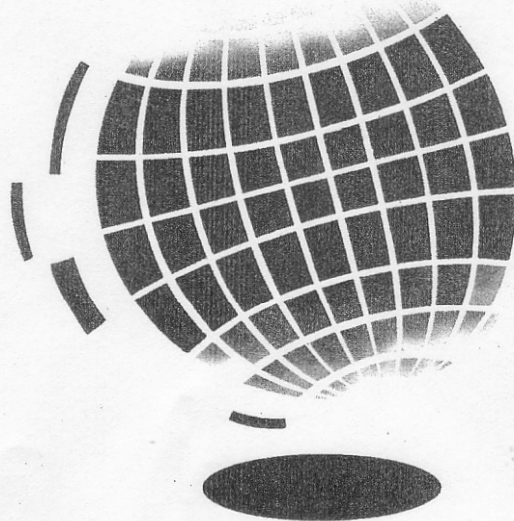


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SOIL REDOX WINDOW WITH MINIMUM NITROUS OXIDE AND METHANE PRODUCTION: ITS APPLICATION IN A RICE FIELD

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Abstract: In this paper, we present the results of (1) a laboratory experiment using a soil microcosm system to study nitrous oxide (N₂O) and methane (CH₄) production at different redox potential (Eh) conditions and their relative contributions to cumulative global warming potential (GWP), and (2) a field experiment under four different irrigation and organic matter (OM) managements to find a best management practice to mitigate cumulative GWP from CH₄ and N₂O emission in a irrigated rice field. In the laboratory study, we found a redox "window" of +180 to -150 mV where both N₂O and CH₄ productions were low. Soils in this redox window were reducing enough to favor a complete denitrification with nitrogen gas (N₂) as end product, but the reducing intensity was not enough to initiate significant methanogenesis. The field study showed that non-flooding conditions maintained by controlled irrigation effectively reduced CH₄ emission from the fields with (by 79 %) and without (by 71 %) OM addition. Strictly reducing zones (Eh < -150 mV) that were favorable for methanogenesis developed at the lower depths of the soil in the non-flooded fields, which facilitated less CH₄ production and more CH₄ oxidation through the soil surface. Stimulated N₂O emission in the non-flooded soils offset part of the CH₄ emission reduction, especially in the fields without OM addition. In conclusion, non-flooding treatment reduced the cumulative GWP by only 47 % in the field without OM addition, but by 73 % with OM addition. The best management practice proposed in this study is to keep the field non-flooded with OM addition. It shows a minimum GWP from the rice fields, and no decrease in rice yield.

Key words: greenhouse gases, global warming potential, rice field, redox, mitigation

Introduction

Agriculture accounts for about 20 % of the projected anthropogenic greenhouse effect, producing about 50 and 70 %, respectively, of overall anthropogenic CH₄ and N₂O emissions (IPCC, 2001). Only next to carbon dioxide (CO₂), CH₄ and N₂O are the most important atmospheric trace gases contributing to global warming. Updated GWP calculation from the Intergovernmental Panel on Climate Change (2001) showed, in a 100-year time horizon, that 1 kg of CH₄ and N₂O are equivalent to 23 and 296 kg of CO₂, respectively, in radiative forcing of the global warming.

Nitrous oxide can be produced from nitrification under aerobic conditions, and denitrification under moderately

reducing conditions. Significant CH₄ production generally needs strictly reducing conditions. The intensity of soil reducing condition can be instrumentally measured as soil oxidation-reduction potential (redox, Eh). Aerobic (high Eh) and anaerobic (low Eh) conditions may be dominant for a certain period in rice soils depending on irrigation and drainage practice, making rice fields a major source of CH₄ during the flooded season, and an important source of N₂O during the non-flooded season (Abao *et al.*, 2000; Cai *et al.*, 1997; Chen *et al.*, 1997; Tsuruta *et al.*, 1997).

