straw application soil, **d** 12 ton $ha^{-1}$ straw application soil, ●–● planted soil, ★–★ unplanted soil

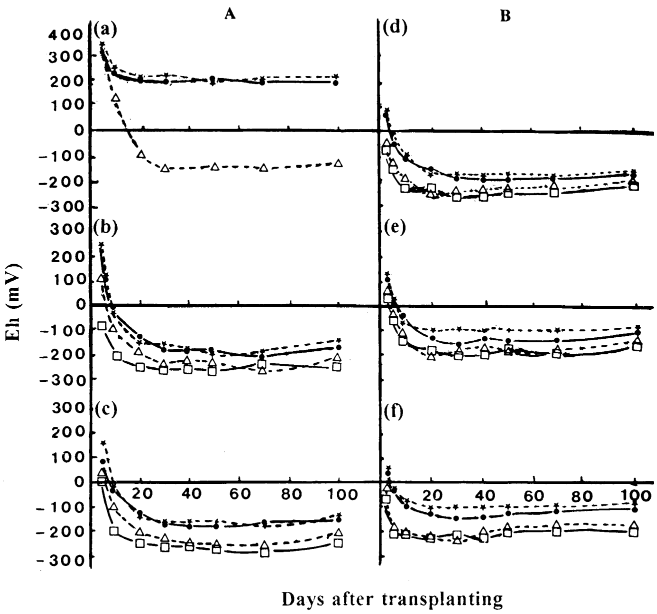


**Fig. 3 a–f** *Eh* values of test paddy soils of 1993. *A* first crop season, *B* second crop season. **a** Linkou planted soil at 20 cm depth, **b** Tzawchyau planted soil at 20 cm depth, **c** Jiaushi planted soil at 20 cm depth, **d** Jiaushi planted soil at 20 cm depth, **e** Jiaushi planted soil at 10 cm depth, **f** Jiaushi unplanted soil at 20 cm depth. ★–★ nonfertilized soil, ●–● chemically fertilized soil, △–△ 6 ton $ha^{-1}$ straw application soil, □–□ 12 ton $ha^{-1}$ straw application soil

**Table 1** Methane production in and emission from different soils planted to Japonica rice (Tainung No. 67)

| Fertilizer application | Rice cultivation | Linkou soil | | Tzawchyau soil | | Jiaushi soil | |
|---|---|---|---|---|---|---|---|
| | | MP | ME | MP | ME | MP | ME |
| First crop season | | | | | | | |
| None | + | ND | ND | 6.8 | 12.3 | 6.5 | 22.1 |
| | – | ND | ND | 7.6 | 7.9 | 7.4 | 14.7 |
| Chem. | + | ND | ND | 10.8 | 15.3 | 9.3 | 29.5 |
| | – | ND | ND | 11.8 | 13.2 | 11.5 | 25.6 |
| 6 ton | + | 18.0 | 7.3 | 54.3 | 77.4 | 49.4 | 91.5 |
| | – | 20.2 | 6.5 | 86.1 | 54.3 | 53.7 | 62.5 |
| 12 ton | + | ND | ND | 98.6 | 186.9 | 76.3 | 201.2 |
| | – | ND | ND | 118.9 | 98.3 | 80.5 | 153.4 |
| Second crop season | | | | | | | |
| None | + | | | | | 4.2 | 5.7 |
| | – | | | | | 4.7 | 4.6 |
| Chem. | + | | | | | 5.0 | 7.6 |
| | – | | | | | 5.3 | 6.4 |
| 6 ton | + | | | | | 33.4 | 51.8 |
| | – | | | | | 43.8 | 41.7 |
| 12 ton | + | | | | | 43.5 | 133.1 |
| | – | | | | | 40.0 | 104.8 |

*MP* methane production (mg $cm^{-3}$), *ME* methane emission (g $m^{-2}$), *None* non-fertilizer application, *Chem.* chemical fertilizer application, *6 ton* 6 tons of rice straw application per hectare, *12 ton* 12 tons of rice straw application per hectare, + with rice plants, – without rice plants, ND not determined

Maximum methane production was during the tillering and maturity stages. Although the difference in daily average soil temperature between the first crop season and the second crop season was not significant, the high temperature in the first crop season had vailed during the flowering and maturity stages, which favored methane production, while the high temperature in the second crop season was noticed during the transplanting and tillering stages. Methane production in the second crop season was only 15–50% of the first crop season (Table 1). Similar results were also reported by Yang et al. (1994). Methane production was high during the flowering stage with chemical fertilizer application, while the high values were during the tillering and flowering stages with organic fertilizer application. The amount of methane production during the flowering stage with chemical fertilizer application ranged between 50 and 60% of the total amount of methane production, and the value was between 25% and 35% with organic fertilizer application. Cicerone et al. (1983) indicated that the amount of methane production during the flowering stage was in the range of 76–92% of the total amount of methane production in the rice paddy of California. This percentage was higher than that in the Taiwan area. Methane production increased from the flowering to the maturity stage due to excretion from the roots (Sass et al. 1991). The methane production increase in the tillering stage with organic fertilizer application might be due to the anaerobic digestion of organic amendments. Methane production increased with the depth of soil from 6 to 30 cm and the highest production was in the deeper zones (18–30 cm). The *Eh* value of soil core samples was lower in the deeper zone than that of the shallower zone. Patrick (1981) and Yagi and Minami (1990) also reported that the lower *Eh* value had the higher methane production.

