7-1-2008

Methane and Nitrous Oxide Emissions From Subtropical Rice Agriculture in China

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Citation Details

Khalil, M. A. K., M. J. Shearer, R. A. Rasmussen, L. Xu, and J.-L. Liu (2008), Methane and nitrous oxide emissions from subtropical rice agriculture in China, J. Geophys. Res., 113, G00A05.

Methane and nitrous oxide emissions from subtropical rice agriculture in China

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Received 5 April 2007; revised 10 October 2007; accepted 17 October 2007; published 22 July 2008.

[1] Emissions of methane and nitrous oxide, two significant greenhouse gases, were measured from rice fields at Qingyuan in Guangdong Province, China. The region has a subtropical climate which allows two crops of rice to be grown every year. The prevailing agricultural practices create a complex interaction between factors known to have a major effect on methane and nitrous oxide emissions from rice fields, namely, intermittent flooding and use of organic fertilizers. In this region, the farmers depend on nitrogen fertilizers and, at least in recent years, have only intermittently flooded their fields during the growing seasons. These factors tend to reduce methane emissions. But the rice straw and crop residues from the first crop of the year are plowed into the fields, providing a large addition of organic material under hot weather conditions favorable to quick decomposition during the second crop period. This, and the addition of farmyard manure, increases emissions of methane emissions from these fields. The results of the present study show that the effect of these competing factors and their timing lead to an average rate of emissions of 5 ± 2 and 6 ± 2 mg/m^2/h from the first crops for the 2 years when measurements were taken (2003 and 2004), and 12 ± 8 and 13 ± 8 mg/m^2/h from the second crop. Further, production measurements showed that during the 2 years of these experiments, the average production rates were about 27 mg/m^2/h for the first crop and 22–34 mg/m^2/h for the second crop, resulting in estimated oxidation rates of about 80% for the first crop and 50–60% for the second crop. The higher fluxes in the second crop therefore appear to be caused more by reduced oxidation than higher production. Nitrous oxide emissions, when they were detected, usually occurred within a few days after the application of nitrogen fertilizers. The seasonally averaged emissions were between 0.01 and 0.02 mg/m^2/h except in the first year when large emissions over one short period pushed the average upward.

Citation: Khalil, M. A. K., M. J. Shearer, R. A. Rasmussen, L. Xu, and J.-L. Liu (2008), Methane and nitrous oxide emissions from subtropical rice agriculture in China, *J. Geophys. Res.*, *113*, G00A05, doi:10.1029/2007JG000462.

1. Introduction

[2] Rice fields are a major anthropogenic source of methane, a greenhouse gas that can cause global warming. Over the years many field studies of emissions have been reported that have ranged from nearly no emissions to seasonal average rates of 40 mg/m^2/h, and hence the global emissions from rice fields have been estimated to be 20–100 Tg/a, although this range has narrowed in recent years [*Sass et al.*, 1999; *Khalil*, 2000; *Yan et al.*, 2003]. Two major factors that affect emissions are the amount of organic fertilizer applied and duration of inundation of the fields. We will report the results of a study conducted near Qingyuan, (23.68°N latitude, 112.95°E longitude), a city some 75 km NNW from Guangzhou, the capital of Guangdong Province in China.

[3] The fields we studied were under the ownership of local farmers who grew rice under the normal practices of the time and place. In this type of agriculture, fields are normally inundated in the early stages of growth. After the plants have grown, water is not always supplied, but rain can keep the fields intermittently flooded. Rice agriculture in the subtropical Qingyuan area follows this cycle with the additional feature of two rice crops a year. The first crop is usually planted in early April and harvested in mid July. During this time inorganic fertilizers are applied once or twice and organic manure is applied as available. The rice straw and stubble from the first crop of the year is plowed into the fields before the second crop is planted. This adds a considerable amount of organic material to the soils. The second crop is planted in early August and harvested in mid-November. Usually fertilizers, both nitrogen based and organic are applied 2–3 times during the growing season. These are

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0148-0227/08/2007JG000462

the prevailing conditions in the fields we studied. The agricultural practices of the region lead to a complex interaction between factors that tend to increase methane emissions such as the incorporation of rice straw from the first crop and factors that decrease emissions, such as lack of inundation during the later part of the second crop. The use of inorganic fertilizers and intermittent flooding, while reducing methane releases, must increase nitrous oxide emissions, another, even more potent greenhouse gas. We therefore also measured the emissions of N_2O which we will report here.