Agriculture can play an important role in mitigation of greenhouse gas production and emission, especially for CH₄ and N₂O, because of the accessibility of direct

management of this ecosystem. Both CH_4 and N_2O production are functions of microbiological activities under different soil redox potential (Eh) conditions. Mid-season drainage has been shown the most effective approach to reduce CH_4 emission from flooded rice fields, but with a potentially adverse effect of stimulating higher N_2O emission (Bronson *et al.*, 1997, Wassmann *et al.*, 2000). The different Eh conditions required for N_2O and CH_4 formation and the trade-off pattern of their emissions as found in rice fields makes it a great difficulty to abate the production of one gas but not to enhance the production of the other. Cumulative GWP from both N_2O and CH_4 emission needs to be considered in evaluating the efficiency of different mitigation approaches proposed.

Two studies are included in this paper. In the laboratory study, eight different rice soils were incubated using the soil microcosm technique to determine an optimum soil Eh range with minimum N_2O and CH_4 production. In the field study, different managements of irrigation, OM application, and fertilization were evaluated to propose a best management practice with minimum GWP contribution from the rice field.

Table 1. Selected characteristics of the sample soils (Yu and Patrick, 2004)

| Soil | pH | mg kg ⁻¹ | | | | |
|-------------|-----|---------------------|-------|-----|-----|-----|
| | | OM | T. N. | Fe | Mn | S |
| Arkansas | 6.0 | 14.6 | 0.7 | 134 | 105 | 13 |
| California | 6.7 | 40.8 | 1.6 | 224 | 107 | 45 |
| Louisiana | 7.3 | 16.7 | 0.7 | 68 | 19 | 11 |
| Mississippi | 7.7 | 25.3 | 1.0 | 71 | 9 | 12 |
| Texas | 5.1 | 25.4 | 1.1 | 115 | 35 | 38 |
| China | 5.6 | 46.4 | 2.7 | 190 | 102 | 66 |
| Indonesia | 5.3 | 23.7 | 1.0 | 211 | 280 | 65 |
| Thailand | 4.7 | 25.8 | 1.2 | 173 | 40 | 190 |

Soil pH was measured in soil-water (1:1) slurry. Soil total OM was measured colorimetrically after oxidizing with $\text{K}_2\text{Cr}_2\text{O}_7$ and concentrated sulfuric acid. Soil total N (T. N.) was analyzed in dry combustion by a Leco N analyzer. Soil extractable Mn, Fe, and S contents were analyzed by inductively coupled plasma (ICP) after extracting with DTPA (diethylene triamine pentaacetic acid) solution (for Mn and Fe in form of soluble and labile solid phases), and with ammonium acetate and acetic acid solution (for S mostly in form of sulfate), respectively.

Materials and methods

Laboratory Microcosm Incubation Study

Sample soils

Eight rice soils (top 20 cm) were collected for this study, including from five from major rice-cultivating states in US (Arkansas, California, Louisiana, Mississippi, and Texas), and three from international regions (Hangzhou/China, West Java/Indonesia, and

Pathumthani/Thailand). The soils were air-dried, sieved (1mm), thoroughly mixed and stored at room temperature (20 °C) before the experiment. Soil characteristics of interest were analyzed and are provided in Table 1.

Soil incubation and measurement

Soils were incubated using a soil microcosm system where soil Eh and pH could be monitored, and gas samples could be taken as needed (for detailed description, see Patrick *et al.*, 1973, Yu and Patrick, 2003). For each of the eight rice soils, a single soil suspension was made by adding 400 g soil into a 2300 ml Erlenmeyer flask with 1600 ml of deionized water. To each soil suspension, 4 g ground rice straw was amended as an additional source of OM, and potassium nitrate (KNO_3) was amended at 50 mg $\text{N}\cdot\text{kg}^{-1}$ soil to provide an additional source of nitrate. Soil incubation was started at oxidized conditions with no further O_2 supply thereafter. Gas samples were frequently taken, whenever Eh in the microcosm system changed by more than 10 mV, to monitor the dynamics of gas production under different Eh conditions. Gas production rate was determined by the gas accumulation in a 1-hr enclosure of the microcosm after flushing it with pure N_2 gas for 2 hrs.

