## B-2.3 Emission of Methane and Nitrous Oxide from Agricultural Lands

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**Abstract** Methane ($CH_4$) emission from rice paddy fields in Japan and Thailand were studied to estimate the annual emission rate of $CH_4$, and to find major factors controlling the CH4 emission. Nitrous Oxide ($N_2O$) emission from fertilized soils and rice paddy fields were also studied to estimate the $N_2O$ emission rate during a cultivated season. Annual $CH_4$ emission rate from rice paddy fields in Thailand was estimated to be 3.7 $TgCH_4$/year from the flux measurements in the rice experiment stations between 1991 - 1994 under the collaborative study with the DOA, Thailand. The annual $CH_4$ emission rate in rice paddy fields of Japan was also estimated to be 0.26 $TgCH_4$/year. Major factors controlling the $CH_4$ emission from rice paddy fields were soil biochemistry, water management, and the amount of organic matter application. A modelling of $CH_4$ transport from paddy soils to the atmosphere was proposed by field and incubation experiments. $N_2O$ was emitted to the atmosphere from fertilized soils mainly through nitrification rather than denitrification. $N_2O$ was also emitted after final water drainage through a fallow season in rice paddy fields, and which were identified as one of the sources of atmospheric $N_2O$. Nitric Oxide (NO) was also emitted from the soils with the application of nitrogen fertilizers, and much higher in emission rate than $N_2O$.

**Key Words** Methane, Rice Paddy Fields, Nitrous Oxide, Fertilized Soils, Emission Rate

### 1. Introduction

Atmospheric methane ($CH_4$) and nitrous oxide ($N_2O$) are greenhouse gases, and their concentrations in the atmosphere have been increasing at the rates of 0.9, and 0.3 % per year, respectively. The $CH_4$ emission rate from rice paddy fields has been estimated to be 12 % of the total emission of the world, and the maximum emission rate of $N_2O$ from fertilized soils has been estimated to be about 30% of the total emission[1]. The uncertainty of the estimation, however, is large because of few data of flux in the field measurements.

The intensive researches on the emission of $CH_4$ from rice paddy fields, and of $N_2O$ from fertilized soils, have been carried out for these five years, collaborating with the researchers in the Asian countries. We gave priority to the $CH_4$ emission study over the $N_2O$ emission study. In this report, the major scientific results of these studies will be shown.

## 2. Research Objectives

### (1) Methane emission from rice paddy fields

One of the major purposes is to measure $CH_4$ flux from rice paddy fields in Japan and China in temperate region, and Thailand in tropical region to make clear the diurnal and seasonal variations of the $CH_4$ flux, and to estimate the total $CH_4$ emission rate in Japan and Thailand. We have been making a collaborative study with Department of Agriculture in Thailand since 1991, and Institute of Soil Sciences, Chinese Academy of Sciences since 1992. The another purpose is to find the major factors controlling the emission of $CH_4$, and to develop a model of the $CH_4$ emission from paddy soils to the atmosphere, by field and incubation experiments.

### (2) Nitrous oxide emission from fertilized soils and rice paddy fields

One of the major purpose is to measure $N_2O$ flux from fertilized soils in Japan, and to estimate the emission rate from different fertilizers. We also planed to measure the flux of nitric oxide (NO), which recently has been found to be emitted from fertilized soils as well as $N_2O$. It is well known that NO is one of precursors of photochemical smog and acid precipitation. The another purpose is to measure $N_2O$ flux from rice paddy fields as well as $CH_4$ through all the seasons, because $N_2O$ is expected to be emitted from rice paddy fields under the oxidized condition in soils through nitrification.

## 3. Experimental

### (1) Methane emission from rice paddy fields

The flux of $CH_4$ from rice paddy fields has been measured by closed chamber method every week during rice cultivated period. The sampling sites are the Ryuhgasaki agricultural experiment station in Japan, 9 rice experiment stations in Thailand, and 6 experiment stations in China. An automated flux measurement system, developed by our group, was set at the Ryuhgasaki station during 3 years (1991-1993) to measure diurnal and seasonal variation of the $CH_4$ flux in detail, and to compare the $CH_4$ emission rates between a continuously flooded plot and intermittently irrigated plot. Soil redox potential, soil temperature were also measured at the time of the flux measurement. The chemical analysis was made of paddy soils collected in the world.

