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Rate of accumulation and emission of N_2, N_2O and CH_4 from a flooded rice soil

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Abstract

In a field experiment using microplots, a flooded Crowley silt loam (Typic Albaqualfs) rice soil was fertilized with ^{15}N labelled (60–74 atom %) urea and KNO_3. Emission of N_2, N_2O and CH_4 and accumulation in soil were measured for 21 d after fertilizer application.

Emission of $^{15}N_2$–N measured from the urea and KNO_3 treated plots ranged from <15 to 570 and from 330 to 3,420 g ha^{-1} d^{-1}, respectively. Entrapped $^{15}N_2$–N in the urea treated microplots was significantly lower (<15 g to 2.1 kg ha^{-1}) on all sampling dates compared to the $^{15}N_2$–N gas accumulation in the KNO_3 treated plots (6.4 to 31.5 kg ha^{-1}). Emissions of N_2O–N were low and did not exceed 4 g ha^{-1} d^{-1}. Fluxes of CH_4 from the fertilizer and control plots were low and never exceeded 33 g ha^{-1} d^{-1}. Maximum accumulation of CH_4 in the flooded soil measured 460 and 195 g ha^{-1} for the urea and KNO_3 treatments, respectively.

Introduction

Lowland rice soils are characterized by the absence of O_2 and by the unique chemical and biological transformations N and C undergo compared to well drained upland soils (Bouldin, 1986; Mikkelsen, 1987). After submergence a thin oxidized soil surface layer and a reduced subsurface layer are developed. The presence of these two distinct soil layers in close proximity favors the simultaneous occurrence of nitrification-denitrification reactions (Reddy and Patrick, 1984). The oxidized zone promotes nitrification and the reduced soil layer supports the reduction of NO_3^- to N_2 and N_2O. Denitrification has been recognized as a major gaseous loss mechanism of added inorganic N causing poor uptake of fertilizer N by lowland rice (DeDatta *et al.*, 1989). Flooded rice soils exhibit low redox potentials (Eh) and oxidation-reduction values as low as −250 to −300 mV can occur (Patrick, 1981). These low Eh values can be reached within 2 weeks after submergence if organic matter and low concentrations of NO_3^-, Mn^{4+} and Fe^{3+} are present (Patrick and DeLaune, 1977; Ponnamperuma, 1981). At these low redox potentials CH_4 can be produced by anaerobic bacteria in addition to N_2 and N_2O (Mikkelsen, 1987; Ponnamperuma, 1984).

Soil submergence can drastically reduce the gas exchange between the soil and atmosphere leading to the accumulation of N_2, CO_2, CH_4 and H_2 in the soil. The continual soil gas production causes a buildup of pressure which is eventually released as bubbles to the atmosphere (Ponnamperuma, 1984). Harrison and Aiyer (1913) analyzed gas bubbles from a flooded rice field and found N_2, CO_2 and CH_4 compositions to vary between 1–95% during the growing season. Ponnamperuma (1984) stated that up to

2.7 Mg of CO_2 ha^{-1} may accumulate in a flooded soil.

Only a limited number of studies have been published on direct emission measurements and soil entrapment of N_2, N_2O and CH_4 from flooded rice soils (Buresh and Austin, 1988; Cicerone and Shetter, 1983; Holzapfel-Pschorn *et al.*, 1986; Katyal *et al.*, 1989; Mosier *et al.*, 1989; Schutz *et al.*, 1989).

Direct emissions of N_2 and N_2O must be measured to help improve N management practices for improved uptake of fertilizer N by lowland rice. In addition, CH_4 and N_2O fluxes from flooded rice fields to the atmosphere need to be quantified because of their role in global warming and atmospheric ozone depletion (Bouwman, 1989). The significance of soil entrapment of these gases needs to be assessed with respect to soil production and evolution from flooded rice systems (Lindau *et al.*, 1988).

The objectives of the study were to 1) determine the emission rates and 2) measure soil accumulation of N_2, N_2O and CH_4 from a flooded rice field after fertilizer application.

Materials and methods

Field procedures

The field study was conducted at the Rice Research Station, Crowley, Louisiana, USA. This experiment was done in conjunction with a field study investigating the affect of rice plants and fertilizer type on gas emissions from flooded soils (Lindau *et al.*, 1990). The soil at the test plots was a Crowley silt loam (Typic Albaqualfs) with 7.0 g total C kg^{-1} and 0.8 g total N kg^{-1}. Its cation exchange capacity was 9.4 $cmol_c$ kg^{-1} soil and it had a pH of 5.8 (1:1, soil/water). The soil contained 11% clay and 71% silt.