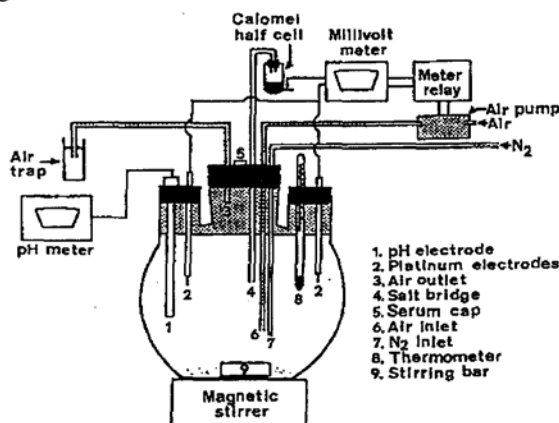


Fig. 1. Soil redox microcosm system (modified from Patrick *et al.*, 1973)

Rice Field Study under Different Managements

Study site and treatment

The field experiment was conducted at Shenyang Experimental Station of Ecology, Chinese Academy of Sciences (41°32'N, 122°23'E). Soil OM contents were 2.12 and 1.51 % for the field with and without annual application of organic manure, respectively. For about 10 years, organic manure (mixed manure of pig and poultry)

has been applied at 30 t·ha⁻¹ each year before rice cultivation in spring. Soil (top 20 cm) pH values fluctuate within 6.4 and 6.7 regardless of OM addition. Other soil characteristics were reported in the previous publications but were highly variable at different rice growing seasons (Chen *et al.*, 1995, 1997).

A major regional cultivar of rice, Liao Kai 79, was cultivated for this study with a single growing season of about 120 days. Rice seedlings were transplanted the second day after flooding the fields (late May). The fields received basal fertilization of (NH₄)₂HPO₄ at 290 kg·ha⁻¹ within 3 days after transplanting. Urea was broadcasted at tillering (156 kg·ha⁻¹, late June) and again at heading (73 kg·ha⁻¹, late August) stage. The fields were drained in late September for rice harvest in early October. Four treatments with 2 replicates were applied to eight experimental plots (4 × 6 m each): (A) No OM addition, flooded, (B) No OM addition, non-flooded, (C) OM addition, flooded, and (D) OM addition, non-flooded. Major management practice in this region is as treatment (C). The flooded fields kept 5 to 10 cm standing water, while the soil surface in the non-flooded fields was wet with water table fluctuating between soil surface to approximately 5 cm below ground. The same inorganic fertilization practice was applied to all study plots.

Soil Eh measurement

Redox potential (Eh) in the soil profile of top layer (22 cm), the plow layer where most of the microbial activities take place, was measured using a platinum (Pt) electrode cable with a calomel reference electrode connected to a portable millivolt meter. Before installation in the fields, all Pt electrodes were pre-checked (deviation < 10 mV) with standard Eh values at pH 4.0 and pH 7.0 buffer solutions saturated with quinhydrone (Bohn 1971). A Pt electrode cable consisted of 6 single Pt electrodes that were located at depths of 1, 2, 4, 8, 14, and 22 cm below the soil surface. Two electrode cables were permanently installed in each differently treated plot during the study period.

CH₄ and N₂O emission in the fields

Methane and N₂O emissions in the rice field were measured at least once a week using a static chamber technique. The chamber was made of Plexiglas in dimension of 1 (H) × 0.8 (L) × 0.8 (W) m. The chamber was manually operated to cover a base unit that was permanently installed in the field, which was sealed with

water and covered both rice plants and soil. Gas samples were collected using a 30 ml syringe at 0, 20, and 40 min upon chamber closure.

Gas Sample Analysis

For both the laboratory and field studies, CH₄ and N₂O concentrations in the gas samples were analyzed using gas chromatography (GC) with a flame ionization detector (FID) for CH₄, and an electron capture detector (ECD) for N₂O, respectively, in calibration with certified standard gases.

Calculations and Statistical Analysis

For both the laboratory and field studies, CH₄ and N₂O emissions were determined by the slope of a linear regression of their concentrations against the enclosure time. Redox potentials (Eh) were reported as relative values to the standard H₂ electrode by adding 247 mV (the correction factor for calomel reference electrode at 20 °C) to the observed instrument readings. All Eh data were further adjusted to their corresponding values at pH 7.0 that were calculated according to the inverse relationship of Eh and pH as described by the Nernst equation (59 mV per pH unit, Bohn, 1971). Global warming potentials in CO₂ equivalents over a 100-year time horizon were calculated by taking mass conversion factors of 23 for CH₄, and 296 for N₂O, respectively (IPCC, 2001).

