

Sources of methane in China

Wang Mingxing

Institute of Atmospheric Physics, Chinese Academy of Sciences,
Beijing 100012, China

Abstract— Continuous measurements of the methane flux from rice fields in Hangzhou, eastern China, and Leshan, Sichuan Province, were carried out for 3 years. The results show that the methane flux from rice fields has a strong diurnal variation which may be explained by the soil temperature effects and rice plant growing activities, and that the day-mean flux has a large seasonal variation which may be related to soil temperature and the availability of organic substrate in the soil.

An overall sources of methane for China are estimated while the rice fields are major sources, and the sewage treatment is an important source of methane in China that has not been measured. Sewage in China is often treated in cesspools, which are similar to the biogas pits. However, no methane is recovered from such cesspools. We estimate that the total flux of methane from such sources may be of the order of 10 Tg/a. The total emissions of methane from sources in China are estimated to be about 40 ± 10 Tg/a.

Keywords: methane source; methane emission; rice field; biogas.

1 Introduction

While the concern about the greenhouse effect of increasing atmospheric CO₂ concentrations has stimulated numerous research programs in atmospheric, oceanic and biospheric sciences already in the 1970s, little was known about the increase of atmospheric methane (CH₄) until the early 1980s. Recent investigations have shown that the atmospheric CH₄ is increasing with a rate much faster than that of CO₂ and that its effect on climate is already as important as that of CO₂ and may become even more important in the near future (Schneider, 1989). Therefore, there is an urgent need to quantify the sources and sinks of methane and to investigate the global methane cycle more comprehensively.

The long continuous record of atmospheric CH₄ concentration measurements dates back to 1976 (Seiler, 1982). Further measurements with wider geological locations and higher frequency have been carried out since the early 1980s (Rasmussen, 1981;

Steele, 1987; Blake, 1988; Wang, 1990). Based on these measurements it has been calculated that atmospheric CH_4 is increasing globally with a rate ranging from 0.8%—1.7% per year and that its present globally averaged concentration is about 1.7 ppm with higher value in northern hemisphere. Analyses of air bubbles trapped in ricecores have indicated that the concentration of atmospheric CH_4 has more than doubled in the last 200 years, and that its concentration remained between 0.6ppm and 0.8 ppm before 1800 through past 3000 year (Rasmussen, 1984).

Like CO_2 , atmospheric CH_4 is a very important radiatively active gas that governs the earth's climate. The presence of 1.5ppm CH_4 in the atmosphere causes the globally averaged surface temperature to be about 1.3 K higher than it would be with zero CH_4 (Donner, 1980). The effect on climate of the increase of atmospheric CH_4 , from 0.7 ppm to 1.7 ppm is about half the effect of the simultaneous increases of atmospheric CO_2 from 275 ppm to 345 ppm. As atmospheric methane increases at a rate faster than atmospheric CO_2 , its contribution to future climate changes will become even more important.

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 as 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 $\text{CO}_2\text{-C/a}$ 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 Cl 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 some of individual sources and sinks are still not sufficiently quantified. The most important source of atmospheric CH_4 is the anaerobic decay of organic matter. Anaerobic environments with a high potential for CH_4 production are natural wetlands, rice paddies, waste dumps, and the rumen of livestock and other animals. CH_4 flux measurements for these sources have revealed the complex nature of CH_4 production. Regional disagreements among the measured CH_4 fluxes for

the individual sources give all the estimates of global CH_4 sources strengths a significant degree of uncertainty. The source strength estimates of rice paddies, in particular, suffer from the lack of data from Asia where more than 90% of the world's total rice fields are harvested. The limited information on the emission of CH_4 from rice paddies reported so far are based on measurement in the USA and in Europe. These measurements showed that incubation experiments in the laboratory are not suitable for calculating methane fluxes from rice paddies in the field (Holzapfel, 1986; Schutz, 1989). A better estimate on the emission of CH_4 from rice paddies can therefore only be achieved by field measurements of CH_4 emission rates in the most important rice-growing regions.

