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Seasonal Variation of Methane Flux From a California Rice Paddy

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To allow increased understanding of the global budget of atmospheric methane, individual methane sources require investigation. We have measured methane emissions from a California rice paddy during the entire 1982 growing season. A very strong seasonal dependence was observed. Methane emissions were highest in the last 2–3 weeks before harvest; daily emissions reached 5 g CH_4/m^2. Over the 100-day season, daily emissions averaged about 0.25 g CH_4/m^2, higher than our previously reported values. Attempts to estimate global rice paddy emissions must recognize the possibility of seasonal variations. Soil temperature at 10-cm depth correlated poorly with our measured fluxes; soil redox potential was a more reliable indicator.

INTRODUCTION

Deducing the identities and sizes of the sources of atmospheric methane has been a subject of interest to atmospheric chemists and geochemists for about 20 years. Carbon 14 data reviewed by *Ehhalt and Schmidt* [1978] demonstrate that over 80% of atmospheric methane is biogenic. Of this biogenic methane, 33–49% was attributed to release from the world's rice paddies. In making these estimates, Ehhalt and Schmidt were forced to employ estimates of methane emission rates from laboratory incubations of rice paddy soil [*Koyama*, 1963, 1964]; no field measurements were available. The first such measurements in rice paddies were reported by *Cicerone and Shetter* [1981]. They found lower methane emission rates than Koyama measured in the laboratory. Cicerone and Shetter also reported that the principal means of escape of methane is through the rice plants and not through diffusion or escape of bubbles across the air-water interface and that nitrogen fertilization rates affect the methane escape.

Questions about methane sources have acquired new importance now that it has been shown that atmospheric methane concentrations are increasing globally [*Rasmussen and Khalil*, 1981; *Blake et al.*, 1982; *Craig and Chou*, 1982; *Ehhalt et al.*, 1983]. Thus it has become important to quantify the individual methane sources and the temporal changes in these sources, including the most recent suggestions of potentially significant sources, for example, termites [*Zimmerman et al.*, 1982; *Rasmussen and Khalil*, 1983] and biomass burning [*Crutzen et al*, 1979].

In this paper we present new field measurements of methane emissions from rice paddies. The data show strong dependence of the methane emission rate on time elapsed in the growing season and an apparently complicated dependence on nitrogen fertilization rate, factors that must be recognized in any attempt to extrapolate to global emission rates.

EXPERIMENTAL PROCEDURES

The experimental rice paddy is located on the grounds of the University of California, Davis (40.2°N, 122.1°W). The rice (*oryza sativa, cultivar* M 101) was planted by drilling in rows 25 cm apart on May 11, 1982. The field (area about 0.5 ha) was irrigated as needed to maintain water depth at about 11–15 cm through the growing season, and the soil was a vertisol (Capay clay). Temperatures and other soil and water conditions are given below. Prior to planting, the field received an application of ammonium phosphate–ammonium sulfate (16-20-0) at the rate of 220 kg/ha. An additional top dressing of urea was applied at the time of planting at the rate of 166 kg/ha for a total of 113 kg N/ha. A separate subplot (P1) received an additional application of 166 kg/ha of urea (77 kg N/ha) on July 1, 1982. Water inflow was stopped on September 28, and the remaining water was drained on October 2, 1982.

Field sampling was performed with a saran bag collector with stainless steel tubing and flasks as described by *Cicerone and Shetter* [1981]. The collectors were placed over the rice plants with the lower rim into the water surrounding the rice, and gas samples were extracted from inside the collectors after 15 min. All other analytical procedures were as those in the work of *Cicerone and Shetter* [1981] except that methane concentrations were determined by peak area (HP3390 miniintegrator) rather than by peak height. During each such collection for flux measurements, we also measured temperatures of ambient air, air inside the collector, soil (at 10-cm depth), and rice height. Soil redox potential, E_h, was measured by means of a platinum electrode referred to a calomel half cell.

We attempted to place the collectors over equal numbers of rice plants, but because counting stalks was difficult physically and because we sought to avoid bending and jostling the rice stalks, this goal was not attained reliably.