Statistical analysis ($\alpha = 0.05$) was conducted using SAS (SAS Institute Inc.). Analysis of variance (ANOVA) using PROC GLM was conducted to calculate the least significant difference (LSD) between the means of different treatments. Simple linear regressions using PROC REG were conducted to test if the slope of a regression was significantly different from zero.

Results and Discussion

Redox Window with Minimum CH₄ and N₂O Production

The studied rice soils showed a large variation in pH and Eh change during the incubation from aerobic to anaerobic conditions (data not shown). However, productions of N₂O and CH₄ in the soils showed a quite similar pattern when they were plotted against Eh, even though their production rates varied significantly under the similar incubation conditions due to large variations in soil characteristics (Table 1). Nitrous oxide production, probably from both nitrification and denitrification, began immediately after the incubation started, but was mostly

produced in an Eh range of +400 to +200 mV. Only a small amount of N_2O was present below +180 mV, due to stronger reduction of N_2O to N_2 at lower Eh (Masscheleyn *et al.*, 1993). The critical Eh value to initiate a significant CH_4 production was about -150 mV at neutral pH (Masscheleyn *et al.*, 1993; Wang *et al.*, 1993; Yu *et al.*, 2001). Although significant CH_4 production occurred at different time of the incubation for each soil, for all soils it happened only when the soil Eh decreased below -150 mV. Thus, major CH_4 production occurred in a narrow Eh range of -150 to about -300 mV, and the production rate increased greatly with Eh decrease within this Eh range. The results delineated a wide Eh range where the cumulative GWP from N_2O and CH_4 reached a minimum (Fig. 2). In this Eh range, soils were reducing enough to favor complete denitrification with N_2 as end product, but were still oxidizing enough to inhibit significant methanogenesis. The Eh "window" with minimum GWP contribution slightly varied for each soil, but generally located between +180 and -150 mV at pH 7.

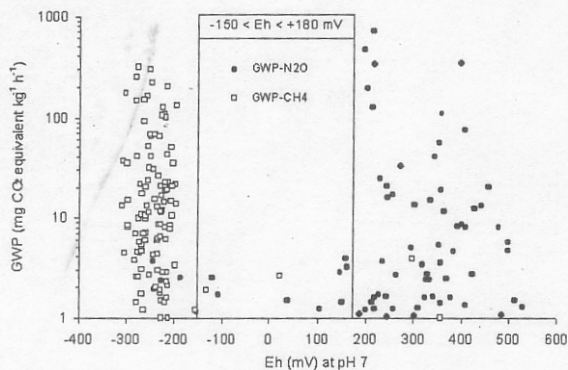


Fig. 2. Soil Eh range with minimum GWP contribution (modified from Yu and Patrick, 2004)

Relative contributions of N_2O and CH_4 in the cumulative GWP at different Eh range were highly variable for each soil. On average of the eight soils, 57 % of the total GWP was produced when Eh was higher than +180 mV, and 38 % when Eh lower than -150 mV. Only 5 % of the total GWP was produced in the Eh range of +180 to -150 mV that accounted for about 40 % of the entire Eh range studied. The results provide an optimistic perspective in mitigating GWP in rice soils if the soils could be maintained in this favorable redox range. Therefore, a field study including management of irrigation, OM addition and fertilization was setup to test such a hypothesis.

Effect of Field Management on Soil Redox Status

Soil Eh in the studied rice fields generally spanned a range of +700 to -300 mV. Unlike homogeneous soil suspensions, both oxidizing and reducing conditions existed simultaneously in the rice fields, due to the heterogeneous nature of the field, slow diffusion of oxygen (O_2) in water and soil, and presence of rice plant. Soil redox status under the different treatments showed a similar seasonal pattern (Fig. 3). Flooding the field (A and C) and adding OM (C and D) facilitated the development of reducing conditions in the soils. After drainage, soil Eh in the upper layers of the field increased up to +450 mV in just a few days. Strictly reducing conditions (Eh < -150 mV) that were favorable for methanogenesis generally developed at 3 periods after rice transplanting: day 50 to 60 (early), day 67 to 77 (middle), and day 95 to 105 (late). Non-flooding conditions (B and D) provided more aeration to the top layers of the fields than the flooded fields (A and C), and consequently resulted in the strictly reducing zones (Eh < -150 mV) being developed 4 or 5 cm deeper than in the flooded fields.