In this paper, we report the first field measurements of CH_4 emission rates from rice field in Asia, and the measurements and estimates of CH_4 emission rates from other sources. Continuous measurements of CH_4 emission rates were carried out on a rice field near Hangzhou, a typical rice growing region where two crops of rice are harvested each year, and a rice field near Leshan, Sichuan Province, another type of growing region where single crop of rice is harvested each year. Chamber technique was used for the determination of CH_4 flux from rice fields.

Similar technique was used for the measurement of CH_4 leakage from bio-gas pits. The emission rates of CH_4 from other sources were estimated based on other data sources.

2 Emission rates of CH_4 from rice paddy fields

2.1 Measurements in Hangzhou

An automatic sampling and analyzing system was used in this study.

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 River, in Zhejiang Province, China. The Soil of the field used for the present study is representative for the plain of the middle and lower reaches of Yangtze River, which covers more than one fourth of the total sown area of rice paddy field 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 varied between 6.5 and 7.5 during the vegetation periods.

The experimental sites were routinely managed using the local farming procedures. The field was usually planted with green manure crops or vegetable 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 harvest 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 seedling had recovered and begun to grow, water level is 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 chemically fertilizer, e.g. urea, ammonium sulfate, was surface applied during the reproductive phase of the rice plants.

The experimental site used for measurements had a size of $24 \times 14 \text{ m}^2$ and was parceled into 4 narrow fields of $24 \times 3 \text{ m}^2$ 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 7.5 kg K_2SO_4 , (approximately 1050 kg/ha), the third one received 5 kg rape cake (700kg/ha) in addition to 7.5 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 thermocouple. Two thermocouple measured the air temperatures inside and outside of one of the gas collector boxes, respectively. Another two thermocouple were placed into the paddy water at about 5 cm depth with one inside and one outside of a gas collector box. Four additional thermocouple were used to measure the soil temperatures at the surface, 5cm, 10cm, 15cm depth in an area adjacent to the boxes. Both air and soil temperatures recorded inside and outside the boxes always showed comparable values.

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 using Equation 1:

$$F = Hp \frac{dm}{dt}, \quad (1)$$

where H is the height of the box, p is the density of CH_4 at the pressure and temperature recorded inside the box, and $\frac{dm}{dt}$ 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 if the mixing ratio reached values of 300 ppmv within the 90 minutes 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 techniques was suitable for the determination of the rate of increase of the CH_4 mixing ratios in the present study.

CH_4 emission fluxes from the 4 spots of each of the four narrow experimental fields were determined 8 times a day. The CH_4 emission fluxes from the 4 measuring spots of each field were averaged to give the CH_4 emission flux for that particular experimental field at a specific time.

2.1.1 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 different significantly for the early rice and late rice, as shown in Fig. 1. Moreover, the magnitude of the fluctuations is also different for early rice and late rice, and varies significantly during the entire growing season.

For the early rice, the most pronounced peaks were generally found in the early afternoon, 13:00–16:00, 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 (Schutz, 1989a; 1989b). 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 (Corad, 1989). Whenever the CH_4 production rate is low and/or the transport is inefficient, most of the

