

**UC Davis**

**The Proceedings of the International Plant Nutrition Colloquium  
XVI**

**Title**

Carbon Sequestration and Gas Emissions in Paddy Field Ecosystem Affected by Nitrogen Application in Purplish Soil, Southwest China

**Permalink**

<https://escholarship.org/uc/item/7kj5f940>

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**Publication Date**

2009-07-05

Peer reviewed

## 1. Introduction

In cropland ecosystems, plants transform solar energy into biotic energy through photosynthesis, fixing CO<sub>2</sub> and releasing O<sub>2</sub>. The contribution of these ecosystem services to human welfare may be comparable to that of food and fibre production (Pimentel et al., 1997). But because most of these ecosystem services are indirect, they are seldom recognized by the general public (Lillemor and Söderqvist, 2002). Therefore, there is a need to develop an effective way of valuating the croplands ecosystem services (Björklund et al., 1999).

In rice paddy ecosystems, CH<sub>4</sub> and N<sub>2</sub>O are emitted throughout the growing season. Plants and soil organisms release CO<sub>2</sub> through respiration. CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O are all greenhouse gases (GHGs). Many studies have shown that a large amount of GHGs is released from paddy fields and that a substantial quantity of CO<sub>2</sub> is sequestered by plants in paddy fields (Bruce et al., 1999; Liou et al., 2003; Zou et al., 2003). These two processes constitute the GHGs regulation by rice paddy ecosystems. China has the second largest amount of the paddy fields in the world in terms of the total area, and the gas exchange by these fields may be of global importance (Li et al., 2004). Although there has been much information from field experiments on the cropland ecosystems, only few studies have been conducted to estimate the economic values of these ecosystem services. Therefore, a field experiment was conducted to systematically value the gas regulation services of purple paddy fields in Sichuan province, Southwest China. We investigate the effects of applications of nitrogen fertilizer and wheat straw on the economic values of gas regulation in paddy field.

## 2. Materials and Methods

### 2.1. Location and soil characteristics

The field site (31°16'N, 105°27'E), locating in the Yanting County, Sichuan Province, Southwest China, belongs to the Yanting Station for Agri-Ecology Observation, Chinese Academy of Sciences. It has a subtropical monsoon climate with annual mean precipitation of 826 mm and temperature of 17.3 °C over the past 30 years. The purplish paddy soil is classified as Regosol in the FAO taxonomy. Some chemical characteristics of the soil surface (0~20 cm) were: the total nitrogen, the total organic matter, the available nitrogen, phosphorus and potassium were 0.14% and 2.07%, 119.60, 10.42 and 74.72 mg · kg<sup>-1</sup>, respectively.

### 2.2. Field experiments

The field experiment (four treatments, three replicates each) was conducted in 12 plots (5 m × 6 m each) with a randomised complete block design in 2005. The treatments and the amounts of fertilizer applied were described in Table 1.

Table 1 Paddy field treatments / kg·ha<sup>-1</sup>

Treatments	Straw addition	Fertilization			
		N	Base fertilizer P <sub>2</sub> O <sub>5</sub>	K <sub>2</sub> O	Topdress fertilizer
N0	0	0	0	0	0
N150	0	90	45	45	60
N75-S	10620	45	45	45	30
N150-S	10620	90	45	45	60

### 2.3. Gas sampling and measurements

The fluxes of GHGs were determined using the techniques of static opaque chamber and gas chromatography (Hutchinson et al, 1993) during 1 June to 14 September 2005. A stainless steel base (0.5 m length × 0.5 m width × 0.2 m height) with a water groove on the top was installed at each mini-plot. The portable sampling chamber (0.5 m length × 0.5 m width × 1 m height) covered six hills of rice, and the plant density inside the chamber (hill spacing was 0.25 m × 0.17 m) was the same as that outside the chamber. All chambers were equipped with fans and thermometers. There were heat preservative foams on the surface of portable sampling chambers. Removable wooden boardwalks were set up at the beginning of the rice season to avoid soil disturbances during sampling. Gas samples were collected twice a week between 9:00 AM and 11:00 AM within an 18-min-period. Duplicate gas samples were taken at 0, 6, 12 and 18 min with 100 ml syringes. The gas samples were analyzed by a gas chromatography (GC, HP-5890 II, The Hewlett-Packard Company, Palo Alto, California, USA) with separate electron capture and flame ionization detectors (ECD at 330 °C and FID at 200 °C) for N<sub>2</sub>O and carbon GHGs measurements, respectively. Gas emission flux was calculated as:

$$F = T_0 / (T_0 + T) \cdot \rho \cdot h \cdot (\Delta c / \Delta t) \quad (1)$$

Where F is the GHGs fluxes (mg·m<sup>-2</sup>·h<sup>-1</sup>), T<sub>0</sub> is the absolute temperature of 273.15 K at standard state (0 °C and 1013 hPa), T is the air temperature inside the chamber (°C); ρ represents the gas density at standard state (kg·m<sup>-3</sup>) (0 °C and 1013 hPa), h is the headspace height of the chamber (m); c is the gas mixing ratio concentration (ppm), t is the time for chamber closure (h), Δc is the volume mixing ratio of gas increased (or decreased) in the chamber during Δt (Zheng, et al., 2000).

Leaf net photosynthetic rates were determined once every 2 hours from 8:00 AM to 18:00 PM by using a portable Li-6400 photosynthesis system (Li-Cor Inc., Lincoln NE, USA) during jointing-booting, anthesis and milking stages, respectively. Rice biomass and yield were measured in harvest time.

### 2.4 Carbon (C) fixations

The Total GHGs emissions in the whole season were calculated as follows:

$$Y_{CH_4 \text{ or } N_2O} = \sum_{i=1}^n X_i \times 24, \quad Y_{CO_2} = \sum_{i=1}^n X_i \times 12, \quad n = 1, 2, 3, \dots, 106 \quad (2)$$

Where Y (kg·ha<sup>-1</sup>), X<sub>i</sub> (kg·ha<sup>-1</sup>·h<sup>-1</sup>) and i are the cumulative GHG emission, the average GHG emission rate and the number of days during the rice growth stage, respectively.

The IPCC global warming potential coefficients based on a 100 year time horizon (GWP<sub>100</sub>) 21 and 310 for CH<sub>4</sub> and N<sub>2</sub>O, respectively, were used to convert emissions to CO<sub>2</sub> equivalents (UNFCCC, 2004). Root biomass was assumed to be ≈ 0.1 of the total above-ground biomass (TAGB) (Yoshida, 1981), the C fixation and emission quantities were calculated as follows (Huang et al., 1997, 1998):

$$C_{CO_2} = 0.27 \times Q_{CO_2} \quad (3)$$

$$C_{CH_4} = 0.75 \times Q_{CH_4} \quad (4)$$

$$NPP = 1.1 \times TAGB \quad (5)$$

$$C_{NPP} = 0.27 \times NPP / (0.68 \times 0.90) = 0.44 \times NPP \quad (6)$$

$$\Delta C = C_{NPP} - C_{CO_2} - C_{CH_4} \quad (7)$$

Where  $C_{CO_2}$  and  $C_{CH_4}$  ( $\text{kg}\cdot\text{ha}^{-1}$ ) are the total C contents of the  $CO_2$  and  $CH_4$  emissions respectively;  $Q_{CO_2}$  and  $Q_{CH_4}$  ( $\text{kg}\cdot\text{ha}^{-1}$ ) are the total  $CO_2$  and  $CH_4$  emissions respectively.  $NPP$  ( $\text{kg}\cdot\text{ha}^{-1}$ ) is the net primary production,  $C_{NPP}$  ( $\text{kg}\cdot\text{ha}^{-1}$ ) is the total C amount of the NPP. The coefficient 0.44 was derived from three assumed factors: 0.27, the C content of  $CO_2$ ; 0.68, the conversion of  $CO_2$  to carbohydrate ( $[CH_2O]/[CO_2]=0.68$ ); and 0.90, the conversion of carbohydrate to rice biomass ( $[C_6H_{10}O_5]/[C_6H_{12}O_6]=0.90$ ) (Photosynthesis equation).  $\Delta C$  is the net carbon fixation quantity.