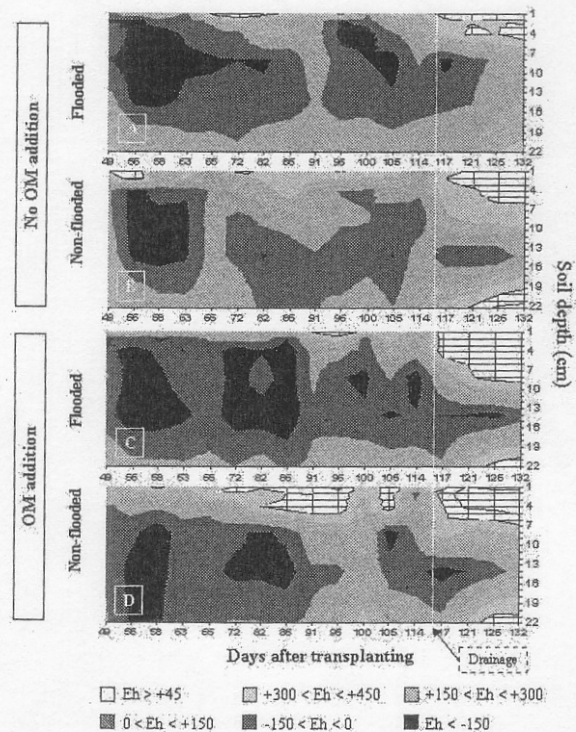


Fig. 3. Soil Eh under different treatments (modified from Yu *et al.*, 2004)

Treatment: (A) No OM addition, flooded; (B) No OM addition, non-flooded; (C) OM addition, flooded; (D) OM addition, non-flooded.

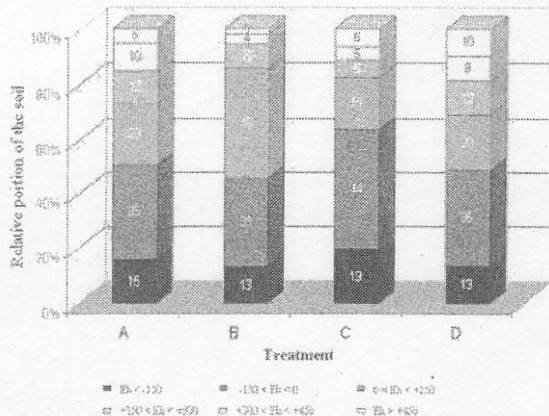


Fig. 4. Relative portion of the soil at each Eh range (Yu *et al.*, 2004)

Treatment: (A) No OM addition, flooded; (B) No OM addition, non-flooded (C) OM addition, flooded; (D) OM addition, non-flooded.

Irrigation and OM management practice showed a significant impact on the soil redox status. Under the flooding conditions, the bulk of the soil with Eh < 0 mV accounted for 63 and 50 % of the field (top 22 cm) with (treatment C) and without (treatment A) OM addition, respectively. The non-flooding management enlarged the bulk of the soil with higher Eh, and in compensation reduced the portion with lower Eh (Fig. 4). The lower water table in the treatment B and D aerated the soil surface layers, thus strictly reducing conditions developed at deeper layers of the soil profile where reducing intensity was strong enough to initiate a significant CH₄ production.

