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Temporal patterns of methane emissions from wetland rice fields treated by different modes of N application

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Abstract. Methane emission rates from wetland rice fields were determined in Los Baños (Philippines) using an automatic system that allows continuous measurements over time. Methane emission was monitored in an irrigated Aquandic Epiaqualf planted to rice cultivar IR72. Urea fertilizer was applied using four modes: (1) broadcast 10 days after transplanting, (2) broadcast at transplanting, (3) broadcast and incorporated at final harrowing, and (4) deep placement as sulfur-coated granules. The treatments were laid out in a randomized complete block design with four replicates. Measurements were done in the 1991 wet season, 1992 dry season (four treatments), and the 1992 wet season (only treatment 3). Methane emission rates from the experimental plots showed pronounced seasonal and diel variations. The diel pattern of methane emission rates followed a consistent pattern, with highest rates observed in the early afternoon and lowest rates in the early morning. Methane emission rate was generally highest at the ripening stage. The average methane emission rate during the 1992 dry season (190 mg CH_4 m^{-2} d^{-1}) exceeded the average flux rates of the 1992 wet season (79 mg CH_4 m^{-2} d^{-1}) by a factor of 2.4. The total methane emitted from these flooded rice fields amounted to 19 g CH_4 m^{-2} in the dry season with rice yields of 5.2–6.3 t ha^{-1} and 7 g CH_4 m^{-2} in the wet season with rice yields of 2.4–3.3 t ha^{-1} regardless of the mode of N application. Significant amounts corresponding to 20% of the methane released under waterlogged conditions were released when the soil was drained after harvest. Emission rates increased sharply when the floodwater receded and macropores started to drain. Emission of methane stopped only when the soil became fully aerated.

Introduction

The tropospheric abundance of methane has been steadily increasing in the past decades. At present the mixing ratio of methane is described to vary from approximately 1.7 parts per million by volume (ppmv) in the northern hemisphere to approximately 1.6 ppmv in the southern hemisphere [*Rasmussen and Khalil*, 1986]. These levels are roughly 3 times higher than the concentration 100 years ago [*Craig and Chou*, 1982]. Because of the long atmospheric residence time and the high-infrared absorption capacity, the warming efficiency of CH_4 is 20–32 times that of CO_2 [*Blake and Rowland*, 1988]. In addition to the radiative forcing, methane is an important agent for various chemical reactions in the troposphere as well as for the stratospheric cloud formation in polar regions [*Rowland*, 1988].

Since the first records of the increase of tropospheric methane, the causes have been intensively discussed. It has been suggested that the increase is due to a decline of the main sink mechanism in the troposphere initiated by reaction with OH radicals. However, a reduction in OH radicals should result in a simultaneous increase in tropospheric concentrations of other trace gases. Such relationship could not be verified in empirical studies [*Seiler*, 1985]. Therefore the elevated CH_4 concentration in the atmosphere is likely to be caused by increased methane release from different sources.

The total annual emission of CH_4 is estimated to be 505 (± 105) Tg CH_4/yr [*Crutzen*, 1991], 70–80% of which is of microbial origin [*Cicerone and Oremland*, 1988]. Methanogenic bacteria are active in anaerobic environments which include wetland soils. The source strengths of anaerobic environments increased in recent decades due to the expansion of wetland rice areas [*Bouwman and Sombroek*, 1990; *Schütz et al.*, 1990]. Global extrapolations of emission rates from wetland rice fields based on few reported measurements imply great constraints [see *Wassmann et al.*, 1993a]. These extrapolations are also hampered by uncertainties regarding the influence of rice cropping and management practices and the temporal and spatial variability of CH_4 emissions [*Rennenberg et al.*, 1992; *Neue and Roger*, 1993]. A previous field study in Italy demonstrated the influence of the fertilizer application mode on methane emission [*Schütz et al.*, 1989], but this factor has not yet been investigated under conditions in the tropical rice-growing areas of Asia.