### (2) Nitrous oxide emission from fertilized soils and rice paddy fields

The flux of $N_2O$ from agricultural fields was measured by closed chamber method at the lysimeter in National Institute of Agro-Environmental Sciences, and at the Ryuhgasaki experiment station where the $CH_4$ flux was measured. Soil water content, ammonium and nitrate ion in soils were also analyzed. NO flux was also measured in the fertilized soils.$N_2O$.

## 4. Results and Discussions

### (1) Seasonal variations of $CH_4$ flux during rice cultivation

The $CH_4$ flux in a continuously flooded plot of rice paddy field through a cultivated season in Japan showed a gradual increase after flooding with a maximum in the final stage of rice plant growth as shown in Fig. 1. Soil redox potential (Eh), a good indicator of oxidized or reduced conditions in soil, was closely correlated with the $CH_4$ flux. The mid-summer drainage, which is a typical water management performed for several days in summer by farmers of Japan, caused a drastic decrease of the $CH_4$ flux because the soil condition shifted to be oxidized. In rice paddy fields in Thailand, the another seasonal variation of $CH_4$ flux was observed that the $CH_4$ flux was already high within one month after transplanting, resulting from fast decomposition of organic matter in paddy soils which was flooded during about one month before transplanting.

(2) Diurnal variation of methane flux

The $CH_4$ flux from rice paddy fields, as shown in Fig.2, showed a clear diurnal variation with a maximum in early afternoon, and with a low and constant value during the nighttime, when the weather was fine and the difference of temperature in ambient air and soil between day and night was large in Japan and Thailand. The amplitude of the $CH_4$ flux in Thailand was, however, twice as much as that in Japan, and which was supposed to be the emission of $CH_4$ through bubbles from paddy soils due to higher temperature in Thailand.

(3) Methane emission rate in Japan and Thailand

In Ibaraki Prefecture of Japan where the Ryuhgasaki station is located, the $CH_4$ flux in a range of 5.9–44.8 $gCH_4/m2$ in one cultivated season, was measured in rice paddy fields with three different soil types and with four different applications of organic matter[2]. Using the data, the $CH_4$ emission rate from the rice paddy fields in Japan was estimated to be 0.26 $TgCH_4/year$. In Thailand, the $CH_4$ flux has been measured at 9 rice experiment stations in wet and dry seasons for these 5 years, as shown in Fig. 3. The $CH_4$ emission rate calculated from the flux data was shown in Table 1. Using the data, the $CH_4$ emission rate from rice paddy fields in Thailand was estimated to be 3.7 $TgCH_4/year$[3,4]. Because this value was calculated from the flux in the rice experiment station with the different management from farmers, we are now recalculating the $CH_4$ emission rate in Thailand, by measuring the biomass of rice plant in the rice paddy fields of farmers.

Since about 90 % of the total cultivated area of rice plant in the world is concentrated in Asia, many Asian countries have been recently measuring the $CH_4$ flux from rice paddy fields. India and China have the rice cultivated area of about 30% and 25% of the total area. The $CH_4$ emission rate in India and China was reported to be 4 and 15 $TgCH_4/year$, respectively, on the basis of their flux measurements from rice paddy fields. In Indonesia, The $CH_4$ emission rate was estimated to be 2–4 $TgCH_4/year$. IPCC[1] estimated the total emission rate in the world to be 60(20–150) $TgCH_4/year$, although it had a large uncertainty. Hence, to resolve the large differences among these emission rates, the more field measurements are expected to be made, and the development of a modelling of $CH_4$ emission is also expected, making clear the major factors controlling the $CH_4$ emission.

(4) Major factors controlling methane emission

Major factors have been listed up controlling the $CH_4$ emission, production and oxidation, such as soil chemistry, rice cultivar, root activity, water management, organic matter application, soil temperature, cultivation practice. In this study, the influence of soil chemistry and water management on $CH_4$ emission were investigated.

Takai et Al.[5] Already Defined, Reducing Capacity (Rc) as the Amount of Easily Decomposable Organic Matter (Or Available Nitrogen), and Oxidizing Capacity (Oc) as the Sum of Oxygen, Nitrate, Free Iron, Easily Reducible Manganese and Sulphate. The $CH_4$ emission is supposed to be high in the soil which has high reducing capacity and low oxidizing capacity. we found the excess reducing capacity of many paddy soils in the world calculated by chemical analyses, which equals to the difference between RC and OC, was positively correlated with the $CH_4$ production rate of soils obtained by incubation experiments as shown in Fig. 4. The correlation was not good, however, between RC and the $CH_4$ emission rate measured in the rice fields in the world. That strongly suggests there should be another important factor influencing the $CH_4$ emission from the rice paddy fields.