Methane production mostly occurs in soil microenvironments where the Eh values are lower than what is normally measured (Neue, 1997). However, soil Eh measurement can qualitatively indicate the redox status in the soil microenvironment, especially in flooded soils where soil aggregates tend to break down. When measured soil Eh is lower (lower O₂ partial pressure in the soils), the soil microenvironment is more reducing, and vice versa (Tiedje *et al.*, 1984). Soil OM is the major electron donor in various soil redox reactions, and is the driving force of developing soil-reducing conditions. Release of new OM from the rice root and degradation of the dead rice roots significantly contributed to developing the middle and late strictly reducing zones, respectively (Schutz *et al.*, 1989). Studies using isotopes indicated that a large fraction of the OM that supported methanogenesis was derived from recently fixed carbon (Minoda and Kimura 1994). In the fields without receiving OM where the rice was of poor growth (with less rice yield, see Table 2), less reducing

zones developed in the middle season, probably due to less root exudates or dead root tissues from the rice plants. Oxygen diffusion through the rice plant might play a significant role in elevating the soil Eh level between the three strictly reducing periods of the soils.

Effects of Field Management on CH₄ and N₂O Emission, and on Rice Yield

Major periods with higher CH₄ and N₂O emission remained the same under the different treatments (Fig. 5), which also agreed quite well with the previous measurements in the same rice field where more complete seasonal variations of CH₄ and N₂O emission were recorded (Chen *et al.*, 1995 and 1997). The three periods with major CH₄ emission in the rice fields corresponded to the seasonal development of the strictly reducing conditions in the soils (Fig. 3), indicating a close relationship between soil Eh and methanogenesis activity. The highest CH₄ emission was found in the treatment C (OM addition, flooded), and the lowest in the treatment B (No OM addition, non-flooded). Flooded fields showed low N₂O emission, and occasional consumption of ambient N₂O. Nitrogen fertilization during the rice-growing season stimulated higher N₂O emission, especially in the non-flooded fields (Fig. 5). Drainage at the end of the season also resulted in higher N₂O emission, but meanwhile terminated CH₄ emission in the fields.

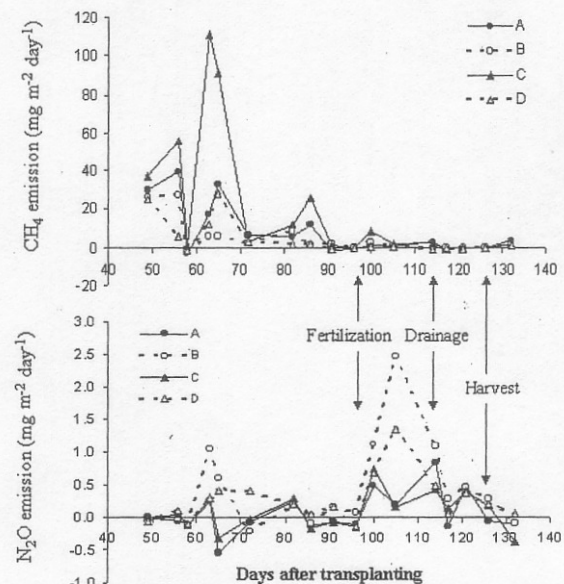


Fig. 5. CH₄ and N₂O emissions in the rice field (modified from Yu *et al.*, 2004)

Treatment: (A) No OM addition, flooded; (B) No OM addition, non-flooded; (C) OM addition, flooded; (D) OM addition, non-flooded.

Major results of this field study are summarized in Table 2. When the rice fields were flooded, no addition of OM reduced the CH₄ emission by 57 % with no difference in average N₂O emission. Without OM addition, non-flooding management reduced the cumulative GWP from both CH₄ and N₂O by 46 %, but about one third of the CH₄ emission reduction (176.6 CO₂ equivalent m⁻²·d⁻¹) was offset by the increase of N₂O emission (56.2 CO₂ equivalent m⁻²·d⁻¹). In the OM added fields, non-flooding management reduced the cumulative GWP by 72 % as the result of the CH₄ emission reduction by 458.2, and the N₂O emission increase by 29.6 CO₂ equivalents m⁻²·d⁻¹. Although the local traditional management (treatment C) showed the highest GWP, appropriate irrigation (e. g. treatment D) could effectively reduce the cumulative GWP by a significant reduction of CH₄ emission with little enhancing N₂O emission from the rice field. More O₂ was available for the soils under the non-flooding conditions, thus a larger portion of the soil OM converted to CO₂, instead of converting to CH₄ by methanogenesis under the strictly anaerobic conditions.

