



# Effects of increasing fertilization rates on nitric oxide emission and nitrogen use efficiency in low carbon calcareous soil



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## ARTICLE INFO

### Article history:

Received 3 November 2014  
Received in revised form 19 January 2015  
Accepted 28 January 2015  
Available online xxx

### Keywords:

Calcareous soil  
Emission factor  
Partial factor productivity of applied nitrogen  
Nitrogen deposition  
Optimized management

## ABSTRACT

Global nitrogen fertilizer consumption is expected to continue to increase. To explore effective mitigation strategies, a deeper understanding of the responses of nitrogen use efficiency, nitric oxide (NO) emission and the NO direct emission factor (EF<sub>d</sub>) to increasing fertilization rates is needed. A gradient of fertilization rates (0, 135, 270, 430, 650 and 850 kg N ha<sup>-1</sup> yr<sup>-1</sup> in the form of urea, hereafter referred to as N0, N135, N270, N430, N650 and N850, respectively) was used to fully represent the nitrogen application levels in the wheat–maize rotational cropping system that has been widely adopted in China. The annual NO emissions ranged from 0.43 ± 0.04 (N0) to 2.64 ± 0.35 kg N ha<sup>-1</sup> yr<sup>-1</sup> (N850) and linearly increased with increasing fertilization rates ( $P < 0.01$ ). The high pH and low carbon availability in the calcareous soil limited NO production; thus, low EF<sub>d</sub> values (0.26–0.36%) were observed. The partial factor productivity of applied nitrogen (PFP<sub>N</sub>) rapidly decreased with increasing fertilization rates; the relationship could be characterized by a rectangular hyperbolic function ( $P < 0.01$ ). The expected trade-off between EF<sub>d</sub> and PFP<sub>N</sub> was not observed. The on-farm PFP<sub>N</sub> was only 33 ± 3 kg grain kg<sup>-1</sup> N (N430), highlighting the necessity of optimizing current management strategies. Based on a review of previous studies, a comprehensive optimized management strategy is recommended to obtain the maximum benefits for multiple goals of a wheat–maize cropping system. However, consecutive field observations and model studies are still needed to validate the long-term effects of this management strategy.

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## 1. Introduction

The winter wheat and summer maize rotational cropping system has been extensively adopted in northern China (Henan, Hebei, Shandong, Shaanxi and Shanxi Provinces) and has provided more than half of the food supply in China (high grain yields of ~5.5 and 6.0 t ha<sup>-1</sup> yr<sup>-1</sup> for wheat and maize, respectively; Cui et al., 2010). The soils in the wheat–maize rotational area are usually low in organic carbon content (SOC of <11 g kg<sup>-1</sup>) and are often alkaline (pH >8, Ju et al., 2009, 2011; Cui et al., 2010, 2012; Liu et al., 2011; Zhang et al., 2011). The high pH and low SOC contents can significantly affect ammonia (NH<sub>3</sub>) volatilization, nitrate (NO<sub>3</sub><sup>-</sup>) leaching, microbial nitrification and denitrification, which influence the nitrogen use efficiency (NUE) of crops. The high soil pH is beneficial to gaseous NH<sub>3</sub> formation and diffusion after applications of ammonia-based and ammonia-forming fertilizers, which are the dominant fertilizers applied in wheat–maize fields.

Therefore, NH<sub>3</sub> volatilization is the primary fertilizer-N loss pathway in the wheat–maize rotational system (Ju et al., 2009; Cui et al., 2010, 2014). Soils with pH values between 7 and 8 are optimal for microbial nitrification (Nieder and Benbi, 2008). The very high soil pH (>8) and low SOC contents (<11 g kg<sup>-1</sup>) in wheat–maize rotational fields may limit microbial nitrification and denitrification, respectively (Ju et al., 2011; Cui et al., 2012; Liu et al., 2012). The commonly high fertilization and irrigation amounts (fertilization: 500–600 kg N ha<sup>-1</sup> yr<sup>-1</sup>; irrigation: 90–690 mm yr<sup>-1</sup>; Cui et al., 2010; Liu et al., 2014) plus the carbon (C)-limited denitrification process certainly lead to significant NO<sub>3</sub><sup>-</sup> accumulation in soils and leaching losses (Ju et al., 2009; Liu et al., 2014). The overuse of synthetic nitrogen (N) fertilizers, the high soil pH and low C peculiarities, and the nonfeasance toward significant N losses produce the extremely low N recovery efficiencies for the wheat–maize rotational system in China (16–18%; Cui et al., 2010).

