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Effects of Production and Oxidation Processes on Methane Emissions from Rice Fields

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Abstract. The emission of methane from rice fields is the difference between the amount produced in the anaerobic zone below the soil and the amount oxidized in the root zone. Plants can also contribute to methane production by exuding organic compounds that may be utilized by methanogenic bacteria. We measured methane emissions from rice fields at Tu Zu in China between 1988 and 1994, which gave average emissions of about 30 mg m^{-2} h^{-1}. We estimate that 45-60% of the methane produced was oxidized before reaching the atmosphere; and root exudates may have contributed of the order of 10% of the methane that was produced. The fraction of methane oxidized is low compared to experimental studies at other locations (60-85%). At Tu Zu, methane production is enhanced by continuously flooded fields and the use of large amounts of organic fertilizers; in addition, the lower oxidation rate may also contribute to the higher methane emissions observed compared to other locations. In the past, most of the attention has been devoted to the factors that affect methane production and transport, but it seems that the factors that affect methane oxidation are equally important in determining the flux, if not more so. The comparison of methane fluxes observed at different locations and the extrapolation of field measurements to accurately estimate global emissions will require a better understanding of the rate of methane oxidation in the soils and the factors that control it.

1. Introduction

The production and release of methane from rice fields is a complex process in which the rice plants play an active role. The plants can add to methane production below the soil, aid in the oxidation of methane before it can be released to the atmosphere, and act as conduits for the remaining methane so that it is rapidly released from the soil to the atmosphere. Between 1988 and 1994 we conducted extensive experiments of methane emissions from rice fields at Tu Zu in the Sichuan Province of China. The experiments included direct flux measurements with different planting densities in the same fields and many replicates (0, 1, 2, and 4 plants per plot). The observed differences of emissions for different planting densities are indicators of the action of plants on the methane flux. In this paper we will use these observations to estimate the role of oxidation, production of methane by root exudates, and the transmission of methane through the plants to the atmosphere.

The main observational results, relevant to this study, are the methane emission rates during the growing season for different planting densities as shown in Figure 1. It is a composite of some 5000 flux measurements taken over small plots (0.06 m^2). One plant per plot represents the normal planting density in the rice fields of this region. The experimental results from the different years were very similar, and the fields were managed in the same manner each year. This allows us to pool the data from all the years and to represent it as a function of time in days from transplanting. The pooled data were then smoothed by a 40-point moving average filter to eliminate any period that is dominated by data from a single year and to reduce the variability. This amounts to averaging over 11±2 days and somewhat higher averaging times for the beginnings and ends of the growing season (14-20 days). The database and the environmental conditions under which it was obtained are described in detail by *Khalil et al.* [this issue (a)].

In subsequent sections we will discuss the empirical observations on methane emissions from rice fields. These observations will be represented by a model that takes into account the oxidation and production processes. We will then apply this model to the observations shown in Figure 1 and deduce the amount of methane oxidized and the possible role of root exudates in methane production.

2. Processes and Mechanisms of Rice Field Methane Emissions

2.1. Empirical Observations and Theoretical Ideas on Methane Emissions

Methane is produced below and around the root zone by methanogenic bacteria. These bacteria utilize the carbon from organic fertilizers, crop residues, dying roots, and possibly from
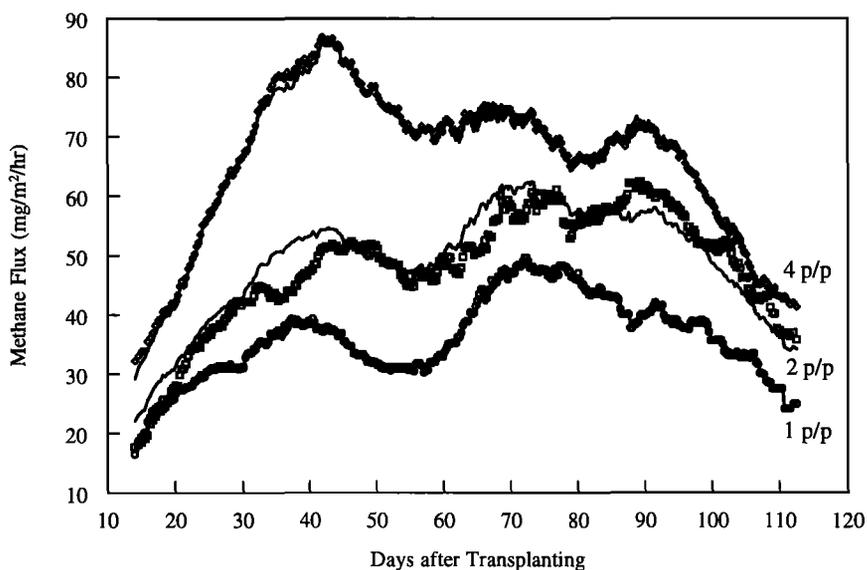
Figure 1. Emissions of methane from rice fields with the normal planting density (1 plant/plot) and with 2 and 4 times the normal planting density. The differences of fluxes are attributed to the changes that planting density causes for oxidation of methane in root zone, the production of methane from root exudates, and the transport of methane through the plants. Results are based on the composite of 7 years of data from Tu Zu in China (1988-1994). The solid lines are the calculated fluxes as discussed in the text. When the lines are not visible, it is because the calculated and measured fluxes are almost identical, which they are for most of the data. [x p/p = x plants per plot (x = 1, 2, 4)].