RESULTS AND DISCUSSION

Dates and times of methane flux measurements are shown in Table 1 along with soil temperatures (at 10-cm depth), soil redox potentials, rice heights, and the methane fluxes themselves. Data are shown for each of the two subplots, P1 and P2. Figure 1 shows the methane fluxes graphed versus time during the 1982 growing season.

A dramatic variation of the methane emission rate during the growing season is apparent in Figure 1. Although chamber methods for flux measurements such as ours are not guaranteed to yield absolute accuracy, relative measurements over an

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TABLE 1. Dates and Results of Methane Flux Measurements From Rice Paddy Near Davis, California, in 1982

				P1		P2	
Date	Time	Soil Temperature, °C	E_h, mV	Rice Height, cm	Flux, g CH_4/m^2 d	Rice Height, cm	Flux, g CH_4/m^2 d
June 16	1150				$<10^{-3}$		$<10^{-3}$
June 23	1210		275		$<10^{-3}$		$<10^{-3}$
July 1	1210	19.5	−134		1.2 (−3)		$<10^{-3}$
July 8	1030	20	−166		4.28 (−3)		2.81 (−3)
July 9	1100				7.15 (−3)		5.70 (−3)
July 14	1200	26	−156	50	1.57 (−2)	50	1.11 (−2)
July 21	1210	20		58	2.23 (−2)	58	1.04 (−2)
July 28	1210	23	−161	67	6.83 (−2)	67	2.41 (−2)
Aug. 4	1215	23	−176	70	6.52 (−2)	68	1.43 (−2)
Aug. 18	1140	23.5	−181	73	2.26 (−2)	70	9.41 (−3)
Sept. 8	1140	23	−191	80	6.35 (−1)	75	6.04 (−1)
Sept. 16	1130	20		81	9.16 (−1)	76	4.73 (−1)
Sept. 22	1330	23		82	1.96 (−1)	77	1.02
Sept. 26	2030	21	−141	82	2.81	78	4.94
Sept. 27	1600	18	−241	82	2.47	78	4.24
Sept. 28	0430	14	−226	82	1.72 (−1)	78	5.71 (−1)
Sept. 28	1130	17	−221	82	1.78 (−1)	78	2.87 (−1)
Sept. 28	1430	17	−236	82	1.86 (−1)	78	2.18 (−1)
Sept. 28	2000	18	−216	82	1.23 (−1)	78	1.63 (−1)
Sept. 29	0530	13.5	−206	82	7.14 (−2)	78	1.41 (−1)
Sept. 29	1735	15.5	44	82	7.40 (−2)	78	1.30 (−1)
Sept. 30	0200	15	44	82	5.30 (−2)	78	1.07 (−1)
Sept. 30	1130	15	94	82	3.07 (−2)	78	4.58 (−2)

Soil temperatures, redox potentials, and rice heights are also shown. Subplot P1 was more heavily fertilized than subplot 2 (see text). Numbers in parentheses indicate powers of 10; for example, read 1.2(−3) as 1.2×10^{-3}.

entire season should be valid. For both P1 and P2 fluxes were highest late in the season. The total amounts emitted between June 23 and September 30 (100 days) were 28 g CH_4/m^2 and 21.8 g CH_4/m^2 for P2 and P1, respectively. For both subplots, most of the total flux was emitted during September: 96% for P2 and 88% for P1. Note also that the more heavily fertilized subplot, P1, showed a higher methane flux through July, August, and early September, but because P2 emitted more methane September 10–30, the total emission from P2 exceeded that of the more heavily fertilized P1. Because our data show such a large variation of methane flux with time of growing season, the daily average methane flux is not a particularly meaningful figure. Our largest fluxes, 2.5 to 5 g CH_4/m^2 d, are 14 to 27 times those (0.18 g CH_4/m^2 d) reported by *Cicerone and Shetter* [1981]. Averaged over 100 days, the daily emission rate was 0.28 g and 0.22 g CH_4/m^2 d for P2 and P1. These figures can be compared with the *Cicerone and Shetter* [1981] value of 0.18 g CH_4/m^2 d.