[4] There are few data on emissions from the complete double rice crop cycle under normal agricultural practices although they constitute a considerable fraction of the global rice production. In China during the 1990s it was estimated that 66% of the rice was grown in the double crop system, however, more recent estimates have put it at 56% [*Li*, 1992; *Frolking et al.*, 2002; *Xiong et al.*, 2002]. On a global scale such practices are common in the tropical regions where the conditions permit multiple crops and may account for a third of the rice grown (on the basis of data from *Xiao et al.* [2006] and *Frolking et al.* [2002]). Although the same conditions may not prevail in all regions where a double crop of rice is grown, the fields we have studied are likely to represent a significant category of rice agriculture with unique emission rates of the greenhouse gases because of the complex interactions of the variables mentioned. The flux and production measurements, environmental data and plant growth dynamics are included in the auxiliary material[1] for future use by the readers.

2. Experiments

[5] The experiments are similar to established methods at other sites [*Khalil et al.*, 1998a, 2008]. In essence the fluxes of gases are measured by placing a chamber over a small prescribed section of the rice field in which a base has been embedded before the rice is planted. Access to plots away from the edge of the field is by a boardwalk that is installed. This is necessary to prevent agitation of the soil that may compromise the flux measurements. The chamber is sealed at the bottom by the water in the base. Air samples are taken periodically from the chamber at intervals of 3, 6, 9, and 12 min for methane measurements and at 0, 30 and 60 min for nitrous oxide measurements. Samples for methane analysis were taken in plastic syringes and were analyzed using a Gow Mac GC/FID instrument located at the nearby laboratory of the local meteorological bureau (Meteorological Agency, Chinese Meteorological Administration). There was no instrument available for the measurements of nitrous oxide. Air samples were taken by pumping air into 0.8L stainless steel containers that have been used for many years in our experiments. The flasks were sent back to our laboratory at the Oregon Graduate Institute where N_2O was measured with a GC/ECD instrument [*Rasmussen and Lovelock*, 1983].

[6] Production of methane was determined by taking a soil core. The coring device has holes at various depths 2.5 cm apart from which we can extract a soil sample. This sample is placed in a glass flask and emulsified with paddy water if needed. The headspace is flushed and replaced with nitrogen to keep it anaerobic. Periodic samples of the headspace air are taken to determine the methane content over a period of a few hours to 24 h. The buildup of methane is taken to represent production [*Khalil et al.*, 2008].

3. Methane Emissions From the Two Crop Cycle

3.1. Emissions

[7] The observations of methane emissions are shown in Figure 1. Also shown are simultaneous measurements of production and water levels, both of which are related to emissions. In the first year (2003), three adjacent fields were sampled with three plots in each field. The results shown are the average emissions from each field on each day samples were taken. In 2004, samples were taken in one field with three replicate plots. The number of plots sampled and the frequency of measurements is constrained by the financial arrangements with the local contractors.

[8] There were two periods when the fields were fallow. The first was before the first crop in 2003 and the second is between the first and second crops of 2004. During the first fallow period in 2003 we see that there was practically no flux and no production for most of the time. There was also no standing water in these particular fields, which contributed to the lack of emissions. Soon after the fields were flooded, just before the crop was planted, methane production and flux were observed. The second fallow period shows the same results but is brief and there are only 3 days of measurements (2 weeks between 14 and 28 July 2004).