Test plots were drill seeded (April 8) with the long-grain rice cultivar Lemont and permanent flood was established 40 days later. Phosphorus and K were applied preflood at a rate of 67 kg^{-1} ha^{-1} and fertilizer N (urea and KNO_3) were broadcast on the floodwater at a rate of 120 kg ha^{-1}.

Thirty-six treatment and 2 control microplots were established in the flooded field between plant rows. Polyvinyl chloride (PVC) cylinders (30.0 cm length by 10.2 cm i.d.) were driven 10 cm into the soil down to the hardpan 1 d after permanent flood. A floodwater depth of 3–4 cm was maintained within the microplots throughout the experiment. Two days after permanent flood, labelled urea and KNO_3 (74.2 and 60.2 atom% ^{15}N, respectively) were dissolved and added to the microplot floodwater. Nitrogen was not added to the control plots and surrounding areas. Two replications per N treatment were established in the flooded field. At 3, 5, 7, 9, 11, 13, 15, 17 and 21 d after fertilizer application two PVC cylinders of each N treatment were sealed (gasketed PVC end caps) and accumulation and evolution of N_2, N_2O and CH_4 measured. Treatment and control chambers were sealed 1 hour before sunset and headspace gas samples were taken early the next morning. This minimized pressure or temperature buildup within the chamber which could affect gas evolution from the flooded soil. This length of collection was required to detect $^{15}N_2$ in the chamber headspace. Headspace heights were recorded prior to end cap placement and zero-time gas samples were collected immediately after cap placement. Gas samples were removed through a rubber septum sealed in the end cap with a gas-tight syringe and hypodermic needle. Collected headspace gas samples were immediately transferred into evacuated glass Vacutainer tubes (100 mm length by 16 mm i.d.) and the septum needle puncture sealed. A slight over pressure of sample gas was injected into each Vacutainer. This prevented atmospheric gases from leaking into the tube and contaminating the sample. Following the morning gas sampling, internal chamber temperatures were recorded and the sealed cores were removed from the field and the chamber bottoms sealed. Cylinders were vigorously shaken to release soil entrapped N_2, N_2O and CH_4 bubbles. The headspace gases were sampled a second time and transferred to evacuated Vacutainers. Sampling the headspace gases before and after shaking allowed an estimation of the concentration of entrapped gases to be made. Gas accumulation in the control plots was only measured on the last sampling date. Soil redox potential was not measured for fear of dislodging

soil entrapped gases during placement and removal of Pt and calomel electrodes.

Laboratory procedures

Nitrous oxide, CH_4 and $^{15}N_2$ sample concentrations were determined by mass spectrometry of gas chromatography. The N stable isotopic distribution (28, 29 and 30 masses) was determined on a Finnigan Mat Delta E gas isotope ratio mass spectrometer fitted with a dual inlet and a triple collector assembly. The inlet system was constructed to permit direct injection of a sample gas (2 cm^3) into an evacuated glass manifold. A LN_2 trap was used to remove N_2O, CO_2 and H_2O vapor and a hot Cu furnace removed O_2 from the sample gas (Siegel *et al.*, 1982). The emissions of N_2 from the soil to the atmosphere were calculated using the equations described by Mulvaney and Boast (1986) for triple collector mass spectrometers.

Nitrous oxide gas concentrations were determined on a Perkin-Elmer 8410 gas chromatograph equipped with an electron capture detector. A 2-cm^3 aliquot was injected onto a Chromosorb 106-80/100 mesh column (0.006 × 1.8 m) and the carrier gas was an Ar/CH_4 mixture (30 cm^3 min^{-1}). The detector was set at 395°C and the column oven at 70°C (Lindau *et al.*, 1988).

A Perkin-Elmer 900 gas chromatograph equipped with a flame ionization detector was used to measure the sample CH_4 concentrations. A 1.5 cm^3 gas sample was injected into a stainless steel column (0.003 × 2.4 m) packed with Haye-Sep D polymer (100/120 mesh). Column temperature was 40°C and the injector and manifold set at 110°C.

