

## NO and N<sub>2</sub>O fluxes from agricultural soils in Beijing area<sup>\*</sup>

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**Abstract** Chinese agriculture represents one of the most intensively managed agroecosystems in the world. Typical nitrogen fertilization rates are more than three times those of the world's average, resulting in dramatically accelerated nitrogen cycling in China. In this study, we have examined NO and N<sub>2</sub>O exchange in the upland agricultural systems of Beijing area. Inorganic and organic fertilizer treatments were arranged in order to evaluate their impact on the magnitude and proportion of trace gas emissions. Increasing inorganic fertilization rates showed a highly significant impact upon emissions of both NO and N<sub>2</sub>O. Organic matter amendment did not have a statistically significant impact on the N-gas fluxes examined here. Overall losses of added nitrogen by NO and N<sub>2</sub>O emission averaged 1.24% and 0.22% respectively over the range of treatments in this study. Results from our field study indicate that compared with other studies done elsewhere, emissions of reactive nitrogen from agricultural systems in Beijing area are not so large as expected before.

**Keywords:** N<sub>2</sub>O, NO, emission, chemical nitrogen input, organic amendment.

The magnitude of global nitrogen cycling has more than doubled under anthropogenic influence, impacting even what are considered the most pristine of the earth's ecosystems<sup>[1]</sup>. Anthropogenic activity is currently believed to increase the magnitude of reactive nitrogen (N<sub>R</sub>) present in the environment by 140 Tg yr<sup>-1</sup>. This dramatic acceleration in nitrogen cycling has had measurable impacts through its most basic ecosystem functions<sup>[2]</sup>. The continued increase of nitrogenous fertilizer application rates in China has raised concern over regional and global environmental impacts.

Among the most important results of accelerated nitrogen biogeochemistry are those which result in changes to atmospheric composition. Although in the developed countries of the west the creation of N<sub>R</sub> tends to be associated with the combustion of fossil fuels, this source accounts for only approximately 20 Tg N<sub>R</sub> production per year globally. Far more important in magnitude and distribution is the agricultural source of N<sub>R</sub>, particularly that portion resulting from the consumption of synthetic nitrogen fertilizers. Fertilizers account for approximately 80 Tg N<sub>R</sub> production per year, well over half of the anthropogenic nitrogen budget. Inorganic nitrogen in the soil is subject to chemical conversions by microbial ni-

trification and denitrification. These enzymatically mediated processes are not entirely efficient, and inadvertently emit the trace gases NO and N<sub>2</sub>O<sup>[3]</sup>. Atmospheric nitrogen oxides (NO<sub>x</sub> = NO + NO<sub>2</sub>) and nitrous oxide (N<sub>2</sub>O) are often studied for their atmospheric chemistry and climatological qualities<sup>[4,5]</sup>. Skiba et al.<sup>[6]</sup> estimated a 10 Tg NO-N soil source, 41% of which originated from agricultural soils<sup>[7]</sup>. IPPC<sup>[8]</sup> estimated that cultivated soils were responsible for more than 60% of anthropogenic N<sub>2</sub>O production.

China's contribution to global mobilization is currently second only to that of the U.S. Unlike the U.S., the rate of biogeochemical nitrogen cycling in China is expected to grow continuously in the foreseeable future as a result of unprecedented fertilizer application rates<sup>[9,10]</sup>. Of the 80 Tg mobilized by fertilizer annually, China is itself responsible for more than 22 Tg<sup>[11]</sup>. It is, however, still difficult to accurately estimate China's contribution to global N<sub>R</sub> mobilization although fertilizer N application in agriculture is widely considered a major contributor to the N<sub>R</sub> mobilization. According to some studies conducted in China, the total N<sub>2</sub>O emission from agricultural soil ranged 0.31<sup>[12]</sup>~0.398 Tg N yr<sup>-1</sup><sup>[13]</sup>. But very few data of NO or NO<sub>x</sub> fluxes from agricultural fields are

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available in China<sup>[14]</sup>. Considering that China shifted from primary organic fertilizer sources 40 years ago to synthetic fertilizers, which for example has resulted in 77.1% reduction in soil organic matter in upland arable soils<sup>[15]</sup>, we think that it is necessary to examine the trace N gas emissions from main cropping systems in China. Matson et al.<sup>[16]</sup> have shown that management is the dominant control of trace gas losses. Any evaluation of the global nitrogen cycle should fully consider China's specific characteristics.