Because it is well known $CH_4$ is produced by bacteria under absolutely reduced conditions in soil, water management is also an important factor controlling the $CH_4$ emission. In Japan, mid-summer drainage in rice paddy fields, is usually performed by farmers, in which the paddy water is drained in the mid-stage of rice growth to activate the plant root by giving oxygen. Using the automated flux monitoring system, the detailed $CH_4$ flux was measured in detail at the two plots, one was continuously flooded through all the season, and the other was irrigated as farmers usually made. The result showed that the $CH_4$ flux in the mid-summer drainage, drastically decreased to be zero as shown in Fig. 5, and that the $CH_4$ flux after re-flooding was lower than in the continuously flooded plot. As a result, the $CH_4$ emission rate in the irrigated plot was lower by 45% than in the continuously flooded plot.

### (5) Modelling of methane emission

Rice plants are considered to play a role as conductance pipes for $CH_4$ transport from the rhizosphere to the atmosphere[6,7]. We assume that methane is emitted by molecular diffusion in rice plants by concentration gradient, and that the $CH_4$ flux F could be expressed as follows:

$$F=(Cs-Pa/H)D \qquad (1)$$

where Cs is the $CH_4$ concentration in the soil water; Pa is the $CH_4$ concentration in the atmosphere; H is the Henry's constant for CH4 solubility in water; and D is the conductance (the reciprocal of resistance) due to biomass of rice. In rice paddy fields, Pa is usually negligible compared with Cs. In the lysimeter experiment, we found a strong relationship between the conductance per shoot and air temperature at the time of day flux measured in the rice straw plot as shown in Fig. 6. Therefore, the equation (1) was modified as follows:

$$F=Cs\times(23\times Sn\times0.308\times\exp(0.126\times T)/1000 \qquad (2)$$

where Sn is shoot (tiller) number per hill, and 23 is hill number per $m^2$, and T is the air temperature. Using the $CH_4$ concentration measured in the soil water as Cs, the simulated $CH_4$ flux from the rice straw plot in 1994 is shown in Fig. 7, using the data of the conductance in the experimental regression equation(2). The observed $CH_4$ flux was nearly equal to the calculated $CH_4$ flux. The good relationship was also obtained in the mineral fertilizer plot.

### (6) Emission rate of nitrous oxide from different fertilizers

$N_2O$ flux was measured from the andosol soils filled in the lysimeter with the application of three different fertilizers, which were organic matter, ammonium sulphate, and slow-release fertilizer(super IB) every three days for one month. The maximum and lowest emission rates were found in the organic matter plot, and the plot of slow-release fertilizer, respectively. The ratio of emission rate in the organic matter plot to in the ammonium sulphate plot was about 15.

### (7) Emission of nitrous oxide and nitric oxide from ammonium and nitrate fertilizers

In an another lysimeter experiment, the flux measurement of NO and $N_2O$ was made at the plots with the application of two different fertilizers, ammonium sulphate and potassium nitrate[8]. The emission rate of NO-N to $N_2O$-N at both plots was in the range of 3-10 as shown in Table 2, indicating that NO was more emitted than $N_2O$ from nitrogen-fertilized soils. The emission rate of NO and $N_2O$ was higher in the ammonium plot than in the nitrate plot. Within a week after the application in the ammonium plot, the flux of $N_2O$ and NO reached maximum, and ammonium concentration in the soil decreased and nitrate concentration increased. These results indicate that the major process of producing $N_2O$ and NO from fertilized soils is through nitrification rather than denitrification.

### (8) Nitrous oxide emission from rice paddy fields during non-flooded period

The low flux of $N_2O$ has been measured from rice paddy fields during flooded period. During non-flooded period, however, no measurement was made of $N_2O$ flux from rice paddy fields, although rice paddy soils have a potential of emission of $N_2O$ through nitrification in oxidized condition. We have started to measure the $N_2O$ flux in addition to $CH_4$ flux, from rice paddy fields at the Ryuhgasaki station through all the seasons[8,9]. After the final drainage for harvest, $CH_4$ emission stopped and $N_2O$ began to emit, due to the decomposition of organic matter accumulated in the soils followed by nitrification. During non-flooded period of 8 months,$N_2O$ was continuously emitted until the day of re-flooding in the following spring as shown in Figs. 8 and 9. That means rice paddy fields emit $CH_4$ during flooded period, and $N_2O$ during non-flooded period, and the emissions of $CH_4$ and $N_2O$ are in a relationship of "trade off".

