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Nitric oxide emissions from black soil, northeastern China: A laboratory study revealing significantly lower rates than hitherto reported

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ABSTRACT

Nitric oxide (NO) is an important component of biogeochemical cycling of nitrogen, produced via biologically mediated processes of nitrification and denitrification in soils. The production and consumption processes of NO in black soils are not fully understood. We established how moisture and temperature affect NO dynamics for black soil samples of maize land in the temperate zone of northeastern China. The optimum soil moisture for the maximum NO production and emission was determined to be 41% water-filled pore space (WFPS), based on laboratory experiments and modeling. For a given moisture, NO fluxes increased exponentially with soil temperature at any given soil moisture. The optimum soil moisture for the maximum NO emission was constant and independent of soil temperature. The NO consumption rate constant (k) in the studied soil (range 9.31×10^{-6}–15.1×10^{-6} $m^3\ kg^{-1}\ s^{-1}$) was in the middle of the range of similar k values published to date. The maximum NO emission potential for black soils at 25 °C and 15 °C were about 18.6 and 9.0 ng N $m^{-2}\ s^{-1}$, respectively. Based on laboratory results and field monitoring data of soil water content and soil temperature, the average NO fluxes from black soils in the region were estimated to be 10.7 ng N $m^{-2}\ s^{-1}$ for an entire plant growth period. NO emissions likely occur principally in July, associated with optimum soil moisture. The present study suggests that NO fluxes from black soil are much lower than the previous reports from cropland in southern parts of China.

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1. Introduction

Nitric oxide (NO) is a frequent subject of ongoing studies due to its importance to the production and the destruction of tropospheric ozone (Cicerone, 1987; Crutzen, 1979) as well as soil nitrogen (N) loss to the atmosphere. NO is a precursor in the photochemical formation of gaseous nitric acid (HNO_3) and thus contributes to the acidity of clouds and precipitation (Liu et al., 1987). It is also an important precursor to ozone (O_3) formation in the lower atmosphere, which can lead to undesirable air quality and detrimental effects on human health and may decrease crop yields (Tabachow et al., 2002).

Soils are a major source of atmospheric NOx (NOx = NO + NO_2) (Delmas et al., 1997; Yienger and Levy, 1995). Worldwide, the budget of NOx source from soils is around 8.9 Tg N y^{-1} or about 20% of the global total source (IPCC, 2007). Thus, soils contribute to the global budgets of NO sources; their contribution as sinks is likely but considered to be small (Meixner and Eugster, 1990). Nitric oxide in soil is produced through biological processes of nitrification and denitrification, as well as chemical decomposition of HNO_2 (Firestone and Davidson, 1989; Remde and Conrad, 1991; Conrad, 1996; Gödde and Conrad, 2000; Russow et al., 2009). Temperature, soil moisture, soil texture, fertilization and land-use have all been shown to be key factors controlling NO emission from soils (Gut et al., 1999, 2002; Pilegaard et al., 1999; Venterea and Rolston, 2000; Venterea et al., 2005; Williams and Fehsenfeld, 1991; Yu et al., 2008). The approximate ranking of NO emission levels sources, *viz.* fertilized agricultural fields > grasslands > forests > other

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natural systems is in broad accordance with the nitrogen status of corresponding soils (Ludwig et al., 2001). According to present knowledge, NO is produced in soils nearly ubiquitously; therefore soil emissions constitute a continuous background flux of NO to the atmosphere (Williams et al., 1992). Although soils are usually net sources for atmospheric NO, they can also act as sink, at least temporarily (Slemr and Seiler, 1984). Most studies to date have concentrated on production of NO and neglected consumption processes. This is mainly because that production rates are usually larger than consumption rates and therefore, net production is the overall measure. NO can be both oxidized and reduced by soil microorganisms (Remde and Conrad, 1991; Rudolph et al., 1996). The oxidative NO consumption has much lower affinity for NO than the reductive consumption, but seems to be dominant in many soils (Koschorrech and Conrad, 1997).

Although soils are a considerable source of atmospheric NO*x*, there still exists huge uncertainty and controversy about the total contribution. A probable cause of this uncertainty is that the disproportionate numbers of previous studies of NO emissions have been carried out for only a few regions. For example, most studies about NO fluxes in cropland have been carried out in Europe and USA (Aneja et al., 1995; Civerolo and Dickerson, 1998; Davidson, 1992; Gut et al., 1999; Harrison et al., 2002; Li et al., 1999; Russow et al., 2008; Skiba and Ball, 2002; Tabachow et al., 2001; Thornton and Valente, 1996; Venterea and Rolston, 2000). Only four reports on NO emissions from Asian cropland soils are apparent to our knowledge, (Fang and Mu, 2006; Li and Wang, 2007, 2008; Zheng et al., 2003b) and none of these involve Asian black soil. However, the controversy cannot necessarily be resolved by integration of fluxes over larger areas and longer time periods, since most of the observed uncertainties and problems with NO flux data can be traced (Meixner and Yang, 2006). Some researchers have found that net fluxes of NO, which derive from soil samples in the laboratory, agreed well with dynamic chamber measurements at corresponding field sites (Ludwig et al., 2001; Otter et al., 1999; van Dijk et al., 2002). In this study, we present NO emission results from laboratory studies on black soil samples in northeastern China, a maize distribution region with heavy fertilizer application. Our specific aims were: (i) to study NO production, NO consumption and NO emission in black soils from maize distribution zones as functions of soil temperature and soil moisture, and (ii) to estimate the biogenic NO emission from black soils in the studied region based on results obtained using a laboratory incubation technique.