[9] In both years the first crop emitted much less methane than the second crop, a result similar to the findings reported earlier [*Wassmann et al.*, 1993; *Khalil et al.*, 1998b; *Cai et al.*, 2000]. We attribute this observation in part to the additional organic material available because the rice straw and residues from the previous crop are plowed into the fields before the second crop is planted. Air and soil temperatures are high when the second crop is planted thus accelerating the decomposition of organic matter in the soils. It is noteworthy that average temperatures during the entire growing periods for the two crops were similar but the seasonal change, shown in Figure 1 has a major effect on the emissions of methane (average soil temperatures: 29°C and 28°C for the two crops of 2003, and 27°C and 22°C for 2004). In both years the fields were intermittently inundated thus reducing emissions, especially in the later part of the second crop. The water levels were generally higher in the second year (2004) and may have contributed to the higher fluxes observed for both crops. Although, it can be seen that after a dry spell, there was water in the fields during the later part of the second crop in 2004, the flux never recovered. This may be due to the lags in the time it takes for methanogenic bacteria to grow and perhaps also due to the reduced readily decomposable organic material present at that stage of the annual cycle under cooler temperatures.

[10] There is considerable spatial variability. The precision of the measurements and the sampling methodology is high enough that the observed spatial variability can only come from the differences in the production, oxidation and transport process at different nearby locations. The coherence of emissions in the same field is measured by the

[1] Auxiliary materials are available at ftp://ftp.agu.org/apend/jg/2007jg000462.

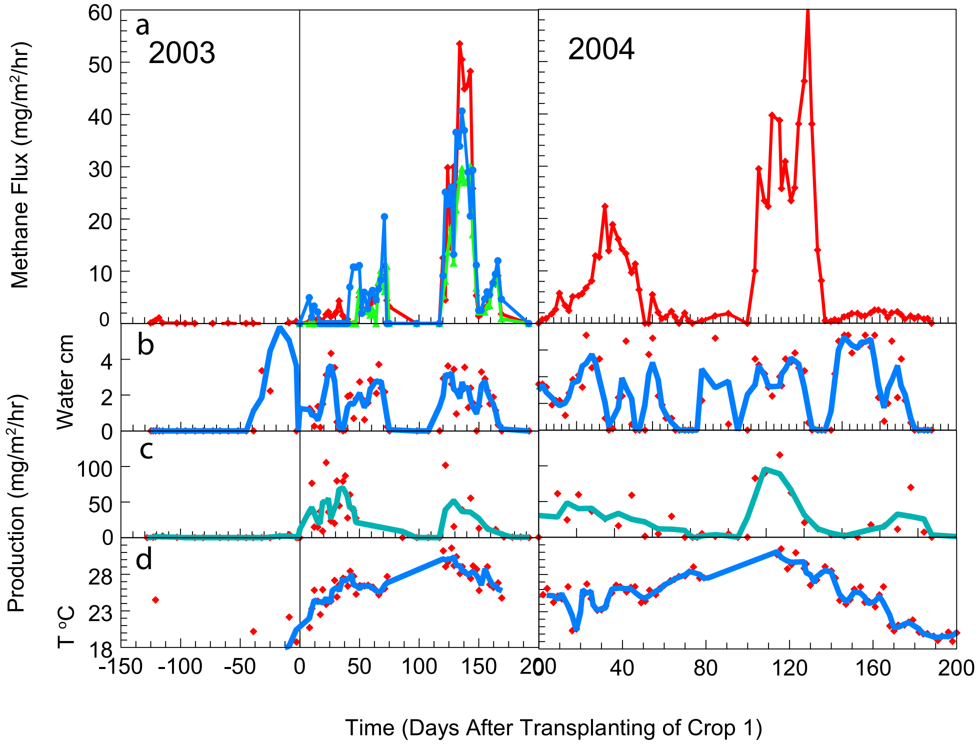

Figure 1. Methane emissions from rice fields in Qingyuan, Guangdong, China for (left) 2003 and (right) 2004. (a) Emissions from each field sampled. The points are an average of three plots in each field. Time is plotted in days after transplanting (DAT) which refers to the time when the plants are taken from the seedling beds and planted in the fields. The early period is 2 December 2002 to 3 April 2003 when the fields were fallow. The next period is when the first crop was grown (6 April to 13 July 2003; DAT = 0–98) and the period after that is for the second crop (1 August to 15 October 2003; DAT = 117–192). For the top graph on the left, the two periods are for the first and second crops (19 April to 9 July 2004 and 1 August to 15 October 2004; DAT = 0–81 and 103–207). (b) Water levels. The points are actual measured depths of water (centimeters), and the solid line is a smoothed version (three-point running average representing a span of about 5 days). (c) Methane production rates ($mg/m^2/h$) during the growth of the two crops in each year. The solid line is a smoothed version calculated as by a three-point running average. (d) Average soil temperatures measured at 5 and 10 cm depths.

