# Mitigation Options for Methane, Nitrous Oxide and Nitric Oxide Emissions from Agricultural Ecosystems<sup>①</sup>

Zheng Xunhua (郑循华), Wang Mingxing (王明星), Wang Yuesi (王跃思), Shen Renxing (沈壬兴), Li Jing (李 晶)

LAPC, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029

J. Heyer, M. Koegge, H. Papen Fraunhofer Institute for Atmospheric Environmental Research. Garmisch-P, Germany

> Jin Jisheng (金继生), Li Laotu (李老土) Wuxian Institute of Agricultural Sciences, Suzhou 215128

(Received December 22, 1998; revised October 14, 1999)

#### ABSTRACT

An experimental study on mitigation of greenhouse gas  $(CH_4, N_2O \text{ and } NO)$  emission has been conducted in a typical cropping system of Southeast China for 4 years. By simultaneous measurement, the  $CH_4$ ,  $N_2O$  and NO emission fluxes from rice-wheat rotation fields, effects of fertilization, water management, temperature and soil moisture were investigated. Temperature, fertilization and water status were found to be the key factors to regulate  $CH_4$ ,  $N_2O$  and NO emissions. Based on the experimental results, some agricultural measures were recommended as technical options to mitigate greenhouse gas emissions from rice-wheat rotation ecosystems. These mitigation measures are reducing mineral N input, coupling organic manure with chemical fertilizers, applying fertilizers which release available N slowly during periods with intensive plant activity, and applying dry fermented organic manure and well management of water and fertilizer.

Key words: Mitigation options, Emission, Greenhouse gases, Ecosystems

#### 1. Introduction

Nitrous oxide  $(N_2O)$  and methane  $(CH_4)$  are the most important greenhouse gasses in the atmosphere with its contribution to global warming just lower than  $CO_2$ . Their concentrations in atmosphere have been noted to increase currently at the rate of 0.25% yr<sup>-1</sup> and 1.02% yr<sup>-1</sup>, respectively (IPCC, 1995). At present, the increase of N<sub>2</sub>O and CH<sub>4</sub> in the atmosphere has been estimated to account for 20–25% of the global warming (FAO & IAEA, 1992; Batjes & Bridges, 1992). NO does not absorb radiation directly in the atmosphere, but the increasing concentration of NO may arouse strong greenhouse effects indirectly by

This study was supported by projects "Experimental and Modeling Study on N<sub>2</sub>O Emission from the Rice-Wheat Rotation Fields of Southeast China" and "Experimental and Modeling Study on NO Emission from Croplands", which were granted by the National Natural Science Foundation of China, the State Key Fundamental Research Project "Predicting the Future (20-50 years) Trend of Environmental Change in China", and the project of Chinese Academy of Sciences "Theory and Methodology on Air Pollution Prediction".

promoting photochemical production of  $O_3$  in the troposphere.  $O_3$  is a greenhouse gas, with global warming potential much higher than  $CO_2$ . Besides arousing strong greenhouse effects directly or indirectly,  $CH_4$  and NO are chemically active in atmosphere, and  $N_2O$  is photochemically active in the stratosphere. So the increase of their concentration in the atmosphere may change atmospheric chemistry and then impact the global climate. Therefore, trying to stabilize the concentrations of these greenhouse gases is a great but difficult task for mankind today.

Biological sources are responsible for 70–90% of the atmospheric CH<sub>4</sub>, N<sub>2</sub>O and NO. Most of the biogenic CH<sub>4</sub>, N<sub>2</sub>O and NO are emitted from croplands. Because of the increasing demand for food supply, the cultivated area of flooded rice and the number of ruminant livestock will be further enlarged and, the input of nitrogen chemical fertilizers into fields will be further increased. As a result, the impacts of CH<sub>4</sub>, N<sub>2</sub>O and NO from agriculture on the global warming are expected to be more serious in the near future. Therefore, it is urgent to develop practical measures or techniques for mitigating the emissions of CH<sub>4</sub>, N<sub>2</sub>O and NO from agricultural production. For these purposes, a field experimental study was launched in the Taihu region located in Southeast China in 1994 and lasted for more than 4 years. In this experiment, the emission fluxes of CH<sub>4</sub> and N<sub>2</sub>O were measured simultaneously since June of 1994, but the measurement of NO emission was started in November of 1996. Based on the experimental results, the characters of CH<sub>4</sub>, N<sub>2</sub>O and NO emissions from the rice—wheat cropping ecosystem which is widely distributed in South China are outlined and some mitigation strategies and measures are discussed in this paper.