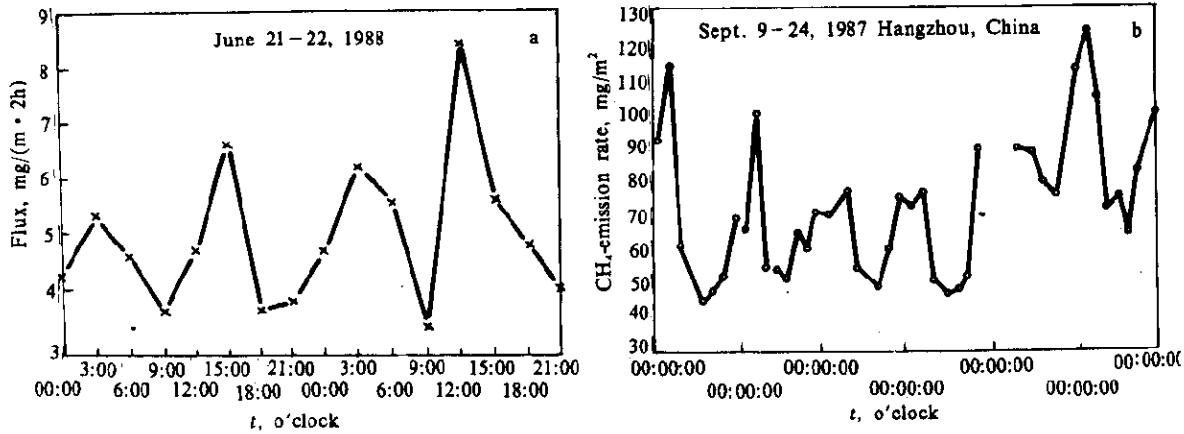


Fig.1 Diurnal variation of CH₄ emission flux from double-crop rice paddies
a. early rice; b. late rice

produced CH₄ 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 CH₄ production rate is high, and/or the gas transport is efficient, a substantial amount of the CH₄ 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 patterns of the CH₄ emission flux reported here. Generally, the higher temperature in the paddy soil in the afternoon is in vapor of higher CH₄ 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 produced CH₄, reoxidized or emitted to the atmosphere (Corad, 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 CH₄ emission flux from soil to atmosphere is significantly reduced, although the CH₄ 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 O₂ 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 sun-set the reopening of the plant stomata may enable transport of accumulate 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 sub-tropical region, it further depends strongly on the variety of rice soil.

2.1.2 Seasonal variation of 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. 2). During the three growing seasons, the CH_4 emission fluxes showed three major maxima with minor short-term fluctuations. The first 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, shortly before and 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 the soil and the 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 .

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 reflect 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.