## 2.5 Gas regulation values

The value of carbon fixation can be estimated by the afforestation cost method or the carbon tax method and we adopted their average value:

$$E_c = 0.27 \times \alpha_{GWP} \times E \quad (8)$$

$$v_c = \frac{1}{2} (C_{f-CO_2} + C_t) \times E_c \quad (9)$$

Where  $E_c$  is the carbon content ( $\text{kg}\cdot\text{ha}^{-1}$ ); 0.27 is the carbon content of  $CO_2$ ;  $\alpha_{GWP}$  is the global warming potential of GHG;  $E$  is the total emission quantity of GHG ( $\text{kg}\cdot\text{ha}^{-1}$ );  $v_c$  is the gas regulation value ( $\text{USD}\cdot\text{ha}^{-1}$ );  $C_{f-CO_2}$  is the afforestation cost which adopted is  $0.0380 \text{ USD}\cdot\text{kg}^{-1}\text{C}$  (State Forestry Administration, 1990);  $C_t$  is the carbon tax which adopt is  $0.1500 \text{ USD}\cdot\text{kg}^{-1}\text{C}$  (Current carbon tax in Sweden).

Based on photosynthesis equation, dry matter per kilogram produced can supply 1.19kg oxygen. Therefore, the value of oxygen supply can be estimated according to the afforestation cost or the industrial oxygen production cost and their average value was used:

$$v_{O_2} = \frac{1}{2} (C_{f-O_2} + C_p) \times 1.19 \times M_{NPP} \quad (10)$$

Where  $v_{O_2}$  is the value of oxygen produced ( $\text{USD}\cdot\text{ha}^{-1}$ );  $C_{f-O_2}$  is the afforestation cost which adopted is  $0.0514 \text{ USD}\cdot\text{kg}^{-1} O_2$  (State Forestry Administration, 1990);  $C_p$  is the industrial oxygen production cost which adopt is  $0.0583 \text{ USD}\cdot\text{kg}^{-1} O_2$  (National Bureau of Statistics of China, 1992).  $M_{NPP}$  is the dry matter weight per hectare produced by rice paddy ecosystem.

## 2.6 Data analyses

All the data were subjected to statistical analysis (SPSS 13.0). Differences between treatments were analyzed using ANOVA, followed by LSD at the 0.05 probability level and two-tailed T-test.

## 3. Results and Discussions

### 3.1 GHGs flux rates and cumulative emission quantities

The seasonal flux rates and cumulative emission quantities of CH<sub>4</sub>, CO<sub>2</sub> and N<sub>2</sub>O were listed in table 2. Results showed: the seasonal CH<sub>4</sub> flux rate of N150-S was the greatest and significantly higher than that of N0 and N150; the seasonal CO<sub>2</sub> flux rate of N150-S was the greatest and significantly higher than that of the others; the seasonal N<sub>2</sub>O flux rate of N150 was the greatest and significantly higher than that of the others (P<0.05). The total CH<sub>4</sub> emission quantity increased with the increase of nitrogen applications and straw additions. The total CH<sub>4</sub> and CO<sub>2</sub> emission quantities of N150-S were the greatest in all treatments. The total N<sub>2</sub>O emission quantity of N150 was the greatest in all treatments.

Our results were consistent with other studies that straw application stimulated CH<sub>4</sub> emission in paddy fields (Watanabe et al., 1999; Zou et al., 2004; Ma et al. 2007, 2008). The results supported the finding that nitrogen application reduces CH<sub>4</sub> emission (Wassmann et al., 1994; Yang and Chang, 1998). Nitrogen application can increase soil pH, and may have further inhibited methane emission in the paddy fields (Wang et al., 1993, Xiao Y. et al., 2005). The average flux and seasonal emission of CO<sub>2</sub> increased significantly with increasing straw and nitrogen application rates, this might have been caused by the increase of soil microbial activities. The excessive available N produced more CO<sub>2</sub> through respiration and the excessive organic matters which came from straw decomposition could result in increase of soil microbial activities (Ma et al. 2007, 2008). The average flux and seasonal emission of N<sub>2</sub>O increased with nitrogen application rates; whereas decreased with straw application rates. The concentration of soil NH<sub>4</sub><sup>+</sup> likely increased with nitrogen application, which enhanced nitrification and denitrification processes to produce more NO<sub>2</sub> (Xu et al., 1998; Suratno et al., 1998). Straw decomposition had decreased N mineralization (Yan et al., 2007), a net immobilization of N occurred with the addition of straw (Azam et al., 2004), the higher amount of immobilized N could thus depress nitrification and denitrification in paddy fields, resulting in a lower N<sub>2</sub>O emission.