2. Materials and methods

2.1. Site description

The studied region is located in the middle of Jilin province and Heilongjiang province at 41°–49° N, 124°–127° E (240–300 m above sea level) in northeastern China. The Chinese black soil region, which is one of three major black soil regions in the world, covers an area of 11.02 M Ha (The Institute of Soil Science, Chinese Academy of Sciences, 1978). The studied region is a semi-humid continental monsoon climate region in the temperate zone; it is cold and arid in winter and hot and rainy in summer. The annual average temperature is 1.5 °C, ranging from 32 °C in the summer to −37 °C in the winter. Annual precipitation ranges from 500 to 600 mm, with about 90% of the precipitation falling as rain between April and September (Xiong and Li, 1987). This region is considered to be particularly sensitive to global climate change (Wang et al., 2002). Recently, the climate in this region has tended towards warm with drought conditions. In the past 50 years (1950–2000), the mean annual and winter temperatures have increased 1.3 and 2.1 °C, respectively (Guo et al., 2005). Mean annual precipitation, especially in the summer season, increased during the early 1980s, and then decreased obviously at the end of 1990s (Zu et al., 2004). The black soil region has been one of the major grain production areas for corn and soybean in China because of its high fertility and arability. Thus, intensive tillage is an important factor in declining soil nutrients and variation of soil properties in this area (Shen, 1998; Yang et al., 2003).

The studied sites were located at Hailun National Research Station of Agroecology and Dehui Black Soil Demonstration Station for Agriculture, Northeast Institute of Geography and Agricultural Ecology, Chinese Academy of Sciences, at approximately N47°26′, E126°38′ and N44°12′, E125°32′, respectively (Fig. 1). The soil is typical black soil (Luvic Phaeozem, FAO) developed on loess-like parent material of the Quaternary period. The predominant land-use type is cropland for maize. Nutrient concentrations in soil layer at depths of 0–20 cm were 20.9–39.8 g kg^{-1} for TOC, 1.73–5.93 g kg^{-1} for TN, 0.24–1.60 g kg^{-1} for TP, 14.8–24.3 g kg^{-1} for TK, 51.2–542 mg kg^{-1} for available N, 0.9–144 mg kg^{-1} for available P, 79.2–443 mg kg^{-1} for available K (Wang et al., 2004; Yu et al., 2006; Zhang et al., 2007). Soil pH ranges from 5.90 to 6.98, and average bulk density is 0.98 g cm^3.

2.2. Laboratory experiments for NO measurement

2.2.1. Dynamic laboratory incubation system

The NO production and consumption at each sequential moisture change were measured under temperatures of 15 °C and 25 °C, respectively, in laboratory using a fully automatic laboratory dynamic incubation system. The system includes four sub-systems: air purification system, gas dilution system, dynamic chamber system and measurement system (described in detail by Yu et al., 2008). The CLD 780TR Chemiluminescence NO Analyzer (detection limit 0.052 ppb and precision ±0.026 ppb, Eco Physics AG., Switzerland) was used for NO measurement and Binos (Rosemount, Germany) for vapor signal capture. For more detailed information, please refer to van Dijk et al. (2002).

2.2.2. Treatment of soil samples and experimental layout for NO measurements

Black soil samples from maize land in the Hailun National Research Station of Agroecology and Dehui Black Soil Demonstration Station for Agriculture were collected from the surface soil (0–20 cm depth) in early October, 2005 for NO production measurements (Fig. 1). Soil temperature, air temperature, soil bulk density and soil water content were measured *in situ* when soil samples were collected. The dry soils from different sites were sieved through a 2 mm coarse stainless steel sieve, then mixed and kept in sealed plastic bags at 5 °C to limit microorganism activities until the time of the NO emission analysis. Roots and other organic matter were removed to homogenize the sample. The soil samples were stored for no more than two months before study.

An 80 g soil sample was weighed and spread evenly across the bottom of the chamber. Soil samples were incubated three days after saturation. Soil moisture was measured before NO analysis.

During the experiment, the dynamic chambers were kept in a thermostat cabinet to maintain certain soil temperatures. The purified air with or without NO standard gas flowed from the gas dilution system into a main Teflon tube with 5 T-connections to supply air, via 5 MFCs (mass flow controllers), to each chamber at a flow rate of 2.5 L min^{-1} (Fig. 2 in Yu et al., 2008). The NO analyzer was set to measure the NO mixing ratio in the headspace of the chambers every 10 s. The measuring process for one chamber was 1.5 min. The average value of nine measurements within 1.5 min was used for data analysis. The NO mixing ratios in the headspace of the chambers were determined by a chemiluminescence NO