The fluxes of N_2O and CH_4 from the soil surface were estimated using the closed chamber equation (Rolston, 1986)

$$f = (V/A)(\Delta C/\Delta t)$$

where f = gas flux, V = volume of chamber air, A = soil area and $\Delta C/\Delta t$ = change of gas concentration per unit of time. Emission rates of $^{15}N_2$ and N_2O reported represent fluxes above control data. All data were subjected to statistical analysis of variance (SAS, 1988).

Results and discussion

Denitrification measurements

Soil emissions of $^{15}N_2$–N from the urea treated microplots ranged from <15 (detection limit) to 570 g ha^{-1} d^{-1}. Evolution of labelled N_2 was first detected (43 g ha^{-1} d^{-1}) 7 d after urea–N application and steadily increased to day 15 and decreased thereafter (Fig. 1). Fluxes of labelled N_2 from the KNO_3 treated microplots were much higher and steadily increased to 3,420 g ha^{-1} d^{-1} at day 21. Emission of N_2 from the KNO_3 treated plots was first detected (360 g ha^{-1} d^{-1}) 3 d after N addition (Fig. 1). Analysis of variance showed that the $^{15}N_2$ emissions from the KNO_3 plots were significantly higher (0.05 level) at all sampling dates except day 11.

The percent of the total N_2 evolved due to fertilizer addition can be calculated using the fertilizer atom % ^{15}N enrichment and the $^{15}X_N$ value (mole fraction of ^{15}N in NO_3^- pool) calculated from the equations of Mulvaney and Boast (1986). Approximately 58 to 84% of the total N_2 evolution from the urea treated plots was due to fertilizer over the 21 d sampling period. Dinitrogen derived from the native soil N pool ranged from <15 to 130 g ha^{-1} d^{-1} on day 15 (Table 1). Even though N_2 fluxes measured from the KNO_3 treated plots were much higher, the soil-derived N_2 contribution was much less compared to urea, and accounted for less than 3% of the total evolved N_2. At all sampling dates greater than 97% of all N_2 evolved was due to KNO_3 addition. Soil-derived N_2 never exceed 65 g ha^{-1} d^{-1} (Table 1).

Urea must undergo hydrolysis and nitrification prior to the denitrification reactions (Reddy and Patrick, 1984). These additional steps may help explain the lower N_2 fluxes and the lag time observed before detection of labelled N_2 from the urea treated microplots.

The cumulative effect of $^{15}N_2$ accumulation in the flooded rice soil is shown in Figure 2. Soil entrapped N_2 was first detected (95 g ha^{-1}) 5 d after urea addition and increased to 2.1 kg ha^{-1} on day 17. The production of labelled N_2 from urea–N in the flooded soil preceded N_2 emissions to the atmosphere by 2 d (Fig. 1 and Fig. 2). Labelled N_2 gas entrapped in the soil after KNO_3

