

Seasonal characteristics of nitric oxide emission from a typical Chinese rice–wheat rotation during the non-waterlogged period

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Abstract

Nitric oxide emissions from a typical rice–wheat rotation system in southeastern China were continuously measured with an automatic system in 1996–1997. The seasonal pattern of the NO emissions was characterized for the non-waterlogged period of a rotation cycle. Nitric oxide emissions during the period from March through June were 3.9–6.3 folds for the fertilized plots and 1.6 folds for the unfertilized plot larger than those from November through December. Nitric oxide emissions were not detectable during the winter period from January through February. Amendment of synthetic fertilizer N significantly enhanced the NO emission by a factor of 6.5, but the enhancement was significantly mitigated by 25% through substituting ca. 16% of the synthetic fertilizer N with organic N from fermented crop residues or by 21% through deep tillage. The NO–N emission factor, defined as the amount of NO–N released per unit of synthetic fertilizer N input, was determined to be $0.025 \text{ kg NO-N kg}^{-1}$ of N applied for the non-waterlogged period, which was reduced by 32% through substituting part of the synthetic N fertilizer with fermented crop residues or by 24% through deep tillage. In addition, the NO emission factor, defined as the amount of NO–N emitted from unit unfertilized area per day, was observed to be ca. $3.8 \text{ g N ha}^{-1} \text{ d}^{-1}$. Approximately $0.55 \text{ Tg N yr}^{-1}$ was likely released as NO from Chinese cultivated lands.

Keywords: continuous measurement, cultivated land, emission factor, emission flux, nitric oxide

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Introduction

Gaseous nitrogen oxides ($\text{NO} + \text{NO}_2 = \text{NO}_x$) are well known as atmospheric trace constituents. These gases play a critical role in regulating the oxidation capacity (e.g. O_3 , OH) of the troposphere (Crutzen, 1983) and are directly involved in the production and depletion of other greenhouse gases, e.g., ozone (O_3) (Duxbury *et al.*, 1993). Global estimates of the NO_x amount released per year range from 20 to 100 Tg N ($1 \text{ Tg} = 10^{12} \text{ g}$) (Conrad, 1990; Lee *et al.*, 1997). Soils have been recognized as an important source of tropospheric NO_x since 1978 when Galbally & Roy (1978) reported the first field measurement of nitric oxide (NO) emissions from grassland. The production of NO in soils is tightly coupled to the microbial processes of nitrification and denitrification (e.g.

Firestone & Davidson, 1989) and, if the soil pH is lower than 5.0, chemodenitrification may also contribute (Van Cleemput & Baert, 1984; Galbally, 1989). There is no evidence for biological production of nitrogen dioxide (NO_2) in soils, but the year-round deposition of atmospheric NO_2 into the ground of forest ecosystems was detected (Gasche & Papen, 1999). There may be a small degree of $\text{NO} + \text{O}_3 = \text{NO}_2$ occurring within the top few mm of the soil leading to apparent NO_2 emission. However, more than 90% of NO_x is released from soils as NO (Conrad, 1990; Williams *et al.*, 1992). The global total of NO emissions from soils were estimated to be within a range from 4 to 21 Tg N yr^{-1} (Yienger & Levy, 1995; Davidson & Kingerlee, 1997). The great uncertainties of these global estimates were thought to come from wide variations of field observations (Hutchinson & Davidson, 1993) and the unknown effects of plant canopy on the oxidation and re-deposition of NO, which may prevent up to 50% of soil-released NO from escaping into the ambient atmosphere (Lovett & Lindberg, 1993;

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Robertson, 1993; Yienger & Levy, 1995). Field measurements of NO emissions have shown a high degree of temporal and spatial variability (e.g. Butterbach-Bahl *et al.*, 1997; Gasche & Papen, 1999). Nitric oxide fluxes from cultivated soils measured in North and South America (e.g. Matson *et al.*, 1996), Europe (e.g. Jambert *et al.*, 1994, 1997), Australia (e.g. Galbally *et al.*, 1987) and China (Zheng *et al.*, 1998a) range from 1 to 556 ng N m⁻² s⁻¹ with a mean of 40.4 ng N m⁻² s⁻¹ for fertilized fields, from 0.3 to 21.7 ng N m⁻² s⁻¹ with a mean of 4.3 ng N m⁻² s⁻¹ for unfertilized fields, and 0.2 ng N m⁻² s⁻¹ for rice paddy fields. The high variability is mainly due to variations in the physical, chemical and biological properties of soils and parameters including soil temperature, soil moisture, farm practices, vegetation cover, plant uptake, season, surface wind speed, and NO_x concentrations in the air above the soil (Williams *et al.*, 1987; Kim *et al.*, 1994). In order to reduce the uncertainties and to develop mitigation strategies of anthropogenic NO emissions from cultivated lands, Matson (1997) suggested there was a need (1) to quantify the effects of canopy on NO_x-exchange between the biosphere and the atmosphere, (2) to investigate the impacts of management practices, and (3) to understand the consequences of changes in N cycling, e.g., due to N-deposition and/or N-fertilization.