This study was aimed (1) to provide the first CH_4 emission data from wetland rice fields in Southeast Asia, which is one of the most important rice-growing regions, and (2) to

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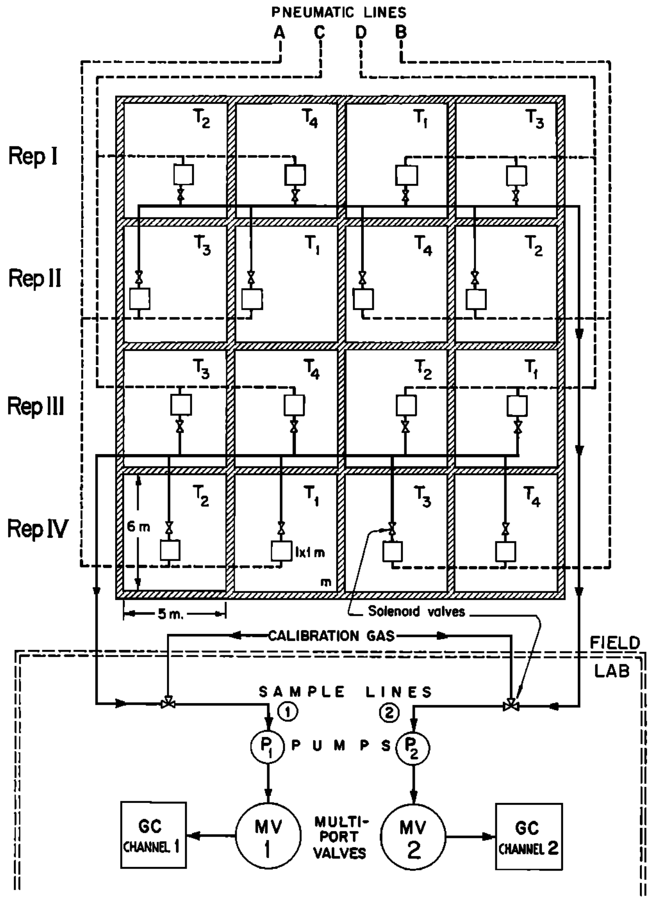

Figure 1. Field design (Rep, replicate; T, fertilizer treatment) and schematic sketch of sample and pneumatic lines.

determine the influence of different agricultural practices on methane flux rates. Special emphasis was given to four modes of basal nitrogen application: three different timings of surface application of urea and deep placement of urea granules coated with sulfur.

A further objective of this field study was to determine methane emission after harvest in a dry fallow rice-rice system. Significant amounts of methane are entrapped in the soil when the field falls dry at harvest. Previous measurements were limited to the flooded growing seasons or the sequence of measurements was not appropriate to monitor the fate of the entrapped methane.

Method and Site Description

Methane emission rates were determined by an automatic measurement system based on the "closed chamber technique" in rice fields at the Research Farm of the International Rice Research Institute, Philippines. The principles of the sampling and analytical procedure were given by *Schütz et al.* [1989]; technical details of the system used in these measurements were described by Wassmann (in *International Atomic Energy Agency* (IAEA) [1992]). The sampling device consists of two components (Figure 1): (1) a pump-driven air transfer from the chambers to a multiport valve through a tube/valve system and (2) an aliquot transfer from the sample loop mounted to the multiport valve to the gas chromatograph (GC). The analytical components are a gas chromatograph equipped with two FID channels and a two-channel integrator. The system was calibrated using gas samples from a tank filled with compressed air of a known CH_4 concentration which was connected to the sample loop.

Two sampling units consisting of eight chambers each were set up. Both units were subdivided into four pairs of boxes that could independently be opened and closed by the pneumatic lines A, B, C, and D (Figure 1). One pair of chambers was closed for a 24-min interval, facilitating four measurements of the methane mixing ratio inside each chamber. The flux rates were derived from the temporal increase of the mixing ratio [e.g., *IAEA*, 1992]. Sampling of gases from the chambers was conducted in a constant 2-hour cycle with a persistent sequence of operations which included three calibrations and one determination of emission rates for each box. All operations were triggered by a computer equipped for time control functions; another computer was used for the automatic data acquisition. The individual CH_4 emission rates were computed by regression analysis using the four readings obtained during each closing period.

The dimensions of the 16 gas collector boxes were 1 m × 1 m basal area and 1.2-m height. The four basal corners of the gas collector boxes were placed and fixed in the rice field on PVC tubes (50-cm length, 15-cm diameter) that had been sunk vertically into the soil prior to flooding and remained at the same position during the entire vegetation period. The floodwater separated the volume of the closed boxes from the ambient atmosphere, but the water could exchange with the surrounding water body. Floodwater was maintained at 5- to 7-cm depth during the whole growing seasons. During phases of receding floodwater and upland conditions after harvest, the boxes were pressed into the soil to achieve a basal sealing.

