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N_2O emissions from agricultural soils in the North China Plain: the effect of chemical nitrogen fertilizer and organic manure

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Abstract: An enclosed chamber technique was used to measure N_2O emissions from intensively agricultural soils of the North China Plain during the periods of 1995—1996 and 1997—1998, to reflect distinct components of winter wheat and summer maize growing seasons. The results showed that the continuous application of fertilizer in agricultural soils increased N_2O emissions by a factor of 24.1—28.1, the calculated annual chemical N fertilizer-transformed N_2O-N emissions was 0.67%. Our results indicated that the application of organic manure also had a significant influence on soil N_2O emissions, which combined with the use of chemical N increased about 20% in a year. It was calculated that there were about 0.11% N of organic manure transformed as N_2O-N. Annual mean N_2O emission from our study area of fertilized soils was estimated to be 57.1 $\mu gN_2O/(m^2 \cdot h)$. A weak correlation was also found between N_2O emissions and soil available nitrogen content NH_4^+.

Key words: N_2O; emissions; the North China Plain; agricultural soils; chemical nitrogen; organic manure

Introduction

Nitrous oxide (N_2O) is a very important greenhouse gas in the atmosphere. Interest in the increase of atmospheric N_2O has been recently stimulated by the understanding that N_2O gas plays an important role in the chemistry and ozone layer destruction of the stratosphere. The radiative forcing of N_2O has, on a molar basis, about 220—290 times greater global warming potential than that of CO_2 (Lloyd, 1995). The principal mechanism for N_2O destruction is photolysis in the stratosphere, followed by reaction with exited single oxygen atoms, which represents the major natural source of stratospheric nitrogen oxides; this leads to a significant effect on the chemistry of the stratosphere and the ozone layer destruction (Crutzen, 1971).

Although there are many studies have been made on N_2O emissions from natural soils and from agricultural soils, however, N_2O emissions from Chinese agricultural soils are still poorly defined and the budget of global atmospheric N_2O is still uncertain (Bouwman, 1995). Particularly it is still need more data from China about the effect of chemical nitrogen and local fertilizer (organic manure) on the N_2O emissions from agricultural soils. More than 7% of the total Chinese land surface area is intensively agricultural soils with a considerable amount of nitrogen fertilizer consumed by agriculture soils annually, being more than 20% of the world total chemical N in 1990, which should serve as an important regional N_2O source. A better understanding of this source is important to estimate and control the regional and global climate changes caused by the increasing N_2O concentration. The objective of this study was to compared the N_2O emissions from chemical N treated soils, chemical N combined with organic manure treated soils, and non-fertilized soils under traditional agricultural land management system. The effect of application of organic manure prior to summer maize and winter wheat seed time on soil N_2O emissions was also explored.

1 Materials and methods

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1.1 Experiment site and soils

The measurements were carried out at Yucheng Integrated Experimental Station (36°57′N, 116°38′E, above sea level 23m) of Chinese Academy of Sciences, which is located on the alluvial flat of the North China Plain with representative and typical of the natural conditions and the average level of agricultural productivity. The dominant crop rotation system is summer maize and winter wheat with grain productivity of about 10.4 t/(hm^2·a). The area has a semi-arid and sub-humid monsoon climate with a mean annual precipitation of 608 mm and average air temperature of 13.1℃ (30 years).

Soil in the measurement area is classified as aquatic series with sandy loam texture, which contains organic matter (0.78%), total nitrogen (0.049%), and available nitrogen (58 ppm) in top 20 cm layer. The soil pH value of top 20 cm is 7.9 (in a 1:5 soil-to-water ratio) with bulk density 1.45 g/cm^3 and water filled pore space 44.9%.