Global synthetic N fertilizer consumption exceeded 100 million tons in 2012, and it is expected to increase threefold by 2050 to meet the food demand (Tilman et al., 2002; FAO, 2012). China accounts for approximately one-third of the global synthetic N

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fertilizer consumption. The average growth of synthetic N fertilizer consumption was approximately 2% over the last 10 years in China (China Statistical Yearbook, 2012). With the rapid increase in N fertilizer consumption, N use efficiencies of global crops have decreased over the last half century (Tilman et al., 2002). The current recovery efficiency of N fertilizers by crops is typically less than 50% (Tilman et al., 2002; Ladha et al., 2005). The intensively managed wheat–maize rotational system in northern China presents much lower N recovery efficiencies (16–18%; Cui et al., 2010) compared with the global means (22–55%; Ladha et al., 2005).

The applied N that is not recovered by crops either accumulates in soils or is lost to the environment. Nitric oxide (NO) emission is an important loss pathway of N fertilizer to the atmosphere through microbial nitrification and denitrification. Global fertilizer-associated NO emissions are approximately  $0.6 \text{ Tg N yr}^{-1}$ , representing 0.5% of the applied N (FAO and IFA, 2001). Nitric oxide exchange between the biosphere and atmosphere is difficult to measure due to its short atmospheric lifetime of a few seconds. Global measurements of NO emissions from fertilized fields, particularly for more than two levels of N fertilizer treatment, are scarce. Available studies indicate a strong increase in NO emissions with increasing fertilization rates (Bouwman et al., 2002). Veldkamp and Keller (1997) presented a significant linear relationship between NO emissions and N fertilizer applications in temperate areas through the statistical analysis of 6 studies with different fertilizer types, soils, crops and climate conditions. On a global scale, no clear relationship has been found between fertilization rates and NO emissions, but soils with high organic C content, good drainage, coarse texture and neutral pH are prone to high emissions (FAO and IFA, 2001; Bouwman et al., 2002).

The nitric oxide direct emission factor ( $EF_d$ ) is defined as the cumulative emission differences between fertilized and unfertilized treatment expressed as a percentage of the applied N.  $EF_d$  represents the loss rate of N fertilizer to the atmosphere through NO emissions, and it is used extensively when compiling national emission inventories for croplands. High N use efficiencies indicate effective fertilizer management strategies and the reduction of N overuse for NO production. We may expect a trade-off between NUE and  $EF_d$ . However, the dependencies of NUE, NO emission and  $EF_d$  on increasing N fertilizer consumption and the relationship between NUE and  $EF_d$  remain unclear on local and global scales.

To evaluate the influences of changing N fertilizer application rates on NUE, NO emission and  $EF_d$ , NO fluxes, crop yields, and major environmental, soil and field management factors were investigated for treatments with six N fertilizer levels in an irrigated wheat–maize rotational cropping system. The aims of the study were to (a) quantify the dependencies of NUE, NO emissions and  $EF_d$  on changing N fertilizer application rates, (b) determine the potential relationship between NUE and  $EF_d$ , (c) present the particularities of N fertilizer transformation, NUE, NO emission and  $EF_d$  in low C calcareous soils, and (d) explore optimized N management strategies for simultaneously achieving high grain yields and N use efficiencies in a wheat–maize rotational system.

## 2. Materials and methods

### 2.1. Experiment site

The experiment site ( $34^{\circ}55.51'N$ ,  $110^{\circ}42.59'E$ ) is situated within Dong Cun Farm in Yongji City, Shanxi Province, northern China. One-year winter wheat and summer maize rotation and cotton are the primary cropping systems. The two cropping systems are usually rotated every 3–5 years to avoid negative monoculture effects on grain yields. The experimental field had been planted with cotton for 5 years prior to October 2005. Subsequently, the

field was cultivated with maize (*Zea mays* L.) between early June and mid-October and with wheat (*Triticum aestivum* L.) during the remainder of the year.