correlation coefficient between time series of emissions from one plot to the next. There is generally good coherence but it varies from crop to crop and across the years. In 2003 the average correlation between all plots for each crop was 0.8 with a range of 0.5 to 0.99 ($r > 0$, at $p < 0.01$). In 2004 the average correlation was 0.6 for the first crop with a narrow range and 0.7 with a range of 0.5 to 0.8 for the second crop. This variability is manifested also in the integrated seasonal averages.

[11] The time series of fluxes shown in Figure 1 can be integrated over the growing season to obtain the seasonal average emission rate. The integrated flux is obtained by the following formula that is applied to all the sampled variables (V) of interest:

$$\langle V \rangle = {}^{1}\!/_{T} \int_0^T V dt \approx \left\{ \sum_{i=1}^{n} [V(t_i + \delta t_i) + V(t_i)] \left({}^{1}\!/_{2}\delta t_i\right) \right\} \Big/ \sum \delta t_i, \quad (1)$$

where t_i for i = 1... n are the times when samples are taken during the growing season and δt_i is the times between sampling.

[12] In 2003 samples for the entire growing season were obtained only from field 1. In field 2 the measurements were taken only during the last part of the season and hence could not be integrated to obtain a seasonal average. In field 3 there was a gap in sampling but with interpolation a seasonal average flux could be calculated. No significant emissions were observed during the fallow periods. The average emissions from the crops in 2003 in $mg/m^2/h$ were 5 ± 2 and 12 ± 8. In 2004 the average emissions were 6 ± 2 and 13 ± 8 $mg/m^2/h$ for the two crops. The ± values are the standard deviations of the data from the 9 plots in each crop during 2003 and the 3 plots in 2004. The details of fluxes in $mg/m^2/h$ given as triplicates for each field representing the plots sampled are: For 2003, fallow conditions, Field 1 = 0.1, 0, 0; Crop 1, 98 days, Field 1 = 4, 2.6, 2.1; Field 3 = 4.9, 6.6, 8.5; Crop 2, 90 days, Field 1 = 11.1, 19.5, 10.5; Field 2 = 7.3, 8.8, 10.8; Field 3 = 8.3, 15.9, 14.1. For 2004, Crop 1, 81 days, Field 1 = 7.9, 4.0, 7.3: Fallow between crops: 0.9, 0.4, 0.4; Crop 2, 97 days, Field 1 = 21.9, 7.5, 8.9. The results for the 2 years are therefore quite similar.

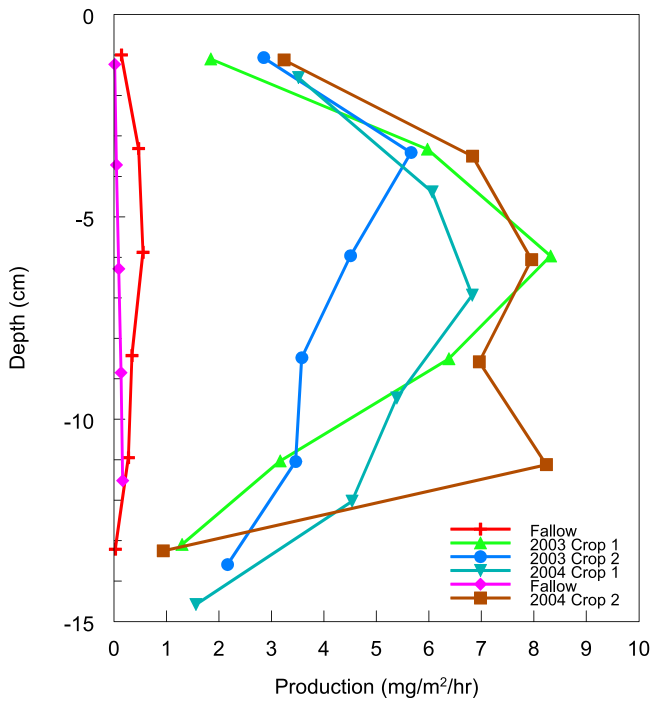

Figure 2. Methane production rates with depth for the periods studied during 2003 and 2004.