Winter wheat-corn double cropping is a major rotation system in North China Plain<sup>[17]</sup>. Excessive use of fertilizer N is very often in the cropping system, particularly in regions with high population densities. In Beijing area, for example, the average N application rates are  $309 \text{ kg} \cdot \text{N} \cdot \text{ha}^{-1}$  for winter wheat and  $256 \text{ kg} \cdot \text{N} \cdot \text{ha}^{-1}$  for corn<sup>[18]</sup>. Clearly, such high N rates greatly exceed the N requirements of both crop species at local yield levels and will inevitably lead to large N losses particularly trace gas losses. Thus we have conducted a field experiment to identify: (1) the impact of fertilizer additions on the magnitude and relative composition of trace gas flux under the extreme application rates and traditional land management practices in Beijing area; and (2) how organic matter changes trace gas fluxes.

## 1 Materials and methods

### 1.1 Agriculture in Beijing area

Summer corn-winter wheat rotations account for 80% of northern China's agriculture land use including Beijing area<sup>[19]</sup>. Fertilization rates typically exceed  $300 \text{ kg} \cdot \text{N} \cdot \text{ha}^{-1}$  each crop<sup>[10,18]</sup>, with synthetic fertilizers accounting for more than 64% of total fertilizer applied in China. Organic fertilizers consist of animal manures, night soil, and green manures<sup>[20]</sup>. Approximately two-thirds of the synthetic fertilizer applied is urea, with ammonium bicarbonate accounting for the majority of the remainder.

### 1.2 Experimental site and design

The study was conducted at the campus of China Agricultural University,  $39^{\circ}57'$  north latitude,  $116^{\circ}18'$  east longitude, 16.5 km northwest of downtown Beijing. The experimental soil was an Aquic Cambisol according to Chinese Soil Taxonomy. Soil pH averaged 8.17 over the top 30 cm, with initial organic carbon values of 0.87% and nitrogen content of 0.072%. Soil was a loam texture with a bulk density

of  $1.32 \text{ g} \cdot \text{cm}^{-3}$ . Every attempt was made to simulate typical management practices. This included two separate fertilization events over the course of the corn cropping cycle. The synthetic fertilizer applied was urea, and organic matter was a mixed farmyard manure containing 2.95% N by dry weight. First, at planting, a "base coat" fertilization was applied, consisting of all of the organic matter to be applied to that crop, and 50% of the synthetic nitrogen; 35 days following planting the remainder of the synthetic nitrogen was applied, which is termed the topdressing.

Studies were conducted over a 7-week period at the beginning of the corn portion of the crop rotation. Four replicates were maintained for each of the six treatments, consisting of three levels of urea addition (0, 150 and  $300 \text{ kg} \cdot \text{N} \cdot \text{ha}^{-1} \cdot \text{crop}^{-1}$ ) and two levels of organic matter amendment (0 and  $3000 \text{ kg} \cdot \text{C} \cdot \text{ha}^{-1} \cdot \text{crop}^{-1}$ ). The size of each plot was  $2.5 \text{ m} \times 2.5 \text{ m}$ . NO measurements were made 2~5 times per week using a close chamber method according to the method described by Williams and Davidson<sup>[21]</sup>. A luminol-based chemiluminescent nitrogen oxides detector, Unisearch model LMA-4, was used over 5-minute measurement periods of the soil headspace gas in monitoring NO flux rates.  $\text{N}_2\text{O}$  was monitored twice per week according to the method given by Mosier<sup>[22]</sup>. Samples were taken three times over a 30-minute period and analyzed within 24 hours by GC-ECD.

Ambient and soil temperature, soil moisture, inorganic nitrogen (ammonium and nitrate nitrogen) were measured at depths of 0~10 cm, 10~20 cm, and 20~30 cm each time gas monitoring occurred. Inorganic nitrogen content was extracted by  $1 \text{ mol} \cdot \text{L}^{-1}$  KCl solution and measured by a Continuous Flow Analysis (TRAACS2000 from Germany) instrument. Precipitation was recorded during gas monitoring period as well. Nitrogen ( $\text{NH}_4\text{-N}$  and  $\text{NO}_3\text{-N}$ ) input from rainfall and irrigation was measured during corn growing period using the same instrument.