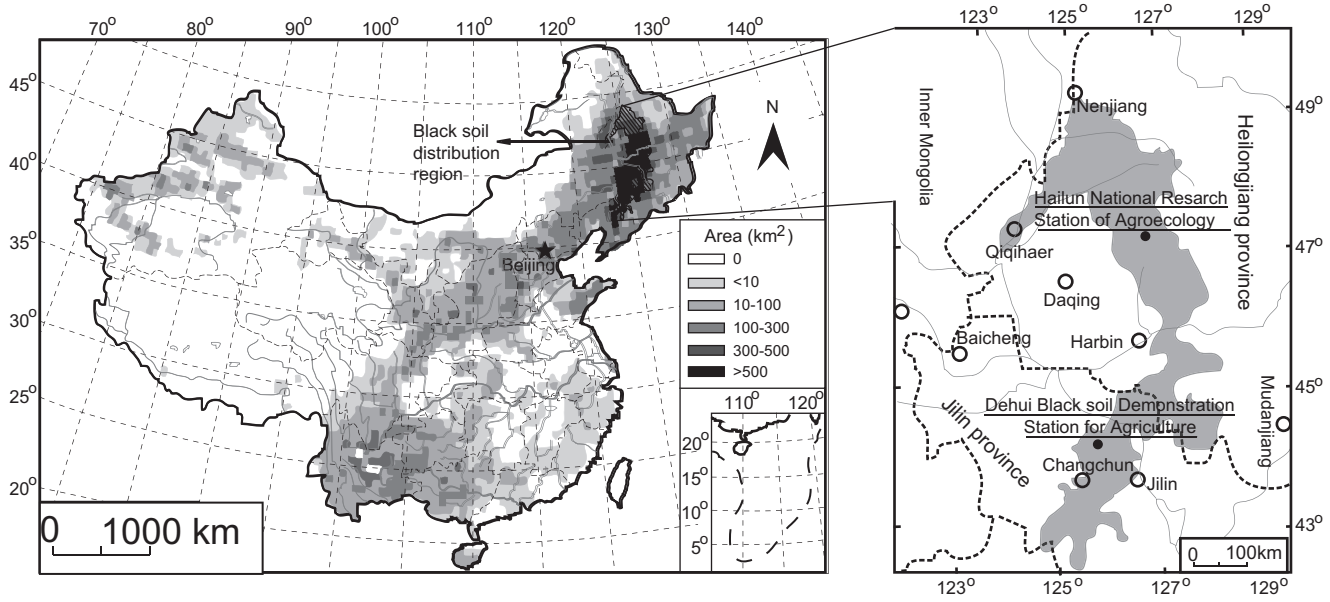

Fig. 1. Map of Chinese maize distribution (per 0.5° grid cell) showing the location of the studied region and sampling sites.

analyzer. Calibrations were performed once a week with a NO standard (200 ppm NO in N_2, Messer-Griessheim Germany).

2.2.3. Field soil temperature and soil moisture monitoring

The field soil temperature and soil moisture of the surface soil at a depth of 10 cm were monitored in Dehui Black Soil Demonstration Station for Agriculture from middle April to early October in 2005, 2006 and 2007. The Watchdog model 2700 Weather Station (Spectrum Technologies, Inc. USA) with temperature probe and water content sensor was used to measure soil temperature and soil moisture, respectively. The soil temperature and soil moisture were measured and recorded automatically 24 times a day at 1 h intervals.

2.3. Data calculation

2.3.1. Calculation of NO release rate J

The NO release rate *J* was calculated using the difference of the NO mixing ratio between the outlet and inlet of each dynamic chamber described in Equation (1).

$$J = \frac{Q}{m_{\mathrm{sdx}}} \times (\mathrm{NO}_{\mathrm{outlet}} - \mathrm{NO}_{\mathrm{inlet}}) \times \frac{M_N}{V_N} \quad (1)$$

where *J* is the release rate of NO (ng N kg^{-1} s^{-1}), *Q* is gas flow rate through chambers (m^3 s^{-1}), m_{sdx} is the soil dry weight (kg), $\mathrm{NO}_{\mathrm{outlet}}$ and $\mathrm{NO}_{\mathrm{inlet}}$ are NO mixing ratios at chamber outlet and inlet (ppbv), respectively, M_{N} is the molecular weight of nitrogen (14.0067 kg $kmol^{-1}$) and V_{N} is the molecular normal volume (22.4 m^3 $kmol^{-1}$).

2.3.2. Calculation of volumetric NO uptake rate k

To determine NO uptake by the soil, the dynamic chambers were flushed with air mixed with NO standard gas. The obtained inlet NO mixing ratios using the gas dilution system range from 0 to 200 ppbv. The NO release rates at inlet NO mixing ratios of 0 ppbv and 60 ppbv were measured. To obtain the volumetric NO uptake rate *k*, a linear regression is used. The volumetric NO uptake rate *k*, NO production rate*P*and NO compensation mixing ratio, NO_{c} were calculated as Equations (2)–(4).

$$J = P - k \times \mathrm{NO}_{\mathrm{amb}} \quad (2)$$

$$k = -\frac{J_{[0\mathrm{ppbNO}]} - J_{[60\mathrm{ppbNO}]}}{(0ppb - 60ppb)} \times \frac{M_A \times 0.001}{\rho_A \times M_N} \quad (3)$$