Methane emission was monitored in an irrigated rice field planted to cultivar IR72. The soil was an Aquandic Epiaqualf. For land preparation the field was flooded 14 days before transplanting. The four N fertilizer treatments were (1) broadcasting 10 days after transplanting, which is mostly practiced by local farmers; (2) broadcasting of prilled urea into floodwater at transplanting which some farmers prefer; (3) broadcasting and incorporation of prilled urea to the wet soil without standing water which is recommended to reduce N losses due to volatilization of NH_3; and (4) deep placement (5-cm depth) of sulfur-coated urea granules at transplanting which represents an advanced technique to achieve high fertilizer efficiency (granules are not yet commercially available). The experimental plots were laid out in a randomized complete block design with four replicates (Figure 1).

Results

Fertilizer Treatments

Grain and straw yields were significantly higher in the dry season but only marginal deviations occurred between yields from the different treatments of a given season. The rice yields varied between 5.2 and 6.3 t ha^{-1} in the dry season and between 2.4 and 3.3 t ha^{-1} in both wet seasons.

The effect of the fertilizer treatments on methane fluxes during the 1992 wet season is shown in Figure 2. The observation period started 22 days after transplanting and ended 104 days after transplanting. Methane emissions increased during the growing season, with highest rates observed during the maturity stage. In the 1992 dry season,

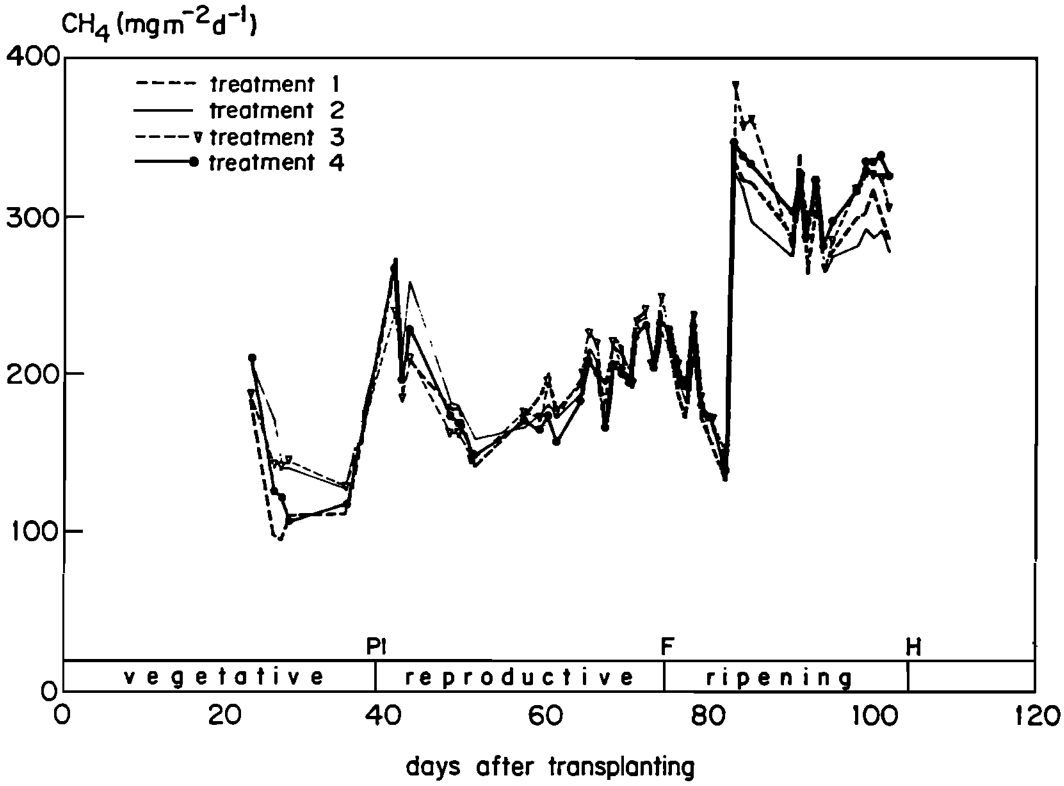

Figure 2. Methane emission rates (mg CH_4 m^{-2} d^{-1}) during the 1992 dry season from four different fertilizer treatments (see text). PI, panicle initiation; F, flowering; H, harvest.

methane emissions were higher and showed pronounced peaks at the early and late growth stage. The pattern and rates of methane emission were not significantly different among N fertilizer treatments.