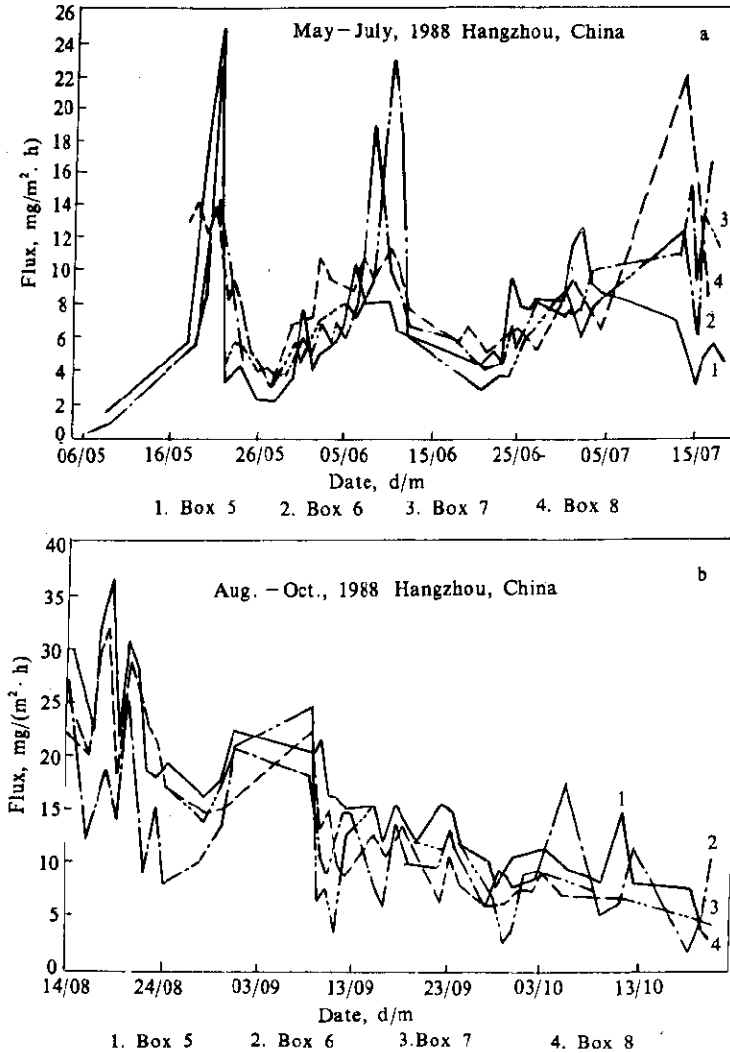


Fig. 2 Seasonal variation of CH_4 emission flux from double-crop rice paddies
a. early rice; b. late rice

2.1.3 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 2.1, the experimental field was divided into four narrow plots by reinforced concrete boards, and were then treated with

different fertilizers. As shown in Fig.2, 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 bacteria. 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 (Corad, 1989). On the other hand, fertilization caused an enhancement of plant growth, provides additional substrate for the methanogenic bacteria. As a consequence, CH_4 production during the late period of vegetation may be enhanced.

2.2 Measurements in Sichuan

South-west of China represents another unique agricultural practice, where it is common that winter green manure or vegetables is grown in addition to a single crop of rice. Four conventional rice fields, each with an area of 0.1 hectare, along a small valley at Tuzu, near Leshan, Sichuan Province, were selected for the measurements of CH_4 emission rates from single crop rice fields. The fields were managed according to local tradition. Vegetables were grown in winter time. Animal and human waste and green manure were ploughed into the soil right before flooding in late April. The rice seedlings were raised on a separate field in early April and transplanted in the flooding fields in early May. Harvest begun at late August and early September, giving a total growing season of about 120 days.

In each of the four fields, 6 chambers were placed on mud. Flux measurements were carried out by taking 4 samples from each chamber at a particular time interval. Measurements were so arranged that two fluxes were determined for each chamber each day and four days a week.

Measurements have been carried out at the same spots in 1988–1990. Total 3500 flux estimates have been obtained. There is a general tendency that emission rates higher in late afternoon than that in the early morning. In addition to the diurnal variations, there is also a pronounced day-to-day variations, as shown in Fig.3.

The day mean CH_4 emission rate can be related to day-mean soil temperature, except for the late part of the growing season when the CH_4 emission rates drop significantly mainly due to the water draw off for harvest (Fig.3).

There is also a significant year-to-year variation. Overall emission rates in 1989 are lower than that in 1988 (Fig. 4). This variation may be related to climate

changes, as in 1989 the area experienced a lower temperature summer.

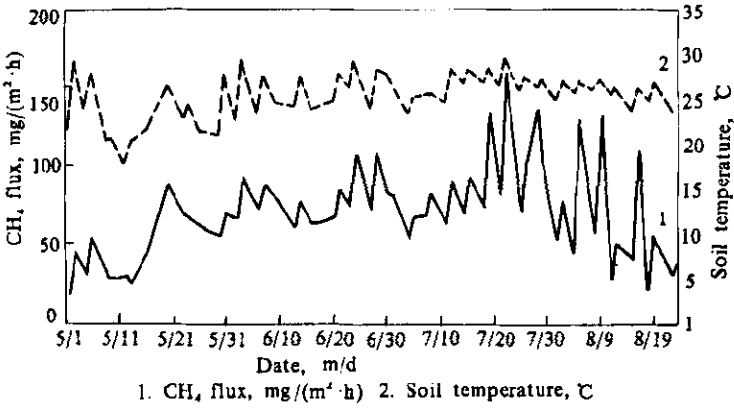


Fig. 3 Seasonal variation of CH₄ emission flux from single crop rice paddies as related to soil temperature