Diurnal variations of the methane flux appear to be insignificant. For the 5-day period September 26–30, the average ratio of the measured daytime flux to that at night for each date is 1.01 ± 0.34 (one standard deviation) for P1. For P2 it is 0.81 ± 0.37. When we take each individual day/night ratio from these studies and those of *Cicerone and Shetter* [1981], a total of 14 points, we obtain a day/night ratio of 1.00 ± 0.43.

These data demonstrate several facts and they raise several important questions. First, it is clear that there is the potential for a strong seasonal dependence of methane release rates in all rice paddies; attempts to determine emissions from paddies of other world regions must recognize this. But is our seasonal pattern (Figure 1) typical of other regions or even of California, and why is there such a strong variation? The studies of *de Bont et al.* [1978] are relevant here. They showed, even before *Cicerone and Shetter* [1981], that the presence of rice plants enhances the escape of methane from soil. We interpret one of their experiments as showing that older rice plants (at the ripening stage) released about 20 times more soil methane than 2-week-old rice seedlings did (this conclusion assumes that the initial soil methane concentrations were comparable). Is the increase of methane flux with elapsed time in growing season due to a buildup of soil methane or instead to the

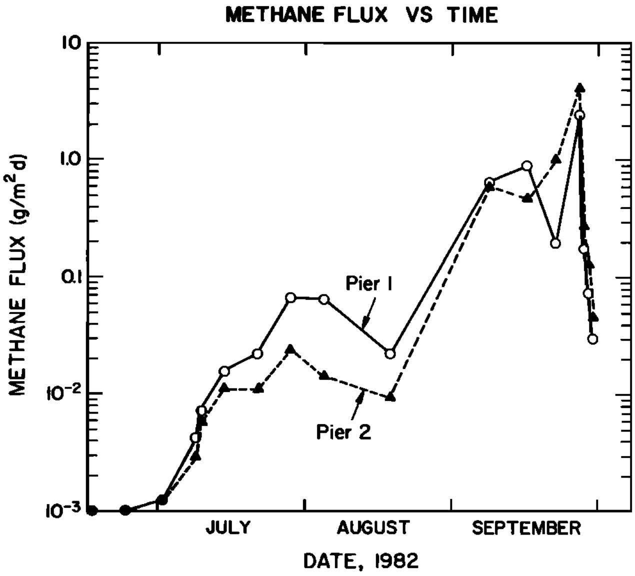

Fig. 1. Methane fluxes from two subplots, P1 and P2, of a rice paddy near Davis, California, during the 1982 growing season. Fertilization rates and dates and information on irrigation are in the text. Individual data points and related data are shown in Table 1. Note logarithmic scale.

effects of maturation of rice? Does fertilizer accelerate the growth of roots and allow the cortex of roots to conduct gases earlier in the season? This last question relates to our finding that nitrogen fertilizer appears to stimulate methane release rates during early and mid-season but does not influence the cumulative amount of methane released over the entire growing season. Clearly, more data are needed to determine the range of N fertilizer effects.

Further questions about the mechanism of methane escape and how to extrapolate field data to global conditions are apparent when one notices the lack of any clear correlation between methane flux and soil temperature at 10-cm depth (Table 1). Note, for example, that methane fluxes rose 10–100 fold during July and August with soil temperatures always near 23°C. On the contrary, *Seiler et al.* [1983] have found a positive correlation between methane emissions and soil temperature at 0.5-cm depth. It is important to determine if methane emissions to the atmosphere, like methane production in soils, rise with soil temperature at some specified soil depth.

Table 1 does show a positive correlation between soil E_h and methane emissions, with largest fluxes when $E_h \lesssim -190$ mV. The onset of methanogenesis for $E_h \lesssim -200$ mV has been observed previously in flooded soils; methanogenesis appears to follow the sequential usage of oxygen, ferric ion, nitrate, and sulfate [*Mah et al.*, 1977].