Seasonal Variation and Annual Emission

Since application modes of N fertilizer did not affect the pattern and rates of methane fluxes, this discussion on seasonal variations focuses only on one treatment. Treatment 3 was chosen because the incorporation of urea is not affected by N volatilization, by harrowing reduces N losses, as compared to broadcasting. Results available for this treatment comprise emission rates of three vegetation periods (Figure 3). The 1991 wet season and the 1992 dry season lack records for the initial 42 and 21 days, respectively. A complete record of emission rates was accomplished for the 1992 wet season. The emission rates of the three seasons shown in Figure 3 show pronounced seasonal differences. The average methane emission rate during the 1992 dry season (190 mg CH_4 m^{-2} d^{-1}) exceeded the average flux rates of the 1992 wet season (79 mg CH_4 m^{-2} d^{-1}) by a factor of 2.4.

In the 1992 wet season, diel mean emissions were lower than 40 mg CH_4 m^{-2} d^{-1} during the vegetative stage and the initial reproductive stage. Emission rates increased to 70 mg CH_4 m^{-2} d^{-1} at the middle of the reproductive phase. Emission rates at the first half of the 1992 wet season were similar to fluxes of the 1991 wet season. During the second half of the 1992 wet season, emission rates showed pronounced fluctuations and reached diel means of 240–390 mg CH_4 m^{-2} d^{-1} during the last two weeks before harvesting.

Emission rates during the 1992 dry season were generally higher than corresponding rates of the wet seasons and varied in the range of 150 to 240 mg CH_4 m^{-2} d^{-1}. Maximum methane emission (390 mg CH_4 m^{-2} d^{-1}) in the 1992 dry season occurred during the ripening stage, and it remained on a high level (>270 mg CH_4 m^{-2} d^{-1}) until harvest.

Figure 4 shows cumulative methane emission for the dry and wet season in 1992 until harvest. Results for the 1991 wet season were omitted in Figure 4, because only 60% of the vegetative period was recorded. The inset in Figure 4 displays the approximation conducted for the methane emis-

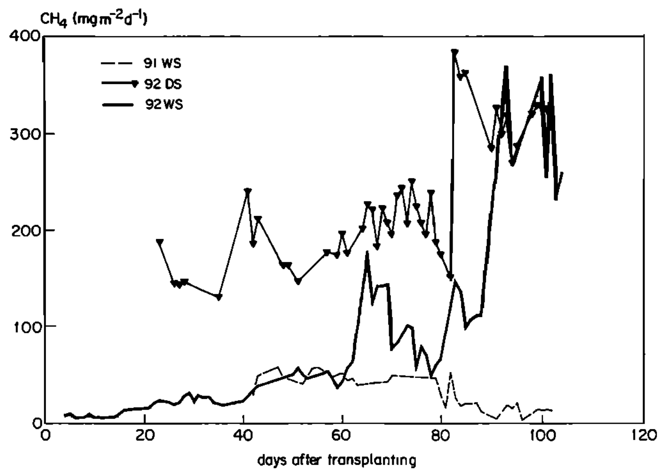

Figure 3. Methane emission rates (mg CH_4 m^{-2} d^{-1}) during the 1991 wet season, 1992 dry season, and 1992 wet season, as recorded for treatment 3 (broadcasting and incorporating at final harrowing).

