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CH_4 EMISSION FROM A CHINESE RICE PADDY FIELD

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ABSTRACT

CH_4 emission rates have been measured continuously for the early rice of 1988 and late rices of 1987 and 1988 during entire growing seasons in a rice paddy field in Hangzhou, Zhejiang Province, China, by using an automatic sampling and analyzing system. During most parts of the seasons CH_4 emission rates showed strong diurnal variations. Bi-mode patterns with the highest value in the afternoon and a second peak at mid-night were generally found for the early rice, while the highest values were almost always found in the night for the late rice. Bi-mode patterns with a second peak in the afternoon were also found during the reproductive phase of the late rice plants. These diurnal variations may be explained by the diurnal variations of the soil temperature and the activity of rice plants. Strong seasonal variations with one peak in the tillering stage and two during the reproductive phase of rice plants were observed for all the three growing seasons. The seasonal variations may be explained by the activity of rice plants, availability of organic substrates in the soil, and the activity of soil bacteria related to soil temperatures. Fertilization did not show significant effects on the total seasonal CH_4 emissions but slightly changed the pattern of the seasonal variations of the CH_4 emission rates. Averaged over the measuring periods and 8 spots, CH_4 emission rates of 7.8 mg m^{-2} h^{-1} for the early rice and 28.6 mg m^{-2} h^{-1} for the late rice were obtained. Based on these measured data, the total global emission of CH_4 from rice paddies is estimated to be about 90 Tg/yr ranging from 70 to 110, accounting for 20% of the total source of CH_4.

I. INTRODUCTION

Recent investigations have shown (1) that atmospheric CH_4 is increasing with a rate much faster than that of CO_2, and (2) that its effect on climate is already as important as that of CO_2 and may become even more important in the near future (Schneider, 1988). Therefore, there is an urgent need to quantify the sources and sinks of methane and to investigate the global methane cycle more comprehensively.

Like CO_2, atmospheric CH_4 is a very important radiatively active gas that governs the earth's climate.

Unlike CO_2, atmospheric CH_4 is also a chemically active gas. Therefore, an increase of atmospheric CH_4 will also change chemical reactions in the atmosphere and hence exert indirect effects on the earth's climate in addition to the direct effect on the radiative energy balance mentioned above. The most important reactant that destroys CH_4 is the gas phase hydroxyl radical, OH, a key radical in atmospheric photochemistry. CH_4 oxidation may reduce OH thereby increasing atmospheric CO_2, CO, H_2O, H_2, O_3 and CH_2O, which in turn affects the chemistry of other atmospheric gases. About 0.34×10^{15}g CO_2 C/yr is produced by oxidation of atmospheric methane on a global scale, which is about 6% of the total CO_2 release due to human activities (Cicerone, 1988). Moreover, stratospheric CH_4 reacts with atoms, so that an increase of atmospheric CH_4 may play an important role in the ozone chemistry in the stratosphere.

Despite the importance of atmospheric CH_4, the causes responsible for its current increase are not yet fully understood. One possible explanation is a continuous increase of the global CH_4 sources, while a reduction of CH_4 sink strength can not be excluded. The globally important CH_4 sources and sinks have probably been identified, but the individual sources and sinks are still not sufficiently quantified. In this paper we report the first continuous field measurements of CH_4 emission rates performed in Asia during the late season, August to November, 1987, and the two growing seasons in 1988 in a rice paddy field near Hangzhou, a typical rice growing region in China, where rice cultivation procedures and climatic conditions are basically different from those in the USA and Europe.