## 5. References

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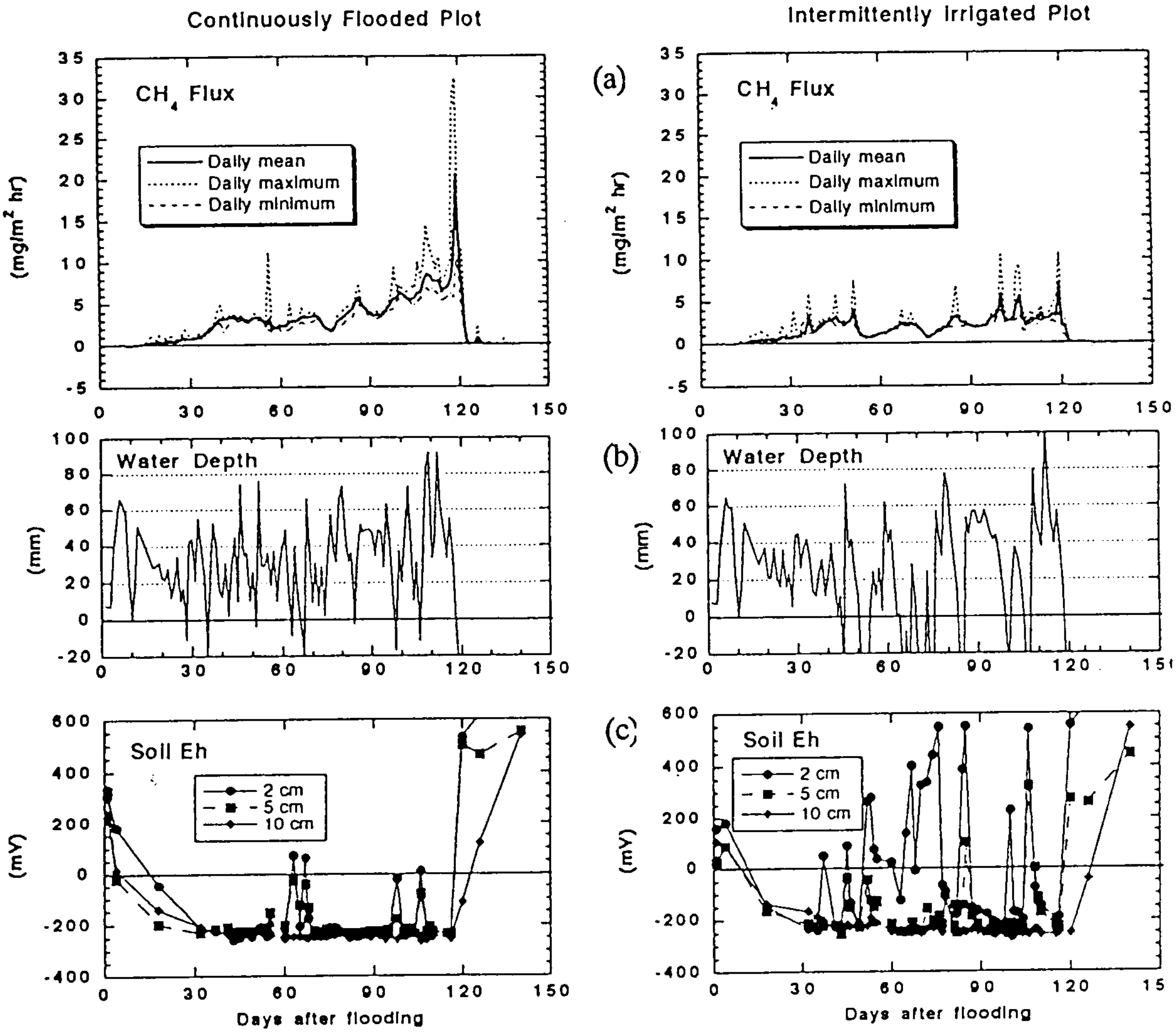

Fig. 1. Seasonal variation of (a)CH4 flux, (b)water depth, and (c)soil redox potential from the Ryuhgasaki rice paddy field in Japan measured by automated CH4 flux monitoring system. The left and right figures show the data from the continuously flooded plot and from the intermittently irrigated plot, respectively.