3.2. Production

[13] Production measurements were taken once a week, at one site that was moved around over the three fields under study. So the production record should be compared with the composite or average flux from all the fields during each year. In 2003 the last production measurement was taken on 23 May 2003 well before the harvest date of 13 July 2003. This causes the seasonally integrated production for the first crop of 2003 to be limited to about half of the growing season. For the second crop the sampling for production measurements ended about a month before harvest, but this has perhaps a minor effect on the results based on the patterns seen in 2004. The same sampling protocol was followed in 2004 except that samples span the full growing seasons of both crops. The sparse sampling was due to financial and labor constraints and some of the stoppages in 2003 were caused by the spread of the SARS virus at that time. The results therefore, are not as detailed as desired and only rough conclusions can be drawn.

[14] In Figure 2 we show the production of methane with depth. These profiles are constructed by taking the average production over the entire sampling period at each depth using equation (1). It is apparent that there is very little methane production during the fallow period even though water is present some of the time. This is due to a combination of factors including the intermittent rain fed flooding, the effect of plants and available organic matter. During the times when rice crops are in the fields, there is significant methane production that tends to follow the general features of the emissions, that is, low in the beginning and end of the growing season and high in the middle. This is reflected in the time series of depth profiles (not shown in the figures, but available in the data archive). Most of the production takes place between 3 and 10 cm although there is some depth uncertainty of ±2 cm because of the sampling methods. The production rates are similar for all crops except the second crop of 2003 when the production rates are lower for reasons not known.

[15] The production can be further summed over each depth to arrive at the time average production rate in the rice fields whereas in Figure 1 we showed the total production rate over all depths but for each sampling time (spatial average). For 2002–2003, during the fallow period, the first and second crops the production rates are respectively: 0.6, 40, 25 mg/m^2/h. For the first crop of 2004, the fallow period and the second crop the rates are: 27, 2 and 34 mg/m^2/h. It should be noted that in 2003 the value of 40 mg/m^2/h is the average production rate for only the first 47 days of the crop which we believe to be accurate for this period. If we calculate the average production rate for the first 47 days during the first crop of 2004 we get 36 mg/m^2/h which is in agreement with the value for 2003. The temporal pattern of the production is consistent with the periods of observed fluxes (Figure 1), although the exact relationship between production at a given time and the flux at the same time is complicated by the transport and oxidation processes.

3.3. Oxidation

[16] Oxidation is calculated from the seasonal average fluxes and production rates as: $f = [1 - \Phi/P]100\%$. Here f is the percent of methane that is oxidized relative to the amount produced, and Φ and P are the seasonally averaged flux and production rates. The results are shown in Figure 3.

[17] Oxidation rates during the growing seasons are quite high at about 80% for the first crops but lower for the second crops (50–60%). In 2003 we have used the averaged flux only for the first 50 days to be consistent with the production measurements, which as mentioned earlier, were only taken over this period. This result is consistent with our findings at Tu Zu and Jinsha which suggest that the oxidation rate is reduced when there is a large quantity of organic matter in the fields, or perhaps more specifically, rice straw and stubble [*Khalil et al.*, 2008]. In the Qingyuan experiments we see that the seasonally averaged production rates are not much different between the first and second crops, but the fluxes are. Since oxidation is a calculated quantity in our experiments that balances the production and the emissions, we attribute most of the increase of emissions during the second crop, compared with the first, to be due to lower oxidation rather than higher production. A similar conclusion was reached for the work at Jinsha which has different agricultural practices [*Khalil et al.*, 2008]. This finding is unexpected and the possible mechanistic explanations are still under investigation in greenhouse studies.