Table 2 GHGS flux rates and cumulative emission quantities

Treatments	GHGs flux rates (mg·m <sup>-2</sup> ·h <sup>-1</sup> )			Cumulative GHGs emission quantities (kg·ha <sup>-1</sup> ·yr <sup>-1</sup> )		
	CH <sub>4</sub>	CO <sub>2</sub>	N <sub>2</sub> O	CH <sub>4</sub>	CO <sub>2</sub>	N <sub>2</sub> O
N0	9.937(1.203)c	431.850(55.861)c	0.128(0.013)b	291.100	5652.004	3.190
N150	10.649(2.611)bc	817.452(25.753)b	0.231(0.003)a	292.332	11316.817	5.286
N75-S	19.807(3.123)ab	761.774(35.429)b	0.098(0.010)b	505.729	9597.755	2.235
N150-S	22.413(3.865)a	1041.618(32.042)a	0.092(0.016)b	553.598	13087.650	2.380

Means followed by the same letter are not significantly different (P<0.05) within columns. Standard deviations of the means are shown in parentheses.

### 3.2 Daily carbon sequestrations in growth stage

Daily carbon sequestrations at different rice growth stage were listed in table 3. Results showed: in the joint-booting stage, with the increase of nitrogen application the C emission and the ratio of C emission to fixation increased. The photosynthesis C fixation sequenced as N0>N150>N150-S>N75-S and N150-S>N75-S >N0>N150 in the Joint-booting and anthesis stages, respectively. The C emission of N150 was the greatest in all treatments both in the anthesis and milking stages, and the ratio of C emission to fixation of N150 showed the same

trend. In the milking stage, the photosynthesis C fixation sequenced as N75-S>N150-S>N0>N150.

Table 3 Daily carbon sequestrations in growth stage / kg C·ha<sup>-1</sup>·day<sup>-1</sup>

Treatments	Joint-booting stage			Anthesis stage			Milking stage		
	C emission	Photosyn. C fixation	Emission / fixation	C emission	Photosyn. C fixation	Emission / fixation	C emission	Photosyn. C fixation	Emission / fixation
N0	13.431	103.352	0.130	23.351	81.669	0.286	25.166	80.253	0.314
N150	28.725	95.312	0.301	41.008	77.607	0.528	38.654	74.453	0.519
N75-S	31.267	94.063	0.332	25.463	86.226	0.295	20.299	97.971	0.207
N150-S	47.340	94.283	0.503	38.336	90.568	0.423	27.902	93.706	0.298

### 3.3 Carbon sequestrations

The carbon sequestrations in rice paddy ecosystems were listed in table 4. Results showed with the increase of nitrogen application, the carbon amount of the total CH<sub>4</sub> increased. The carbon amount of the total CO<sub>2</sub>, the TAGB, the NPP, the C<sub>NPP</sub> and the net carbon fixation amount showed the same trend and all sequenced as N150-S>N150 >N75-S>N0. During the rice cultivation period, all plots were carbon sinks (uptake > emission). Our results supported the finding that nitrogen application increase C sinks (Xiao Y. et al., 2005).