Anthropogenic NO emission from cultivated lands was estimated to be within a range from 1.4 to 5.3 Tg N yr⁻¹, accounting for about 40% of the global soil source of NO (Yienger & Levy, 1995; Davidson & Kingerlee, 1997; Lee *et al.*, 1997), and could be a dominant source of NO in some less developed regions (Matson, 1997). Field measurements also indicated that farming practices including fertilization, irrigation and tillage are important factors in determining the NO emission (e.g. Jambert *et al.*, 1997; Sanhueza, 1997). However, few studies to date have dealt with the quantitative relationship between NO emissions and the parameters of soil, climate and crop farming practices.

China's cropland area in accounts for ca. 7% of the global total and the annual amount of synthetic fertilizer N used contributed ca. 27% to the global consumption in the mid 1990s (FAO, 2001). Though the cropland acreage in China has recently decreased by 0.14% per year, the synthetic fertilizer N consumption has annually increased by ca. 6.5%, being higher than the value of 4.8% for other Asian regions (FAO, 2001). In addition to the high rate of synthetic fertilizer N amendment, China has a great diversity of agricultural practices. Because of a lack of field measurements, little knowledge of NO emissions from the cultivated land in China has been available so far. To accurately estimate the NO emission from Chinese croplands, and hence to develop feasible NO mitigation strategies, it is essential to get first hand data sets *in situ*.

In this paper, we report our field measurements of NO emission from a typical Chinese rice-wheat rotation during the non-waterlogged period of 1996–1997. The seasonal fluxes and NO–N emission factors are presented and the total emission of NO from the croplands of China is discussed.

Materials and methods

Experimental site and field treatments

Field experiments during the non-waterlogged period of a rice-wheat rotation, which includes the whole wheat-growing season and its following fallow period, were performed in 1996–1997 on a typical permeable paddy soil at a suburb of Suzhou city (31°16'N, 120°38'E), Jiangsu province, China. Zheng *et al.* (2000) described the location, climate, soil properties and farming practices of the experimental sites.

The experimental field was divided into four plots, 12 m × 8 m each. Winter wheat, *Triticum aestivum* L. cv. Yangmai 5, a local prevailing cultivar, was sown in all plots on November 1st, 1996 and harvested on May 27th, 1997. Then the field remained bare until the rice transplanting on June 18th. No irrigation was performed during the wheat-growing period. The plots were wetted through irrigation for 1 week before flooding and plowing on June 17th.

Three treatments of fertilization and two treatments of tillage were applied. The control, i.e., no fertilizer applied, was assigned to plot A. The treatment of organic manure plus synthetic fertilizers was assigned to plot B and that of synthetic fertilizers was assigned to plots C and D. The rate of fertilizer application was 191 kg N ha⁻¹ for the wheat-growing season. For plot B, wheat straw aerobically fermented for about 5 months was raked into the surface soil at a rate of 10 t ha⁻¹ (ca. 30 kg N ha⁻¹) before wheat sowing. Meanwhile, compound fertilizer (N : P₂O₅ : K₂O = 12% : 6% : 7%) and urea were simultaneously applied at a rate of 30 and 6 kg N ha⁻¹, respectively, as basal fertilizers. For C and D, the basal fertilizer N was applied at the same rate of 66 kg N ha⁻¹, at 30 kg N ha⁻¹ of compound fertilizer and 36 kg N ha⁻¹ of urea. Thereafter, urea was top-dressed for all the fertilized plots on December 29th, 1996, February 25th and March 13th, 1997 at 29, 48 and 48 kg N ha⁻¹, respectively. Before wheat sowing, all the plots were tilled. Plot D was tilled to a depth of 15 cm, and the others 5 cm.