1.2 Soil treatments

Three field treatments of chemical N treated soil (CNT), chemical N combined with organic manure treated soil (CNOT), and non-fertilized soil served as control experiment (CK) were used for this study. The control experiment of our study was a very special field treatment which has been no fertilization since 1985 for about 12 years and it is continuously planted with a rotation system of winter wheat and summer maize year by year for a long term fertilizer experiment. The organic manure was local animal excreta composted with straw organic matter with a rate of 10 $th^{-1}m^2$. The effect of application of organic manure prior to summer maize and winter wheat seed time on soil N_2O emissions was also measured. Chemical N was applied at the same rate of 210 $kgNh^{-1}m^2$ urea for each crop of fertilizer treated soils, which was split into 2 applications for maize and 2 or 3 applications for wheat in a year. The area of fertilized soil experimental was larger than 1 hm^2 and the control experiment plot with a small area of 1200 m^2. Summer maize was planted in late June and harvested in middle October, and then winter wheat was planted in late October and harvested in middle June.

1.3 Flux measurements and techniques

N_2O emissions measurements were conducted weekly using an enclosed chamber method, except biweekly intervals during winter time. The enclosed chamber was made by 8 mm thick acrylic material with about 0.23 m^2 surface area. An acrylic material lid was placed on the top of the chamber with rubber sealed and removed between measurements, which was fitted with an air mixture fan, temperature sensor and 3-way sampling stopcock. The chambers with 30 cm height were directly inserted into the soil to a depth of 8 cm and used for most of flux measurements. A chamber with 70 cm height was used during the later growing time of winter wheat to include higher plants. During each of the flux measurement (generally between 09:00 to 12:00 h), headspace gas was extracted by a teflon-covered membrane pump at 0, 10, 20 and 30 min with 0.5L into polyethylene-coated aluminum bag following placement of the chamber lid, respectively.

Air samples were analyzed in one or two weeks by a GC (type: Hewlett-Packard 5890 II) with electron capture detector (ECD). The detector temperature was maintained at 330℃. The GC had a backflush system with 1.84m × 3.2 mm o.d. stainless steel pre-column and 3.68m × 3.2 mm o.d. analytical column packed with PORAPAK Q, both 80—100 meshes, and held at 90℃ oven temperature. The carrier gas (5% CH_4 in Ar) flow was adjusted to 26 ml/min through the analytical column, and for backflush gas at a flow rate of 40 ml/min through the pre-column. Analytical reproducibility was less than 0.8% for repeated analysis of ambient air compared with standard of N_2O 315 ppbv in compressed air (Deuste Steininger) during 8 hours.

2 Results and discussion

2.1 N_2O emission from unfertilized soil

Monthly mean of measured N_2O emissions from unfertilized soils (CK) show a large scatter and extremely low flux rate (Table 1). The annual mean emission rate was 2.2 ± 0.6 $\mu gN_2O/(m^2 \cdot h)$ ($n = 86$, $\pm SE$) from unfertilized soils, with a relatively small amount annual emission of 19.2 mgN_2O/m^2 contrasted with fertilized soils (Table 2). The N_2O fluxes from unfertilized soils were significantly less than those of chemical N and chemical N combined with organic manure treated soils (t-test, $p = 0.002$ and $p = 0.001$, respectively), which was also near three times less than a natural unfertilized temperate forest soils (Dong, 1998). These lower emissions might be due to continuous non-fertilization.

Table 1 Monthly mean N_2O fluxes from non-fertilization (CK), chemical N (CNT), and chemical N combined with organic manure (CNOT, CNOT*) treated soils, the estimates (average values for each treatment ± standard error) are based on individual measurements in the month, number of measurements in parenthesis