root exudates from the rice plants. This methane is transported upward toward the surface where it encounters the root zone of the plants. Around the roots is an aerobic region created by the exchange of oxygen between the plant and the air above. This zone harbors methanotropic bacteria that can utilize methane for food when high concentrations are available, as is the case in the rice fields. Methane is driven across the gradient toward the roots. Studies have shown that possibly 60-85% of the methane that is produced is oxidized in this part of the rhizosphere [Holzapfel-Pschorn et al., 1985; Schütz et al., 1989; Sass et al., 1990]. The fraction of the methane that is left is taken up through the roots and transported by the plant stems out to the atmosphere. It is released through openings in the sheath, the aerenchyma, where the leaves attach to the stem [Nouchi et al., 1990]. Plant respiration and solar radiation do not appear to affect methane emissions, since the aerenchyma do not open and close with changing environmental conditions (for a review, see Conrad, 1996).

So far, two parameters, namely, organic fertilizer use and water levels, have been shown to definitely influence methane emissions. Apparently, both factors affect the flux by changing the production of methane below the soil. Addition of certain types of organic manure and rice straw can greatly increase the emission of methane relative to fields which are otherwise the same but without organic fertilizers [Schütz et al., 1989; Yagi and Minami, 1990; Chen et al., 1993; Khalil et al., this issue (b)]. For water levels there is little or no relationship with flux as long as there is standing water. Once the water is drained or allowed to evaporate, methane emissions drop to zero. This effect can be caused by reducing the anaerobic zone and changing the conditions for methane production. These two factors explain some of the diversity of the observational results, but there is still substantial variability that is unaccounted for. When the fields are continuously flooded and organic fertilizers are used, the methane emissions are between 10-40 mg $m^{-2} h^{-1}$; without organic fertilizers, the emissions are 5-20 mg $m^{-2} h^{-1}$; with intermittent water in the fields and low organic matter, the

emissions can be 0-5 mg $m^{-2} h^{-1}$ [Sass et al., 1992; Chen et al., 1993; Husin et al., 1995; Yagi et al., 1996]. Factors that affect the amount of methane oxidized under field conditions are unknown at present. Even a small difference in the oxidation of methane, from one field to another, could greatly affect methane emissions and explain the seemingly contradictory results between studies. Yet these processes are difficult to measure and have rarely been observed directly. In this paper we will estimate the amount of methane oxidized in the soil and the amount possibly produced by root exudates, based only on the observations of the flux.

2.2. A Model to Study the Effect of Planting Density on the Processes That Affect Emissions

The effect of planting density can be used to construct a simplified model that takes into account the major mechanisms involved in the production, oxidation, and transport of methane within the soils, through the plants and to the atmosphere, as discussed in the previous section. The model includes the soil, the rhizosphere, and rice plants, which are conduits for the transport of methane.