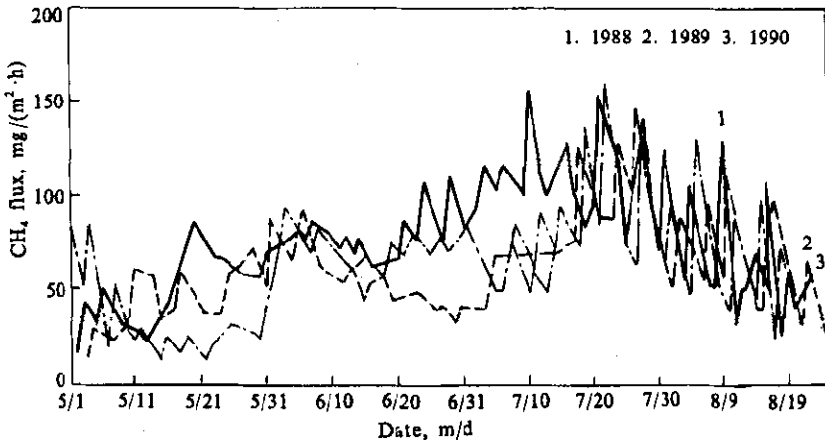


Fig. 4 Year-to-year difference of CH₄ emission flux from single rice paddies

2.3 Estimate of total regional emission rates of CH₄ from rice paddy fields

The field measurement results presented in this paper have demonstrated the complex nature of the CH₄ emission from rice paddy fields, as it has very large diurnal, seasonal, and year-to-year variations with complex patterns. To obtain a reliable total season emission rate from a particular field, continuous measurements during the whole growing season must be carried out for several years. Unfortunately, most of the published data so far are beyond this kind.

Our continuous measurements have resulted in seasonal average CH₄ emission rates of 7.8 mg/m²·h for the early rice and 28.6 mg/m²·h for the late rice in Hangzhou, and 60 mg/m²·h for the single rice in Sichuan. Taking into account of the growing periods, the above results have resulted in total seasonal emission rates of 16.2 g/m² for the early rice and 75.9 g/m² for the late rice in Hangzhou, and 194.4 g/m² for the single rice in Sichuan.

These results from Chinese rice fields differ greatly with the published data obtained from the rice fields in USA, Italy and Spain, i.e. the CH₄ emission rate from rice fields have wide range of spacial variation. Among the many factors that govern emission rates soil types, rice plant varieties and local climate may be most responsible for the observed spacial variations.

Table 1 Estimate of regional CH₄ emission from rice paddies

Regions	Area	Vegetation	CH ₄ emission		Annual CH ₄	References
	harvested	period,	flux, g.m ⁻²	emission	emission	
	10 ¹⁰ m ²	day	per day	per year	10 ¹² g	
China	32.20				16.6-26.3	
Early rice	9.58	75-95	0.19	14.25-18.05	1.4-1.7	This work, Hangzhou, 1988
Later rice	20.30	80-140	0.69	55.5-96.6	11.2-19.6	This work, Hangzhou, 87,88
Single crop rice	2.32	120-150	1.44	172.8-216	4.0-5.0	This work, Sichuan, 88, 89
Japan	2.31	120-150	0.10	12.0-15.0	0.3-0.4	Japan, 1988
Asia						
(ex, China and Japan)	98.22				44-75	
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	California, 1988
Europe	0.38	120-150	0.28	33.6-42.0	0.13-0.16	Italy 1985, 1988
Other regions	13.30	120-150	0.37	44.4-55.5	5.9-7.4	Italy 1985, 1986
Global	147.5				70-110	

Assuming that earlier measurements made in Europe, USA and Japan were representative of those regions (Schutz, 1989; Yagi, 1989; Cicerone, 1983) and the present measurements are representative of China and South Asia, we present here estimates of the regional CH_4 emission rate from rice paddies, as listed in Table 1. Based on these estimates, the total CH_4 emission from the global rice paddies is estimated to be 70–110 Tg/a with a probable number of 90 Tg/a, which accounts for 18% of the global atmospheric CH_4 sources.