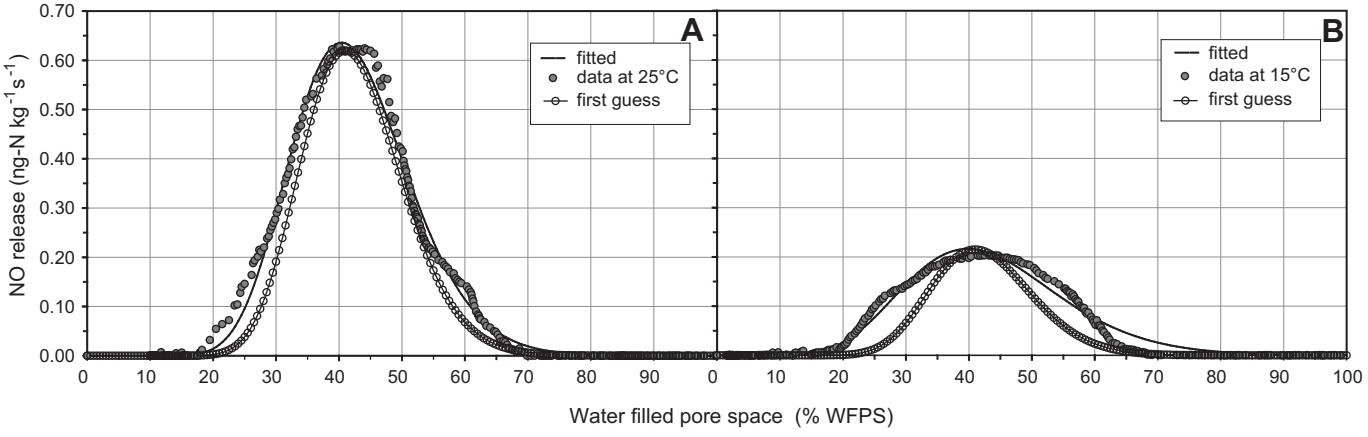

Fig. 2. Guesstimate and fitted relations of NO release from black soil samples in maize land and soil moisture at constant soil temperature of 25 °C (A) or 15 °C (B).

$$NO_c = \frac{P}{k} \quad (4)$$

where J is the release rate of NO (ng N kg^{-1} s^{-1}), P is the NO production (ng N kg^{-1} s^{-1}), k is the volumetric NO uptake rate/NO consumption rate constant (m^3 kg^{-1} s^{-1}), NO_{amb} is the ambient NO concentration (ng N m^{-3}), M_A is the molecular weight of dry air (28.9644 kg $kmol^{-1}$), ρ_A the air density at certain temperature (kg m^{-3}) and M_N is the molecular weight of nitrogen (kg $kmol^{-1}$). NO_c is compensation mixing ratio (ng N m^{-3}). The intercept of the linear regression with the y-axis expresses the compensation point or the compensation concentration, where NO uptake is equal to NO production, which results in a NO release rate $J = 0$. The algorithm used to fit the laboratory data was described by Meixner and Yang (Meixner and Yang, 2006) in detail.

2.3.3. Calculation of NO production rate

NO production can be expressed as the intercept of the linear regression. This value represents the emission of NO by the soil under the condition that the NO mixing ratio in the headspace of the chamber is zero, assuming no NO uptake by the soil.

2.3.4. Determination of NO fluxes

The model of Galbally and Johansson (Galbally and Johansson, 1989) shows an algorithm of transforming the laboratory-derived NO release into a net NO flux. The model considers 3 variables for NO production and NO consumption in soils: (1) the soil bulk density, (2) the NO concentration and diffusive or net gaseous flow through the soil, and (3) two unknown parameters representing NO production and uptake in the soil. We measured the NO flux from soil in the laboratory under 15 °C and 25 °C, thus the NO flux for certain soil with certain nitrogen content can be obtained from Equation (5). A detailed description of the determination of NO fluxes for laboratory measurements is given in van Dijk et al. (van Dijk et al., 2002).

$$F_{NO}(WFPS, T) = \sqrt{BD \times k(WFPS, T) \times D} \times \left[\left(\frac{P(WFPS, T)}{k(WFPS, T)}\right) - [NO]_{amb}\right] \quad (5)$$

Where F_{NO}(WFPS,T) is NO flux (ng N m^{-2} s^{-1}), BD is soil bulk density (kg m^{-3}), k(WFPS, T)is the volumetric NO uptake rate under different water filled pore space (WFPS) (m^3 kg^{-1} s^{-1}) and temperature (℃), D is NO diffusion coefficient in soil (m^2 s^{-1}), $P(WFPS, T)$ is the NO production under different WFPS (ng N kg^{-1} s^{-1}) and temperature. NO diffusion coefficient in soil and WFPS were calculated as Equations (6)–(7), respectively.