Month	CK	CNT	CNOT
October-95	0(1)	37.3±16.1(2)	77.5±21.4(2)
November-95	−1.0±4.3(5)	9.8±2.8(5)	39.0±15.7(5)
December-95	−1.5±1.5(4)	2.1±1.2(4)	5.0±1.2(3)
January-96	0(2)	6.0±1.4(2)	7.2±2.3(2)
February-96	0(3)	7.8±1.3(3)	11.2±0.3(3)
March-96	0(2)	10.6±7.9(2)	8.3±3.9(2)
April-96	0.7±0.7(4)	144.6±24.3(4)	82.9±19.0(4)
May-96	0.5±0.5(5)	92.7±35.0(5)	65.9±35.8(5)
June-96	4.2±4.2(2)	61.6±13.5(4)	107.4±92.6(2)
July-96	7.9±5.3(3)	65.4±28.0(3)	185.5±169.4(3)
August-96	8.3±8.7(4)	91.5±66.7(4)	161.5±147.1(4)
September-96	3.3±1.2(2)	34.4±5.1(2)	24.4±0.4(2)
	CK	CNOT	CNOT*
July-97	3.2±2.5(4)	56.0±42.2(4)	166.2±83.0(4)
August-97	2.4±1.0(5)	126.9±76.8(5)	208.7±112.5(5)
September-97	4.2±2.8(4)	7.1±2.4(4)	18.0±5.8(4)
October-97	1.6±1.6(5)	78.6±49.5(5)	91.2±43.2(5)
November-97	1.8±1.9(6)	128.3±29.0(6)	20.0±9.0(6)
December-97	3.4±1.7(3)	42.4±23.4(3)	1.5±1.5(3)
January-98	0(2)	3.9±0.9(2)	1.4±1.4(2)
February-98	0(2)	9.9±7.8(2)	1.7±0.6(2)
March-98	2.8±1.9(4)	23.3±11.7(4)	68.0±52.5(4)
April-98	1.4±3.9(4)	80.3±42.3(4)	57.6±35.8(4)
May-98	0.2±3.0(5)	18.1±4.0(5)	17.7±3.8(5)
June-98	6.6±2.5(5)	12.5±5.3(5)	1.4±1.4(5)

·CNOT = organic manure is used prior to winter wheat seed time (October). CNOT* = organic manure is used prior to summer maize seed time (June)

Table 2 Average emission rates ($\mu gN_2O/(m^2 \cdot h)$) and yearly weighted emissions ($mgN_2O/(m^2 \cdot a)$) from each of experiments, the estimates (average values for each treatment ± standard error) are based on individual measurements in the year, number of measurements in parenthesis

Treatment of soil	Average emission (1995 − 1996)	Average emission (1997 − 1998)	Average emission (1995 − 1998)	Yearly emission
CK	1.8±1.2(37)	2.5±0.6(49)	2.2±0.6(n=86)	19.2±0.2
CNT	52.8±10.6(40)		52.8±10.6(n=40)	462.2±10.6
CNOT	69.1±21.4(37)	56.6±12.1(49)	61.4±9.4(n=135)	538.0±9.4
CNOT*		60.5±16.7(49)		

2.2 N_2O emissions from fertilized soils

2.2.1 Variability of N_2O emissions

The annual cycle of N_2O fluxes from fertilized soils indicated a quite similar seasonal pattern

with higher emissions in spring and fall months (Fig. 1). This seasonality were more causes by a combination of two critical factors: (1) concentrated fertilizer applications which enhance N_2O emissions (the total amount of annual chemical N was split into 4 or 5 applications in March, June, July and October, and once time application of organic manure used prior to the wheat seed time of October), and (2) soil water content adaptation to increase denitrification and nitrification N_2O production (Schuster, 1992), in general, the agricultural soils of our measurement area were managed in a good water content situation for crop growth with moisture content between 40%—70% of field water capacity.