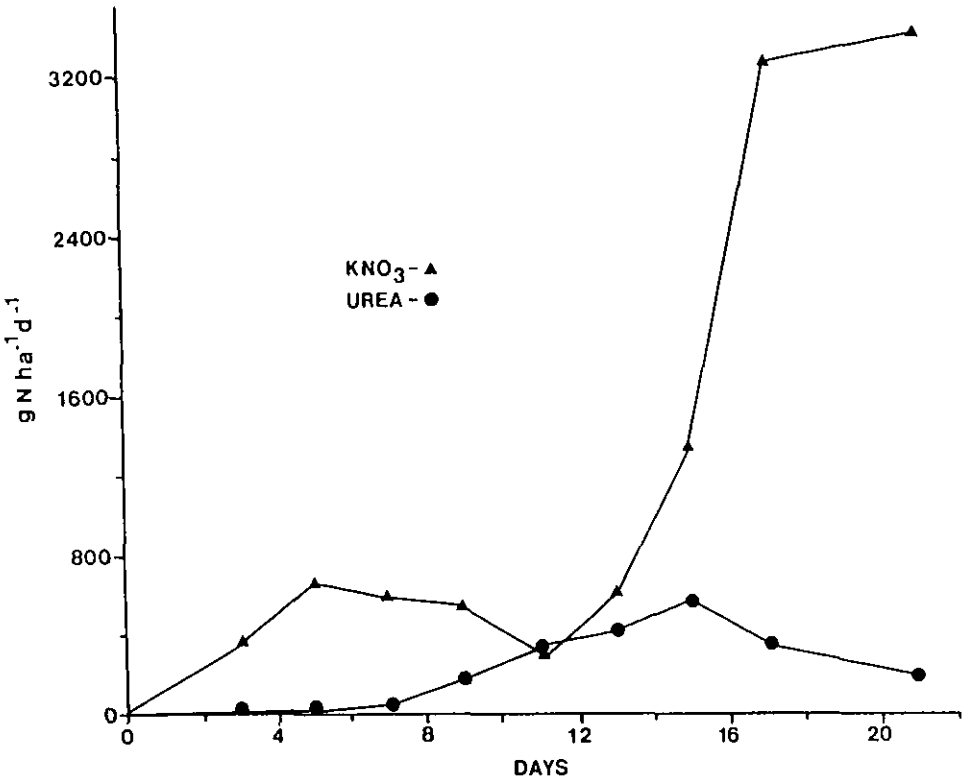

Fig. 1. Daily emissions of $N_2 + N_2O$ from a flooded Crowley silt loam rice soil after fertilizer application.

Table 1. Dinitrogen emissions due to fertilizer N and the native soil

Days after N application	N form	Source of N_2	
		Fertilizer ($g\,ha^{-1}\,d^{-1}$)	Soil ($g\,ha^{-1}\,d^{-1}$)
3	Urea	ND[a]	ND
	KNO_3	350	8
5	Urea	ND	ND
	KNO_3	650	10
7	Urea	25	20
	KNO_3	580	8
9	Urea	160	30
	KNO_3	550	9
11	Urea	260	85
	KNO_3	320	9
13	Urea	300	115
	KNO_3	580	15
15	Urea	440	130
	KNO_3	1,320	20
17	Urea	260	100
	KNO_3	3,180	65
21	Urea	110	75
	KNO_3	3,370	55

[a] Not detected above detection limit ($<15\,g\,ha^{-1}\,d^{-1}$).

addition was much higher and ranged from 6.4 kg ha^{-1} (day 3) to 31.5 kg ha^{-1} on the final sampling day (Fig. 2). Soil entrapped N_2 derived from the labelled KNO_3 fertilizer was significantly (0.05 level) higher compared to the N_2 accumulation in the urea microplots on all sampling dates. Entrapped N_2 measured on day 21 accounted for approximately 1 and 26% of the applied urea and KNO_3^-–N, respectively. These percentages of entrapped N_2 may have been reduced if rice plants had been included in the microplots (Mosier *et al.*, 1989a).

Nitrous oxide emissions attributed to urea and KNO_3 addition were low and never exceeded 3 g N ha^{-1} d^{-1} above control values (about 0.5 g ha^{-1} d^{-1}). The highest N_2O–N fluxes were recorded on day 3 from the KNO_3 treated plots. For the majority of samplings N_2O fluxes from the N treated plots were not detected above control values. Soil entrapped N_2O did not exceed 4 g N ha^{-1} over the 21 days and decreased with time. The low N_2O fluxes may be attributed

