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Two-year simultaneous records of N_2O and NO fluxes from a farmed cropland in the northern China plain with a reduced nitrogen addition rate by one-third

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ABSTRACT

Given the common problem of fertilizer overuse, agronomists are calling for a reduction of the high nitrogen dose by 1/3. We carried out a field experiment over two full winter wheat–summer maize rotations in the North China Plain (NCP) to determine whether this degree of nitrogen reduction will significantly reduce the emissions of nitrous oxide (N_2O) and nitric oxide (NO). Three treatments were investigated in the field trial: a control with no nitrogen application, the conventional practice with nitrogen over-application and the optimal practice with a reduced dose of nitrogen by 1/3. Our observations across all treatments over the experimental period reveal significant correlations of the fluxes of either gas with soil temperature and moisture as well as the concentrations of soil ammonium, nitrate and dissolvable organic carbon. There were strong correlations within the functions of the dual Arrhenius and Michaelis–Menten kinetics, giving apparent activation energy values of 40–97 and 59–92 kJ mol^{-1} for N_2O and NO fluxes, respectively. Our results provide annual direct emission factors of 0.48–0.96% for N_2O and 0.15–0.47% for NO and demonstrate a significant correlation between N_2O emission induced by fertilization and fertilizer nitrogen use efficiency (NUE). The correlation indicates a significant potential of N_2O mitigation via enhancing NUEs. A reduction in the nitrogen dose did not obviously mitigate either the annual NO emission in both rotations or the annual N_2O emission in the second one. However, nitrogen reduction significantly decreased the annual total N_2O emission by 38% during the first rotation. These inconsistencies in the responses of N_2O emission to the reduced nitrogen dose can be attributed to improper fertilization practices, such as broadcasting urea prior to heavy rainfalls or irrigation events during the maize season, which implies a need for further fertilization practice options/techniques in addition to the reduction of nitrogen doses.

1. Introduction

Nitrous oxide (N_2O) is a long-lived greenhouse gas. It significantly contributes to the current period of global warming (IPCC, 2007). The emission of N_2O from agricultural soils as a consequence of nitrogen fertilizer application accounts for approximately 42% (ranging from 25% to 72%) of total anthropogenic sources (IPCC, 2007). Although fossil fuel combustion, industrial processes and biomass/biofuel burning dominate atmospheric NO_x (i.e., nitric oxide plus nitrogen dioxide) sources at the global scale (IPCC, 2007), the contribution of nitric oxide (NO) emissions from nitrogen-fertilized croplands could be significant in rural regions (Bouwman et al., 2002; Butterbach-Bahl et al., 2009). Therefore, mitigation of N_2O and NO emission from improved nitrogen fertilization practices is a significant concern for agricultural areas.

The North China Plain (NCP) is a very important agricultural region for the nation. It has only 23% of China's cropland, but accounts for as much as 39% of annual national grain production (e.g., Ding et al., 2007). To obtain high crop yields, farmers lacking in scientific awareness will often over-apply nitrogen fertilizers. For instance, Ju et al. (2009) reported nitrogen addition rates as high as 550–600 kg N ha^{-1} yr^{-1} were applied to the winter wheat–summer maize rotation regime in this region. Given fertilizer nitrogen use efficiencies (NUE) as low as 16–41% (with a mean of 28%) in this area (e.g., Zhang et al., 2008), only 90–250 kg N ha^{-1} yr^{-1} can be taken up by the crops. High residual nitrogen fertilizer from over-application most likely enhances the soil content of nitrogen (Ju et al., 2004), which then stimulates nitrification and/or

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denitrification (Liu et al., 2003) and may thus promote N_2O and NO emissions (e.g., McSwiney and Robertson, 2005). Ju et al. (2011) and Cui et al. (2012) have reported that N_2O emissions in the NCP are mainly attributable to nitrification, which is also related to NO emissions from unsaturated agricultural soils (e.g., Fang and Mu, 2006, 2007; Li and Wang, 2007). The authors have also indicated that the contribution of denitrification to the production of both gases is severely limited by a lack of carbon substrates in calcareous soils if no additional labile carbon is supplied. Although there are intensive reports on soil N_2O emission in NCP (e.g., Ding et al., 2007; Liu et al., 2012), few have addressed the complex relationship between N_2O and/or NO fluxes and regulating factors in terms of soil environment and carbon and/or nitrogen substrates. In addition, the relationship between N_2O emission and NUE in this region has been seldom investigated in the previous studies.

Zhu (1998) have recommended a reduction of current nitrogen fertilizer application rates by approximately 1/3 for most agricultural areas in China, including the NCP. Reducing nitrogen fertilizer doses to such an extent is hypothesized to significantly mitigate N_2O and NO emissions. Limited evidence from a single-year experiment supports this hypothesis for N_2O (e.g., Liu et al., 2012). However, whether this significant reduction can be sustained inter-annually for both N_2O and NO emissions remains unclear.

This study employed two-rotation field measurements with different nitrogen dose treatments for a winter wheat–summer maize cropping system in the eastern NCP. The experimental goals were to (a) further test the hypothesis that a reduction of fertilizer nitrogen dose by 1/3 of the conventional addition will significantly mitigate N_2O and NO emissions, (b) quantify the direct emission factors of both gases and, (c) understand the underlying processes and mechanisms of substrate availability and other regulating environmental factors with respect to fluxes of both nitrogen gases.