The model is written as follows:

$$\frac{dC_s(N,t)}{dt} = P(N,t) - \frac{1}{\tau_s}[C_s(N,t) - C_r(N,t)] \qquad (1)$$

$$\frac{dC_r(N,t)}{dt} = \frac{1}{\tau_s}[C_s(N,t) - C_r(N,t)]\frac{\delta_s}{\delta_r(N)}$$
$$- [\frac{1}{\tau_0} + \frac{1}{\tau_p(N,t)}]C_r(N,t) \qquad (2)$$

where

C_s, C_r concentrations of methane in the soil and the rhizosphere (g/m^3);

δ_s, δ_r effective depths (m) of the methane production zone in the

soil s and the oxidizing root zone r but do not necessarily represent the actual depths. These are defined as the volumes of the zones in the field divided by the surface area of the field;

P production of methane in the soils $(g\ m^{-3}\ h^{-1})$;.

N number of plants per unit surface area;

τ_s transport time for exchange of methane between the soil and the rhizosphere (hours);

τ_o time that defines the rate of oxidation in the root zone (hours).

τ_p time for transport across the root membranes into the plants. (hours);

τ_b timescale for ebullition, which we assume is proportional to the concentrations in the soil (hours).

These equations are a core subset of a more complete model. Here we consider two zones below the soil: the region where methane is produced (subscripts s) and the rhizosphere (subscripts r) containing the roots and representing a zone where methane can be oxidized before entering the plants. We have written two mass balance equations for methane in the two zones. The processes that change the methane concentration in the zones are on the righthand sides.

In the first equation dealing with the production zone, the first term reflects the amount of methane produced per unit volume of soil. The production could depend on the number of plants if root exudates are a precursor for methane production. The second term is the net transfer of methane between the soil and the rhizosphere driven by the gradient of the concentrations across the two zones that occurs over a characteristic time τ_s. The value of τ_s depends on diffusion and therefore on the physical characteristics of the soil. Two additional processes can also release methane to the atmosphere: bubbling, which transfers methane directly from the production zone in the soil to the atmosphere; and exchange of methane between the water and the air above. Some methane that passes through the water may also be oxidized. The contributions of these processes are expected to be small and are not included here, which greatly simplifies the model [see *Seiler et al.*, 1984].

The second equation represents the mass balance of methane in the rhizosphere. The net amount of methane that is transported from the soil is the source of methane into the rhizosphere as represented by first term. The ratio $\delta_s\ /\ \delta_r$ in the mass balance accounts for the different volumes of the rhizosphere and the methane production zone (for further discussion of these volumetric factors, see *Khalil and Rasmussen* [1984]). The second part of the equation contains two terms. The first represents the oxidation of methane in the rhizosphere that can be expressed as a characteristic lifetime (τ_o) of methane in this zone. It depends on the volume of the zone and the types and populations of methane oxidizing bacteria. The second term represents the transfer of methane across the root membrane into the atmosphere and the characteristic time associated with this cross-media transfer (τ_p). We assume that no methane comes back from the plant into the rhizosphere. We assume further that all the methane that gets into the plant is released to the atmosphere. This last term therefore also represents the flux to the atmosphere, which is much of the flux that we measure in our experiment.

The effect of planting density is reflected in the transport by the plants, the oxidation zone, and in the production rate of methane. These factors are written as

$$\tau_p(t) = \tau_{p_o}(t)/N \qquad (3)$$

$$\delta_r = \delta_{r_o} N \qquad (4)$$

$$P(N,t) = P_o(t)[1 + \gamma(t)N] \qquad (5)$$

where γ is the ratio of the methane production from root exudates for the prevailing planting density of the rice field (one plant per plot for the plot size in our experiments) to the production of methane from precursors other than root exudates, $P_o(t)$ is the production of methane from precursors other than root exudates $(g\ m^{-3}\ h^{-1})$, and δ_{ro}, τ_{po} is the effective depth (m) and exchange time (hours) for planting density one plant per plot corresponding to δ_r, τ_p defined earlier.

If root exudates are a significant source of methane production, then more plants would lead to more methane production. When there are more plants per unit surface area, the volume of the root zone increases, thus increasing the effective depth δ_r. If the transport to the atmosphere is slowed down, more methane is oxidized because it spends a longer time in the oxidation zone. In this model, more plants lead to competition between a larger oxidation zone, which tends to reduce emissions, and faster transport and greater production, which tend to increase emissions.

Many variables, such as P_o, γ, τ_o, and τ_p, change during the growing season. As the rice plant tillers, it increases the root surface area (τ_o decreases), and the many new stems add more conduits for methane transport (τ_p decreases). These factors also depends on the rice cultivar and agricultural management of the fields.