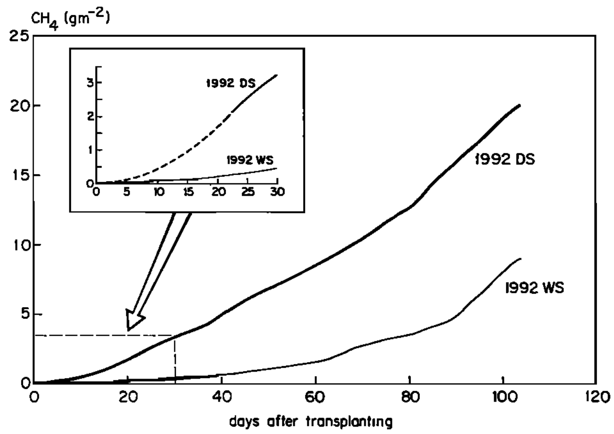

Figure 4. Cumulative methane emission (g CH_4 m^{-2}) in the 1992 dry and wet seasons, as recorded for treatment 3 (broadcasting and incorporating at final harrowing).

sion during the first 21 days of the 1992 dry season when data were missing. In the 1992 dry season, cumulative methane emission rates rapidly increased over the season. In the 1992 wet season, methane emission was considerably lower until 90 days after transplanting. The slope of cumulative methane emission was similar to that in the dry season only during the last 15 days until harvest. The total methane emission for the growing period was 19.2 g CH_4 m^{-2} in the dry season and 9 g CH_4 m^{-2} in the wet season. The mean soil temperatures (at 5-cm depth) in these two seasons were 26.3°C (dry season) and 27.6°C (wet season); the average rainfall was 0.9 and 9.3 mm per day during dry and wet season, respectively (Table 1). The mean values for solar radiation in 1992 were 515 and 441 mJ d^{-1} in dry and wet season, respectively; the differences between both seasons are more apparent by comparing the minimum values indicating periods with very low solar radiation during the wet season (Table 1).

Postharvest Emission

The postharvest release of methane was investigated in the 1992 dry season. At harvest (104 days after transplanting), rice plants were cut 10 cm above the soil surface and irrigation was terminated. The floodwater vanished from the rice field within 6 days after harvesting due to evaporation and seepage. Figure 5 shows the emission rates of treatments 3 and 4 during the last 4 days before harvesting and the following 12 days after harvest. The patterns of methane emission rates were almost identical. Similar results were obtained for treatments 1 and 2 (data not shown). Each data point in the diagram (Figure 5) represents the average value of the emission rates obtained during a single 2-hour cycle on one plot. The temporal resolution clearly reveals the typical diel pattern of emission rates. Prior to harvest, emission rates were in the range between 5 and 30 mg CH_4 m^{-2} h^{-1}. Diel patterns and rates of methane fluxes remained the same until 6 days after harvest. Thereafter, emission rates sharply increased up to 90 mg CH_4 m^{-2} h^{-1} at 8 days after harvest. The postharvest flush of methane emission can be attributed to the escape of methane previously entrapped in the soil. The postharvest flush of methane release coincided with the proceeding aeration of soil macropores after the floodwater receded. In Figure 5 this period of high emission rates is depicted by a peak with the maximum value in the central part and two ripple-shaped vertices at both flanks. These two structures at the flanks as well as the maximum value were recorded in the early afternoons which corresponded to the diel maximum of methane emission rates during the preceding periods. Postharvest emission from the soil became zero when the soil became oxidized 11 days after harvest.

Discussion

Modes of Fertilizer Application

In this study, nitrogen was added to the soil as urea in different application methods and at different times of the growing season. Neither the modes of application nor the timing of basal N fertilization had an impact on the methane emission from rice fields in the experiment. Figure 2 shows the high degree of similarity of the emission rates from all four treatments obtained during the 1992 dry season.

Modes of fertilizer applications were previously compared by *Schütz et al.* [1989]. Compared to a nonfertilized field, they found that deep placement of prilled ammonium sulfate reduced methane emission by 62%, incorporation into the soil by 43%, and surface application by 6%. The reasons for the discrepancy between results obtained by *Schütz et al.* [1989] and results of this study are probably due to the different N sources used, i.e., ammonium sulfate versus urea.

Nitrogen fertilizer is commonly applied in two doses: the first dose applied at the early vegetative growth and the second dose during panicle initiation. This is done to adjust N supply to plant demand and to achieve high N uptake efficiency [*De Datta*, 1981]. The effect of a second nitrogen dose could not be investigated in this study. The similarities between emission rates in the four treatments and during the reproductive stage irrespective of season indicate that nitrogen application after the vegetative stage does not seem to seriously affect methane emission. Topdressing of N fertilizer at panicle initiation increases grain yield but not significantly total biomass, which seems to control methane emission at the reproductive stage.