Table 4 Carbon sequestrations /kg C·ha<sup>-1</sup>·yr<sup>-1</sup>

Treatments	C amount of total CO <sub>2</sub> (C <sub>CO2</sub> )	C amount of total CH <sub>4</sub> (C <sub>CH4</sub> )	Total above ground biomass (TAGB)	Net primary production (NPP)	Total Carbon amount of NPP (C <sub>NPP</sub> )	Net Carbon fixation amount (ΔC)
N0	1541.454	218.247	11812.500	12993.750	5717.250	3957.549
N150	3086.402	219.249	19027.200	20930.250	9209.310	5903.659
N75-S	2617.567	379.297	18261.330	20087.470	8838.485	5841.622
N150-S	3569.356	415.199	23056.000	25361.600	11159.104	7174.549

### 3.4 Gas regulation services and values

The gas regulation services and values were listed in table 5. Results showed: the GHGs regulation values of CO<sub>2</sub>-C emission and CO<sub>2</sub>-C fixation all sequenced as N150-S>N150>N75-S>N0. The GHGs regulation values of CH<sub>4</sub>-C emission ranged as N150-S>N75-S >N150>N0. The GHGs regulation values of N<sub>2</sub>O-C sequenced as N150>N0>N150-S>N75-S. The O<sub>2</sub> regulation values of N0, N150, N75-S and N150-S were 848.122, 1366.149, 1311.139 and 1655.390 USD·ha<sup>-1</sup> respectively. The overall economic values of N0, N150, N75-S and N150-S were 893.218, 1430.580, 1323.262 and 1677.327 USD·ha<sup>-1</sup> respectively.

The economic values of specific gas regulation services (including O<sub>2</sub> emission and GHGs regulation) supported by paddy fields were summed up to obtain the overall economic values for different treatments. The results showed that the overall values were all positive, indicating that these ecosystems actually contribute to environmental sustainability in terms of the production of O<sub>2</sub> and reduction of GHGs. The maximum overall economic value of gas regulation was provided by the paddy fields with treatment N150-S. Our results were consistent with Xiao et al (2005).

Table 5 Gas regulation services and values

Treatments	Carbon content Ec (kg·ha <sup>-1</sup> )				GHGs regulation values Vc (USD·ha <sup>-1</sup> )				O <sub>2</sub> regulation values V <sub>O<sub>2</sub></sub> (USD·ha <sup>-1</sup> )	Overall economic values (USD·ha <sup>-1</sup> )
	CO <sub>2</sub> -C emission	CO <sub>2</sub> -C fixation	CH <sub>4</sub> -C emission	N <sub>2</sub> O-C emission	CO <sub>2</sub> -C emission	CO <sub>2</sub> -C fixation	CH <sub>4</sub> -C emission	N <sub>2</sub> O-C emission		
N0	1541.454	3957.549	1666.609	269.734	-144.897	372.010	-156.661	-25.355	848.122	893.218
N150	3086.402	5903.659	1674.265	446.916	-290.122	554.944	-157.381	-42.010	1366.149	1430.580
N75-S	2617.567	5841.622	2896.444	198.647	-246.051	549.112	-272.266	-18.673	1311.139	1323.262
N150-S	3569.356	7174.549	3170.604	201.214	-335.519	674.408	-298.037	-18.914	1655.390	1677.327

Overall economic value was obtained via direct summation of V<sub>O<sub>2</sub></sub> and Vc.

## 4 Conclusions

The average flux and seasonal emission of CH<sub>4</sub> increased significantly with increasing straw application rates; the differences of CH<sub>4</sub> emissions were not significant with increasing nitrogen application rates. Those of CO<sub>2</sub> increased significantly with increasing nitrogen and straw application rates. Those of N<sub>2</sub>O increased with nitrogen application rates; whereas decreased with straw application rates.

In the Joint-booting stage, with the increase of nitrogen application the C emission and the ratio of C emission to fixation increased. The C emission of N150 was the greatest in all treatments both in the anthesis and milking stages, and the ratio of C emission to fixation of N150 showed the same trend.

During the rice cultivation period, all plots were sinks for carbon (uptake > emission). The overall values were all positive, indicating that these ecosystems actually contribute to environmental sustainability in terms of the production of O<sub>2</sub> and reduction of GHGs. The maximum overall economic value of gas regulation was provided by the paddy fields with treatment N150-S.

## Acknowledgement

This research was supported by the National Nature Science Foundation of China (40805058 and 40331014) and the Knowledge Innovation Program of the Chinese Academy of Sciences (KSCXZ-YW-N-037).

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