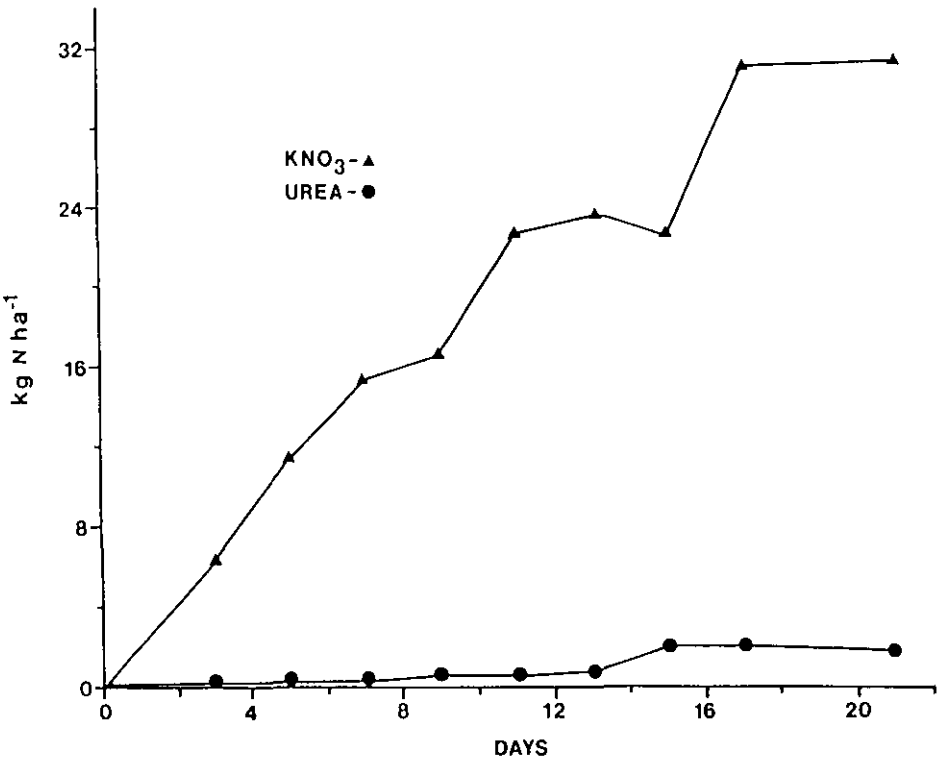

Fig. 2. Accumulation of $N_2 + N_2O$ in a flooded Crowley silt loam rice soil after fertilizer application.

to the flooded soil conditions and/or rapid conversion to N_2.

Figures 1 and 2 demonstrated there may be a direct delayed relationship between soil entrapment and emission of $N_2 + N_2O$ from the soil surface. As gas bubbles accumulate in the flooded soil and are released to the atmosphere then increases in daily emissions are observed. Our results are similar to the published research on direct measurement of N_2 and N_2O emissions and accumulation in flooded rice soils. Following labelled urea–^{15}N applications of 44 and 58 kg N ha^{-1} to a flooded Philippines rice paddy Buresh and Austin (1988) measured the flux of $(N_2 + N_2O)$–^{15}N over a 17 d sampling period. Fluxes over the 17 d period never exceeded 103 g N ha^{-1} d^{-1} except on one sampling date (267 g N ha^{-1} d^{-1}). Samson *et al.* (1989) applied NO_3^-–^{15}N (6.9 kg ha^{-1}) to a puddled Philippines rice soil and measured $(N_2 + N_2O)$–^{15}N emission rates as high as 625 g N ha^{-1} d^{-1} 2 d after N addition. After 7 d flux rates had dropped off to <15 g N ha^{-1} d^{-1}. Mosier *et al.* (1989) measured N_2 and N_2O gas evolution from an Australian lowland rice field for 7 d after urea–^{15}N had been applied at a rate of 80 kg N ha^{-1}. Over the 7 d sampling period N_2 and N_2O gas emissions did not exceed 720 and 5 g N ha^{-1} d^{-1}, respectively. In a later study, Mosier *et al.* (1990) measured the evolution of labelled N_2 and N_2O from rice fields in India. In that study, 99 atom % ^{15}N urea was applied to intermittently flooded rice paddies (100 kg N ha^{-1}) and denitrification was measured for 15 d. Labelled $N_2 + N_2O$ fluxes from the treatment plots ranged between 14–750 (rice plants removed) and 14–980 (with rice plants) g N ha^{-1} d^{-1} (Mosier *et al.*, 1990).

Laboratory incubation studies (without rice plants) by Lindau *et al.* (1988) and Katyal *et al.* (1989) demonstrated that denitrification gases were entrapped and easily released from flooded rice soils. Lindau *et al.* (1988) showed that 28% of the applied urea–^{15}N and 40% of the KNO_3–^{15}N was trapped as N_2 33 d after fertilizer addition to flooded soil columns. Our field research also shows a much larger percentage of the applied KNO_3–^{15}N entrapped as N_2. Sixteen

days after labelled KNO_3 application (100 kg N ha^{-1}) Katyal *et al.* (1989) demonstrated that 41% of the applied N was trapped as N_2 in a submerged Crowley silt loam soil. Mosier *et al.* (1990) measured gas entrapment of $^{15}N_2$ in a greenhouse experiment using rice planted and unplanted soil columns. Four weeks after urea fertilization $^{15}N_2$ accumulation in the unplanted flooded soil columns accounted for 8.4% of the added ^{15}N compared to only 1.2% entrapped for the planted columns. Mosier *et al.* (1990) suggested that rice plants act as a conduit to the atmosphere for soil produced N_2. This transport mechanism has also been demonstrated for CH_4 (Schultz *et al.*, 1989). If rice plants had been included in our experiment accumulation of N_2 and N_2O in the soil would probably have been greatly reduced.