In situ measurements of NO fluxes

During the period from November 1st, 1996 through June 17th, 1997, NO emissions were continuously and automatically measured using a static chamber-based

method. With technical help from the Institute for Meteorology and Climate Research, Karlsruhe Research Center (formerly the Fraunhofer Institute for Atmospheric Environmental Research) (e.g. Zheng *et al.*, 1997, 1998b), scientists at the Institute of Atmospheric Physics, Chinese Academy of Sciences designed an automatic system for simultaneous *in situ* measurement of CH₄, N₂O and NO emissions from croplands, which was used in this study. Figure 1 shows the structure of the automatic system modified from Zheng *et al.* (2000). With this system, air samples were automatically collected from closed chambers installed in the field plots and injected into the NO, CH₄ and N₂O instruments for analysis. As shown in Fig. 1(b), there were 23 3-way valves driven by a set of 24-V direct current supplies. Of these, six valves were dedicated to switch compressed air flow for chamber closure/opening, 12 were used to switch air sample flow from different chambers, three were employed to dry the samples via a cooling water trap, and two were orientated to switch air samples between the NO analyzer and the injection sector of a gas chromatography (GC) for analysis of N₂O and CH₄. A Trace Level 42C NO–NO₂–NO_x chemiluminescent analyzer (Thermo Environmental Instruments Inc., USA), which is called a NO_x analyzer, was included in the automatic system.

A total of nine 70 cm × 70 cm mini-plots, one chamber for each, were simultaneously monitored. Three mini-plots were monitored in plot B, and two were observed in each of plots A, C and D. Each chamber was inserted into the soil to a depth of 10 cm and with an 80-cm high headspace. Two ventilators for each chamber were installed at opposite walls inside to maintain air turbulence. To determine an emission flux, the chamber was programmed to close for 60 min and four air samples were taken during each closure. A diaphragm pump transported the sample air at a flow rate of 250 mL min⁻¹ to the instruments for analysis. It took 5 min for a complete measurement, i.e. automatically taking the sample air, injecting into the GC for determination of N₂O (Zheng *et al.*, 2000) and CH₄, and analyzing NO with the NO_x analyzer. From 0.0 to 4.0 min of each 5 min, the sample air was directed through the NO_x analyzer and the NO volume mixing ratio was automatically recorded at 3.5 min.

The temperature of the air and soil (2 cm depth) inside the chamber was simultaneously measured every 10 min with thermocouples (Fig. 1a). Before the temperature probes were permanently installed in the fields, they were accurately calibrated with a mercury thermometer. The probes for the air temperature measurement were protected against direct sunlight. In addition, the pressure of the ambient air was manually recorded four times per day with a barometer.

The NO flux was calculated with the following equation,

$$F = k \cdot \frac{T_0}{T_0 + T} \cdot \frac{P}{P_0} \cdot H \cdot \rho \cdot \frac{dC}{dt},$$

in which F is the NO emission flux (ng N m⁻² s⁻¹), T is the average air temperature (°C) inside the chamber, P is the daily mean of air pressure (hPa), H is the headspace height of the chamber (cm), ρ is the NO density (g L⁻¹) at T_0 (273 K) and P_0 (1013 hPa), C is the NO mixing ratio by volume (10⁻⁹), t is the time (min) for chamber closure, dC/dt is the increase rate of NO mixing ratio by volume (10⁻⁹ min⁻¹) in the chamber, and the coefficient $k = 1/6 \times 14/30$ is introduced to give units of ng N m⁻² s⁻¹.

Under an inlet flow rate of 250 mL min⁻¹, the NO detection channel of the NO_x analyzer was calibrated in laboratory by calibration gases with concentrations of 0, 100 and 180 × 10⁻⁹ mixing ratio by volume. Then a calibration curve was obtained as $C_a = 3.517C_r$ ($R^2 = 0.996$, $P < 0.001$), where C_a the actual NO concentration of the gas and C_r the instrument reading of NO concentration. It was applied to determine the NO concentrations of gas samples from the instrument readings. A standard gas with a NO volume mixing ratio of 11 × 10⁻⁹ in air, which was very near the NO volume mixing ratio of the local ambient air, was connected with the field sampling tube of a chamber and analyzed for a 1-h period using the automatic system. These calibrations were performed at the beginning and the end of the observation period, and the 1σ value (i.e. standard deviation) for quantifying the NO volume mixing ratio was determined to be 1.4 × 10⁻⁹. Based on this 1σ value, the chamber headspace height (80 cm), and the '3σ statistical principle', the detection limit, i.e., the minimum NO emission flux that the automatic system was able to detect, under 1013 hPa and 273 K, was determined to be 12 μg NO m⁻² h⁻¹ or 1.56 ng N m⁻² s⁻¹. The actual detection limits fluctuated depending on the air temperature and pressure, and ranged from 1.58 to 1.75 ng N m⁻² s⁻¹ over the whole observation period.