These equations contain many unknown parameters. Our experiments do not have sufficient information to estimate most of the variables, nor are there sufficient data in the published papers to estimate the values reliably. We can simplify these expressions in such a way as to be able to estimate the effect of key processes on the methane flux, namely, the fraction of methane oxidized and the fraction that may be attributed to root exudates. An approximate solution (equations (6)-(9), useful for this purpose, can be obtained if we assume pseudo steady state $(dC/dt = 0)$, then

$$F(N,t) = \frac{P_o\ \delta_s(1 + \gamma N)}{(1 + \frac{\lambda}{N})} \qquad (6)$$

$$C_r = P\frac{\delta_s/\delta_r}{(\eta_o - \eta_p)} \qquad (7)$$

$$C_s = C_r + P\tau_s \qquad (8)$$

$$\lambda = \frac{\tau_{p_o}}{\tau_o} \qquad (9)$$

where $F(N,t)$ is the flux of methane $(mg\ m^{-2}\ hr^{-1})$, for N plants per unit plot size at time t in days since transplanting (measured in the experiments), η is the inverse of the transport or lifetime: $1/\tau$ (1/hour), and λ is the dimensionless ratio of the rate of oxidation to the rate of transport in the rice field system based on the prevailing planting density (here taken to be 1 plant/plot with plot size as specified earlier). The pseudo steady state assumption is justified since the transport times are expected to be faster than the timescales over which the methanogen populations and productivity change (P).

Equation (6) is the main result as it relates the production P_o $\delta_s(mg\ m^{-2}\ h^{-1})$, oxidation (λ), and root-mediated production (γ) to the methane flux, which is measured. We have flux measurements at three planting densities (Figure 1), and there are these three unknowns. We will return to the use of these equations in the next section.

We can explain our observations in Figure 1, qualitatively, as follows: In the earliest phases of growth (first few weeks), because the plants are small, without tillers or large root masses, τ_p is long and λ/N is very large, making F ≈ 0 in (6) (in our experiments we have few observations during this phase). The only emissions that take place are through ebullition, and these are independent of the planting density at this stage of the growth. The emissions are therefore transport-limited. As the rice plants grow, λ decreases as the transport efficiency increases until tillering stops and the plants flower and mature. During this period, and to harvest time, the rapid transport prevents large buildups of methane in the soil that can lead to ebullition. From the seed production stage to end of the growing season the transport is very efficient or $\lambda/N \ll 1$, and root exudation is no longer large $\gamma N \ll 1$, so that F ≈ P δ_s and is again not affected by the planting density. In this case the emissions are production-limited. In between, λ has intermediate values, and root exudation may also occur, resulting in the emission patterns with clearly observable difference between the case for one plant per plot and four plants per plot. We have described three periods, transport-limited in the beginning and production-limited at the end. The processes smoothly interpolate between these extreme cases and result in small differences of emissions with planting density in the early and later stages of the crop cycle and large differences in between.

3. Application of the Model: Estimates of Oxidation and Root Exudate Methane

For convenience of discussion we write the following three equations:

$$r(N,M) = \frac{F(N)}{F(M)} = \left[\frac{(1+\lambda/M)}{(1+\lambda/N)}\right]\left[\frac{(1+N\gamma)}{(1+M\gamma)}\right] \quad (10)$$

$$f = \frac{\lambda}{(1+\lambda)} \quad (11)$$

$$g = \frac{\gamma}{(1+\gamma)} \quad (12)$$

where f is the fraction of the methane that is oxidized relative to the amount produced; $0 \le f \le 1$, g is the fraction of the total production of methane that can be attributed to the roots; $0 \le g \le 1$, and r(N,M) is the ratio of emissions from N plants/plot to emissions from M plants/plot. Either N or M may represent the field planting density, while the other represents planting the effect of a higher (or lower) planting density.

3.1. Upper Limits of Production and Oxidation

In most years we have data for only two planting densities; it is therefore not possible to estimate all three variables of interest, namely, the production P_o δ_s (mg m^{-2} h^{-1}), oxidation (λ), and root-mediated production (γ). If we assume, however, that the observed differences in the emissions with different planting densities are due only to the effect of oxidation ($\gamma = 0$), we can then find the fraction that must have been oxidized by using (10) and (11) and the production rate from (6). We can do this for each year, except 1991 when we took measurements at only one planting density. The results are as follows: f (%) (1988, ...,1994) = (54, 60, 59, NA, 73, 44, 75), and production rates Pδ_s (1988,..,1994) = (96, 80, 109, NA, 126, 70, 160) mg m^{-2} h^{-1}. The average oxidation rate is 60 ± 12%,

and the average production rate is 107 ± 33 mg m^{-2} h^{-1} (the plus or minus values are standard deviations). Using the composite data in Figure 1, we get an average oxidation rate of 61% and a production rate of 95 mg m^{-2} hr^{-1} These rates are upper limits for the oxidation.