Methane measurements

Soil CH_4 fluxes from the treatment and control plots over the 21 d sampling period were low and highly variable between replications and fertilizer treatment (Fig. 3). Concentrations at the time-zero collections ranged from 7.8 to 12.4 μl CH_4 L^{-1} air over the 21 days. Methane emissions from the urea fertilized plots ranged from nondetectable (ND) values (above time-zero concentrations) to 33 g ha^{-1} d^{-1} recorded on day 11. The CH_4 fluxes from the KNO_3 treated plots were much lower and ranged from ND to 8.2 g ha^{-1} d^{-1}. Control plot CH_4 emissions were low over the first 11 d but gradually increased to 25 g ha^{-1} d^{-1} at day 21 (Fig. 3). Except for the relative large CH_4 flux recorded on day 11 from the urea treated plots CH_4 fluxes from all the treatment microplots gradually increased over time. The largest incremental CH_4 increases occurred on or after the 15 d. Methane fluxes from the urea treated microplots were significantly higher (0.05 level) compared to the CH_4 emissions from the KNO_3 and control plots on days 7, 9, 11 and 17. On days 13, 15 and 21, CH_4 fluxes from the urea and control plots were significantly higher than recorded CH_4 emissions from the KNO_3 treated microplots. Nitrate tends to buffer soil redox potential slowing soil reduction and methane production (Patrick and DeLaune, 1977).

Accumulation of CH_4 in the soil of the urea treated plots ranged from ND (day 3) to 460 g ha^{-1} on day 21 (Fig. 4). Methane entrapment in the KNO_3 treated microplots was significantly lower (0.05 level) at all sampling dates (except day 3) and slowly increased to 195 g ha^{-1} at day 21. The fertilizer effect on CH_4 accumulation in the flooded soil could not be directly

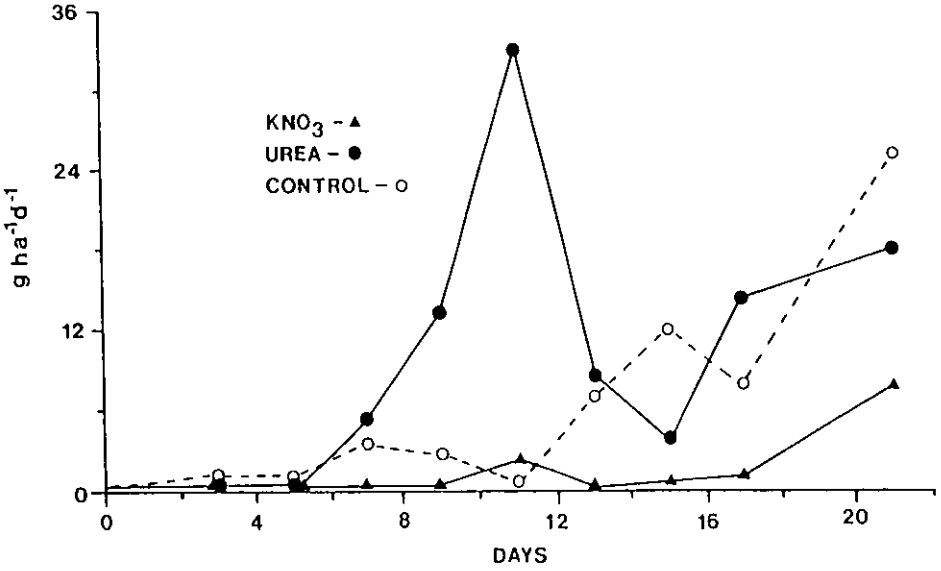

Fig. 3. Daily emissions of CH_4 from a flooded Crowley silt loam rice soil after fertilizer application.