## Methane emission rate

In the first crop season, methane emission rate was high during the flowering stage in Linkou soil and during the maturity stage in Tzawchyau and Jiaushi soils. Methane emission rate in Jiaushi soil in the second crop season is shown in Fig. 5. Methane emission was higher during the flowering and maturity stages in nonfertilized and chemical fertilizer soils, and the values were higher during the transplanting and tillering stages in organic fertilizer soils. These results might be due to the root secretion in the chemical fertilizer application and anaerobic digestion with organic fertilizer application. Methane emission is also listed in Table 1. Rice cultivation not only stimulated methane production, but also enhanced methane oxidation. The amount of methane emitted from paddy fields to the atmosphere was mainly controlled by the balance of the production, oxidation and leaching of methane in paddy soil. The percentage of methane emission to methane production was between 5.4–21.0% in the first crop season in the Jiaushi soil, and the value was between 9.0% and 29.0% in the second crop season. Field measurements showed that the percentage of methane emission to methane production ranged between 3% and 56% (Schutz et al. 1989) between 10% and 81% (Sass et al. 1990). Methane, not accounted for, is considered to be oxidized in the plowed layer of paddy soil or leached to the lower layer. The higher methane emission rate in the pot cultivation than in the field experiment might be due to the continuous flooding with 5 cm water during the crop season, while the fields were subjected to several periods of drainage. Methane emission rate in the paddy soil with continuous flooding with 5 cm water was 2–3 times higher than intermittent irrigation during the tillering, flowering and

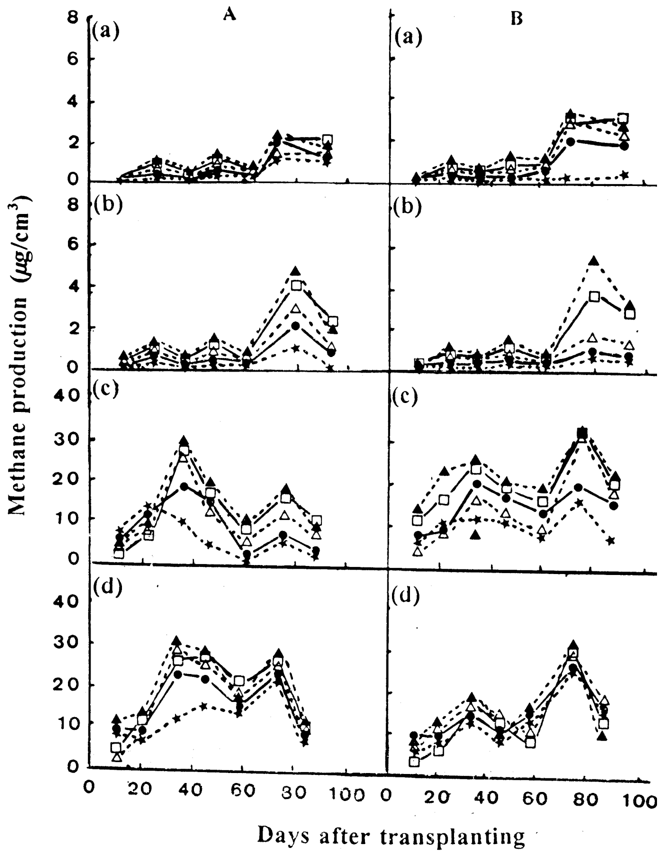


**Fig. 4 a–d** Methane production in Jiaushi soil in the second crop of 1993. *A* Planted soil, *B* unplanted soil. **a** Nonfertilized soil, **b** chemically fertilized soil, **c** 6 ton $ha^{-1}$ straw application soil, **d** 12 ton $ha^{-1}$ straw application soil. ★–★ 6 cm depth, ●–● 12 cm depth, △–△ 18 cm depth, □–□ 24 cm depth, ▲–▲ 30 cm depth