Similarly, if we assume that the observed differences for different planting densities are due entirely to root exudation, which is very unlikely, we can derive the values of γ (since $\lambda = 0$) using the same equations. The results are g(%) (1988,..,1994) = (23, 21, 12, NA, 35, 14, 37), and Pδ_s (1988,..,1994) = (44, 39, 53, NA, 34, 55, 40) mg m^{-2} h^{-1}. The average production by root precursors is 23 ± 10%, and the average total production is 44 ± 8 mg m^{-2} h^{-1}. When the composite data in Figure 1 are used under the same assumptions, the results are an average root mediated production rate of 27% and a total production rate of 37 mg m^{-2} h^{-1}. These values are the upper limit of the amount of methane produced from root precursors.

3.2. Estimates of Oxidation and Production From the Composite Data

With the composite data in Figure 1 it is possible to estimate the simultaneous existence of oxidation processes and root exudates since we have three equations of the form represented by (6) and three unknowns. Since the equations are nonlinear, exact solutions do not always exist, and a number of different types of solutions that may be equally good are representing the data. Nonetheless, the nature of the patterns in Figure 1 is such that no root exudate methane can be accommodated after about 50-60 days (from transplanting), and little or no oxidation can be accommodated in the early phases of the data before 40 days. One of the nearly optimal solutions, which embodies this general characteristic, was derived by a combination of grid search and partial solutions procedures. The results are shown in Figure 2.

In Figure 2a we show the three parameters $P_o\delta_s$, f (equivalently λ by equation (11)), and g (equivalently γ by (12)), from which other variables of interest can be calculated. In Figure 2a we also show the oxidation rate as a function of time in the growing season if no root exudate effect is considered ($\gamma = 0$ case). In Figure 2b we show, in addition to $P_o\delta_s$ (production unrelated to plants), the total production rate Pδ_s. The difference is the production by root-mediated precursors.

If we take the $P_o\delta_s$, λ, and γ from Figure 2 and put them into (6) with N= 1, 2, or 4, we get the solid lines in Figure 1, representing the fit of these functions to the observed data. The rms deviations between observed and calculated fluxes (equation (6)), F(1,t), F(2,t), and F(4,t) are 0.5, 3.6, and 0.8 mg m^{-2} h^{-1} respectively. These results can also be interpreted to say the values of production, oxidation and root exudates, as shown in Figure 2, explain our observations, which are shown in Figure 1. According to these calculations the average production of methane in these experiments was 65 mg m^{-2} h^{-1}, the fraction oxidized was 44%, and the amount of methane produced from root precursors was 7% of the total production, under the prevailing planting density in these fields.