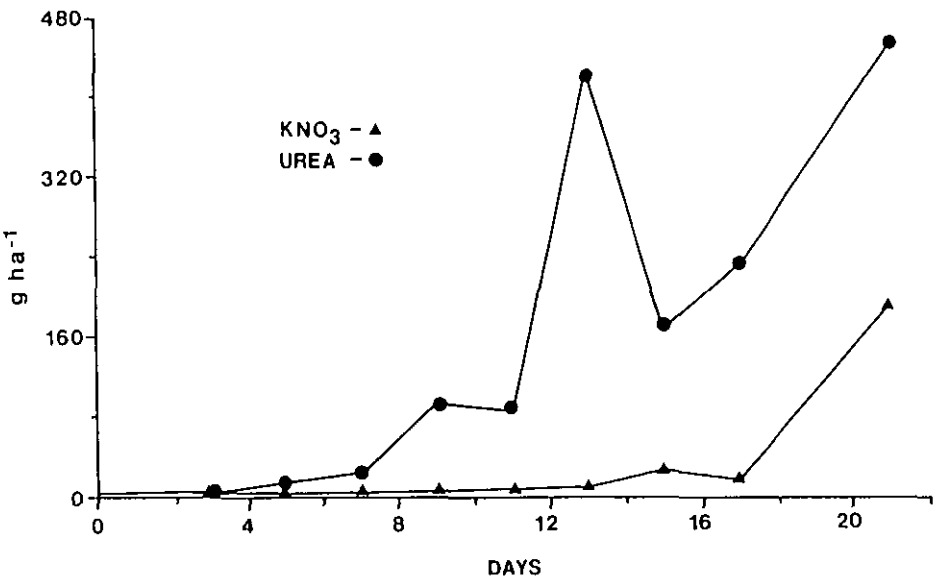

Fig. 4. Accumulation of CH_4 in a flooded Crowley silt loam rice soil after fertilizer application.

assessed. Entrapment of CH_4 in the control plots was not measured until day 21. On this day the average CH_4 accumulation in the control plots was 725 g ha^{-1}. Generalizing the CH_4 accumulation data for day 21, fertilizer addition appears to reduce net soil CH_4 production during the early growing season. The large concentrations of CH_4 bubbles accumulated in the soil over time would appear to support previous studies showing emission by bubbles is much more important than CH_4 emission by diffusion (Seiler *et al.*, 1984; Schutz *et al.*, 1989).

The low CH_4 flux rates recorded from the Louisiana flooded rice field may be due to high soil redox conditions shortly after flooding, low soil organic matter content and/or soil gas accumulation. Cicerone and Shetter (1981) measured CH_4 emissions ranging from 35 g to 1.8 kg ha^{-1} d^{-1} from a California rice field with the fertilized planted plots showing the highest fluxes. In a later study, Cicerone and Shetter (1983) demonstrated a strong seasonal dependence on CH_4 flux with peak emission occurring 2 to 3 weeks before harvest. Methane fluxes ranged from <10 g ha^{-1} d^{-1} (early growing season) to 49.4 kg ha^{-1} d^{-1} (prior to harvest) which supports our CH_4 measurements during the early rice season. Cicerone and Shetter (1983) also questioned whether the CH_4 emission increase with time was due to an accumulation of soil CH_4 or due to rice plant maturity. Seiler *et al.* (1984) investigated CH_4 fluxes from rice paddies in Spain and measured rates ranging from 0.5 to 3.4 kg ha^{-1} d^{-1}. Approximately 95% of the CH_4 released was through the rice plant gas transport system (Seiler *et al.*, 1984). Holzapfel-Pschorn and Seiler (1986) measured CH_4 fluxes from an Italian rice paddy. Fluxes were less than 24 g CH_4 ha^{-1} d^{-1} shortly after flooding but increased to 12.2 kg ha^{-1} d^{-1} at the flowering stage. They also observed no significant differences in soil CH_4 emissions comparing fertilized to unfertilized plots.

The results presented demonstrate that the type of fertilizer applied had significant effects on N_2 and CH_4 emissions and accumulation in a flooded rice soil. If rice plants had been included in our microplots N_2 and CH_4 emission rates may have increased and the buildup of N_2 and CH_4 in the soil sharply reduced which has been previously demonstrated (Cicerone and Shetter, 1981; Holzapfel-Pschorn *et al.*, 1986; Mosier *et al.*, 1989; Lindau *et al.*, 1990). The CH_4 and N_2 flux and soil accumulation data presented in this study was very limited due to the short sampling period and the absence of rice plants. Detailed gas flux measurements must be taken over the entire rice growing season(s) and under a variety

of site specific management practices throughout the world. In addition, accumulation of gases in the soil must also be thoroughly investigated with respect to production and evolution of greenhouse and denitrification gases from flooded rice. This type of detailed data will help scientists predict annual worldwide fluxes of N_2, N_2O and CH_4 from lowland rice to the atmosphere when local management practices are altered.

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