Soil OM played an important role in rice yield (Table 2). When additional OM was provided, rice plants showed a more healthy growth as observed in the field and higher yield at harvest regardless of irrigation conditions. This was probably due to the additional nutrients (e. g. phosphorus) in the organic manure and a generally beneficial effect of OM on soil fertility. Compared with the local traditional management (treatment C), the rice yield was significantly decreased ($P < 0.05$) by 16 % if no additional OM was applied (treatment A), and by another 9 % if the field was non-flooded (treatment B). Therefore, addition of OM should be included in the field management practice, at least for this region, because of the top priority for higher rice yield. With OM addition, non-flooding treatment (D) showed no significant ($P > 0.05$) reduction in rice yield (5 %).

Table 2. Summary of the rice field study results

| Measurement | Treatment ^a | | | | LSD ^b (n) |
|--|------------------------|-----------|------------|-----------|----------------------|
| | A | B | C | D | |
| CH ₄ (mg·m ⁻² ·d ⁻¹) | 10.80 (95) | 3.12 (51) | 25.20 (98) | 5.28 (75) | 13.32 (17) |
| N ₂ O(mg·m ⁻² ·d ⁻¹) | 0.04 (5) | 0.23 (49) | 0.04 (2) | 0.14 (25) | 0.15 (17) |
| GWP | 260 | 140 | 591 | 163 | 306 (17) |
| Yield (t·ha ⁻¹) | 9.7 | 8.8 | 11.5 | 10.9 | 1.6 (4) |

a. Treatment: (A) No OM addition, flooded; (B) No OM addition, non-flooded; (C) OM addition, flooded; (D) OM addition, non-flooded. Data in parenthesis denoted the relative contribution (%) of CH₄ or N₂O in the cumulative GWP.

b. It represents significance in statistics if the difference between the means is greater than LSD.

Proposed Field Management Practice and a Possible Modification Option

Irrigation showed a critical impact on controlling the soil redox status, and on CH₄ and N₂O production and emission. The best management practice proposed in this field study, in order to reduce the cumulative GWP from the rice field without decreasing the rice yield, is to keep the field non-flooded with OM addition (treatment D). This is a minor modification of the current local management practice (treatment C), which makes it more feasible in application. Less water used for the non-flooded fields may provide some additional benefits to the farmers with less labor, water, and electricity expenses. This management approach may be also feasible for the rice fields with no information available on seasonal variation of CH₄ and N₂O emission, because irrigation control is adjusted according to the wetness of the soil surface, instead of any information of instrumental measurement.

Increasing N₂O production and emission can significantly offset CH₄ reduction during the drainage or non-flooded practice in mitigating CH₄ emission, resulting in low efficiency in overall GWP reduction. However, higher N₂O production and emission is always associated with N-fertilization during the rice-growing season. Our results also suggest a possible modification to the currently proposed management practice (treatment D) to reduce the short-term higher N₂O emission by temporarily flooding the fields upon fertilization (only applied to ammonium-based fertilizers). Such temporary flooding condition may prevent the undesirable nitrification activity that makes the fertilizer N unstable, and limits N₂O production and emission as found under the flooding conditions (Fig. 5, and Table 2). This modification will not affect the feasibility of the proposed field management, but how long the field should be flooded after fertilization, without introducing significant CH₄ emission, deserves further investigation.

Conclusions

The wide Eh range (+180 to -150 mV) with minimum N₂O and CH₄ production found in the laboratory study can be used in the rice fields to achieve a maximum reduction of the cumulative GWP from CH₄ and N₂O. Although soil Eh in the entire soil profile of the rice fields cannot be regulated within such an Eh range, proper irrigation management can make the soil Eh distribute in a desirable way to largely reduce CH₄ emission with least enhancing

N₂O emission. Non-flooded management didn't show any water stress to the rice plant growth, and the rice yield was not decreased in this field trial. Best management practices should be field specific in general, however, it is recommended to apply this management approach to the rice fields where no specific information on seasonal variations of CH₄ and N₂O emission is available. Effective mitigation of GWP from rice fields will make rice ecosystems environmentally sustainable.

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