II. GAS COLLECTOR BOXES AND AUTOMATIC SAMPLING AND ANALYZING SYSTEM

The gas collector boxes used for the present experiments (Fig. 1)were made of plexiglass (colorless, smooth, 3 mm thick). The edges were fixed by aluminium-profiles (4 mm thick) and were sealed with silicone on the inside. The boxes covered an area of about 0.4 m^2 (ca. 0.65 m side length) and were 0.9 m high. The inner volume of the box was ca. 380 L. As soon as a measuring cycle was started the boxes were closed with a cover,

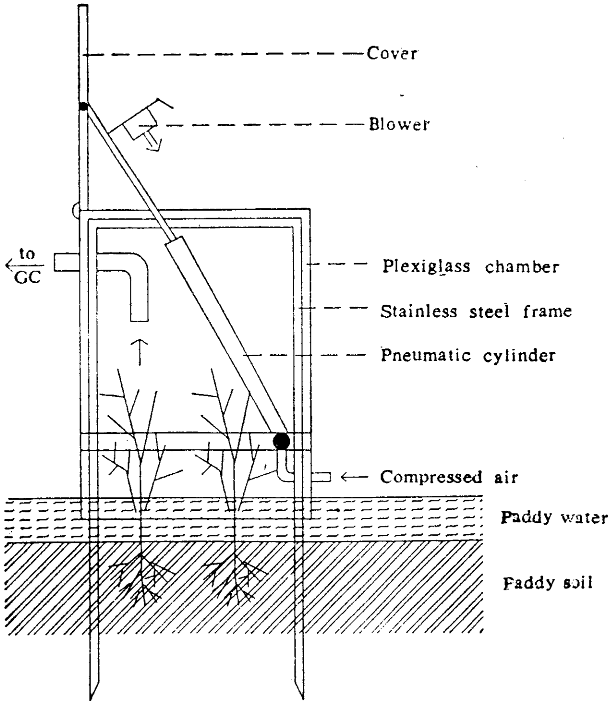

Fig. 1. Schematic diagram of the gas collector box used.

which was opened again with a pneumatic pressure-cylinder at the end of a 90-minute measuring cycle. A fan at the inner side of the cover exchanged the air inside and outside the box when the cover was open, and mixed the air inside the collector box during closure in order to avoid vertical CH_4 gradients. Control measurements were performed routinely by analyzing air samples taken from different heights inside the box by means of syringes; vertical CH_4 gradient were never detected.

The inertness of the box material was checked by filling the box with air containing high (10—100 ppm) CH_4 mixing ratios. The CH_4 mixing ratios did not change significantly during a measuring period of about 1 hour.

The gas collector boxes were mounted on stainless steel frames which had been fixed into the soil prior to flooding and remained at the same position during the whole vegetation period. The height of the frames was adjusted in such a way that the box dipped approximately 5 cm into the water, so that the inner volume of the box was sealed against ambient atmosphere with the surrounding water body.

Rice seedlings reached a maximum height of 85 cm and thus, were smaller than the height of the box. A difference in the growth of plants inside or outside of the boxes was never observed, indicating that the permanent installation of the boxes at the measuring sites did not cause a change in plant development.

Fig. 2 shows the scheme of the automatic setup consisting of 2 completely separated measuring systems with two gaschromatographs and a 2-channel integrator. The operation of the magnetic valves and the storage of raw data was performed by a programmed microcomputer (Hewlett-Packard HP 9835). The system was run by closing boxes 1, 2, 3 and 4 (1st system) as well as 9, 10, 11 and 12 (2nd system) at the same time while boxes 5, 6, 7 and 8 (1st system) and 13, 14, 15 and 16 (2nd system) remained open, and vice versa. From