A fertilization gradient experiment with six N treatment levels (0, 135, 270, 430, 650 and  $850 \text{ kg N ha}^{-1} \text{ yr}^{-1}$  in the form of urea, hereafter referred to as N0, N135, N270, N430, N650 and N850, respectively) was set up on 16 October, 2009. Eighteen experiment plots ( $6 \text{ m} \times 6 \text{ m}$  each) were randomly allocated to six N levels with three replicates each. Detailed information regarding urea applications in each treatment is provided in Table 1. Phosphate (P) and potassium (K) fertilizers in the form of calcium superphosphate and potassium sulfate were applied identically for all treatments during wheat planting ( $60\text{--}30 \text{ kg P-K ha}^{-1} \text{ yr}^{-1}$ ). A sprinkler irrigation system was used to irrigate the crops using bore water (depth: 130–140 m). Climate data, the main soil properties and detailed field management information for the experiment site were previously reported by Liu et al. (2012).

### 2.2. Measurement of nitric oxide fluxes

Nitric oxide fluxes were manually measured between 19 October, 2009 and 15 October, 2010 using a static chamber chemiluminescence measuring system (Liu et al., 2009). The sampling frequency was either daily for seven days after urea applications or two to three times per week during the remaining period. Various dimensions of stainless steel frames (length  $\times$  width  $\times$  height =  $50 \times 50 \times 20$  and  $50 \times 30 \times 20$  cm) and chambers (length  $\times$  width =  $50 \times 50$  and  $50 \times 30$  cm, heights: 0.3, 0.5, 1.0 and 1.5 m) were implemented during different crop growth stages to ensure the representativeness of fluxes (Liu et al., 2012). The frames were inserted in the center of each plot and were temporarily removed for tillage and seeding. The chamber exteriors were covered with styrofoam (thickness: 3 cm) and reflective materials to limit the temperature increase ( $0.6^{\circ}\text{C}$  on average) inside the chamber during chamber closure. Rubber seals ensured the gas-tightness of the joints of the different chamber parts and frames. The concentrations of NO and nitrogen dioxide ( $\text{NO}_2$ ) in the gas samples were analyzed using a chemiluminescence  $\text{NO}\text{--}\text{NO}_2\text{--}\text{NO}_x$  analyzer (Model 42i, Thermo Electron Corporation, Waltham, MA, USA). Multi-point calibration of the analyzer was conducted monthly using a zero gas generator (Model 111 Zero Air Supply), gas phase titrator (Model 146 Series Multi-gas Calibration System) and NO standard gas (10 ppm NO in  $\text{N}_2$ , Air Products and Chemicals, Inc., Beijing, China). A 1.5 L gas sample was needed for detecting reliable NO and  $\text{NO}_2$  concentrations by the analyzer (sampling flow:  $600\text{--}800 \text{ mL min}^{-1}$ ). To alleviate the effects of the huge sampling volume on the air pressure inside the chamber, two procedures were adopted in the present study. First, ambient air was collected at the chamber sampling height immediately before the chamber closure. This sample represented the initial gas concentrations during the chamber closure. Then, another gas sample was taken from the enclosed chamber headspace 1–10 min after closure. The two gas samples (2 L each) were pumped (flow rate:  $\sim 3 \text{ L min}^{-1}$ , NMP830KNDC, KNF Neuberger, Inc., Freiburg, Germany) into opaque gas bags (made of Tedlar polyvinyl fluoride films) and analyzed within 2 h after sampling. Second, a ventilation tube with a simple cut-off valve was installed on the top of each chamber. The cut-off valve was opened temporarily when installing the chamber onto the frame and when gas sampling; it remained closed during the remaining chamber closure time. The dimensions (inner diameter: 6 mm; length: 15 cm) of the ventilation tube should ensure the overpressure headspace air or ambient air coming through it to maintain pressure equilibrium between the inside and the outside of the chamber during the chamber installation and gas sampling (Hutchinson and Mosier, 1981). The measured

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