#### 2. Methods

An automated system based on techniques of static chambers and gas chromatography (GC) was employed for the continuous and simultaneous measurements of N<sub>2</sub>O, CH<sub>4</sub> and NO emission fluxes from a rice-wheat rotation system, which is a typical rotation system in South China. Eight temperature sensors were installed in the automated system to measure soil and air temperature in and out of chambers. Nine chambers in volume of  $70 \times 70 \times 90$  cm<sup>3</sup> were involved in the automated system, experimental site, field treatments and experimental procedure were described in detail in another paper (Zheng et al., 1999b).

Equal amount of pure N was input in every plot at the rate of  $382 \text{ kg} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$ , with 50% for the rice-cultivated season and the remainder for the wheat-cultivated fields. Each field treatment of fertilization or water management was implemented in a field plot of 8 m × 12 m. Duplicate chambers were installed in each plot. All the chambers were sampled and analyzed alternatively. One chamber was measured once every 4 hours. For each gas species, at least 12 data of emission flux could be collected per day from each plot. The average value of the 12 fluxes was considered representative of the daily emission of certain plot. Ecological factors such as soil moisture, N nutrient status and temperature were investigated simultaneously. With the experimental results, the possible measures to mitigate emissions of CH<sub>4</sub>, N<sub>2</sub>O and NO via fertilization and water management were investigated and discussed.

## 3. Results and discussion

## 3.1 Effects of drainage on $CH_4$ and $N_2O$ emissions and mitigation options

Draining the flooding water periodically from the rice fields is a traditional but effective

measure to improve the grain production of rice. It is widely adopted in the rice cultivation in China. One ecological effect of this measure is to effectively reduce methane emission from rice fields. Fig. 1a shows the seasonal variation of methane emission during the rice-cultivated period from a rice-wheat rotation cycle. It is evident that draining the flooding water in the tillering and panicle initiating stage, respectively, may significantly reduce methane emission by 26-46% (shown by the solid line of Fig. 1a), compared with the emission from continuously flooding fields (shown by the dashed line). From the reports of Shangguan et al. (1993), Wang et al. (1993) and Chen et al. (1993), the similar results may be obtained that intermittent irrigation of rice fields may reduce methane emission by 8-70%, with a mean of 32%. Normally, one drainage period lasts for several days until rifts appear in the fields. As shown in Fig.1a, an intensive methane emission usually happens at the beginning of a draining period. Then the emission drops to low level until the fields are flooded again. The intensive emission is likely due to the increase of soil temperature caused by drainage.



Fig. 1. Effects of drainage and fertilization on methane emission from rice fields. Note: a. Measured in rice field at Suzhou from June 21 to October 27. b. Measured in rice fields at Taoyuan, Hunan Province from May 5 to October 25. (i) Astragalus sinicus L. (for early rice) / rice straw (for late rice) + Mineral compound (base) + KCl (base) + Urea (top-dressing), (ii) Astragalus sinicus L.(for early rice) / rice straw (for late rice) + Swine manure, (iv) Mineral compound (base) + KCl (base) + Urea (top-dressing).