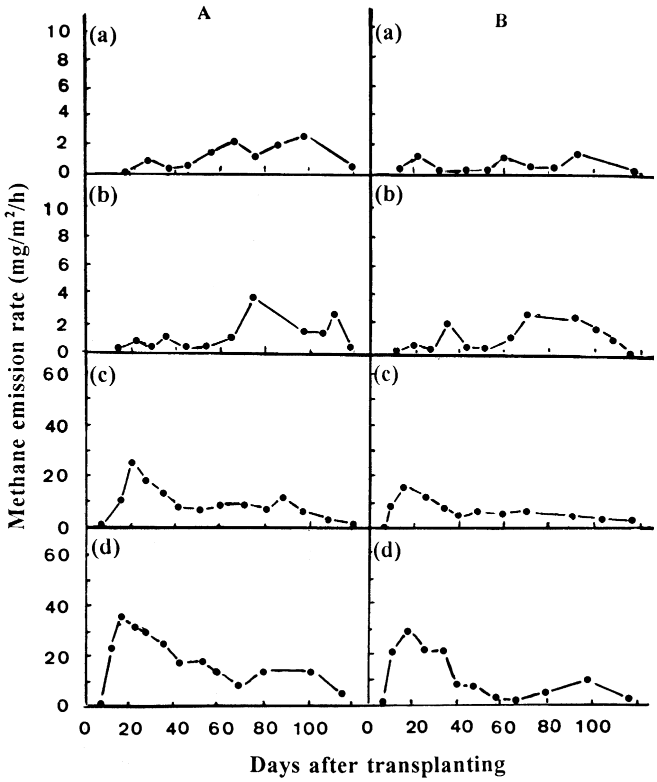


**Fig. 5 a–d** Methane emission rate in Jiaushi soil in the second crop of 1993. *A* Planted soil, *B* unplanted soil. **a** nonfertilized soil, **b** chemically fertilized soil, **c** 6 ton $ha^{-1}$ straw application soil, **d** 12 ton $ha^{-1}$ straw application soil

maturity stages (unpublished data). The methane emission rate in rice paddies has been reported to be 7.92–60 mg $m^{-2}$ $h^{-1}$ in China (Khalil et al. 1991), 4 mg $m^{-2}$ $h^{-1}$ in Spain (Seiler 1984), 6–28.3 mg $m^{-2}$ $h^{-1}$ in Italy (Schutz et al. 1989), 10 mg $m^{-2}$ $h^{-1}$ in California, 2.08–15.0 mg $m^{-2}$ $h^{-1}$ in Texas, 11.25–20 mg $m^{-2}$ $h^{-1}$ in Louisiana (Cicerone et al. 1983; Sass et al. 1990, 1991), 0.42–16.2 mg $m^{-2}$ $h^{-1}$ in Japan (Yagi and Minami 1990), 1.447–27.08 mg $m^{-2}$ $h^{-1}$ in Indonesia (Soedomo and Warko 1993), 15–25 mg $m^{-2}$ $h^{-1}$ in southern India (Lal et al. 1993), 0–49 mg $m^{-2}$ $h^{-1}$ in India (Mitra 1992), 3.8 mg $m^{-2}$ $h^{-1}$ in Australia (Denmead and Freney 1990), and 1.76–32.08 mg $m^{-2}$ $h^{-1}$ in Thailand (Towprayoon et al. 1993; Yagi et al. 1994). In northern Taiwan, methane emission from rice paddies ranged between 0.22 and 2.92 mg $m^{-2}$ $h^{-1}$ in the first crop season, and between 4.00 and 13.72 mg $m^{-2}$ $h^{-1}$ in the second crop season (unpublished data).

## Methane oxidation rate

The methane oxidation rates of the surface layer (5 mm) of Jiaushi soil planted to rice in the second crop season are shown in Fig. 6. Methane concentration decreased with the application of chemical and organic fertilizers. There was no methane oxidation with sterilized soil. With increasing depth of soil, the methane oxidation rate decreased and methane concentration in the soil increased. The methane oxidation rate approached zero at a depth of more than 18 cm.

## Organic matter application and methane production

The effect of straw application on methane production rate is shown in Fig. 7. Methane production rate increased with the amount of organic amendment. In addition, methane production ($y$, μg) and the amount of straw application ($x$, %) had a linear relationship ($y = 369x – 8.0$). The correlation coefficient was 0.9981 ($n = 5$) in the range of 0% and 2.0% of straw application. Each gram of straw produced 1.85 mg methane for 28-day incubation at 25 °C.

