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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.

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