The figure which has been published in another paper (Zheng et al., 1999a) illustrated that 70–90% of nitrous oxide emitted from the rice-wheat system happens during the no-rice period, even though drainage during the rice-cultivated period significantly intensifies  $N_2O$  emission while  $CH_4$  emission is reduced obviously. The data listed in Table 1 show the percentage of the GWP (global warming potential) (Shine et al., 1990) of CH<sub>4</sub>+ N<sub>2</sub>O reduced by drainage in the rice-cultivated season. At a time scale of 20 years, the GWP of  $CH_4$ + N<sub>2</sub>O released from the rice-cultivated season may be reduced by 21% and that from the whole rotation cycle reduced by 19%, compared with those from a rotation cycle with continuously flooding rice-field. At a time scale of 500 years, the reduction percentage is 7% and 5%, respectively. The reduction of GWP is mainly caused by the reduction of  $CH_4$  emission from the rice fields. In other words, the increase of N<sub>2</sub>O emission from rice-cultivated season caused by drainage does not significantly increase the GWP of  $CH_4 + N_2O$ . The increasing  $N_2O$  from the rice season is relatively much lower than the decreasing amount of  $CH_4$  or much lower than the amount of N<sub>2</sub>O emitted during the no-rice period. It implies that the increase of N<sub>2</sub>O emission from the rice fields is not necessary to be taken into account, when intermittent irrigation or periodical drainage is adopted to mitigate CH<sub>4</sub> emission from rice fields. However, the fallow periods and the wheat-cultivated season should be greatly concerned to mitigate N<sub>2</sub>O emission from the rice-wheat rotation ecosystem.

Time scale	GWP of $CH_4 + N_2O$ reduced (%)		
	Rice-cultivated season	Whole rotation cycle	
20 years	21	19	
500 years	7	5	

Table 1. Reduction of GWP of CH<sub>4</sub>+ N<sub>2</sub>O released from rice-wheat fields via drainage

## 3.2 Effects of fertilization on CH<sub>4</sub>, N<sub>2</sub>O and NO emission and mitigation strategies

Some examples of fertilization effects on  $CH_4$ ,  $N_2O$  and NO emissions are shown in Fig.1(b), Fig. 2 and Fig. 3. It is obvious that every gas species is significantly influenced by fertilization.

By reanalyzing the experimental data reported by Shangguan et al. (1993), Wang et al. (1993), Chen et al. (1993), Cai et al. (1994), Chen et al. (1995) and Zheng et al. (1997), some results which reflect effects of fertilization on methane emission from rice fields of China can be outlined in Table 2. As shown in Fig. 1(b) and Table 2, application of pure chemical fertilizers, fermented organic fertilizers or organic manure + chemical fertilizer may mitigate methane emission from rice fields by 19%-59%, in comparison with application of fresh organic manure or fresh organic + chemical fertilizer.

Fig. 2a shows the N<sub>2</sub>O emission from the rice-cultivated period of the rice-wheat rotation system. The dose of N for every plot was the same, but the irrigation regime was different. Total amount of N<sub>2</sub>O released during the rice-cultivated season in Plots-A, B and C was measured to be 2.378, 2.696 and 1.941 kg  $\cdot$  ha<sup>-1</sup>, respectively. It is closely related to the days without water cover in the fields. The relationship may be described with

 $F = (1250.3 \pm 4.69) + (19.51 \pm 0.174)d(n = 3, r^2 = 0.9999),$ 

in which  $F(g \cdot ha^{-1} \cdot yr^{-1})$  represents the seasonal total emission of N<sub>2</sub>O and d (d) is days without water cover. The N<sub>2</sub>O-N released from the rice fields applied with NH<sub>4</sub>HCO<sub>3</sub>

Table 2. Effect of fertilization on methane emission from rice fields of China (percentage of methane emission reduced)

For Chemical fertilizer + Organic manure	Mitigation effect	
Fresh organic manure	0%	
Bio-gas residues	47%	
For pure organic manure		
Fresh organic manure	0%	
Bio-gas residues	33%-59%	
For fresh organic manure		
Organic manure	0%	
Chemical + Organic manure	19%	
Only chemical fertilizer	46%(22-78%)	

accounts for 0.7-0.9% of the seasonal N input. For the whole rice-wheat rotation cycle, shown in Fig. 2b, amount of N<sub>2</sub>O emitted from the control, the organic + chemical fertilizer



Fig. 2. Effects of fertilization on nitrous oxide emission from rice-wheat fields a. Measured during rice grow periods from June 15 to October 31; Plot-A was drained for 54d, Plot-B for 72d and Plot-C for 36d; b. Measured in a whole rice-wheat rotation cycle from June 15 to June 15 of next year. Fertilizer-N was applied in either rice or wheat fields at an equal rate of 191 kg  $\cdot$  ha<sup>-1</sup>.