## 2 Results and discussion

Water filled pore space averaged 44.2% (SD3.7) at the time when samples were made. Average daily ambient temperature ranged from 14.4 to 29.7 °C, and soil temperature averaged  $23.3 (\pm 4.4) ^\circ\text{C}$  over the sampling period. And precipitation (68.1 mm) was mainly distributed in late May and middle June (Fig. 1). And N input from rainfall and irrigation

was  $4.4 \text{ kg} \cdot \text{ha}^{-1}$  during the gas-monitoring period. The increase in soil ammonium and nitrate displayed in Fig. 2 ( $\text{NH}_4\text{-N}$ ) and Fig. 3 ( $\text{NO}_3\text{-N}$ ) corresponds to fertilization events. Compared with  $\text{NO}_3\text{-N}$ , however,  $\text{NH}_4\text{-N}$  changed more rapidly as N fertilization occurred regardless of the amendments of low organic matter or high organic matter (Figs. 2 and 3).

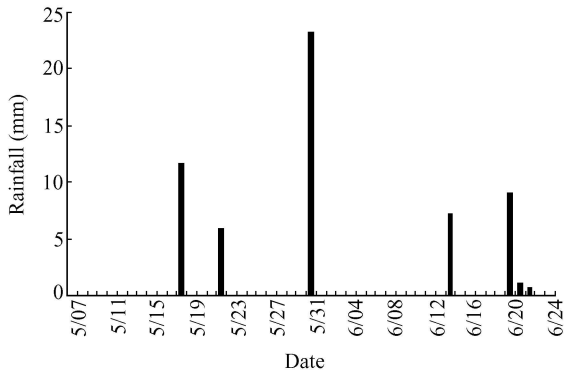


Fig. 1. Distribution of precipitation during the gas-monitoring period.

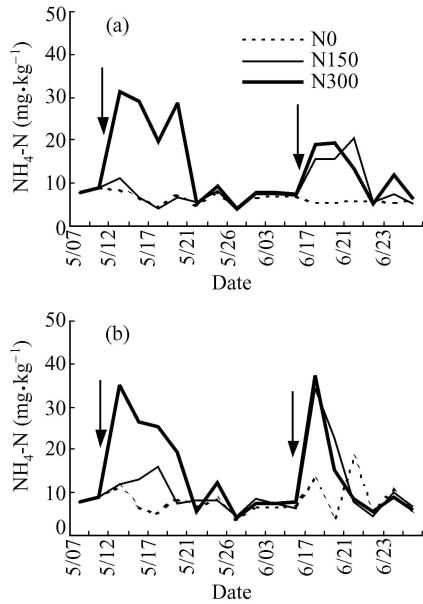


Fig. 2. Effect of N applied on  $\text{NH}_4\text{-N}$  in top 0~30 cm soil under low OM treatments (a) and high OM treatments (b). Arrows denote N fertilization.

Trace gas fluxes are shown in Figs. 4 and 5. Flux values for NO ranged from  $-10.77 \mu\text{g NO-N m}^{-2} \cdot \text{h}^{-1}$  for the control plots to  $174.9 \mu\text{g NO-N m}^{-2} \cdot \text{h}^{-1}$  emissions for the highest N-treatment, with average values ranging from  $-0.68$  to  $54.31 \mu\text{g NO-N m}^{-2} \cdot \text{h}^{-1}$  respectively. Losses over the sampling period were determined by trapezoidal interpolation between sampling days to give the cumulative losses over the sampling period and may be found in Table 1.

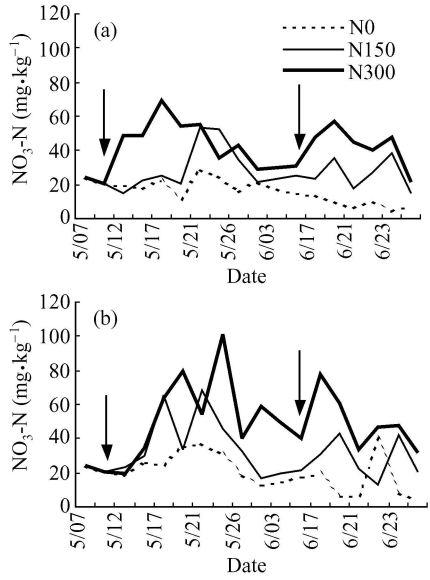


Fig. 3. Effect of N applied on  $\text{NO}_3\text{-N}$  in top 0~30 cm soil under low OM treatment (a) and high OM treatment (b). Arrows denote N fertilization.