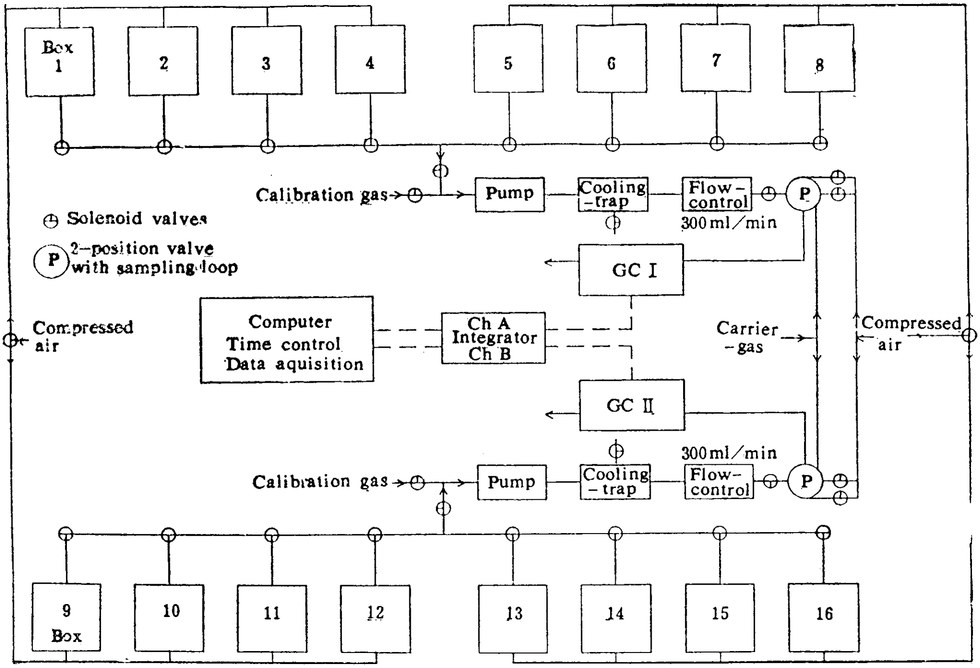

Fig. 2. Automatic sampling and analyzing system,

inside the closed boxes air was drawn by 2 metal bellow pumps MB 158 at regular time intervals, and guided to the laboratory located in a building approximately 40 m away from the field by using stainless steel capillaries (length = 50 m, i.d. = 2 mm). Water vapor was removed by a cooling trap (4°C) before the sampling loop of the gaschromatograph was flushed. CH_4 mixing ratios were analyzed by a gaschromatograph (Shimadzu, GC-mini-2, packed column: 1.5 m length, 4 mm i.d., molecular sieve 13X, 60/80 mesh). Before analysis, the stainless steel tubes were flushed by twice their inner volume with the sampled air.

A measuring cycle began when the box was closed with the cover and ended after 90 minutes when the cover was opened. During this time-period air samples were taken automatically from inside the closed box every 12 minutes. At the beginning and at the end of each 90-minute measuring period the GC-system was calibrated with CH_4 calibration gas (ppm in synthetic air) which was stored in steel cylinders under high pressure and was calibrated prior to the field measurements against a calibration standard in the Fraunhofer-Institute at Garmisch-Partenkirchen, FRG. In this way 7 CH_4 mixing ratios were determined for each of the 8 closed boxes within a 90-minute measuring period. During the following 90-minute measuring period 7 CH_4 mixing ratios were determined for each of the other group of 8 boxes that were then closed. Thus the automatic system produced 8 measurements of methane fluxes per day for each of the 16 measuring spots.

The system was used in the study of an Italian rice paddy field and proved to be reliable. The overall measurement error is less than 10% (Schütz et al., 1989a).

III. EXPERIMENTALS

Field measurements of the emission rates were performed in a rice paddy field of Zhejiang Agricultural University in Hangzhou (30° 19'N, 120° 12'E) located at the alluvial plain of the Qiantang Estuary in Zhejiang Province, China. Zhejiang Province is one of the major rice growing regions in China, covering approximately 7.5% of the total sown area of the Chinese paddy fields. The soil of the field used for the present study is representative for the plain of the middle and lower regions of Changjiang (Yangtze) River, which covers more than one fourth of the total sown area of rice paddy fields of China. The cultivated soil is a silt loam with 8.5% sand, 74.5% silt, and 17% clay. The organic carbon and total nitrogen content prior to flooding was 2.5% and 0.19% respectively. The soil pH measured prior to flooding was 6.3 and varied between 6.5 and 7.5 during the vegetation periods.