$$\frac{D}{D_0} = \frac{\varepsilon^{2.5}}{\Phi} \quad (6)$$

$$WFPS = \frac{\theta_g \times BD}{1 - (BD/PD)} \times 100\% \quad (7)$$

Where D_0 is NO diffusion coefficient in free air (0.0000199 m^2 s^{-1}), ε is the soil air-filled porosity (m^3 m^{-3}), Φ is the soil total porosity (m^3 m^{-3}) (Moldrup et al., 2000), θ_g is gravimetric soil moisture (kg H_2O kg^{-1} oven dried soil) and PD is particle density (assumed to be 2650 kg m^{-3}) (Davidson and Schimel, 1995). An algorithm has been developed to fit observed values from the laboratory flux (Meixner and Yang, 2006), as a function of the WFPS, as shown in Equations (8)–(11). One-way ANOVA was applied for fitted results test.

$$F_{NO}(WFPS) = a WFPS^b \exp(-c WFPS) \quad (8)$$

$$a = \frac{F_{NO}(WFPS_{opt})}{\left[WFPS_{opt}^b \exp(-b)\right]} \quad (9)$$

$$b = \frac{\ln\left[\frac{F_{NO}(WFPS_{opt})}{F_{NO}(WFPS_{upp})}\right]}{\ln\left(\frac{WFPS_{opt}}{WFPS_{upp}}\right) + \frac{WFPS_{upp}}{WFPS_{opt}} - 1} \quad (10)$$

$$c = \frac{-b}{WFPS_{opt}} \quad (11)$$

Where $WFPS_{opt}$ is the soil moisture at which the maximum NO flux is observed, $F_{NO}(WFPS_{opt})$equals the maxF_{NO}(WFPS), and $WFPS_{upp}$ is the soil moisture at which $F_{NO}(WFPS) = F_{NO}(WFPS_{upp}) \approx 0$ for WFPS > $WFPS_{opt}$. The NO fluxes at different temperature were calculated as Equations (12)–(13) (Williams et al., 1992).

$$F_{NO}(T_2) = F_{NO}(T_1) \times \exp[-q \times (T_2 - T_1)] \quad (12)$$

$$q = -\frac{\ln(Q_{10})}{10} \quad (13)$$

Where $F_{NO}(T_2)$ and $F_{NO}(T_1)$ are NO flues (ng N m^{-2} s^{-1}) at different temperature at certain WFPS, q is exponential factor, T_2 and T_1 are different temperature (℃). When $T_2 - T_1 = 10$, $Q_{10} = F_{NO}(T_2)/F_{NO}(T_1)$.

3. Results

The modified algorithm as applied to fit the laboratory results at constant soil temperatures of both 25 °C and 15 °C versus soil moisture for black soils is shown in Fig. 2. The first guess curve is made up of guesstimate results because the parameters of a, b, and c were calculated from three laboratory measurement data of $F_{NO}(WFPS_{opt})$, $WFPS_{opt}$ and $WFPS_{upp}$ based on Equations (9)–(11). The fitted results show that the algorithm was well fitted with the laboratory data ($R > 0.98^{**}$ at the 0.01 level, sum of square for corresponding value difference ranges from 0.05 to 0.10). The optimum soil moisture for black soil NO emission was approximately 25.7%, which is equivalent to 41% WFPS (water-filled pore space) (Fig. 2A, B).

According to Equations (1) and (2), the production P (i.e., NO release J at NO_{amb} is nearly zero) in black soils was evaluated (Fig. 3A). The maximum NO production (in terms of mass of N) at optimum soil moisture was 0.65 and 0.22 ng N kg^{-1} s^{-1} in soils temperatures of 25 °C and 15 °C, respectively. The NO production results at soil temperature of 15 °C and 25 °C in laboratory showed that the optimum soil moisture for NO production did not change significantly with soil temperature (Fig. 3A). Within the range of soil moisture in favour of NO production, the slopes of the average ratio of NO production versus NO release at soil temperatures of 25 °C and 15 °C are about 1.032 (SD = 0.104) and 1.009 (SD = 0.135), respectively. Nitric oxide consumption rate constant k ranges from 9.31 × 10^{-6} to15.1 × 10^{-6} m^3 kg^{-1} s^{-1} (results not shown). The average k value at soil temperature of 25 °C was higher than that at 15 °C within range of soil moisture in favour of NO production. Fig. 3B shows that the compensation concentration varies at soil temperatures of 25 °C and 15 °C. Under optimum soil moisture conditions for NO production, the NO compensation point mixing ratio was about 60.2 and 43.1 μg N m^{-3} (96.3 and 68.9 ppb) for black soils at temperatures of 25 °C and 15 °C, respectively.