Thus the question of the fraction of atmospheric methane due to releases from worldwide rice agriculture is far from settled. Our 100-day averages for methane fluxes of 0.28 and 0.22 g CH_4/m^2 d are 56% higher and 21% higher than the values reported by *Cicerone and Shetter* [1981]. Accordingly, one might extrapolate our 1982 California fluxes to proportionately higher global annual emissions of CH_4 from the world's rice paddies than the *Cicerone and Shetter* [1981] estimate. Even though our 1982 data set is much more complete than that of *Cicerone and Shetter* [1981], we are reluctant to present an estimate of a global emission rate because of many remaining uncertainties such as the effects mentioned above and the effects of varying agricultural practices and because of variable organic contents of soils.

Finally, we note that fall maxima have been observed in atmospheric methane concentrations [*Khalil and Rasmussen*, 1983]. As these authors have discussed, seasonal variations in methane sinks and sources are likely to be involved, but our observed methane emission pattern can partially explain the fall maximum in methane concentration.

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REFERENCES

Blake, D. R., E. W. Mayer, S. C. Tyler, Y. Makide, D. C. Montague, and F. S. Rowland, Global increase in atmospheric methane concentrations between 1978 and 1980, *Geophys. Res. Lett.*, *9*, 477–480, 1982.

Cicerone, R. J., and J. D. Shetter, Sources of atmospheric methane: Measurements in rice paddies and a discussion, *J. Geophys. Res.*, *86*, 7203–7209, 1981.

Craig, H., and C. C. Chou, Methane: The record in polar ice cores, *Geophys. Res. Lett.*, *9*, 1221–1224, 1982.

Crutzen, P. J., L. E. Heidt, J. P. Krasnec, W. H. Pollock, and W. Seiler, Biomass burning as a source of atmospheric gases CO, H_2, N_2O, NO, CH_3Cl and COS, *Nature*, *282*, 253–256, 1979.

de Bont, J. A. M., K. K. Lee, and D. F. Bouldin, Bacterial oxidation of methane in a rice paddy, *Ecol. Bull.*, *26*, 91–96, 1978.

Ehhalt, D. H., and U. Schmidt, Sources and sinks of atmospheric methane, *Pure Appl. Geophys.*, *116*, 452–464, 1978.

Ehhalt, D. H., R. J. Zander, and R. A. Lamontagne, On the temporal increase of tropospheric CH_4, *J. Geophys. Res.*, *88*, 8442–8446, 1983.

Khalil, M. A. K., and R. A. Rasmussen, Sources, sinks, and seasonal cycles of atmospheric methane, *J. Geophys. Res.*, *88*, 5131–5144, 1983.

Koyama, T., Gaseous metabolism in lake sediments and paddy soils and the production of atmospheric methane and hydrogen, *J. Geophys. Res.*, *68*, 3971–3973, 1963.

Koyama, T., Biogeochemical studies on lake sediments and paddy soils and the production of atmospheric methane and hydrogen, in *Recent Researches in the Fields of Hydrosphere, Atmosphere and Nuclear Geochemistry*, edited by Y. Miyake and T. Koyama, pp. 143–178, Maruzen Company, Tokyo, 1964.

Mah, R. A., D. M. Ward, L. Baresi, and T. L. Glass, Biogenesis of methane, *Annu. Rev. Microbiol.*, *31*, 309–341, 1977.

Rasmussen, R. A., and M. A. K. Khalil, Atmospheric methane (CH_4): Trends and seasonal cycles, *J. Geophys. Res.*, *86*, 9826–9832, 1981.

Rasmussen, R. A., and M. A. K. Khalil, Global production of methane by termites, *Nature*, *301*, 700–702, 1983.

Seiler, W., A. Holzapfel-Pschorn, R. Conrad, and D. Scharffe, Emission of methane from rice paddies, *J. Atmos. Chem.*, *2*, in press, 1983.

Zimmerman, P. R., J. P. Greenberg, S. O. Wandiga, and P. J. Crutzen, Termites: A potentially large source of atmospheric methane, carbon dioxide and molecular hydrogen, *Science*, *218*, 563–565, 1982.

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