The experimental sites were routinely managed by using the local farming procedures. The field was usually planted with green manure crops or vegetables in winter and ploughed and raked in late April for the early rice. Flooding was usually done right before transplanting. The early rice seedlings were raised at early April and transplanted to the flooded rice field in early May. The early rice was harvested at the end of July. Immediately after the harvest of the early rice the field was ploughed and raked and flooded again for transplanting of the late rice; the seedlings were raised in seedling beds in the middle of July. The late rice was harvested at early or middle November. Water level was kept at approximately 1 cm above the soil surface for transplanting. After the transplanted seedlings had recovered and begun to grow, water level was kept in the range of 5—10 cm until a few days before harvest. Fresh water was added to the field in amounts needed to maintain this level independent of rainfall, evaporation, transpiration, leaching, and runoff. The rice variety used for the early rice was an early-maturing and short-straw variety with a growing period of 85 days, while that for the late rice was a hybrid rice, "zhefu-3", with a growing period of 100 days. Tillering begun about 10 days after transplanting for the early rice and about one week after transplanting for the late rice. Organic manure, e.g. rice straw, rape seed cake, animal excrement and urine, was ploughed into the field before flooding, while chemical fertlizer, e.g. urea, ammonium sulfate, was surface-applied during the reproductive phase of the rice plants.

The experimental site used for our measurements had a size of 24 × 14 m^2 and was parceled into 4 narrow fields of 24 × 3 m^2 by digging in reinforced concrete boards, in order to study the effect of fertilization. Shortly before flooding the fields were treated as follows: One narrow field was left unfertilized, one was fertilized with 5.0 kg K_2SO_4 (approximately 1050 kg/ha), the third one received 7.5 kg rape seed cake (200 kg/ha) in addition to 5.0 kg K_2SO_4, and the fourth with only 7.5 kg rape seed cake.

Four gas collector boxes were installed in each narrow experimental field in mid-August 1987, and remained in the field ever since. During the vegetation periods the boxes could be reached via small footbridges, which had been built prior to flooding in 1987, to allow work at the boxes without disturbance of the environment of the growing rice plants. Measurements of CH_4 flux were started at the end of August 1987, and have been carried out through the late rice growing period until harvest in mid-November. Continuous measurements have been carried out for both early rice and late rice of 1988 during the entire vegetation periods.

In addition, air and soil temperatures were measured continuously with pt-100 thermocouples. Two thermocouples measured the air temperatures inside and outside of one of the gas collector boxes, respectively. Another two thermocouples were placed into the paddy water at about 5 cm depth with one inside and one outside a gas collector box. Four additional thermocouples were used to measure the soil temperatures at the surface, 5 cm, 10 cm, and 15 cm depth in an area adjacent to the boxes. Both air and soil temperatures recorded inside and outside the boxes always showed comparable values.

IV. RESULTS AND DISCUSSIONS

The CH_4 emission flux from the area covered by a gas collector box was determined from the temporal increase of the CH_4 mixing ratios inside the box in terms of

$$F = h\rho\frac{dm}{dt}, \qquad (1)$$

where h is the height of the box, ρ is the density of CH_4 at the pressure and temperature recorded inside the box, and dm/dt is the rate of increase of the CH_4 mixing ratios inside the box, determined from the 7 measured CH_4 mixing ratios by using a linear regression technique. The measured CH_4 mixing ratios almost always increased linearly with time inside the gas collector boxes, even in the mixing ratio reached values of 300 ppmv within the 90-minute measuring period. A stepwise increase in the measured CH_4 mixing ratios, as reported in the case of gas bubble ebullition, was only rarely found. Thus, the linear regression technique was suitable for the determination of the rate of increase of the CH_4 mixing ratios in the present study.

1. *Diurnal Variation of* CH_4 *Emission Fluxes*

For both early and late rices, the CH_4 emission fluxes showed significant diurnal variations throughout the entire growing seasons. However, the patterns of variations differ significantly for the early rice and late rice, as shown in Fig. 3a and b, respectively. Moreover, the magnitude of the fluctuations is also different for early rice and late rice, and varies significantly during the entire growing season.