Results

Extraction of valid fluxes from observed NO emission data

Fluxes with a value higher than the detection limit were accepted as valid data. Among a total of 8647 flux data points obtained over the whole observation period, 6374 were accepted as valid, and the remainder were rejected as null data. Of the null data, 76% were measured during the winter period from January through February, when NO emission contributed to less than 1% of the total amount over the whole observation period. The negative fluxes, accounting for 1.3% of the observations, were

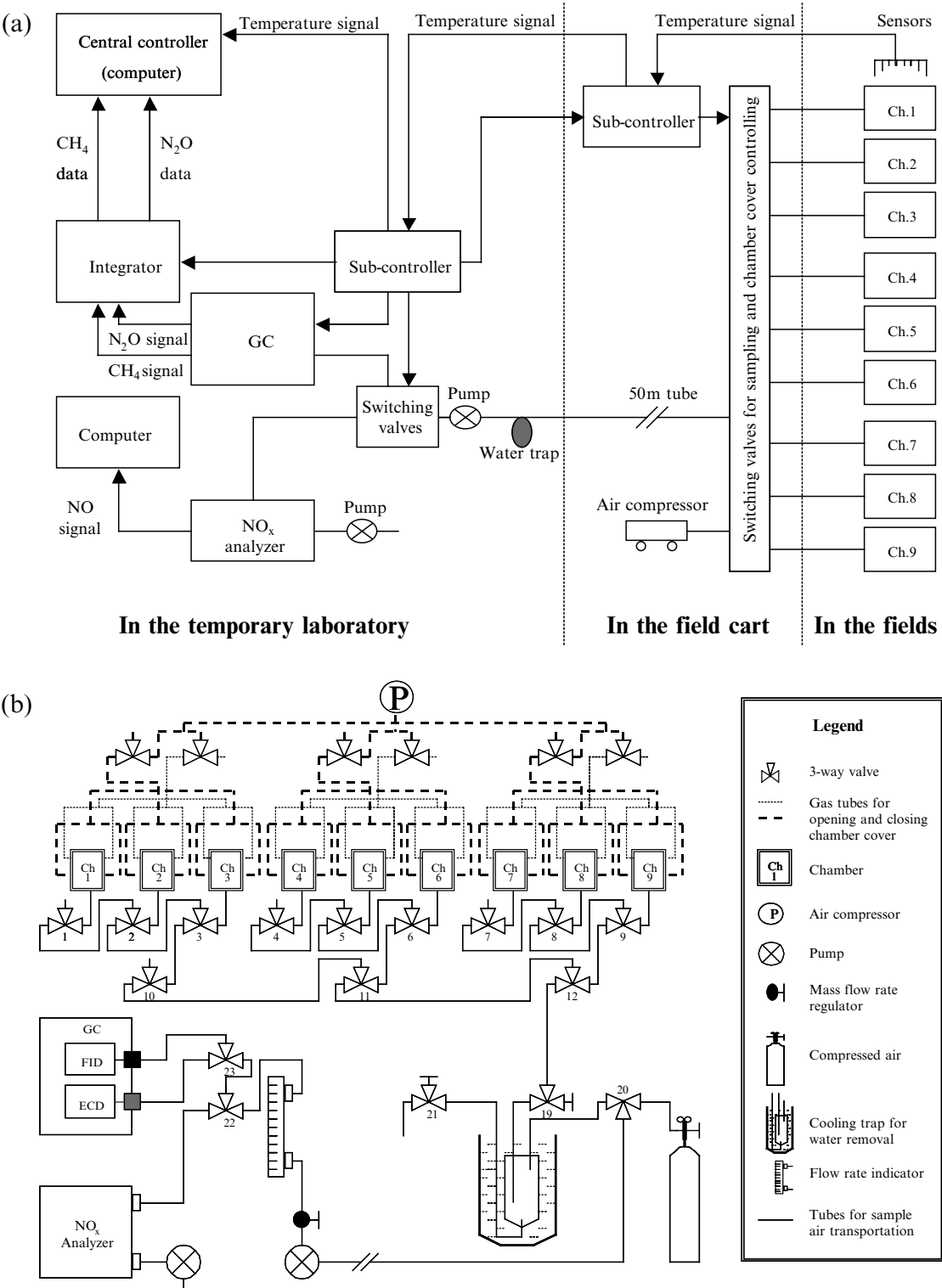


Fig. 1 (a) Structure of the automatic system and (b) Gas flow circuit of the automatic system for simultaneously measuring NO, N₂O and CH₄ emissions from croplands.

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usually null due to sudden disturbances instead of real uptake of NO by soil.