It must be pointed out that the great spacial variability of the measured CH_4 fluxes from rice paddies makes the estimates of regional and global emission a very difficult task. The presently available measured data are far from enough and the present estimates are all questionable. Much more continuous measurements are urgently needed, especially in the major rice growing regions, i.e. South Asia and South-East Asia.

Rice is a high production crop, and with the improvements of water and fertilizer management the production of per unit field may be raised significantly. Therefore, the total world annually harvested rice paddy area (including the multi-crop fields) has almost doubled between 1940–1980, increased from $80 \times 10^{10} \text{ m}^2$ to $145 \times 10^{10} \text{ m}^2$, satisfy the increasing demand for food (FAO, 1985). It is estimated that the harvest area of rice paddy fields in Asia will be increased by 25% by the end of the this century. Assuming that the overall average CH_4 emission rate from rice paddied 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 $49 \times 10^{12} \text{ g/a}$ in 1940 to $90 \times 10^{12} \text{ g/a}$ in 1988 and would increase to $110 \times 10^{12} \text{ g/a}$ in 2000. This at least partly accounts for the observed increase of atmospheric concentration of CH_4 and will be an important factor for the future trend of a atmospheric CH_4 .

3 Emissions of CH_4 from biogas generators

A typical biogas generator consists of two main components (Fig. 5), a large central chamber and a side chamber. On the top of the central chamber there is a hole covered with a concrete plug and sealed with clay and water. Human waste, pig manure, rice straw, and other organic farmyard wastes are put into the bottom of the chamber and the residues are evacuated once or twice a year through the hole. Water is added through the side chamber to a certain level. thus, the organic material forms a sludge under water. The anaerobic processes in the sludge and the liquid above produce biogas, which is mostly CH_4 and CO_2 and small quantities of other gases. These gases bubble up to the empty region at the top of the central chamber. The biogas in the upper part of the chamber is under pressure, so that it is carried to the

kitchen and the house through plastic tubing without the use of pumps.

We identified three areas where methane could leak out. The removable concrete plug in the top hole is potential source of leaks (6 experiments). The second place where methane and other gases may leak out is the top of the side chamber, which is covered with a concrete board (3 experiments). The water level is often about 0.6 m below the top, and it is not sealed and gas is freely exchanged with the atmosphere. Methane released from this area is not recoverable. However, since most of the biogas is produced in the sludge at the bottom of the central

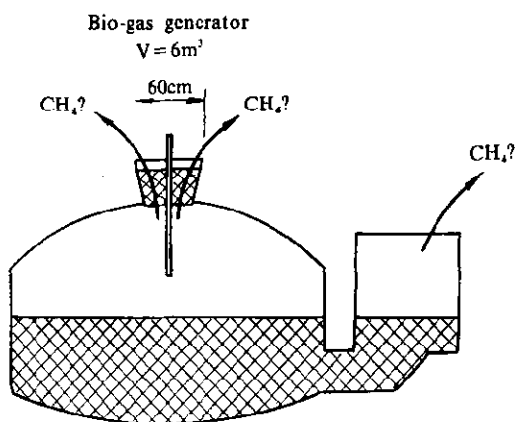


Fig. 5 Possible leakage from a bio-gas generator

chamber and bubbles rise straight to the top into the effectively sealed region at the top of the chamber, very little methane makes its way out to the side chamber. Finally, we also considered losses of methane from the unburned biogas in the kitchens (1 experiment).

In these experiments, the chamber was a clear plastic bag that was placed on the top of the side chamber cover or the plug. At the beginning of the experiment a measured amount of a tracer (CF_3Br or SF_6) was injected into the bag (Typically 30–100 ml of 0.1 ppmv tracer). The bag was shaken gently to mix the tracer. Samples were taken with a pump into the 0.8 L stainless steel containers. Each experiment consisted of a background sample and 4 samples drawn about 5 minutes apart.