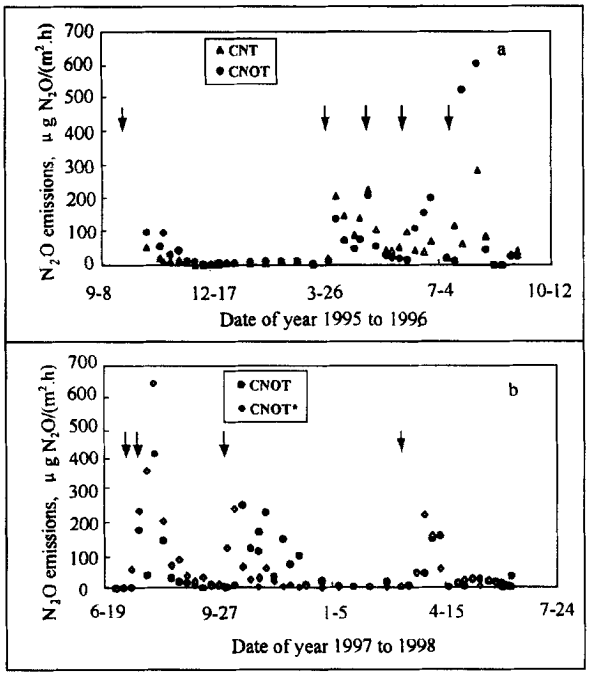

Fig.1 Variability of N_2O emissions from the chemical N (CNT) and chemical N combined with organic manure (CNOT) experiments. CNOT* was organic manure applied prior to summer maize seed time (June 25, 1997). Chemical N application days are indicated by arrows (↓)

Fig. 1a shows a significant influence of organic manure application time on N_2O emissions (t-test, $p < 0.05$). Since organic manure was applied prior to summer maize seed time on June 25, the emissions from CNOT* experiment were always higher than CNOT experiment between June and September, it critically changed to the emissions of CNOT experiment higher than CNOT* experiment between October to December when organic manure applied prior to wheat seed time on October 10. Fig. 1b also illustrated that the organic manure has a long period influence on soil N_2O emissions, but it is not obvious, since the effect of chemical fertilizer is often seen in a short period of time (about 3 weeks).

2.2.2 Soil available nitrogen content and N_2O emission

Since August to December 1997, in a separate experiment the soil flux measurements and soil samples from chemical N treated soil were analyzed to determine the effect of available nitrogen (NH_4^+, NO_3^-) content on N_2O fluxes. No significant changes in N_2O emissions were observed with changes of soil available nitrogen content of NO_3^- from this study (Fig. 2), however, a weak linear correlation ($R^2 = 0.28$, $n = 12$, $\alpha < 0.01$) was found between N_2O fluxes and soil available NH_4^+ content. This might be soil water content to make oxic condition in soil and may suggest that the nitrification process (NH_4^+-NH_2OH-N_2O-NO-NO_3^-) was the main source of N_2O emissions from upland agricultural soils. Our observation agrees with the result that nitrification is generally the main N_2O source under oxidic conditions from agriculture soils (Davidson, 1992; Skiba, 1993).

2.2.3 The influence of N_2O emissions by fertilizer

Fig.1 and Table 1 show that N_2O emissions were significantly influenced by both chemical N and organic manure applications. N_2O emissions from fertilized soil were significantly higher than unfertilized soils (t-test, $p < 0.002$). Also, N_2O emission rates from CNOT experiment were mostly higher than CNT experiment (Fig.1a), except April and May of 1996, which is due to a little more chemical N applied for CNT experiment to supplement less chemical N at seed time. Although the annual mean emission rate and most of individual flux measurements of CNOT

experiment were higher than CNT experiment, this difference was not significant (t-test, $p = 0.5$). Annual mean emission rates from chemical N and from chemical N combined with organic manure treated soil were 52.8 ± 10.6 and 61.4 ± 9.4 $\mu gN_2O/(m^2 \cdot h)$, respectively. The annual average N_2O emission rate from fertilized soils is estimated to be 57.1 $\mu gN_2O/(m^2 \cdot h)$ with a maximum mean emission rate of 69.1 $\mu gN_2O/(m^2 \cdot h)$ from this agriculture system.

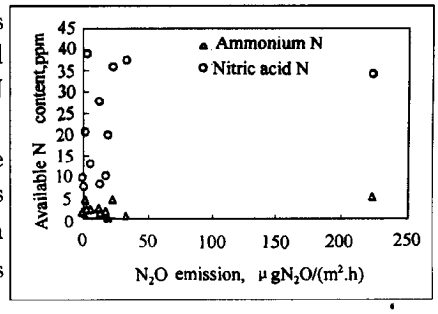

Fig. 2 Relations between N_2O emission and soil available nitrogen content from chemical N treated soils

The annual total flux measured over the whole observation period from different fertilizer treated soils are given in Table 2. In general, this result indicated that each fertilizer treatment had a significantly different amount of N_2O emission. The higher weight of N_2O emission was obtained from chemical N combined with organic manure experiment with mean annual emission of 538.0 ± 9.5 $mgN_2O/(m^2 \cdot a)$, and 462.2 ± 10.6 $mgN_2O/(m^2 \cdot a)$ emitted from chemical N treated soils. The annual maximum emission of this agriculture system was observed to be 605.3 $mgN_2O/(m^2 \cdot a)$ from chemical N combined with organic manure treated soils.