4. Nitrous Oxide Emissions

[18] There are several key differences between the strategies for measuring nitrous oxide emissions from rice fields compared with the measurement of methane emissions. The flux of nitrous oxide is often some 2–3 orders of magnitude lower than methane. This requires a longer exposure time with the chambers to properly measure the accumulation rates. Long exposures have the potential for affecting the plants due the increase of temperature inside

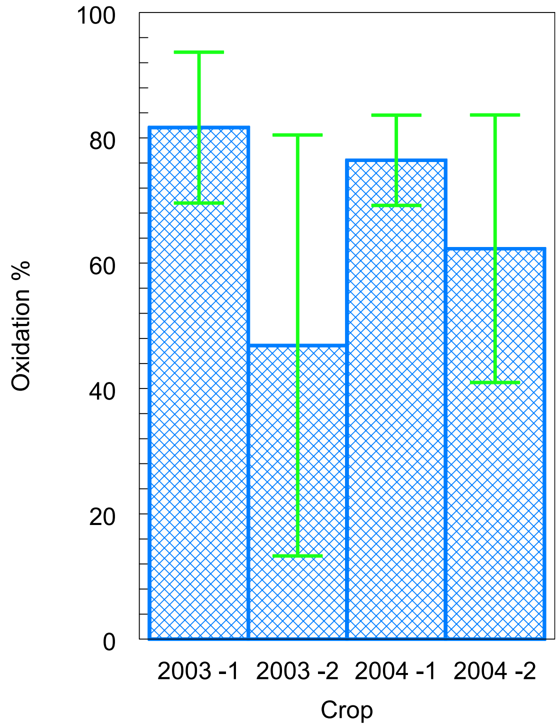

Figure 3. Calculated fraction of methane oxidized during the rice growing seasons of 2003 and 2004. The bars represent the ranges of results from fields where both production and flux measurements were taken.

the chamber and the accumulation of gases. In our experiments the exposure times were about 60 min. Second, experiments have shown that nitrous oxide emissions tend to be sporadic and generally high soon after the application of nitrogen containing fertilizers. Therefore the frequency of sampling has to be high around fertilization to capture the impact of these events. And third, the nitrogen applied to the rice fields during the growing season can produce and release nitrous oxide after the crop has been harvested. Thus measurements taken only during the growing season may not reflect the full effect of the fertilizer used for that crop. The data from our experiments does not completely resolve the second issue, and does not address the third issue at all. Because there was no field instrument available, only a limited number of samples could be collected for shipment back to our laboratory. Nonetheless, we took samples as soon as it was practical for a few days after the fertilizer applications and indeed high fluxes were observed only during these times as shown in Figure 4; we have included the methane fluxes for perspective. The results show that emissions of nitrous oxide are high following fertilizer applications as represented by the "spikes" in Figure 4.

[19] For the first crop of 2003 a compound fertilizer (N:P:K, 15%:6%:8%), urea, and additional potassium was applied on 15 April, which led to the small peaks seen in the early times in Figure 4. Heavier doses of this fertilizer were applied on 1 and 13 May. The earliest sample close to the time of the fertilizer application is 3 May but on this day no significant flux of nitrous oxide was observed. The big peak is on the day after the 13 May application (15.8 mg/m^2/h) but by 16 May, the date of the next sampling the fluxes had dropped to low levels (0.1 mg/m^2/h). This is the nature of the nitrous oxide emissions. For the second crop of 2003 the fertilizer applications were not recorded and also our sampling did not catch any periods of significant emissions. In 2004, urea was applied on 26 April but the closest sample 2 days later shows no nitrous oxide flux. The next application was on 2 May and included the compound fertilizer (NPK 15:15:15), and although the nearest sample is on 5 May, modest fluxes are observed as seen in Figure 4 (0.5 mg/m^2/h). On 9 May, compound fertilizer was again applied, sampling later on the same day shows the peak of 0.2 mg/m^2/h in Figure 4. In the second crop, urea was applied on 13 August but samples were collected later on 16 and 18 August, both of which show significant emissions as seen by the highest value of 0.14 mg/m^2/h in Figure 4. Some modest fluxes are seen after the application of urea on 22 August. These results show qualitative relationships between the application of nitrogen fertilizers and emissions over the next several days. There is no consistent pattern of how long the flux lasts or how large it is. At most other times almost no or very low emission of nitrous oxide was observed. The average emissions in μg/m^2/h were found to be 100 for the fallow period of 2003, 480 and 1180 for the first and second crops. In 2004 the average emissions were 640 and 1270 for the two crops and 60 for the intermediate fallow period. There is good agreement between the 2 years. It is likely that the estimates of emissions during the growing season for these fields are fairly accurate because we have captured the aftermath of fertilizer applications with our sampling even though we have sparse data in between when there are generally no emissions.