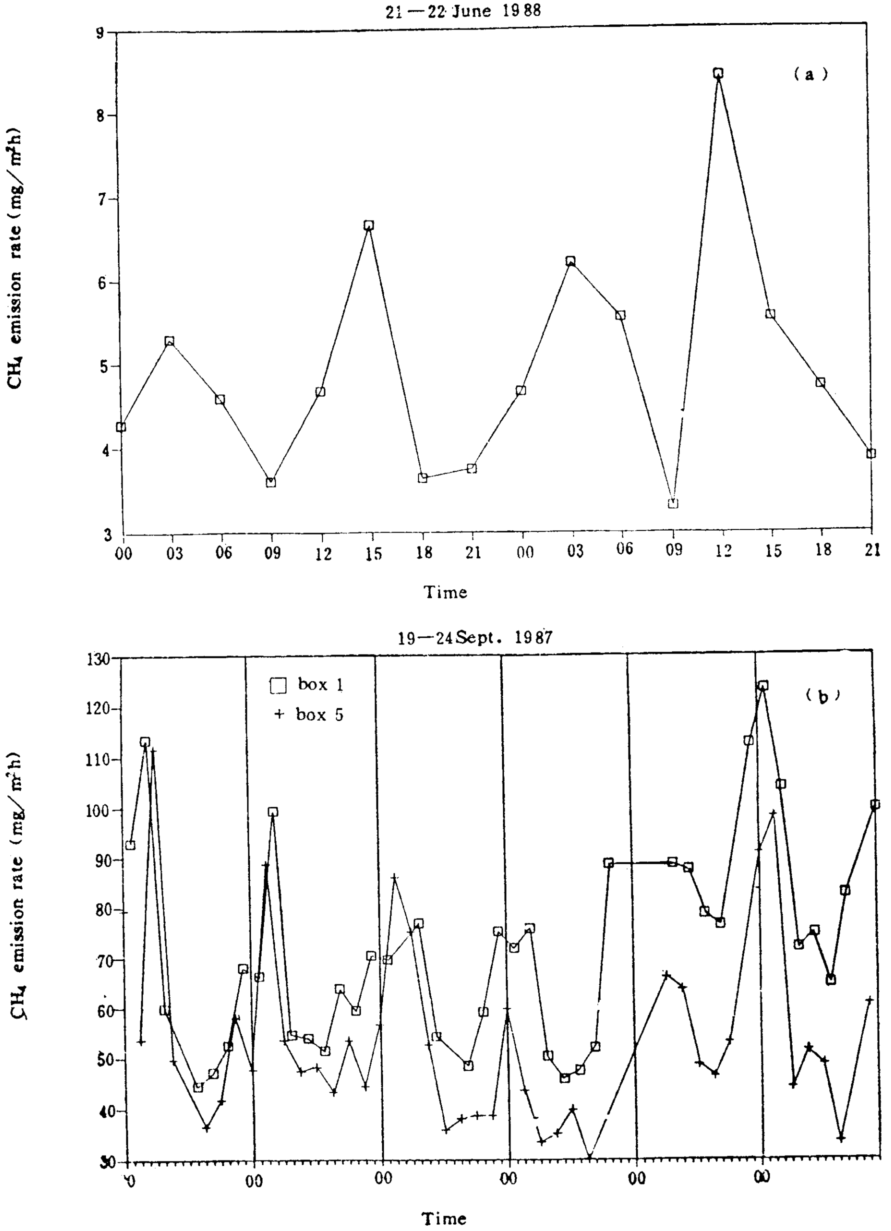

Fig. 3. Diurnal variation of CH_4 emission rate for the early rice (a) and late rice (b).

For the early rice, the most pronounced peaks were generally found in the early afternoon, 12:00—15:00, with the maximum to minimum ratios ranging from 1.26 to 12. A second peak was often found at mid-night, 0:00—3:00. For the late rice, however, the highest values were almost always found in the night, while the lowest values appeared during mid-day. Bi-mode patterns with the second peak in the afternoon were also found during the reproductive stage of the rice plants for the late rice. These findings in a Chinese rice paddy differ significantly from diurnal patterns of CH_4 emissions reported in the literature (Schütz et al., 1989 a,b). These patterns have revealed the complex nature of the processes governing the CH_4 emission flux. The CH_4 emission from planted paddy soil is mainly determined by three different processes, i.e. CH_4 production, CH_4 reoxidation, and gas transport (Conrad, 1989). Whenever the CH_4 production rate is low and/or the transport is inefficient, most of the produced CH_4 in the soil will be reoxidized on its way to the atmosphere, and hence the emission flux will be low. On the other hand, when the CH_4 production rate is high, and/or the gas transport is efficient, a substantial amount of the CH_4 produced in the soil will be able to reach the atmosphere, and hence the emission flux may be high.