## Temperature and methane production

Methane production was undetectable when the soil temperature was lower than 12 °C. The methane production rate increased with soil temperature from 15 to 40 °C, and a significant correlation existed between methane produc-

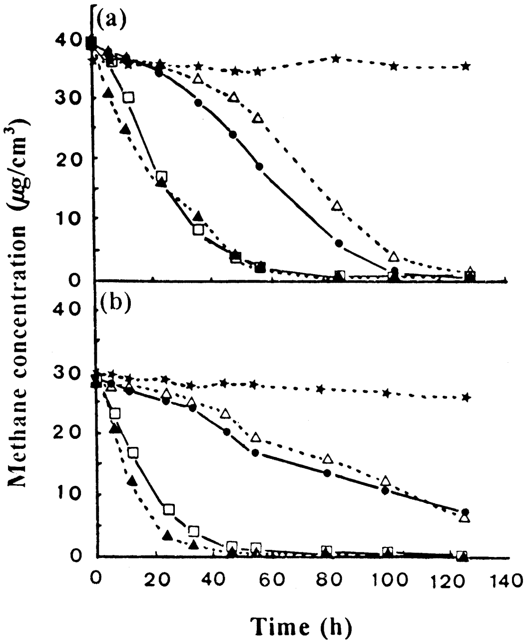


**Fig. 6 a, b** Methane oxidation of Jiaushi soil in the second crop season of 1993. **a** Planted soil, **b** unplanted soil. ★–★ sterilized soil, ●–● nonfertilized soil, △–△ chemically fertilized soil, □–□; 6 ton $ha^{-1}$ straw application soil, ▲–▲ 12 ton $ha^{-1}$ straw application soil

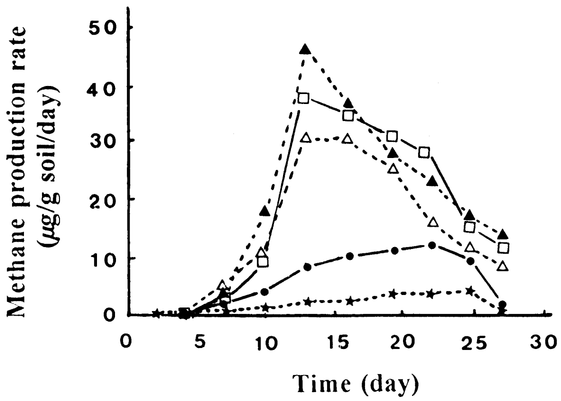


**Fig. 7** Effect of straw application on methane production rate in Jiaushi soil at 25 °C. ★–★ 0%, ●–● 0.5%, △–△ 1.0%, □–□ 1.5%, ▲—▲ 2.0%

tion rate ($y$, μg $h^{-1}$) and soil temperature ($x$, °C) ($y = 0.0557x – 0.8105$, $r^2 = 0.9989$ for $n = 6$). Each gram of soil produced 1.43 μg methane at 40 °C for 1 h incubation.

## Diurnal variation of methane emission

Diurnal variation of methane emission rate in the second crop in Jiaushi soil at the maturity stage is shown in Fig. 8. Methane emission rate was high at noon to 3 p.m. and it was low at 3 a.m. to 6 a.m. The fluctuation of methane emission rate in unplanted soil was larger than that in planted soil. This might be due to the difference in

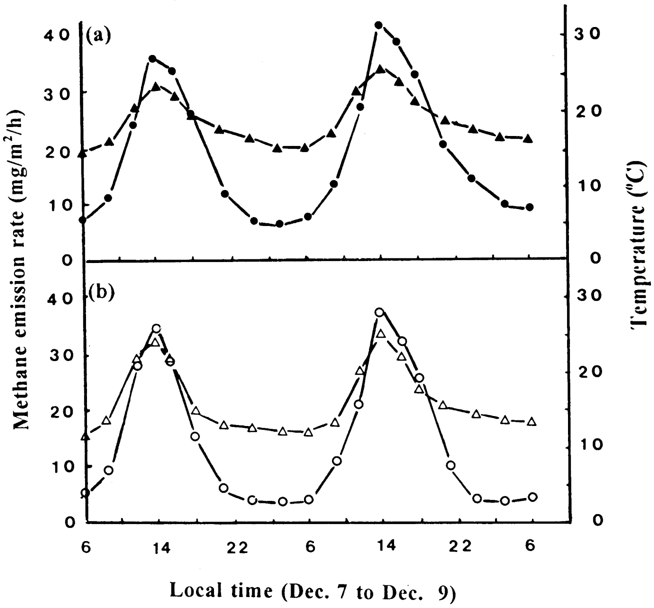


**Fig. 8 a, b** Diurnal variation of methane emission rate and soil temperature in Jiaushi soil at maturity stage during 7 and 9 December 1993. **a** Planted soil, **b** unplanted soil. ● and ○ Methane emission rate (mg $m^{-2}$ $h^{-1}$), ▲ and △ soil temperature (°C)

the soil temperature fluctuation between planted and unplanted soils.

This study demonstrates the influence of organic matter application on the methane production in and emission from paddy soils. The contribution of paddy soils to global atmospheric methane concentrations is well documented. Rice plants serve as a vehicle for transport of methane from the site of production in the soils to the atmosphere.

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