Table 1. Soil Temperatures (at 5-cm Depth), Rainfall, and Solar Radiation During 1992 Dry Season (DS) and Wet Season (WS)

	Temperature, °C		Rainfall, mm d^{-1}		Solar Radiation, mJ m^{-2}	
	DS	WS	DS	WS	DS	WS
Mean	26.3	27.6	0.9	9.3	515	441
Minimum	22.9	25.3	0.0	0.0	150	63
Maximum	29.8	29.8	37.8	24.0	695	754
s.d.	1.4	0.8	4.7	16.1	121	154

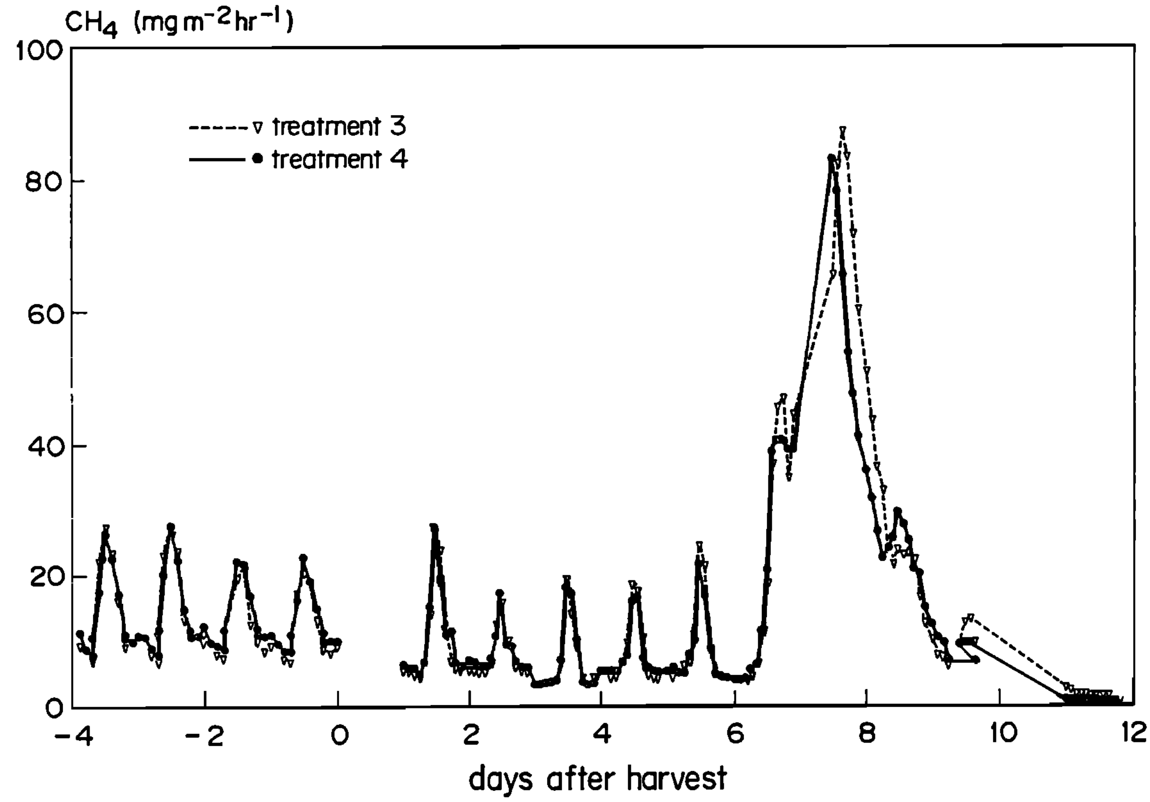

Figure 5. Methane emission rates (mg CH_4 m^{-2} h^{-1}) during the preharvest and postharvest periods of the 1992 dry season, as recorded for treatments 3 (broadcasting and incorporating at final harrowing) and 4 (deep placement of sulfur-coated urea granules).