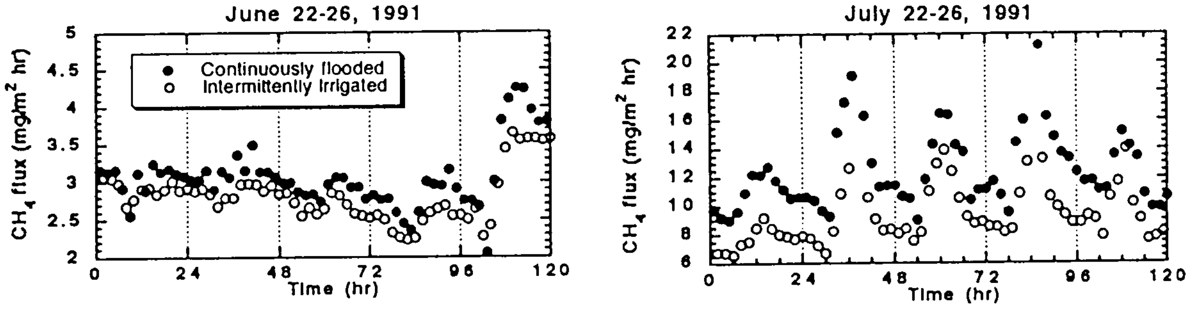

Fig. 2. Diurnal variation of CH4 flux in cloudy or rainy days, and (b) fine days measured by automated CH4 flux monitoring system at the Ryuhgasaki rice paddy field.

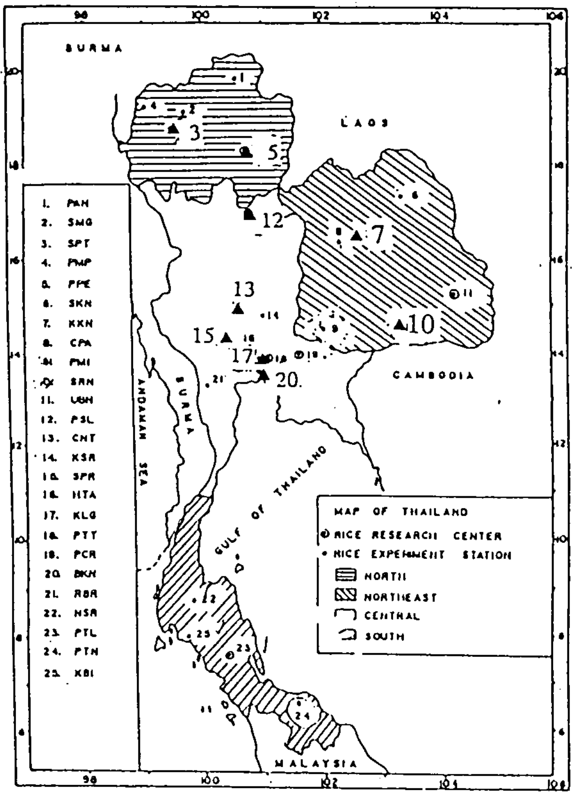

Fig. 3. Field sites of CH4 flux measurement from rice paddy fields in Thailand.

Table 1. CH4 emission rates in wet(W) and dry(D) seasons in the rice paddy fields of Thailand.

| | Location | Daily average (g/m2/day) | Flooding period (days) | Season total (g/m2) |
|---|---|---|---|---|
| | Thailand | | | |
| 15 | Suphan Buri (D) | 0.47 | 109 | 46 |
| | Suphan Buri (W) | 0.77 | 97 | 63 |
| 17 | Khlong Luang (D) | 0.09 | 83 | 6 |
| 13 | Chai Nat (W) | 0.04 | 94 | 3 |
| 20 | Bangkhen (W) | 0.49 | 98 | 48 |
| 12 | Phitsanulok (W) | 0.18 | 98 | 17 |
| | Phitsanulok (D) | 0.09 | 92 | 14 |
| 5 | Phrae (W) | 0.67 | 129 | 87 |
| | Phrae (D) | 0.40 | 127 | 51 |
| 3 | San Pa Thong (W) | 0.31 | 124 | 38 |
| | San Pa Thong (D) | 0.25 | 101 | 25 |
| 10 | Surin (W) | 0.32 | 129 | 41 |
| 7 | Khon Kaen (W) | 0.55 | 137 | 75 |