Fig. 3. Effects of fertilization on NO emission from wheat fields measured from November 1, 1996 to June 17, 1997 at Suzhou.

and the chemical plot was observed to be 8.68, 12.55 and 17.15 kg  $\cdot$  ha<sup>-1</sup>, respectively. It is obvious that application of pure chemical or aerobically fermented organic manure + chemical fertilizer increases N<sub>2</sub>O emission significantly from the rice-wheat field. But coupling organic manure may significantly mitigate the emission of N<sub>2</sub>O caused by chemical fertilizer amendment. The amount of N<sub>2</sub>O-N increased by fertilization from the organic + chemical and the pure chemical plot accounted for 0.65% and 1.41% of total N input, respectively. The latter emission factor is larger than the former one by a factor of 2.2. In a word, application of organic manure may significantly mitigate N<sub>2</sub>O emission from the rice-wheat rotation ecosystem. Such result was well inter-annually duplicated in this experimental study.

The seasonal distribution of NO emission from wheat fields was shown in Fig. 3. It is obvious that 83-94% of the NO emission during the period given by Fig. 3 happens during the period from the beginning of the spring till field flooding for rice planting, with the emission from fertilized plots much more intensive than that from the control plot. The total amount of seasonal NO emission from the control, pure chemical and chemical + organic plot during the no-rice period was measured as 1.481, 11.510 and 8.907 kg  $\cdot$  ha<sup>-1</sup>, respectively. The emission from the pure chemical plot was 29% higher than that from the chemical + organic plot. The NO-N released from these plots accounted for 1.81%-2.45% of the seasonal total N input, with the data from chemical plot higher than that from the organic + chemical plot by 35%. It implies that coupling organic manure with chemical fertilizer may significantly mitigate the NO emission from wheat fields. This result is quite similar to that of N<sub>2</sub>O emission.

## 3.3 Seasonal emission of $N_2O$ and NO emission from wheat fields and mitigation options

The seasonal variation of  $N_2O$  and NO emission during the wheat-cultivated period is shown in Fig. 4. It is observed that the  $N_2O$  emission in the seedling stage of wheat from the beginning of November to the middle December is much more intensive than NO emission. The amount of  $N_2O-N$  released during this period is about 7 times higher than that of NO-N. During the spring period from the beginning of March till field flooding in the middle June, the amount of NO-N is about 1.5 times higher than that of  $N_2O-N$ . The NO-N / N<sub>2</sub>O-N ratio in the wheat-seedling stage and the spring season was 0.125 and 2.33, respectively. The difference in NO-N / N<sub>2</sub>O-N ratios may be partially explained by the obvious difference in soil moisture. The soil moisture, which was expressed by the percentage of soil water holding capacity (abbreviated as %SWHC), in the seedling stage and in the spring season was 96%  $\pm$  10% and 81.55%  $\pm$  14%, respectively. The former is significantly higher than the latter. Mechanically, NO emitted from wheat fields is mainly produced via nitrification which occurs under aerobic condition, whereas N<sub>2</sub>O released from the wheat fields is produced via either nitrification or denitrification. Denitrification is a microbial process occurring under anaerobic condition. Thus, the emission rates of both gas species are usually closely related to soil moisture, which determines the O<sub>2</sub> availability for microbial activities. As a result, more N<sub>2</sub>O may be produced and released from the soil with relatively higher soil moisture and the ratio of NO-N / N<sub>2</sub>O-N may significantly decrease with the soil moisture relatively higher.

Before flooding and ploughing for rice planting, the fields are usually wetted to near or over saturated with water but not flooded by irrigation for about 5 days. During this short period, an explosive emission of N<sub>2</sub>O intensively happens. The amount of emitted N<sub>2</sub>O during this period may account for about 60% of the total observed from the beginning of November till rice planting. The maximum N<sub>2</sub>O emission flux from the fertilized plots may be up to more than 13 mg  $\cdot$  m<sup>-2</sup>  $\cdot$  h<sup>-1</sup>, which is 2–3 orders of magnitude larger than the usual emission level. The NO emission during this period is obviously intensified, too. But its amount only accounts for about 8% of the seasonal total. It implies that some effective measures should be adopted to avoid such water status, under which explosive emissions of NO and N<sub>2</sub>O happen. For example, the wetting period before ploughing for rice planting should be controlled as short as possible. Nevertheless, the mechanism for explosive emission of these gases still remains uncertain. Further research is quite necessary to understand the influencing mechanism of soil moisture on explosive emissions of NO and N<sub>2</sub>O, for it is of great significance to the developing mitigation measures.