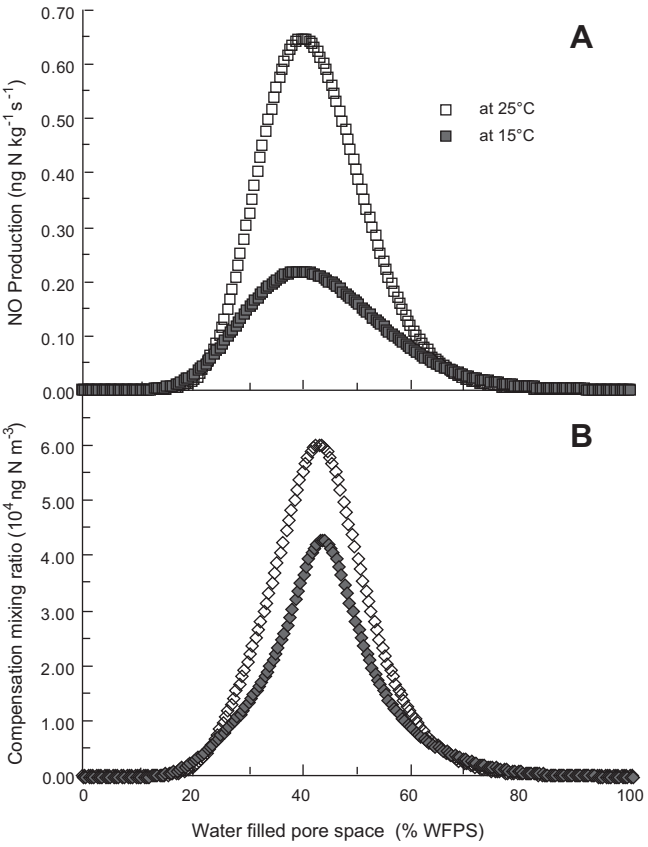

Fig. 3. NO production (A) and compensation concentration (B) in black soils as a function of soil moisture at soil temperature of both 15 °C and 25 °C.

We applied the modified algorithm (Galbally and Johansson, 1989) on NO potential fluxes from black soils in maize land in northeastern China. With increased soil moisture, the NO fluxes increased gradually and peaked at soil moisture about 41% WFPS, then decreased gradually and were up to nearly zero below 80% WFPS (Fig. 4A, B). As soon as soil moisture was no longer the limiting constraint (Fig. 4B), NO emission strongly depended on temperature ($Q_{10} = 2.02$). NO fluxes increased exponentially with soil temperature at any given soil moisture (Fig. 4C). The maximum NO potential fluxes for black soils at 25 °C and 15 °C were 18.6 and 9.0 ng N m^{-2} s^{-1}, respectively.

Based on our laboratory NO flux parameterizations and field data (soil temperature and soil moisture monitoring data), we estimated net field NO fluxes from black soil planted maize in different months (Fig. 5). The results showed that NO emissions increased gradually from April to July; then peaked in July followed by gentle declines. The NO "pulse" emissions were observed in July because of high temperature and high precipitation, which led to optimum soil moisture, and then NO fluxes decreased with soil temperature and moisture. The peak of NO fluxes from black soils was 16.5 ng N m^{-2} s^{-1}. The average of NO fluxes was about 10.7 ng N m^{-2} s^{-1} for an entire plant growth period.

4. Discussion

4.1. Effects of soil moisture and temperature on NO emissions

Many studies have shown that an optimum soil moisture does exist for soil NO emissions (Otter et al., 1999; Verchot et al., 1999; Yang and Meixner, 1997; Yu et al., 2008), but the value varied with soils. For example, a laboratory study showed an optimum soil moisture of about 20% WFPS (Yang and Meixner, 1997), but in another study conducted in saline sodic soil of northeastern China, maximum NO fluxes was observed at approximately 9.5%–18% WFPS (Yu et al., 2008). The optimum value was even discovered to be approximately 50% WFPS in a seasonally dry forest of the eastern Amazon (Verchot et al., 1999). In this study, the optimum soil moisture for black soil NO emission was approximately 25.7%. This phenomenon can be plausibly interpreted by that diffusion of NO through pore spaces to atmosphere is limited under high soil moisture (Skopp et al., 1990), while substrate diffusion through water films to microbial active cells is limited under low soil moisture (Linn and Doran, 1984; Russow et al., 2009). For all soils, we observed that the relationship of NO production and soil temperature at a given soil moisture was similar to previous results (van Dijk et al., 2002; van Dijk and Meixner, 2001; Yu et al., 2008). Compared with NO production (25 °C) in pasture soil (maximum value of 0.26 ng N kg^{-1} s^{-1}) (van Dijk and Meixner, 2001), our results for NO production in black soils are more than twofold higher.

4.2. Nitric oxide consumption rate and compensation mixing ratio

Nitric oxide consumption rate constant k is determined largely by the type of microbial consumption that takes place in different soils. The $k$values range in this study (ranging from 9.31×10^{-6} to15.1×10^{-6} m^3 kg^{-1} s^{-1}) are in the middle of the range of k values so far published (ranging from 0.5×10^{-6} to 60×10^{-6} m^3 kg^{-1} s^{-1}) (Baumgartner and Conrad, 1992; Bollmann and Conrad, 1997; Saad and Conrad, 1993; van Dijk and Meixner, 2001; Yu et al., 2008). Van Dijk and Meixner (2001) found that the value of the NO consumption rate constant, k, evidently depends on soil moisture and soil temperature, i.e., k value decreased with soil moisture and increased with soil temperature. In agreement with those findings, we observed that the average k value at soil temperature of 25 °C was higher than that at 15 °C at certain soil moisture. The dependence on soil temperature can be explained by the microbial nature of the consumption processes both through reduction (Bender and Conrad, 1994) and through oxidation (Gödde and Conrad, 1999).