The complex combination of the above processes is responsible for the complex diurnal variation pattern of the CH_4 emission flux reported here. Generally, the higher temperature in the paddy soil in the afternoon is in favor of higher CH_4 production and may be responsible for the peak values found in the afternoon for both early and late rices. On the other hand, the gas transport efficiency, which is mainly determined by the vascular gas transport through the plant in the planted paddy soil and the changes in stomatal conductance, will determine the fate of the produced CH_4, reoxidized or emitted to the atmosphere (Conrad, 1989). Therefore, it is the plant activity that may be responsible for higher emission fluxes found in mid-night and lower values found in mid-day. We hypothesize here that the stomata of the rice plant may close during mid-day due to strong solar radiation. Thus the gas transport pathway is blocked, and the CH_4 emission flux from soil to atmosphere is significantly reduced, although the CH_4 production rate in the soil will be high due to high temperatures. At the same time, the closure of plant stomata may also block the transport of O_2 from the atmosphere to the plant root system and cause a significant reduction in the reoxidation of the CH_4 produced in the anoxic part of the soil. Thus, the produced CH_4 may be accumulated in the deeper layer of the paddy soil during the day. After sunset the reopening of the plant stomata may enable transport of the accumulated CH_4 to the atmosphere, thereby forming the peak of CH_4 emission observed in the night. This mechanism is most pronounced in mid-summer, June—August, when the solar radiation is stronger in the subtropical region; it further depends strongly on the variety of rice used.

2. *Seasonal Variation of the* CH_4 *Emission Fluxes*

In addition to the diurnal fluctuations strong seasonal variations of the CH_4 emission fluxes have also been observed for the early rice of 1988 and the late rices of 1987 and 1988 (Fig. 4). During the three growing seasons, the CH_4 emission fluxes showed three major maxima with minor short-term fluctuations. The first and largest maximum occurred about 10 days after transplanting at the beginning of the tillering stage of the rice plants. Two other maxima were found during the reproductive phase, short before and/or after the flowering stage of the rice plants. These patterns of seasonal variations of the CH_4 emission rates are likely to be correlated with the availability of organic substrates in the paddy soil and the activity of rice plants, releasing organic substrates into the soil and exchanging gases between soil and atmosphere. The first maximum of CH_4 emission rates is apparently due to the decomposition of organic matter left in the soil before flooding and the efficient transport of the produced CH_4 by the rice plants, growing most vigorously during the tillering stage. The second and third maxima are most probably due to the activity of the rice plants which have developed their root system during the tillering stage. The root system may provide the soil bacteria with organic substrates in the form of root exudates or root litter during the reproductive phase of the rice plants (Yoshida, 1981). Root exudates of rice plants consist mainly of carbohydrates, organic acids and amino acids (Boureau, 1977), which are readily decomposed by fermentative soil bacteria to CO_2, H_2, and acetate. These compounds are then the substrates used by methanogenic bacteria to produce CH_4.