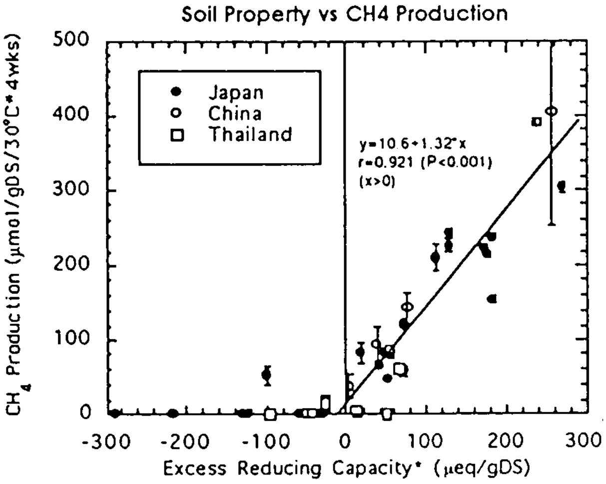

Fig. 4. Relationship between excess reducing capacity(ERC) and CH4 production rate in different paddy soils of the world by incubation experiment.

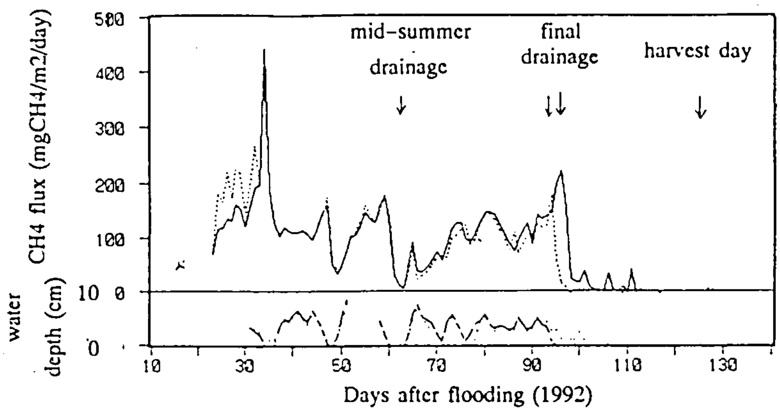

Fig. 5. Significant decrease of CH4 flux during mid-summer drainage for a week in 1992 measured by automated CH4 flux monitoring system at the Ryuhgasaki station.

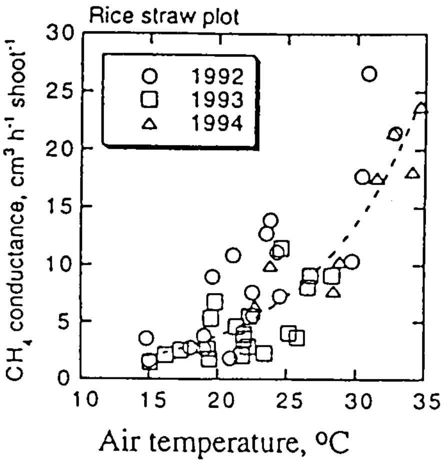

Fig. 6. Relationship between the conductance per shoot and air temperatude in the rice straw plot of the lysimeter for 3 years(1992, 1993 and 1994). Data were fitted by the following experimental curve: D=0.308exp(0.126T), r=0.86, n=46.

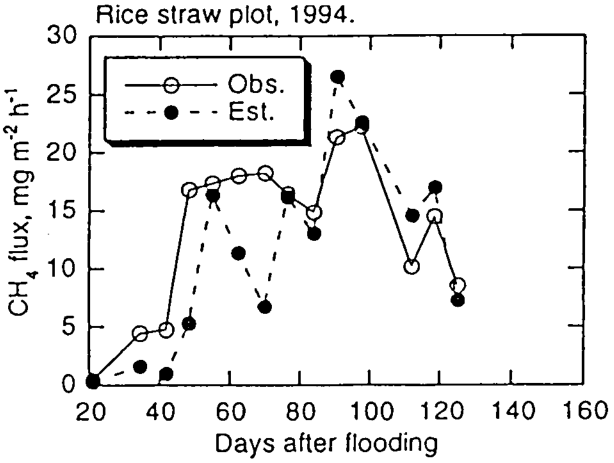

Fig. 7. Observed and simulated CH4 flux from the rice straw plot of the lysimeter in 1994. The simulated CH4 flux was calculated by the modified diffusion equation (5).