Fig. 4. Emission of nitrous oxide and nitric oxide from wheat fields. Measured during the no-rice period of a rice-wheat rotation cycle at Suzhou from November 1, 1996 to June 13, 1997.

No. 1

The released N in the form of NO and N<sub>2</sub>O directly caused by N fertilization during the wheat-cultivated season and the fallow period before rice planting was  $5.381-7.848 \text{ kg} \cdot \text{ha}^{-1}$ , which accounted for 2.8%-4.1% of the seasonally applied N. The amount of (NO+N<sub>2</sub>O)-N emitted from the chemical plot was 46% higher than that from the chemical + organic plot. Such result indicates that N fertilization may greatly intensify NO and N<sub>2</sub>O emission from croplands, but the emission may be mitigated by about 46% via applying compost organic manure.

## 3.4 Impacts of plant growth on diurnal patterns of NO emission and mitigation options

Two typical patterns of NO diurnal emission from wheat fields were discovered. They are the day-peak pattern and night-peak pattern. For the former one, the maximum emission happens during the period of 12-15 o'clock in the early afternoon and the emission at the night keeps a relatively low but a stable level. This pattern was continuously observed in the seedling stage of wheat excluding the winter period (Fig. 5a), in the ripening stage and during the fallow period before rice planting (Fig. 5b). For the night-peak pattern, the maximum emission occurs during the period of 18-24 o'clock while the minimum emission happens during the period of 12-15 o'clock in the early afternoon. This pattern was continuously observed in the fields with normal plant density during the period from the reviving stage in early spring till the ripening stage (Fig. 5c). However, the NO emission also occurs in a typical



Fig. 5. Diurnal variation of nitric oxide emission from wheat fields. a. Measured from November 16 to 22 (in wheat seeding stage); b. Measured from May 14 to 19 (in wheat maturing stage); c. Measured from April 7 to 14 (in wheat earring stage); d. Measured from April 16 to 25 (in a field with 1/4 of the normal wheat density).

day-peak pattern if there is a quite lower wheat density (Fig. 5d), such as 25% of the normal, even though it is in the same growing periods of wheat as that of Fig. 5c. Further analysis indicates that the diurnal emission of NO in the day-peak pattern is closely related to temperature. For the NO emission in the night-peak pattern, however, temperature was found to have no obvious influence (Zheng et al., 1999c). It implies that there are factors other than temperature to effectively influence the NO emission in the night-peak pattern. Intensive up-take of  $NH_4^+$  -N via wheat plant is likely to be one key influencing factor. Physiologically, diurnal up-take of  $NH_4^+$  -N closely relates to photosynthesis intensity of plant. During the periods when wheat grows prosperously, more  $NH_4^+$  -N may be up-taken by wheat plant during the day time so that less  $NH_4^+$  -N is nitrified to NO by microbes in the soil. In contrast, more  $NH_4^+$  -N is used for nitrification so that the maximum NO emission happens in late afternoon or at night. If the up-take of plant is very weak, for instance in the seedling or ripening periods of wheat growth, the availability of  $NH_4^+$  -N may be always suitable for nitrification. In this case, other factors, such as temperature, are likely to determine the pattern of diurnal emission.

The above experimental results imply that applying N fertilizers during the periods when plant grows prosperously may significantly mitigate NO emission from croplands.

# 4. Conclusions

From above experimental results and detail analysis, some conclusions on mitigation of  $CH_4$ ,  $N_2O$  and NO emissions could be drawn. They are: a) The amount of NO and  $N_2O$  aroused by N fertilization during no-rice periods accounts for 2.8% and 4.1% of the applied N, respectively. b) Intermittent irrigation or periodical drainage of rice fields may reduce methane emission by about 32%. c) Drainage of rice fields can obviously intensify  $N_2O$  emission while it significantly reduces  $CH_4$  emission. But the increase of  $N_2O$  emission from the rice fields is not necessary to be concerned when intermittent irrigation or periodical drainage is adopted to mitigate methane emission from rice fields. Nevertheless, the fallow periods and the wheat-cultivated season should be greatly concerned to mitigate  $N_2O$  emission from the rice-wheat rotation ecosystem. d) Application of chemical N fertilizers may greatly intensify NO and  $N_2O$  emissions from wheat fields, but coupling organic manure with chemical fertilizers may mitigate emissions of NO and  $N_2O$  by about 46%. e) Applying N fertilizers during the periods when plant grows prosperously may significantly mitigate NO emission from croplands.