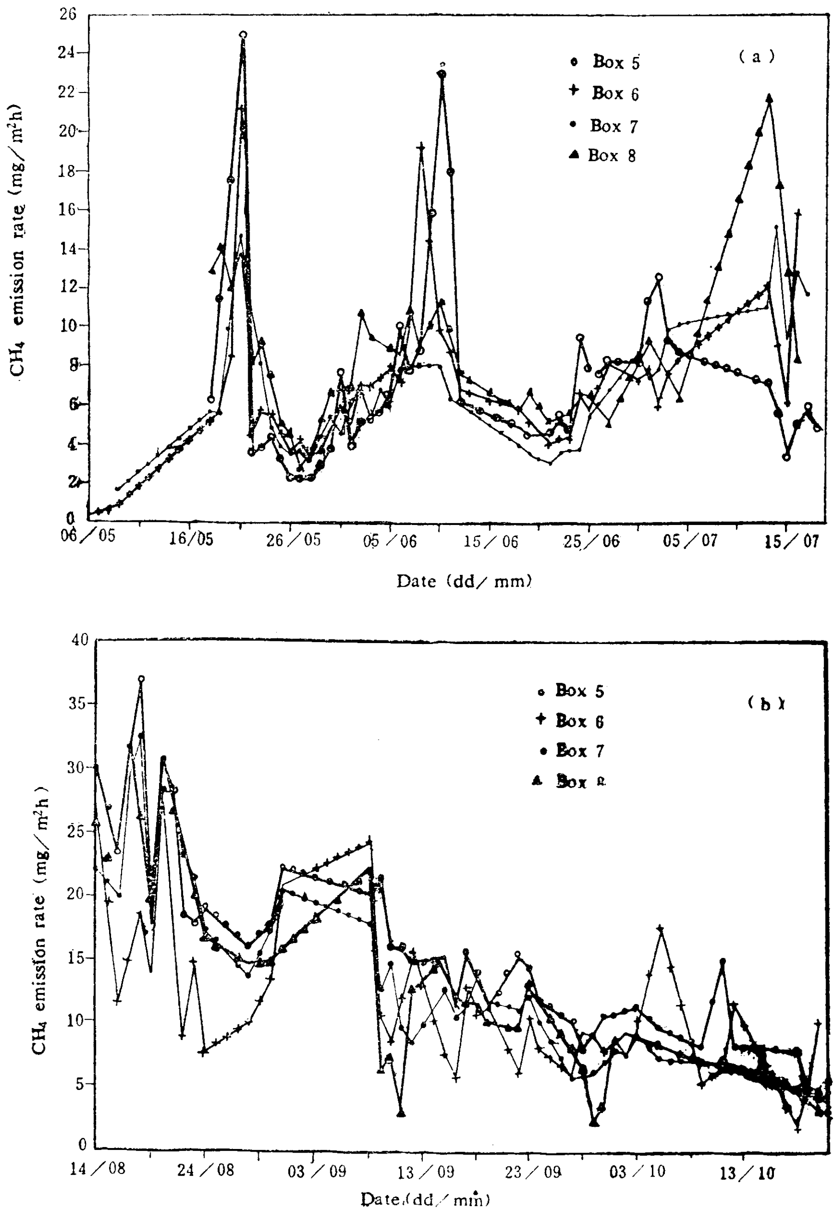

Fig. 4. Seasonal variation of CH_4 emission rate for the early rice (a) during May-July 1988 and late rice (b) during August—October 1988.

The observed seasonal fluctuations may also be the consequence of changes in the activity of soil bacteria due to soil temperature fluctuations. Poor correlations were found between the in-situ measured soil temperatures and the CH_4 emission fluxes. However, if the measured soil temperatures were shifted two days after the real measuring dates, much better correlations between the soil temperatures and the CH_4 emission rates were obtained. This two-day time lag of the CH_4 emission fluxes reflects the time needed for the development of the methanogenic bacteria plus the time needed for the transport of the produced CH_4 through rice plants.

3. *The Effect of Fertilization*

The present field experiments were designed to study the influence of the application of different types of fertilizers on the CH_4 emission fluxes from the rice paddy fields. As described in Section Ⅱ, the experimental field was divided into four narrow plots by reinforced concrete boards, and was then treated with different fertilizers. As shown in Fig. 4, SO_4^{2-} containing fertilizer slightly reduced the CH_4 emission fluxes during the early period of vegetation, but enhanced the CH_4 emission during the late period of the growing seasons. However, no significant effect was found on the seasonal averages of the CH_4 emission rates.