Seasonal variability in NO emission fluxes

Figure 2 shows the seasonal distribution of NO emission from each plot and the daily average temperature in the surface soil (2 cm) inside the chamber. The observed NO emission for each treatment is summarized in Table 1. As shown in Fig. 2, NO emissions from all the plots appeared in a similar seasonal pattern over the non-waterlogged period, which was roughly consistent with the variation pattern of surface soil temperature. This seasonal pattern was characterized as being higher during the period from March until the flooding of the fields for transplanting rice in mid June (MJ), lower during the period from

November through December (ND), and almost undetectable during the period from January through February (JF). Evidently, the NO emission during the MJ period was more intensive than during the ND period, and the difference of NO emission between the two periods in the fertilized plots was even larger than in the unfertilized plot. The seasonal average flux for the fertilized plots was $38.6\text{--}54.6\text{ ng N m}^{-2}\text{ s}^{-1}$ in the MJ period and $8.7\text{--}9.9\text{ ng N m}^{-2}\text{ s}^{-1}$ in the ND period, yielding an average factor of 4.9 with a range from 3.9 to 6.3. For the unfertilized plot, NO was released at a seasonal average flux of $6.5\text{ ng N m}^{-2}\text{ s}^{-1}$ in the MJ period and 4.0 in the ND period, yielding a factor of 1.6. During the JF period, NO emission flux was too low to be detected with the automatic equipment. The average flux of this period, when no valid measured data was available, was estimated