The calculated chemical N fertilizer-transformed N_2O emissions in whole year was 0.67% or 6.7 gN_2O-N/(kgN·a) of inputted 420 $kgN/(hm^2 \cdot a)$ of urea. This result is comparable to the recent report by Yamulki *et al*. (Yamulki, 1995). They found 0.93% of fertilizer N was transformed to N_2O from wheat field. Eichner (Eichner, 1990) summarized global available data where fertilizer-derived N_2O emissions from arable land with a range between 0.04%—1.7% of inputted fertilized N. To compare with unfertilized soils for 12 years period, our results also showed application of chemical N with organic manure increased N_2O fluxes by a factor of 24.1—28.1 (Table 2). This result was higher than the results of N_2O emission from grass land which increased by a factor of 2—3 following N-fertilizer application of 22 $kgN/(hm^2 \cdot a)$ over 13 years period (Mosier, 1991), and it was also higher than those of Bishal *et al*. (Bishal, 1995), who found application of N-fertilizer increased N_2O flux from forest soil by a factor of 1.2—7 with N-fertilizer range between 30—90 $kgN/(hm^2 \cdot a)$. This suggested that the amount of fertilizer and fertilizer application history are factors influencing N_2O release from soils.

Both Table 1 and Fig. 1 show a significant influence of traditional organic manure application on N_2O fluxes, which is estimated to increase by about 20% of the N_2O emission in a year. If we count 4.5% as mean nitrogen content inorganic manure, there were 0.11% N of organic manure transformed as N_2O-N, which showed a lower losses rate in comparison with the report of the excreta application loss of N_2O-N from soils with a range between 0.1%—2.4% (Yamulki, 1998; Vermoesen, 1997). This might be explained by less carbon and nitrogen content of organic manure in for microbial processes of N_2O production, especially the lower extractable nitrogen content.

3 Conclusions

Nitrous oxide emissions from agricultural soils in the North China Plain showed strong yearly temporal variability, which was mainly due to human activities of fertilization. Compared with an unfertilized soil emitted amount of 19.2 $mgN_2O/(m^2 \cdot a)$, continuously chemical N application is a critical factor to enhance N_2O emission from cultivated soils. The organic manure applications have been demonstrated to increase N_2O emissions by about 20% under the condition of combined using chemical N, which should be an another important source of N_2O from agriculture soils. Sustainable and continued agriculture development in China, including continued large scale use of agricultural soils intensive management with increase both of chemical N and organic manure

inputs, will ensure that N_2O emissions from agricultural soils continues to increase in China, affecting stratospheric chemistry and increasing global warming potential.

Comparison with regional reports of field N_2O emissions observed from different locations in China (Wang, 1995; Song, 1996), which indicate that the mean N_2O emissions varied among 9.4—168.1 $\mu gN_2O/(m^2 \cdot h)$. For comparison, the average emission rate of N_2O for these studies is calculated to be 52.7 $\mu gN_2O/(m^2 \cdot h)$, which is in rather good agreement with average N_2O emission rate 57.1 $\mu gN_2O/(m^2 \cdot h)$ from fertilized soils of our study. This is strong support that our study represented a better mean N_2O flux rate from fertilized soils in present time of China, which is useable as a basic emission rate to estimate N_2O distribution from fertilizer applications and cultivated soils for the whole country. Based on our study, assuming nearly all of the cultivated soils receptivity chemical N and half of the area treated with consumption organic manure, we estimate the regional N_2O emissions from cultivated soils in China was about 271.9—332.3 GgN_2O-N/a ($1Gg=10^9g$) in 1997.

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