The reduction of CH_4 emission rates due to the application of SO_4^{2-} containing fertilizer in the early vegetation period may be caused by the competition of sulfate reducers and methanogenic bacterial. The application of sulfate fertilizer may stimulate the growth of sulfate reducers; the acetate-utilizing sulfate reducers are able to outcompete methanogenic bacteria for their common substrate, acetate, thereby reducing CH_4 production in the soil (Conrad, 1989). On the other hand, fertilization caused an enhancement of plant growth, which provides additional substrate for the methanogenic bacteria. As a consequence, CH_4 production during the late period of vegetation may be enhanced.

The application of organic fertilizer showed no significant effect on the CH_4 emission fluxes. This may be due to the insufficient amount of the fertilizer used.

4. *Total Seasonal Emission Fluxes*

The present study has resulted in a seasonal average of CH_4 emission fluxes of 7.8 mg/(m^2 h) for the early rice, and of 28.6 mg/(m^2 h) for the late rice. These resulted in a total-seasonal CH_4 emission of 16.2 g CH_4/(m^2 yr) for the early rice, and of 75.9 g CH_4/(m^2 yr) for the late rice.

A comparison of our results with what has been reported in the literature indicated that CH_4 emission fluxes from rice paddy fields have a wide range of variation. Among the many factors that govern the emission fluxes soil types, rice plant varieties and local climate may be most important. These observations make the estimate of the global CH_4 emission from rice paddies more difficult.

Assuming that earlier measurements made in Europe, USA and Japan were representative of those regions (Schütz et al., 1989a; Yagi et al., 1989; Cicerone et al., 1983) and the present measurement is representative of Asia, we present here a new estimate of the global CH_4 emission from rice paddies for 1987. The details of this estimate are listed in Table 1. The CH_4 emission from rice paddy fields is calculated to be $(65-107)\times 10^{12}$ g per year with a probable number of 90×10^{12} g per year, which accounts for 20% of the global CH_4 sources. This indicates that the rice paddy fields are probably the largest source of atmospheric CH_4 and contribute significantly to the temporal increase of atmospheric CH_4. There is no doubt that the CH_4 emission from the rice paddies had increased during the last decades and will further increase in the following decades. The world annually harvested rice paddy area has almost doubled between 1940 and 1980, from $80\times 10^{10} m^2$ to 145×10^{10} m^2. The harvested paddy area in Asia will increase by 25% until the end of this century (FAO, 1985). Assuming that the overall average CH_4 emission flux from the rice paddies has not changed during the last 4 decades and will remain the same for the rest of this century, the world total CH_4 emission from this source would have increased from 49×10^{12}g in 1940 to 99×10^{12}g in 1987 and will increase to 110×10^{12}g in 2000. This will certainly be an important factor for the future temporal trend of atmospheric CH_4 and for the changing climate.

Table 1. Estimates of Global CH_4 Emission from Rice Paddies

Regions	Area harvested ($10^{10}m^2$)	Vegetation period (days)	CH_4 emission flux ($g\ m^{-2}$) per day	CH_4 emission flux ($g\ m^{-2}$) per year	Annual CH_4 emission ($10^{12}g\ yr^{-1}$)	Ref.
China	32.20					
early rice	9.58	75—95	0.19	14.25—18.05	1.4—1.7	this work, Hangzhou 1988
late rice	20.32	80—140	0.69	55.2—96.6	11.2—19.6	this work, Hangzhou 1987,1988
single crop rice	2.32	120—150	0.44	52.8—66.0	1.2—1.5	this work
Japan	2.31	120—150	0.10	12.0—15.0	0.28—0.35	(Yagi,1989), Japan 1988
Asia (Ex. China and Japan)	98.22					
Dry season	25.26	75—95	0.19	14.25—18.05	3.6—4.6	this work
Wet season	72.96	80—140	0.69	55.2—96.6	40.3—70.5	this work
USA	1.12	120—150	0.25	30.0—37.5	0.34—0.42	(Cicerone, 1983), California 1982
Europe	0.38	120—150	0.28	33.6—42.0	0.13—0.16	(Schütz, 1989a), Italy 1985, 1986
Other regions	13.30	120—150	0.37	44.4—55.5	5.9—7.4	(Schütz, 1989a), Italy 1985, 1986
Global	147.5				64—107	

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