The NO compensation concentration, NO_c, is the concentration of ambient NO where production of NO is equal to consumption; at this point, there is no net uptake or release of NO from the soil. The soil acts as a source for NO only when its NO_c exceeds the atmospheric concentrations of NO. Our results show that the optimum soil moisture for the NO compensation mixing ratio is similar to that for NO production for a certain landscape soil (Fig. 3A, B), because the NO compensation mixing ratio for studied soils is much dependant on NO production.

4.3. Nitric oxide fluxes from black soils

The NO potential fluxes were estimated applying the modified algorithm (Galbally and Johansson, 1989). Compared to previous laboratory results at 25 °C, the maximum NO potential emission in this study (18.6 ng N m^{-2} s^{-1}) is much higher than semi-arid savanna in South Africa (1.3–2.4 ng N m^{-2} s^{-1}) (Feig et al., 2008), and arid and dry/hot semi-arid soils (<0.15 ng N m^{-2} s^{-1}) (Davidson and Kingerlee, 1997; Meixner et al., 1997; Meixner and Yang, 2006), but similar to saline sodic soil in pasture and paddy field (16–30 ng N m^{-2} s^{-1}) (Yu et al., 2008). Fig. 4B shows that the NO emissions are strongly affected by soil moisture. The optimum soil moisture at which maximum NO flux is observed is independent of soil temperature. Statistically sound relationships have been observed between NO fluxes and soil moisture (optimum curves). The effects of soil moisture on biological, physical and chemical processes mediating NO emissions have been proposed in

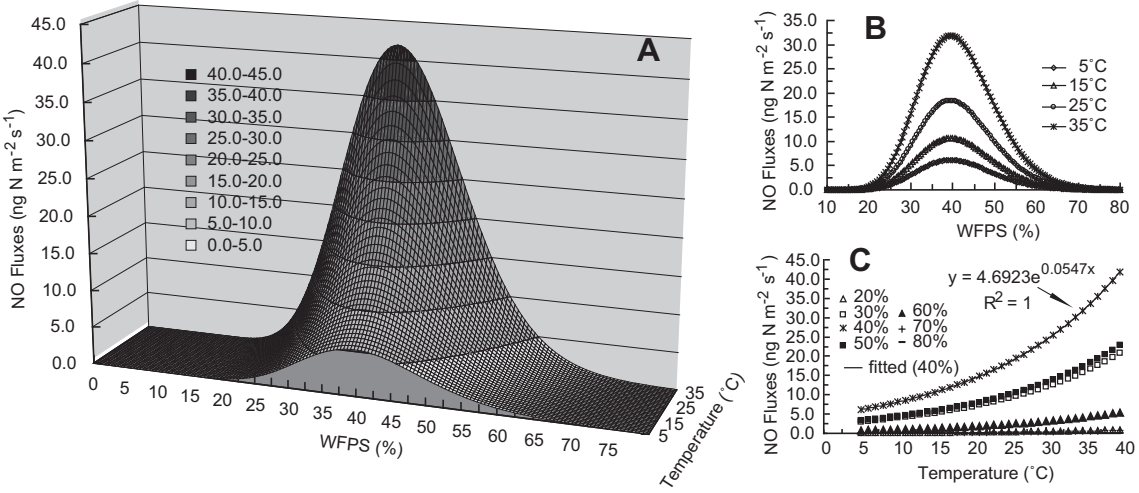

Fig. 4. NO fluxes from black soil samples versus soil temperature and soil moisture (A), the relation of NO fluxes and soil moisture (B) and soil temperature (C).

previous studies (Bollmann and Conrad, 1998; Davidson, 1993; Davidson and Schimel, 1995). Similar results of NO flux and soil temperature relations have also been observed in other studies (Hutchinson and Brams, 1992; Maljanen et al., 2007; Meixner et al., 1997; Yang and Meixner, 1997; Yu et al., 2008). This is because the dominance of soil microbial processes in the production of NO anticipates a marked influence of soil temperature on NO emission rates. The rates of chemical and/or enzymatic processes change exponentially with temperature, as long as others factors (substrate or moisture available) are not limiting.

Compared to previously reported NO fluxes from cropland of different regions (Table 1), our results are generally higher than those from corn land in Mexico, Western Tennessee and Central Pennsylvania of USA, as well as most of croplands in Europe; however, our results are much lower than that from sugar cane land in Hawaiian Islands, soybean land in Spain and cropland in southern China, all of which received high nitrogen input (Matson et al., 1996; Slemr and Seiler, 1984; Walsh, 2001). Peculiarly, high NO fluxes were observed from temperate zone cropland by Matson et al. (1996).

It has been shown by some studies that nitrogen input has a strong impact on NO emission rates since these compounds (NH_4^+ and NO_3^-) serve as substrate for nitrifying and denitrifying bacteria (Akiyama et al., 2000; Williams and Fehsenfeld, 1991; Skiba et al., 1994). However, we cannot find a significant relationship between NO fluxes and nitrogen input based on present reports in different regions, i.e., Matson et al. (1996) observed that NO flux was up to 430 ng N m^{-2} s^{-1} from sugar cane land (loam soil) with 100 kg N ha^{-1} urea application, while very low NO flux (1.5 ng N m^{-2} s^{-1}) was observed from wheat land with 218 kg N ha^{-1} fertilized in Germany (Meixner, 1994). The estimated net field NO flux in this study was also low even though heavy fertilizer was applied (120–180 kg N ha^{-1}) in study region. It indicates that the surface NO exchange is strongly dependent on substrate only if other influential factors such as soil moisture, soil temperature and soil texture are not limiting.