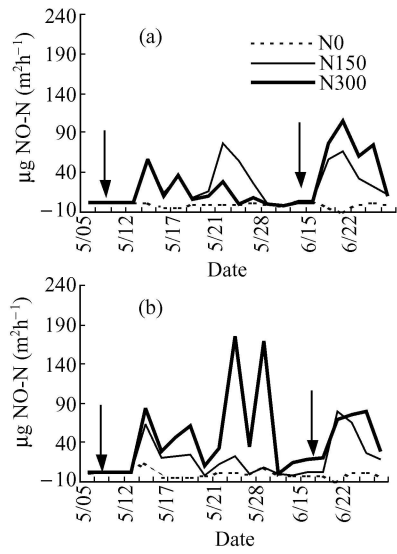


Fig. 4. Effect of N applied on NO fluxes under low OM treatment (a) and high OM treatment (b). Arrows denote N fertilization.

Losses for the highest three treatments, those most representative of typical Chinese agricultural practice, are compared with values in literature (Table 2). The losses compare well with those obtained in agricultural systems in the southeastern United States<sup>[23 24]</sup>, but are quite low compared with values derived from other parts of the world<sup>[25~27]</sup> with comparable fertilization rates. Our emission factors are roughly comparable to those given by Veldkamp et al. in their review<sup>[28]</sup> of 0.5% NO-N in

temperate agriculture. Fig. 5 represents N<sub>2</sub>O emissions. Fluxes over the sampling period range from 5.022 μg N<sub>2</sub>O-N m<sup>-2</sup>·h<sup>-1</sup> for the control plot to 359.5 μg N<sub>2</sub>O-N m<sup>-2</sup>·h<sup>-1</sup> for the 300 kg·N·ha<sup>-1</sup> high organic matter treatment , with average fluxes ranging from 10.24 to 123.3 μg N<sub>2</sub>O-N m<sup>-2</sup>·h<sup>-1</sup> respectively.

Losses over the sampling period were determined by trapezoidal interpolation between sampling days and summed to give cumulative losses over the sampling period , and may be found in Table 1. Losses for the highest three treatments are compared with reported values in Table 3. Our values compare reasonably well to those found in the literature. It has been shown that both the length of time over which measurements performed and the measurement frequency during the sampling period affect the observed magnitude of trace gas emission<sup>[29]</sup>. Additionally it should be noted that in systems such as these , background e-missions , probably associated with N deposition , are

not negligible for their contribution to overall nitrogen fluxes.

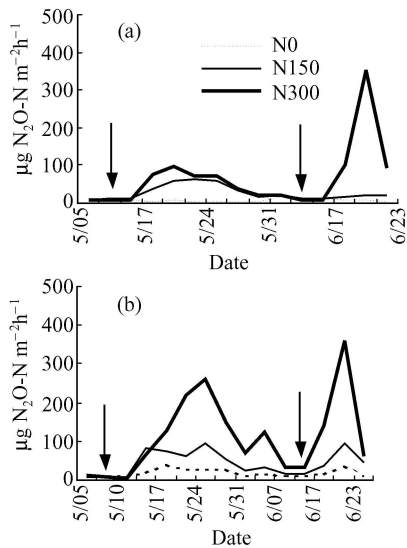


Fig. 5. Effect of N applied on N<sub>2</sub>O fluxes under low OM treatment ( a ) and high OM treatment ( b ). Arrows denote N fertilization.

Table 1. NO and N <sub>2</sub> O fluxes and their accumulative losses as affected by N applied under low OM and high OM conditions							
Treatment	Total N input ( kg·N·ha <sup>-1</sup> )	Average NO-N fluxes ( μg·m <sup>-2</sup> ·h <sup>-1</sup> )	Average N <sub>2</sub> O-N fluxes ( μg·m <sup>-2</sup> ·h <sup>-1</sup> )	Cumulative fluxes of NO-N ( kg·N·ha <sup>-1</sup> )	Cumulative fluxes of N <sub>2</sub> O-N ( kg·N·ha <sup>-1</sup> )	Seasonal loss of NO-N ( % )	Seasonal loss of N <sub>2</sub> O-N ( % )
Low OM							
N0	0	-0.7±3.5	10.2±3.3	-0.0016	0.12	—	—
N150	150	27.9±23.4	30.8±19.0	0.24	0.19	0.16	0.13
N300	300	27.5±34.0	72.8±90.3	0.23	0.60	0.075	0.20
High OM							
N0	88	0.05±5.5	22.0±10.6	0.0038	0.12	0.0043	0.14
N150	238	23.9±27.6	47.4±30.3	0.19	0.35	0.078	0.15
N300	388	54.3±50.6	123.3±103.2	0.52	1.20	0.13	0.30