Seasonal Variation

No explanation can be given at present to the considerable differences of methane emission during the later stages of the vegetation periods between the two wet seasons (Figure 3). Higher methane emission during the dry season could be linked to interseasonal differences in plant growth. The rice yields of the dry season (5.2–6.3 t ha^{-1}) were twice as high as these obtained in the wet season (2.4–3.3 t ha^{-1}), whereas the cumulated methane emission in the dry season exceeded that in the wet season by a factor of 2.4. These results are consistent with the findings of *Sass et al.* [1990] who showed that the aboveground biomass of rice crops are highly correlated to methane emissions. This correlation of both parameters could either be caused by (1) a direct stimulation of methane emission by a high plant biomass, (2) a simultaneous response of plant growth and methane emission on the same external factors (e.g., temperature), or (3) a combination of both mechanisms. The dry season is characterized by high temperatures and strong solar radiation in the later growth stages of the rice plants, which may exert a stimulating effect on both rice yield and methane emission rates. In the wet season these plant stages generally coincide with relatively low temperatures and low solar radiation. A positive correlation between temperature and methane emission rates was shown in several field studies [*Schütz et al.*, 1989; *Butterbach-Bahl*, 1992].

The seasonal averages of methane emission rates in the rice fields at this experimental site in the Philippines amounted to 190 mg CH_4 m^{-2} d^{-1} (1992 dry season) and 79 mg CH_4 m^{-2} d^{-1} (1992 wet season). A compilation of emission rates [*Wassmann et al.*, 1993a] reveals higher emission rates at most other sites. However, mode, source, and quantity of fertilizers applied in the various field studies on methane emissions differed considerably or were not completely reported. Organic amendments, for example, stimulate methane production and, subsequently, increase methane emission significantly [*Yagi and Minami*, 1990]. Variations in soil types, rice cultivars, fertilization, and soil as well as water management represent a major constraint for comparison of data obtained in different field studies [e.g., *Wassmann et al.*, 1993d]. Direct comparison of methane flux rates are only sound if treatments are similar. Such an approach was previously realized in several locations in India [*Parashar et al.*, 1991] and in the Chinese province of Hunan [*Wassmann et al.*, 1993c]. The mean emission rates from the inorganic fertilizer treatments in India varied from 38 to 463 mg CH_4 m^{-2} d^{-1} [*Parashar et al.*, 1991]. In Hunan Province, 156 and 343 mg CH_4 m^{-2} d^{-1} were recorded for the mean emission during the early and late growing seasons of 1991, respectively [*Wassmann et al.*, 1993c]. Methane emission rates recorded here for the Philippines are within the range of other records available from other tropical sites.

Postharvest Emission

The results from this study showed that considerable amounts of methane can be emitted after harvest when the field fell dry (Figure 5). The similarity of methane emission of the preharvest and the initial postharvest phase indicates that removing most of the aboveground biomass and dying roots do not immediately limit methane fluxes. Two deductions on the mechanisms of vertical methane transport from the soil can be derived from this finding: (1) at later growth stages, plant-mediated transport of methane clearly dominates over diffusion in aqueous media and ebullition and (2) at a given growth stage, methane flux rates through rice plants are not controlled by overall stem and leaf properties. Both findings are in accordance with results from previous investigations that revealed the significance of plant-mediated transport [*Holzapfel-Pschorn et al.*, 1986; *Miura et al.*, 1992; *Butterbach-Bahl*, 1992] and identified the root-stem transition as the plant compartment that controls the flux rates of the vertical gas transfer [*Butterbach-Bahl*, 1992]. The diel pattern of emission rates obviously proceeded even without significant photosynthetic activity and was still pronounced during the methane flush after harvest (Figure 5).

Postharvest flush of methane lasted for 3 days and can be attributed to the release of soil-entrapped methane. This phenomenon will probably occur mainly in soils with high amounts of entrapped methane. The flush of methane coincided with the initial aeration of soil macropores. The observation of the postharvest flush indicate that drying periods during and after the growing season of wetland rice have to be taken into account to achieve accurate local methane budgets. Previous measurements were restricted to waterlogged conditions of rice fields and omitted postharvest periods [e.g., *Schütz et al.*, 1989; *Wassmann et al.*, 1993b; *Khalil et al.*, 1990]. In Los Baños the methane emitted in the postharvest period amounted to 20% of the methane emitted during the waterlogged period.

Provided the intense postharvest release of methane proves to be a general phenomenon in rice fields, previous extrapolations based on waterlogged conditions would represent an underestimation of the virtual emission computed over the entire annual cycle.

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