5. Conclusions and Perspectives

[20] Global methane emissions from rice fields are believed to be a major anthropogenic source that has contributed to

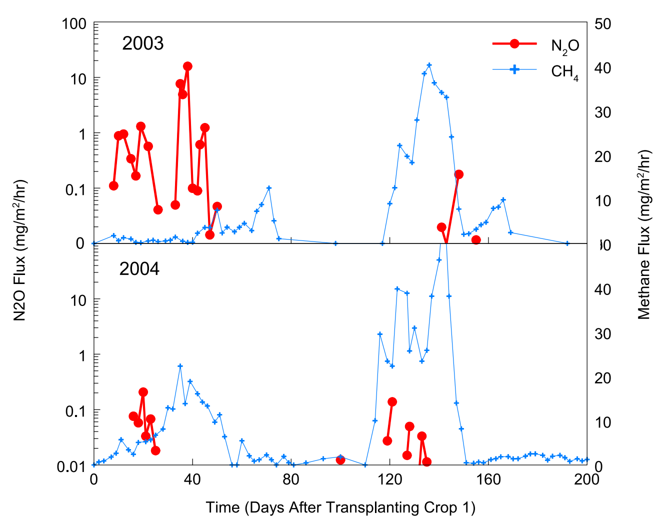

Figure 4. Emissions of nitrous oxide from rice fields at Qingyuan, Guangdong, China (circles). Methane emissions are replotted for comparison (crosses). Owing to logistic constraints, the measurements were taken at a low frequency, but periods soon after the additions of nitrogen fertilizers were covered. The large peaks generally correspond to fertilization times.

the present levels that are some two and a half times higher than pre-industrial times. In recent years however, methane emissions from rice fields are likely to have declined, especially in China where agricultural practices have shifted to intermittent flooding and a greater reliance on nitrogen fertilizers [*Li et al.*, 2002; *Khalil and Shearer*, 2006]. This situation probably exists at Qingyuan, however, organic compost is still used in addition to the nitrogen fertilizers and the second crop gets more organic material from the residues of the first crop, which are plowed into the fields. The organic inputs increase emissions and intermittent flooding decreases them. The interplay in the timing of these factors creates an intermediate level of emissions from rice fields in this area. These dynamics are seen in the data we have presented.

[21] One issue that can affect global estimates is emissions from fallow fields, which is not usually taken into account. In many rice growing areas, there are periods, sometimes several months at a time, when the fields are fallow. In Guangdong Province during the winters the fallow fields may be a source of methane since they are usually inundated owing to rainwater, it is not cold and a secondary crop of rice grows from the stubble of the previous harvest. Other tropical areas also have similar conditions between crops. In the Qingyuan experiments we had hoped to address this issue, but the field and location that was chosen for us did not have the right conditions of water and secondary crop. In the second experiment when the field was fallow between the two crops of 2004 the flux was about 10–15% of the flux from the previous crop. This may be an indicator of the significance of fallow fields.

[22] This experiment is one of a large collection from many independent studies that can be used to better define the global emission rate and the country emissions that are needed to control emissions of methane to limit global warming. The so-called "upscaling" of field measurements to the large scales remains uncertain and complex. The difficulties are reduced as more data are brought to bear on the estimates, especially from locations that have unique characteristics and are likely to contribute significantly to the regional and global budgets, as is the case of Qingyuan.

[23] **Acknowledgments.** We thank Christopher Butenhoff and Zhengqin Xiong at Portland State University for discussions. This work was supported by the Office of Science (BER), U.S. Department of Energy, grants DE-FG03-01ER63262 and DE-FG02-04ER63913. Additional support was provided by the resources of the Biospherics Research Corporation, the Andarz Co., and as part of the Working Group on Quantification of CH_4 emissions from land ecosystems, supported by the National Center for Ecological Analysis and Synthesis, a Center funded by NSF (grant DEB-0553768), the University of California Santa Barbara, and the State of California.

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