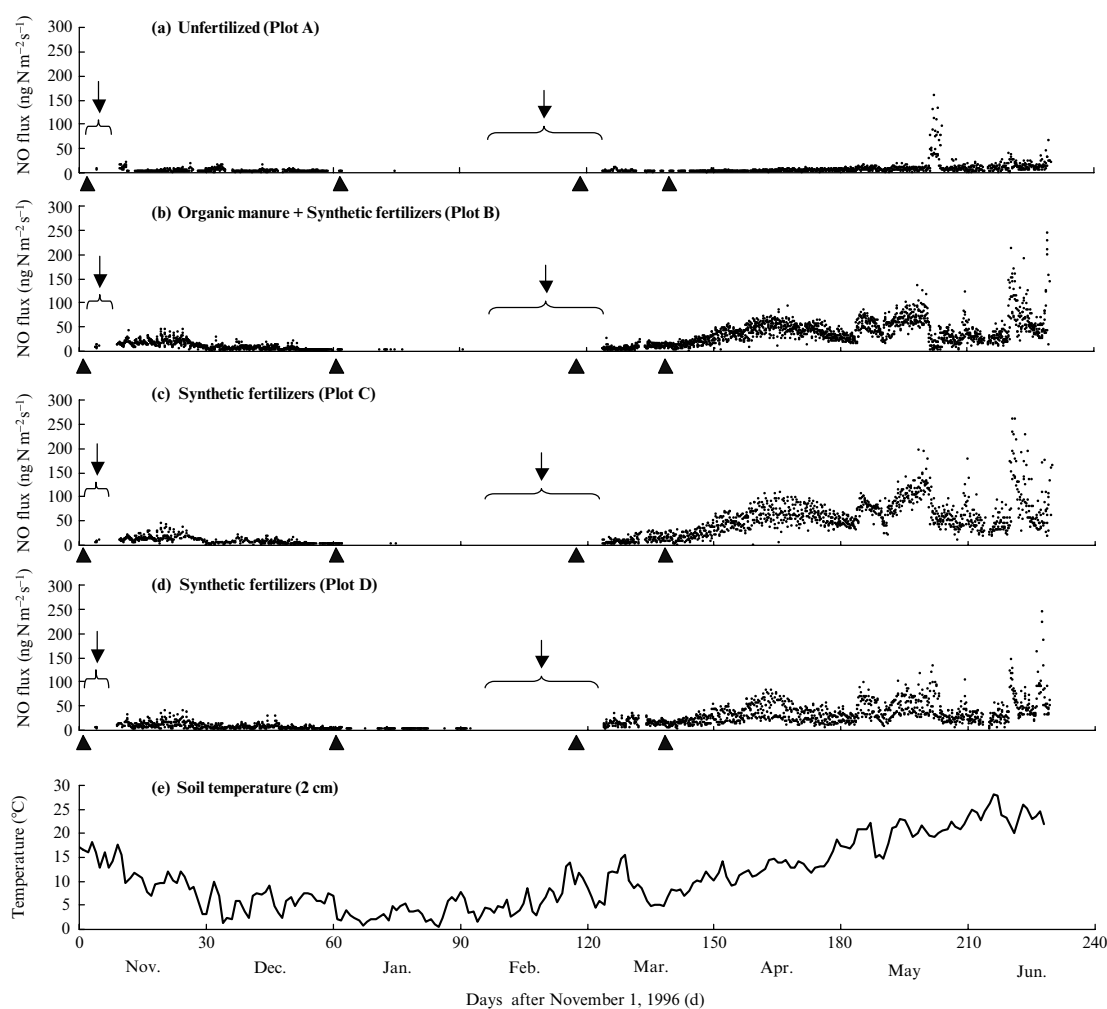


Fig. 2 (a) Nitric oxide emission from the unfertilized plot, (b) Nitric oxide emission from the plot amended with organic manure and synthetic fertilizers, (c) Nitric oxide emission from the plot amended with synthetic fertilizers, (d) Nitric oxide emission from the plot amended with synthetic fertilizers and treated with deep tillage before wheat sowing, and (e) Daily means of surface soil (2 cm depth) temperature. The upward triangles indicate the date on which fertilizers were applied. The downward arrow-braces indicate the periods without data because of equipment failure.

Table 1 Seasonal total amount, average flux and emission factor of NO

Field plots	A	B	C	D ^g
Average flux of ND ^a period (ng N m ⁻² s ⁻¹)	4.0 ± 1.9	9.9 ± 3.4	8.7 ± 3.2	9.5 ± 3.6
Average flux of JF ^b period (ng N m ⁻² s ⁻¹)	-1.6 ~ 1.6 ^e	-1.6 ~ 1.6 ^e	-1.6 ~ 1.6 ^e	-1.6 ~ 2.1
Average flux of MJ ^c period (ng N m ⁻² s ⁻¹)	6.5 ± 4.3	38.6 ± 12.6	54.6 ± 16.0	41.2 ± 9.7
Ratio of the MJ to the ND period	1.6	3.9	6.3	4.4
Sub-total of ND period (kg N ha ⁻¹)	0.22 ± 0.10	0.53 ± 0.18	0.47 ± 0.17	0.50 ± 0.19
Sub-total of JF period (kg N ha ⁻¹)	-0.08 ~ 0.08 ^f	-0.08 ~ 0.08 ^f	-0.08 ~ 0.08 ^f	-0.08 ~ 0.11
Sub-total of MJ period (kg N ha ⁻¹)	0.62 ± 0.52	3.64 ± 2.85	5.14 ± 3.04	3.88 ± 0.91
Total of ND, JF and MJ (kg N ha ⁻¹)	0.87 ± 0.53	4.21 ± 1.37	5.64 ± 1.70	4.43 ± 1.12
Average flux over ND, JF and MJ (g N ha ⁻¹ d ⁻¹)	3.80 ± 2.30	18.38 ± 5.99	24.63 ± 7.44	19.35 ± 4.89
Contribution of MJ period to the total (%) ^d	71	87	91	87
Ratio of B, C and D to A	1.0	4.8	6.5	5.1
Fertilizer-induced emission (kg NO-N ha ⁻¹)	–	3.34	4.77	3.56
Emission factor of fertilizer-N (%)	–	1.75	2.50	1.87

^{a,b,c}Abbreviation of the period from November through December, from January through February, and from March through June, respectively.

^dPercentage of NO released during the MJ period to the total amount over the whole observation period.

^eA value of 1.6 ng N m⁻² s⁻¹ is the detection limit of the automatic equipment.

^fCalculated with the detection limit of 1.6 ng N m⁻² s⁻¹.

^gWheat plant growth at one of the duplicate mini-plots in plot D was abnormal. The biomass production was ca. 70% lower than that of normal mini-plots. The data observed at this abnormal mini-plot would not represent the actuality of D. Therefore, only the data of the other duplicate mini-plot were adopted in the calculation of the seasonal total amount and the average flux for plot D.

between -1.6 and 1.6 ng N m⁻² s⁻¹, the average detection limit. Over the entire non-waterlogged period of the rice–wheat rotation cycle, ca. 84% (ranging from 71% to 91%) of the total NO emission occurred during the MJ period and only ca. 14% in the ND period.

Effects of fertilization and tillage depth

Similarity in the seasonal distribution of NO emission fluxes from all the plots suggests that fertilization and tillage may not influence the seasonal pattern over the non-waterlogged period. However, the magnitude of the seasonal NO amount might be affected by fertilization and tillage. The daily averages of the NO emission fluxes from all fertilized plots were larger than those from the unfertilized plot (*t*-test, $\alpha = 0.05$, $P < 0.001$), suggesting that the application of fertilizers enhanced the NO emission over the investigated period. Total NO emission from the plots of A, B, C and D over the entire non-waterlogged period amounted to 0.87 ± 0.53 , 4.21 ± 1.37 , 5.64 ± 1.70 and 4.43 ± 1.12 kg N ha⁻¹, respectively. In comparison with the unfertilized plot, the amendment of synthetic fertilizer N in the fertilized plots intensified the total NO emission by a factor of 6.5 (Table 1). However, the stimulation of NO emission was significantly mitigated by 25% through substituting ca. 16% of the synthetic fertilizer N with organic N from aerobically fermented wheat residues (*t*-test, $\alpha = 0.05$, $P < 0.001$) or by

21% through deep tillage before wheat sowing (*t*-test, $\alpha = 0.05$, $P < 0.001$).