Thanks are due to Professor Zhang Wen, Dr. Bai Jianhui, Mr. Gong Yanbang, Mrs. Luo Dongmei and Mr. Liu Guangren from the Institute of Atmospheric Physics, Chinese Academy of Sciences for their help in experiments.

#### REFERENCES

- Batjes, N.H. and E.M. Bridges, 1992: World Inventory of Soil Emission, No. 92 / 4. Working Paper and Preprint. p. 4.
- Cai Zucong, Xu Hua, Zhang Hanhui, and Jin Jisheng, 1994: Estimate of methane emission from rice paddy fields in Taihu region, China. *Pedosphere*, 4(4), 279-306.
- Chen Guanxiong, Huang Guohong & Huang Bin, Wu Jie, Yu Kewei, Xu Hui, Xue Xiaohua, and Wang Zhengping, 1995: Emission of CH<sub>4</sub> and N<sub>2</sub>O from rice paddy fields and effects of Azolla cultivation and fertilization.

Chinese Journal of Applied Ecology, 6(4), 378-382. (in Chinese)

- Chen, Z.L., Li, D.B., Shao, K.S., and Wang, B.J., 1993: Features of CH<sub>4</sub> emission from rice fields in Beijing and Nanjing. *Chemosphere*, 26, 239-245.
- FAO & IAEA, 1992: Measurement of methane and nitrous oxide emissions from agriculture. A Joint Undertaking by the Food and Agriculture Organization of the United Nations and the International Atomic Energy Agency. International Atomic Energy Agency, Vienna, p. 1–4.
- IPCC, 1995: Climate Change 1995: Summary for policymakers: the science of climate change, p. 1-24. In: IPCC Second Assessment Report.
- Shangguan Xingjian and Wang Mingxing, 1993: Transportation of methane in paddy soil. Chinese Journal of Earth Science Advances, 8(5), 47-54. (in Chinese)
- Shine, K.P., R.G. Derwent, and D.J. Wuebbles et al., 1990: Radiative forcing of climate. p. 47-48. In: (ed. J.T. Houghton et al.) Climate Change: The IPCC Scientific assessment. Cambridge Univ. Press. Cambridge.
- Wang Mingxing, Shangguan Xingjian, Shen Renxing, Wassmann, R., and Seiler, W., 1993: Methane production, emission and possible control measures in the rice agriculture. *Advances in Atmospheric Sciences*, 10(3), 307-314.
- Zheng Xunhua, Wang Mingxing, Wang Yuesi, Shen Renxing, Shangguan Xingjian, M. Koegge, J. Heyer, H. Papen, Jin Jisheng, and Li Laotu, 1997: N<sub>2</sub>O and CH<sub>4</sub> emission from rice paddies in Southeast China. Chinese Journal of Atmospheric Sciences (English version), 21(2), 167-174.

Zheng Xunhua, Wang Mingxing, Wang Yuesi, Shen Renxing, Li Jing, J. Heyer, Mo. Koegge, Hi. Papen, Jin Jisheng

- and Li Laotu, 1999a: Characters of greenhouse gas (CH<sub>4</sub>, N<sub>2</sub>O, NO) emission from Croplands of Southeast China. World Resource Review, 11(2), 229-246.
- Zheng Xunhua, Wang Mingxing, Wang Yuesi, Shen Renxing, Gou Ji, Li Jing, Jin Jisheng, and Li Laotu, 1999b: Impacts of Soil Moisture on Nitrous Oxide Emission from Croplands—A case study on the rice-based agro-ecosystem in Southeast China, *Chemosphere*. (In press)
- Zheng Xunhua, Wang Mingxing, and Wang Yuesi, 1999c: Temporal Variability of NO emission from Croplands. Chinese Journal of Environmental Science, 20(2), 17-21.