The volume of the chamber is determined by the mass balance of the tracer.

Once the volume of the chamber is known, fluxes of methane leakage are determined by a linear regression method from the increase rate of concentrations of methane inside the chamber that are determined by the analyses of 4 sequential samples.

We found that there is very little leakage on the whole. Most of leakage is from the top of the side chamber. Even though the top of the central chamber is under pressure, little methane leaks out from the top hole because of the clay and standing water seal used to prevent leakages.

The average methane flux rates from the tops of the pits were about 11 mg/h · pit, but the range was 1–170 mg/h · pit. From the top of side chamber the average emissions were 90 mg/h · pit with a range of 60–320 mg/h · pit. These rates are too small to have an effect on the global methane cycle. When extrapolated to the upper limits of the estimated number of pits in China, the annual total emission rates are about 9 gigagram/a with a range of 5–30 gigagram/a.

An experiment was also conducted to test whether there is a significant fraction of methane in the kitchens. This concern arose because the biogas contains large amounts of CO₂ and may not burn as efficiently as pure methane. Four successive samples were collected 2 feet above the flame. The concentrations of methane in all samples were not significantly different from ambient levels. Large amounts of CO₂ and slightly higher levels of N₂O were observed. We concluded that the unburned fraction is not a significant loss of methane.

4 Emissions of CH₄ from other sources

4.1 Urban areas

Methane concentrations in ambient atmosphere have been measured in Beijing and Minqin, a continental background site, and around the rice fields in Tuzu. The results are listed in Table 2. For comparison data from other cities are also listed.

Table 2 Annual average methane concentrations (in ppbv)
in urban and rural areas of China

	Beijing	Minqin	St. Louis	Tuzu	Other Cities
Average	1822	1735	1815	2844	1904
Upper	1835	1739	1818	3001	2073
Lower	1808	1731	1813	2687	1736
Maximum	2069	1796		6414	3420
Minimum	1731	1632		1819	1590

Notes: upper and lower limits are 90% confidence limits of the mean.

We estimated the annual emissions of methane from Beijing by assuming that the ratio of the annual average flux to annual average ambient concentration in excess of background levels is the same in Beijing as in Tuzu near the rice fields (Khalil, 1984). It turns out to be between 5 and 110 mg/m² · h, taking concentrations listed in Table 2. We extrapolate this emission rate to all urban areas of China. The estimated total emissions for urban areas in China is about 0.6 Tg/a.

4.2 Cattle and other ruminants

There is no detailed measurement data for the emission of CH_4 from the Chinese cattle. We use the emission factors given by Lin Erda (personal communications), which were determined by an estimate of methane emission from cows and sheep in relation to their food consumption, and the population of Chinese domestic animals (cows, buffalos, camels and sheep) to calculate the total CH_4 emission from cattle and other ruminants in China, which turns out to be about 5.5 Tg/a in 1988.

4.3 Natural wetland

No accurate data are available on the total area of Chinese natural wetland, and there is no measurements of CH_4 flux from such source. We adopt the averaged emission rate of CH_4 from the world natural wetland, $20 \text{ g/m}^2 \cdot \text{a}$ (Aselmann, 1989) for the Chinese wetland, and the total area of natural wetlands published in the China Agricultural Yearbook, 1985, $1 \times 10^{11} \text{ m}^2$. Thus, we estimate that the total emission from Chinese natural wetlands is $2 \times 10^{12} \text{ g/a}$.

4.4 Coal mines

CH_4 emission from coal mines was estimated based on the limited measurements of gas recycling and venting in the large scale coal mines. The gas with 30% CH_4 or more from coal mines is usually recovered, and that with lower CH_4 content is freely released to the atmosphere. Venting air from coal mines usually contains even less CH_4 (Yu, personal communications). Based on the limited measurement data we estimate that CH_4 emission from such source is about 6 Tg/a in 1988.