Because of missing data caused by two equipment failures at the beginning of November and in February/March (Fig. 2a–d), the immediate responses of NO emission to these two applications of nitrogen fertilizer remain unclear. However, the successful observations following the other two applications of urea, one on December 29th and the other on March 13th, show that no intensive pulse of NO emission was caused by N-fertilizer amendment (Fig. 2a–d), which was likely due to a relatively lower soil temperature of ca. 5 °C (Fig. 2e).

NO–N emission factors

The NO–N emission factor of amended fertilizer N (EF) over the non-waterlogged period was defined as $EF = NO-N / \text{Fertilizer-N}$, where NO–N is the N released as NO from the total amount of fertilizer-N applied during the wheat-growing season. The NO–N was quantified as the difference between the total NO amount released from a fertilized plot and that from the unfertilized plot. As listed in Table 1, the EF for plot B, C and D was calculated as 1.75%, 2.50% and 1.85%, respectively. For all the fertilized plots, there was an average EF of 2.0%. and for plots C and D, where only synthetic fertilizer N was amended, the average EF was 2.19%. Evidently, the EF for plots B and D was lower than that

for plot C by 32% and 24%, respectively, suggesting that substituting part of the synthetic fertilizer with organic manure or deep tillage before wheat sowing significantly reduced the EF (*t*-test, $\alpha = 0.05$, $P < 0.001$).

We also defined the NO emission flux as an emission factor on the basis of area (hereafter a term of area-based emission factor is also used), analogous to the definition of the methane emission factor from rice paddy fields (IPCC, 1997). The seasonal average NO emission factor on the basis of area during the non-waterlogged period of the rice–wheat rotation was quantified as 18.4 ± 5.0 , 24.6 ± 7.4 and $19.4 \pm 4.9 \text{ g N ha}^{-1} \text{ d}^{-1}$ for plot B, C and D, respectively, with a mean of $20.8 \pm 10.2 \text{ g N ha}^{-1} \text{ d}^{-1}$ for the fertilized plots, and 3.8 ± 2.4 for the unfertilized plot (Table 1).

Discussion

The NO emission observed in North and South America, Europe and Australia has an average value of ca. $40 \text{ ng N m}^{-2} \text{ s}^{-1}$ for fertilized agricultural soils and ca. $4 \text{ ng N m}^{-2} \text{ s}^{-1}$ for unfertilized agricultural soils (e.g. Galbally *et al.*, 1987; Jambert *et al.*, 1994, 1997; Matson *et al.*, 1996). In the unfertilized plot, the average NO emission fluxes we observed during both the MJ and ND periods were at a similar level. In the fertilized plots, we also observed a similar average flux of NO emission during the MJ period. During the ND period, however, the NO emission flux we observed appeared much lower, suggesting that periodically flooding the fertilized fields for rice cultivation negatively affected the NO emission during the non-waterlogged period. This effect was likely due to higher soil moisture during the ND period, which was caused by field flooding during the previous rice-growing period, than that of usual dry-land soils free from flooding events. During the MJ period, the soil pores were well developed through the alternation of freezing and thawing and root penetration and so the soil moisture was similar to fertilized dry-land soils. The soil moisture during the ND period was observed to be $41\% \pm 4\%$ (w/w), vs. $34\% \pm 5\%$ (w/w) during the MJ period excluding the 1-week wetting stage. The aeration status of the soil was less developed because of higher soil moisture and thus the NO emission caused by nitrification was likely mitigated to a certain extent.

During the non-waterlogged period of the rice–wheat rotation, the EF of NO–N for each treatment was larger than that of N₂O–N, which was observed to be 1.0%, 1.9% and 1.3% for plot B, C and D, respectively, with an average of 1.4% (Zheng *et al.*, 2000). Coupling aerobically fermented wheat residues with synthetic fertilizers, which was one of the local usual farming practices, or using deep tillage reduced the emission factor of both NO–N and N₂O–N. But the NO–N emission factor was

less effectively reduced in comparison with the reduction of the N₂O–N emission factor. The NO–N emission factor was reduced by 32% (vs. 49% for the EF of N₂O–N) through substituting part of the synthetic fertilizer N with organic manure N or by 24% (vs. 29%) through deep tillage. Nevertheless, both farming practices proved to be effective for simultaneously mitigating the NO and N₂O emissions caused by amendment of synthetic N fertilizers. This result is very important, in particular for the atmospheric environment protection in China, where synthetic fertilizer N is intensively consumed.