The results here suggest that root exudates did not contribute much, if anything, in our experiments. This is because the ratio of fluxes r(N,M) in (10) is a product of two ratios, one for the oxidation and the other for the root exudate production parts of the process. This ratio is fixed by observations (Figure 1), and it turns out that the ratios are small at r(2,1) = 1.33 and r(4,1) = 1.83 that does not exceed 2.5. If there were more active oxidation or greater effect from root exudation, these ratios would have to be much larger than observed since r(4,1) can be up to 16 in this model. The
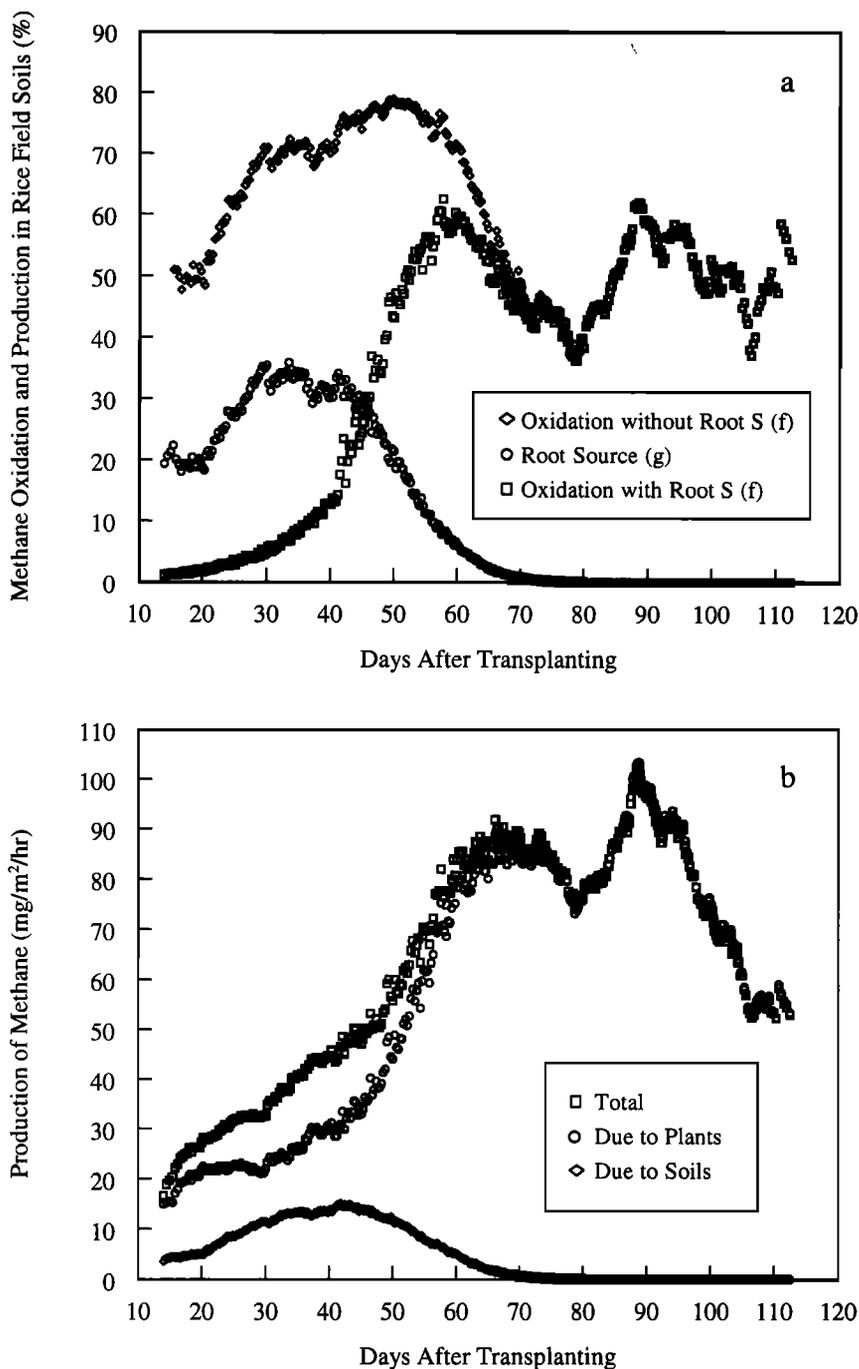
Figure 2. (a) The calculated fraction of methane oxidized in the rhizosphere of the rice fields in the Tu Zu experiments and the estimated fraction of methane that may come from root exudates. (b) The estimated production rates of methane in the experiments at Tu Zu. These results are obtained from the observed differences of emission rates shown in Figure 1 and a mass balance model discussed in the text.

effect of root exudates on methane emissions has not been demonstrated in the field. Rice plants may produce root exudates, but how much of this carbon is emitted as methane under natural field conditions is not known.

3.3. Comparisons With Other Studies

It seems that the root exudation effect is largest during the time between 20 and 60 days after transplanting. This is the period of active plant growth and includes the tillering phase. The fraction of

methane emitted that comes from root exudates is about 24% during this period. These results are in the same range as the laboratory studies reported by *Minoda et al.* [1996] but somewhat lower. The laboratory studies used plants grown in pots, which gave seasonally averaged emissions from photosythesized carbon of about 20-25%. Assuming that root exudates would be the major portion of photosynthesized carbon available to methanogens during the period of active growth, and considering the complexities of extrapolating these laboratory results to field conditions, the agreement is encouraging.