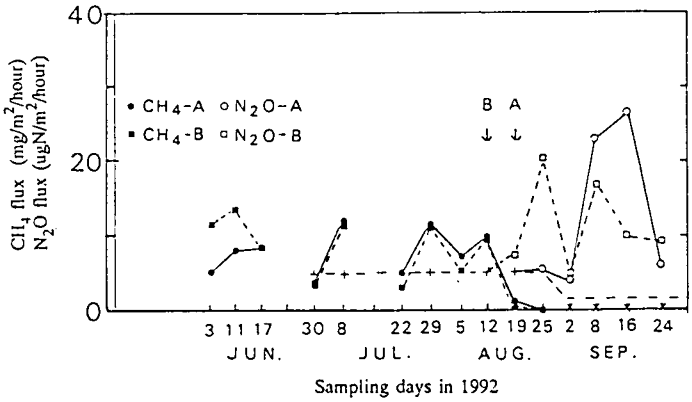

Fig. 8. N2O and CH4 fluxes from two plots(A, B) of the Ryuhgasaki rice paddy field. Two arrows indicate the days of final drainage. The broken line shows the detection limits of N2O flux, and x means the CH4 flux data below the detection limit.

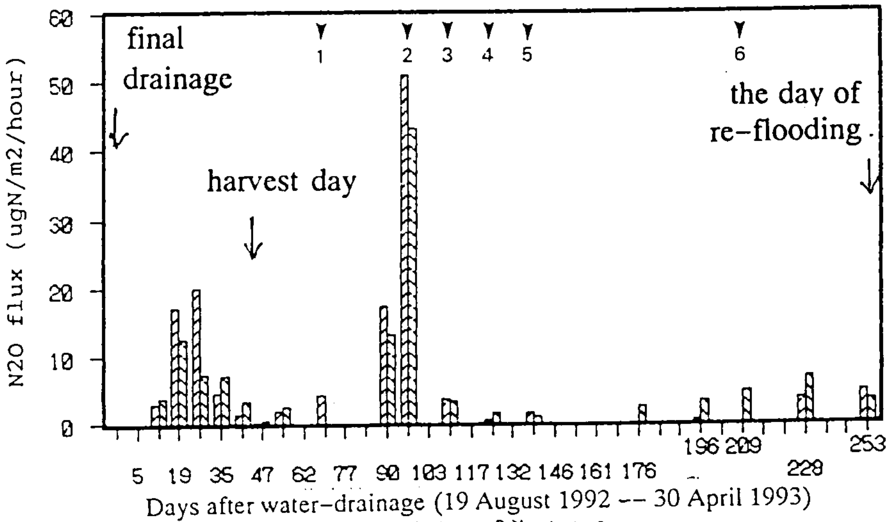

Fig. 9. N2O flux from the Ryuhgasaki rice field for 8 months between the day of final drainage in 1992 and the re-flooding day in the following spring of 1993.

Table 2. $N_2O$ and NO flux measurement from nitrogen-fertilized soils in the summer of 1992.

| Plot No. | N-fertilizer applied Type | (kgN/10a) | Total emission NO (mgN/m2) | Total emission $N_2O$ (mgN/m2) | NO-N / $N_2O$-N | (NO-N)+($N_2O$-N) / N-fertilizer (%) |
|---|---|---|---|---|---|---|
| 2 | $(NH_4)_2SO_4$ | 20 | 84.0 | 12.3 | 6.8 | 0.48 |
| 4 | $(NH_4)_2SO_4$ | 20 | 185.0 | 19.2 | 9.6 | 1.02 |
| Mean | | 20 | 134.5 | 15.8 | 8.5 | 0.75 |
| 1 | $NaNO_3$ | 20 | 18.7 | 5.84 | 3.2 | 0.12 |
| 3 | $NaNO_3$ | 20 | 7.8 | 0.28 | 27.9 | 0.04 |
| Mean | | 20 | 13.3 | 3.06 | 4.3 | 0.08 |