Table 2. NO-N flux rates compared with other studies with comparable fertilization rates					
Study	N application rate ( kg·N·ha <sup>-1</sup> )	Length of study ( day )	Average loss ( μg NO-N m <sup>-2</sup> ·h <sup>-1</sup> )	Proportional loss ( % )	Loss normalized to 1 year( % )
This Study	238	49	23.9	0.078	0.58
This Study	300	49	27.5	0.075	0.56
This Study	388	49	54.3	0.13	1.0
Thornton et al. <sup>[24]</sup>	252	210	22.5	0.20	0.35
Thornton et al. <sup>[24]</sup>	336	365	15.5	0.36	0.36
Veldkamp et al. <sup>[28]</sup>	360	365	441	5.4	5.4
Matson et al. <sup>[26]</sup>	250	150	3000~5000	2.64~4.52	6.4~11
Jambert et al. <sup>[27]</sup>	280	365	50.2	11.3	11.3

Table 3.  $\text{N}_2\text{O}$ -N flux rates compared with other studies with comparable fertilization rates

Study	N application rate ( $\text{kg} \cdot \text{N} \cdot \text{ha}^{-1}$ )	Length of study (day)	Average loss ( $\mu\text{g} \text{N}_2\text{O-N m}^{-2} \cdot \text{h}^{-1}$ )	Proportional loss (%)	Loss normalized to 1 year (%)
This Study	238.5	49	47.4	0.15	1.1
This Study	300	49	72.8	0.20	1.5
This Study	388	49	123.3	0.30	2.2
Thornton et al. <sup>[23]</sup>	336	365	88.5	0.73	0.73
Jambert et al. <sup>[32]</sup>	230	365	179.2	1.1	1.1
Veldkamp et al. <sup>[25]</sup>	360	365	92.9	2.1	2.1
Thornton et al. <sup>[24]</sup>	252	210	138.2	2.6	4.5
Matson et al. <sup>[26]</sup>	250	150	1000~6500	1.8~2.2	4.5~5.5

A highly significant positive relationship was found between increasing urea application and NO flux ( $p < 0.0001$  for both base and top coat fertilizations) and  $\text{N}_2\text{O}$  emissions ( $p = 0.0007$  for base coat and  $p = 0.0431$  for top coat fertilization) by linear regression analysis. No relationship was found between organic matter and flux, and a close relationship ( $p = 0.1423$ ) was identified for  $\text{N}_2\text{O}$  following the base coat fertilization event. This result is contradictory to the findings of Thornton et al.<sup>[23]</sup> and Xing and Zhu<sup>[19]</sup>, who found significant differences in the fluxes from organic and inorganic N-fertilization sources; therefore, further investigation into this matter is warranted.

The length of response in this study appeared to be longer than that in others<sup>[6, 25]</sup>. In this study, elevated emissions were observed for close to 4 weeks following fertilization in the case of  $\text{N}_2\text{O}$ , and 2 weeks in the case of NO, suggesting that this temporal dynamic and causative factors should be taken into account in further studies.

The emission factor for NO was determined to be 1.25%, and 0.22% for  $\text{N}_2\text{O}$  of added nitrogen by linear regression. The proportion of NO is higher than the average in temperate agriculture estimated by Veldkamp et al.<sup>[28]</sup>. The proportion lost as  $\text{N}_2\text{O}$  is lower than the IPCC emission factor of 1.25%<sup>[30]</sup> but comparable to the  $\text{N}_2\text{O}$  emission rate of 0.54% in other region of North China Plain<sup>[31]</sup>, respectively. Although our data represent a small land area and a single land use, we believe that the differences in average emission factors should prompt further inquiry. Because China represents such a large proportion of the world's fertilizer consumption and a unique range of application rates, further studies are recommended in order to determine appropriate emission factors in China, and their incorporation into larger scale examinations of anthropogenically induced trace gas emissions. In general, the preliminary data on NO and

$\text{N}_2\text{O}$  fluxes from this study indicate that trace gas emissions from Chinese agricultural fields might be overestimated before.

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