There are a number of studies that have dealt with oxidation of methane in the rhizosphere of rice plants. Many of these studies are not easily comparable to our work here because of the differences in the environmental conditions under which the studies were done, or because the production rates were studied on only a few days of the growing season. Two sets of studies where production was studied throughout the growing seasons in normal rice fields showed oxidation rates from 60% to 85% during the growing season at sites in Texas [*Sass et al.*, 1990, 1992; *Sass and Fisher*, 1995] and Italy [*Schütz et al.*, 1989]. Further studies of the Texas fields using carbon isotopes gave mean oxidation rates of around 40% [*Tyler et al.*, 1997]. Oxidation rates in the laboratory experiments of *Holapfel-Pschorn et al.* [1985)]were 80%, but an attempt to study the oxidation rate in the field showed lower rates of 10% on 2 days and 40% on 2 days a month later. Other studies have also reported lower oxidation rates. In the Philippines, *Denier van der Gon and Neue* [1995] report a 15% oxidation rate for fields fertilized with urea and around 50% oxidation for fields fertilized with green manure. *Gilbert and Frenzel* [1995] reported a 34% oxidation rate in incubation studies. Our results of 45-60% oxidation are somewhere in the middle of the ranges reported but in the lower part of the range for comparable studies in Texas and Italy [*Sass et al.*, 1990, 1992; *Schütz et al.*, 1989].

It is difficult to compare the studies in further detail because the growing seasons were not the same and the work was done in widely differing environments. Nonetheless, the general patterns observed are similar in that oxidation is low in the beginning of the growing season and picks up around the time of tillering. It is high from then until the harvest time. The seasonal patterns we observed are similar to results reported by *Sass et al.* [1992] but somewhat different from the patterns seen in the work of *Schütz et al.* [1989] where the oxidation rate continued to increase rapidly until harvest time.

The production rates can also be compared among these studies. Since the fraction of methane oxidation is so high in the experiments of *Schütz et al.* [1989], especially toward the end of the growing season, the total production of methane is 100-180 mg m^{-2}/h^{-1} Similarly, *Holzapfel-Pschorn et al.* [1985] report production rates of 200-500 mg m^{-2} h^{-1} in the laboratory studies but only 20-35 mg m^{-2} hr^{-1} in the field study (with few data). Production rates from the work of *Sass et al.* [1990, 1992] are about 20 mg m^{-2} h $^{-1}$, and those from *Denier van der Gon and Neue* [1995] are 5-30 mg m^{-2} h^{-1}. Our results are 40-100 mg m^{-2} h^{-1} and therefore comparable to earlier studies. From these results it would seem that there is a loose correlation between production and oxidation. This may be the case since methane-oxidizing bacteria may grow more readily when larger amounts of methane are available. If this is the case, it would suggest a negative feedback that could limit methane emissions from rice fields, even under conditions prone to high methane production.

4. Discussion

The seasonally averaged emissions calculated from our experiments at Tu Zu tend to be somewhat on the high side of the range of whole season average methane emissions that have been reported in other studies. This is explained by the fact that, at this location, there is heavy use of organic fertilizers and the fields are inundated throughout the growing season. But it is possible that the high observed flux was also due, in part at least, to lesser oxidation at Tu Zu than at other locations. If the oxidation rate at Tu Zu had been 80% instead of 45-60% as calculated here, the average flux would be 10-15 mg m^{-2} h^{-1} instead of 30 mg m^{-2} h^{-1} that is measured.

If the oxidation rate in the world's rice fields varies between 50 and 90%, that alone would cause the emissions to vary over a factor of 5, all else being the same, and the difference of the oxidation rate between 60 and 80% causes a factor of two difference in the flux. This sensitivity of methane flux to the rate of oxidation can easily make different studies, even in similar locations, appear to be contradictory. The available data show a diversity of oxidation rates ranging from the order of 20 to 85%, which could contribute significantly to the variability of the observed emissions.

In this paper we have shown that the empirical observations of the processes that determine the emissions of methane from rice fields can be represented in a simple model. The model provides a framework for explaining observations and calculating the effects of oxidation and the production of methane in the soils. It shows that, by deliberately varying the planting density in the sample chamber, the changes of the observed flux can be used as tools to probe the processes that control methane emissions and the role of the rice plant in these processes. Finally, it suggests that the oxidation rates in the rice fields may differ from place to place, and may be the reason why the results of field studies have been inexplicably different. It is an important parameter that needs to be measured along with fluxes in all field experiments.

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