In China, there were $9.55 \times 10^7 \text{ ha}$ of dry cropland area and $2.56 \times 10^7 \text{ ha}$ of rice paddies in the mid 1990s (ASESP-CNSB, 1996). During this period, China annually harvested $11.29 \times 10^7 \text{ ha}$ of upland crops and $3.08 \times 10^7 \text{ ha}$ of paddy rice, ca. 30% of which was double rice (ASESP-CNSB, 1996; FAO, 2001). In the meantime, China annually consumed 21.30 Tg of synthetic fertilizer N (FAO, 2001). Assuming that the consumption of synthetic fertilizer N was proportional to the land area occupied by paddy rice and dry land crops, then ca. 21% or 4.57 Tg of the synthetic fertilizer N was applied to paddy rice and the remaining 16.73 Tg to dry land crops. The NO emission from croplands usually consists of two parts. One is derived from the immediate release due to the fertilizer N. The other is derived from other N sources including mineralization, atmospheric N deposition, N input from irrigation water, and the remnant fertilizer N from the previous year. The former part can be calculated with the observed EF and the amount of consumed fertilizer N. We define the emission factor on the basis of area for unfertilized agricultural fields as the background NO emission factor, and the later part is the so-called background NO emission. The background NO emissions can be estimated with the cropland acreage and the background emission factor determined through observation of unfertilized fields. With a value of 2.19%, the average EF observed in plots C and D, where only synthetic fertilizers were applied, the NO emission from the consumption of synthetic fertilizer N in the dry-land crop fields of China was roughly estimated to be $0.37 \text{ Tg N yr}^{-1}$ in the mid 1990s. Usually, the non-waterlogged period is about 5 months long for double rice (two rice harvests within one year) fields and 8 months long for single rice (one rice harvest within one year) fields. With a value of $3.8 \text{ g N ha}^{-1} \text{ d}^{-1}$, the background emission factor measured in the unfertilized plot, the background NO emission from the dry-land crop fields and the rice paddies during the non-waterlogged period was estimated to be $0.18 \text{ Tg N yr}^{-1}$ in the mid 1990s. For waterlogged rice paddy fields, an area-based emission factor of $0.2 \text{ ng N m}^{-2} \text{ s}^{-1}$, or $0.17 \text{ g N ha}^{-1} \text{ d}^{-1}$, was reported by Galbally *et al.* (1987). Our measurements presented a NO–N emission factor of 0.05% for the synthetic fertilizer

N applied during the rice growing season and a background area-based emission factor of $0.17 \text{ g N ha}^{-1} \text{ d}^{-1}$ for the same period (unpublished data from a sister measurement campaign of this study). With these emission factors, the NO emission from the waterlogged rice paddy fields of China was estimated to be $0.001\text{--}0.003 \text{ Tg N yr}^{-1}$, which is ignorable in comparison with the magnitude of estimated NO emission from the non-waterlogged farmlands. By summing up the above estimates, approximately $0.55 \text{ Tg N yr}^{-1}$ of NO was likely released from the Chinese croplands, which accounted for 10–40% of the latest estimate, $1.35\text{--}5.5 \text{ Tg N yr}^{-1}$, for global cultivated lands (Yienger & Levy, 1995; Davidson & Kingerlee, 1997).

Conclusions

The NO emission fluxes during the period from March through June were significantly higher than those during the period from November through December by a factor of 4.9 for the fertilized plots and 1.6 for the unfertilized plot. No significant NO emissions were observed during the period from January through February. The seasonal pattern of the NO emission was not affected by fertilization and tillage, whereas application of synthetic fertilizer N significantly enhanced the magnitude of NO emission by a factor of 6.5, in comparison with NO emission from the unfertilized plot. The enhancement was significantly mitigated by 25% through coupling aerobically fermented wheat residues with the synthetic fertilizers or by 21% through deep tillage before wheat sowing. The NO–N emission factor for synthetic fertilizer N applied during the non-waterlogged period of the rice–wheat rotation was determined to be 2.5%, which was reduced by 32% through substituting ca. 16% of the synthetic fertilizer N from aerobically fermented wheat residues and by 24% through deep tillage. The background NO emission factor on the basis of area for non-waterlogged croplands was determined to be $3.8 \text{ g N ha}^{-1} \text{ d}^{-1}$. The cultivated farmlands of China are likely to annually release ca. 0.55 Tg N in form of NO, which accounts for 2.6% of the consumed synthetic fertilizer N.

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