5 Summary

An overall sources of methane for China is given in Table 3. We have shown that rice field are a major source in China and in the global cycle of methane. In China, methane emissions from rice fields are probably greater than that from all other sources combined. We also evaluated the emissions from biogas pits in rural China. These pits are used to generate methane from farm wastes and sewage. The emissions of methane from the 10 million biogas pits do not amount to a significant contribution on a global scale and is certainly less than 0.3 Tg/a. It has been suggested that treatment of other sewage not in biogas pits, could be a more substantial source of methane from China, amounting to about 3 Tg/a. Sewage in rural areas in China is treated in cesspools, which are similar to the biogas generators, but the gases are not recovered. From what we have evaluated, it appears that methane emissions from China are about 9% of the global annual emissions.

Table 3 Source of CH₄ in China

Sources	Emission rate 1988, Tg/a	Percentage of global
Rice field	21 ± 6	25
Cattle and other		
ruminants	5.5	6.9
Urban areas	0.6	<2
Biogas pits	<1	-
Natural wetland	-2	<2
Coal mine	6	13
Sewage treatment	-3	?
Other sources	-	-
Total	40 ± 10	-9

Acknowledgements— Major results presented in this paper are outputs of international cooperations which reflect contributions of 3 science teams led by professors Wang, Rasmussen and Seiler. Financial supports in China are from National Natural Science Foundation of China, and the Chinese Academy of Sciences.

References

- Aselmann I, Crutzen PJ. *J Atm Chem*, 1989; 8:307.
- Blake DR, Rowland FS. *Science*, 1978-1987; 239 1129
- Boureau M. Application de la chromatographie en phase gazeuse leture de L' exsudation racinaire du riz. *Cahiers Orstom*, 1977; 12:75
- Cicerone RJ, Shetter JD, Dellwiche CC. *J Geophys Res*, 1983; 88:11022
- Cicerone RJ, Oremland RS. *Global Biogeochemical Cycles*, 1988; 2:299.
- Corad R. Control of methane production in terrestrial ecosystems, Dahlem conference background paper, West Berlin, 1989
- Donner L, Ramanathan V. *J Atoms Sci*, 1980; 37:119
- FAO. *FAO production Yearbook*, 1985, 39, FAO Rome, 1985
- Holzappel-Pschorn A, Seiler W. *J Geophys Res*, 1986; 91:11803
- Khalil MAK, Rasmussen RA, Wang MX, Ren L. Sources of methane in China: rice fields, biogas pits, cattle, urban areas and wetlands, *Proceedings of ICGREAC*, 1984

- Rasmussen RA, Khalil MAK. *J Geophys Res*, 1981; 86:9826
- Rasmussen RA, Khalil MAK. *J Geophys Res*, 1984; 89:11599
- Schutz H, Holzappel-Pschorn A, Conrad R, Rennenberg H, Seiler W. *J Geophys Res*, 1989a (In press)
- Schutz H, Seiler W, Conrad R. *Biogeochemistry*, 1989b; 7:33
- Schneidder StH. Global climate and trace gas composition. Dahlem konferenzen background paper, West Berlin, 1989
- Seiler W. Zeitliche anderung der konzentration atmospharischer spurenstoffe, BPI-Bericht 4182, 1982:1
- Steele LP, Praser RK, Khalil MAK, Conway TJ, Crawford AJ, Gammon RH, Masarie KA, Thoning KW. *J Atmos Chem*; 1987, 5:125
- Wang Mingxing, Liu Guotao, Rasmussen RA, Khalil MAK. *Chinese Science Bulletin*, 1990; 35:213.
- Yoshida S. Fundamentals in rice crop sicence. The International Research Institute (Ed by Los Banos Lguna), The Philippines, 1981
- Yagi K, Minami K. Effects of organic matter applications on methane emission from Japanese paddy fields, International conference on solid and the greenhouse effect, Wageningen, The Netherlands: ISRIC, 1989

(Received July 14, 1992)