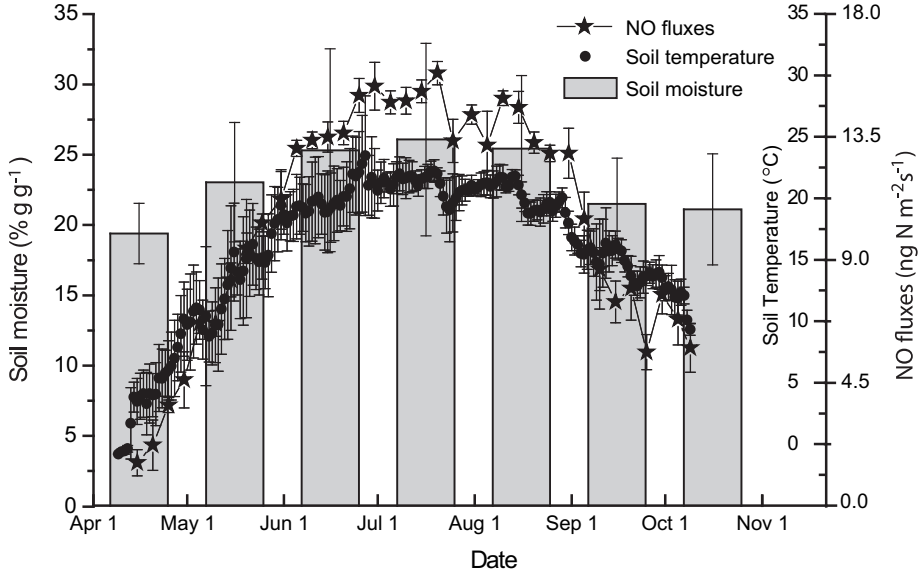

Fig. 5. Seasonal variation of predicted NO fluxes from black soil, 10-cm soil temperature and average soil moisture in a year. Points show means ($n = 120$), standard deviation is indicated by error bars.

Table 1
Reported NO fluxes from cropland of different regions.

Location	Crop	NO fluxes (ng N m^{-2} s^{-1})	Reference
North China	Corn	66.4–150.8	(Walsh, 2001)
Southeastern China	Wheat	21.3–28.5	(Zheng et al., 2003a)
Yangtze Delta, China	Potato, Cabbage, Soybean	11.5–34.2	(Fang and Mu, 2006; Fang and Mu, 2007)
Southern China	Flowering Chinese Cabbage	63.5	(Li and Wang, 2007)
Northeastern China	Maize	10.72	This study
Western Tennessee, USA	Corn	3.06–6.39	(Thornton and Valente, 1996)
Alabama, USA	Cotton	16.11	(Valente et al., 1995)
Central Pennsylvania, USA	Wheat, corn	1.11–9.44	(Williams et al., 1988)
North-Carolina USA	Soybean, cotton, corn	1.67–8.06	(Aneja et al., 1995)
Hawaiian Islands, USA	Sugar Cane	16.4–430	(Matson et al., 1996)
Spain	Soybeans	18.33–94.44	(Slemr and Seiler, 1984)
France	Maize	1.11–1.94	(Jambert et al., 1994)
England	Wheat	6.9	(Skiba et al., 1992)
Canada	Beans	2.1–31.8	(Shepherd et al., 1991)
Germany	Wheat	2.50–4.17	(Meixner, 1994)
Mexico	Corn	2.78	(Davidson et al., 1991)

The seasonal variation of simulated NO fluxes from black soil depends on field soil temperature and soil moisture, which are related to precipitation. The NO flux increased with soil temperature and optimum soil moisture range for NO emission from early April to middle July, and then decreased until October (Fig. 5) because precipitation decreased steadily during the period and thus soil moisture deviated gradually from optimum value for NO production (about 41% WFPS). Consistently, field studies have shown that the soil water content was a very important factor controlling the seasonal patterns of soil NO emissions from ecosystems. For example, Johansson et al. (1988) observed the large increases in NO emission from savannas after small wetting events (<25 mm) following protracted dry periods (Johansson et al., 1988). However, more intensive (>25 mm) wetting events occurring during the wet season appear to prevent NO emission (Cardenas et al., 1993; Davidson, 1993).

5. Conclusions

Up to now, nearly the entire studied region has been changed into cropland from the original natural landscape of wet grassland and the average cultivation age is about 100–200 years. Thus, the black soil distribution in China has been disturbed heavily by human activities. Our findings of biogenic NO emission supports a laboratory study by van Dijk and Meixner (2001) on the relationship of NO production and soil temperature. Statistically sound relationships have been observed between NO fluxes and soil moisture, and NO fluxes also increase exponentially with soil temperature (Q_{10} = 2.02). The estimated NO emissions mainly occur in July when the soil temperature and soil moisture are relatively high. The present study suggests that NO fluxes from black soil are much lower than the previous reports from cropland in southern parts of China.

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