2. Materials and methods

2.1. Field site and experimental treatments

A winter wheat–summer maize rotation field (36°58′N, 117°59′E; altitude: 9 m above the average sea level) at the North China Field Experimental Station for Intensive Agricultural Ecosystems was selected for our study. The field site (in the Xilujia village of the Xincheng township, Huantai county, Shangdong province) was located in the eastern NCP. It is exposed to a typical temperate monsoon climate. The three-year (2008–2010) means of the annual average temperature and annual average precipitation were 14.4 °C and 627 mm, respectively (as derived from the means of observations at seven climate stations located in the different areas of the county). The soil is calcic cambisols. Its cultivated horizon is a silt loam, with a pH (H_2O) of 8.29, bulk density of 1.42 g cm^{-3}, soil organic carbon of 10.28 g kg^{-1} and total nitrogen content of 1.05 g kg^{-1} (Cui et al., 2012). The ground water table is 8–12 m below the soil surface. The rotation regime of winter wheat and summer maize has been applied in the region for more than five decades, with both crops harvested during the same year. Details for the relevant soil properties are referred to Cui et al. (2012).

The field experiment campaign covered two full cycles of winter wheat (*Triticum aestivum* L.) and summer maize (*Zea mays* L.) rotation, which spanned from October 16, 2008 to October 8, 2010 (each rotation covered a full year-round period). Three nitrogen dose treatments were investigated. They consisted of (a) a control without nitrogen application (CK), (b) the farmer's conventional nitrogen dose (FP) of 600 kg N ha^{-1} yr^{-1} (with 45% for wheat, and 55% for maize), and (c) the optimal practice with a reduced nitrogen dose (OPT) that was 1/3 lower than the FP. Three replicated plots (50 m^2 for each) for each treatment were randomly arranged within a uniform area that had been cultivated with the wheat–maize rotation and managed with conventional practices until the time of our experiments. In each of the two winter wheat seasons, the FP and OPT treatments received basal fertilizers of urea plus diammonium phosphate at rates of 162 and 90 kg N ha^{-1}, respectively, at the time of sowing (the sowing dates were October 11 in 2009 and October 13 in 2008). They were top-dressed with urea at rates of 107 and 90 kg N ha^{-1}, respectively, during jointing (the top-dressing dates were April 9 in 2010 and April 13 in 2009). In each of the two maize seasons, FP and OPT were basally amended with urea plus diammonium phosphate at rates of 165 and 120 kg N ha^{-1}, respectively, immediately following sowing (on June 20) in 2009 and one week after sowing (on June 26) in 2010, and top-dressed with urea at 165 and 120 kg N ha^{-1}, respectively, when the crop plants had 10 leaves (on August 5 in 2009 and August 12 in 2010). For all three treatments, phosphorous and potassium were basally applied at 46 kg P ha^{-1}, 50 kg K ha^{-1}, respectively, in each wheat season, and 48 kg P ha^{-1} and 91 K kg ha^{-1}, respectively, in each maize season. The experimental fields were irrigated four and five times in the first and second rotation cycles (Fig. 1a). These events resulted in total water inputs of 240 and 350 mm, respectively, in addition to regular precipitation.

2.2. Gas flux measurement

We used static chambers to collect air samples for measuring the fluxes of both gases. In each replicated field plot, a stainless steel base frame (50 × 50 × 20 cm) was permanently installed during the experimental period by inserting it 20 cm into the soil, although it was briefly removed during the one-week period prior to plowing for each event of wheat sowing. There were nine holes of 2-cm diameter on the lower half of each base frame wall to allow for free water communication. A channel (3 cm wide and 2 cm deep) was inscribed on the upper edge of each base frame for water-sealing. Each chamber was made of a cube, with stainless steel walls and frames, a bottom area of 50 × 50 cm and a height of 50–150 cm that was adapted to the plant size. The chambers were coated with heat-insulating foam to prevent a significant change in the inner air temperature during sampling. Before we collected air samples for N_2O analysis, we filled the channel with a certain amount of water. Then, a chamber was mounted on the top of the base frame by inserting it into the channel filled with water to ensure a gas-tight enclosure. Immediately after enclosure, we took the first air sample using a 60-mL plastic syringe with a three-way valve. Later, we took another four air samples from the same chamber enclosure, at an interval of 15 min (in winter) or 8 min (in the other seasons). During air sampling, the chamber headspace temperature was recorded for gas density correction in flux calculation using a sensor probe (JM624, Jinming Instrument CO., LTD, Tianjin, China) that was installed beside the chamber. After the last air sample was collected, the chamber was immediately opened to establish ambient air condition within the chamber. Approximately 10 min later, we enclosed the chamber again, and took one 3 L air sample from the enclosure after the chamber was closed for 5 min, using a KNF pump (KNF Neuberger GmbH, Freiburg, Germany). At the same time, another 3-L air sample was taken from outside the chamber enclosure to represent the initial NO concentration. Then, the chamber was immediately removed to allow for the least disturbance of the experimental area. Each air sample was stored in a 5-L gas bag (Pilaite Instrument LTD, Dalian, China) before it was analyzed for NO concentration within 4 h after sampling. During the period with less than 150 cm plant height, the base frames were installed on the crop rows to allow for enclosing entire plants in sampling. Details of the air sampling procedures for measurements of both N_2O and NO gases were the same as those described by Mei et al. (2009, 2011). When the maize was higher than 150 cm and the gas fluxes stimulated by the nitrogen top-dressing were still significant, we cut the upper part of maize plant to fit the chamber height. As soon as the significant gas fluxes stimulated by the nitrogen addition disappeared, we moved the chamber base frame to the inter-rows for the latter measurements of the maize season.

The above sampling procedures were conducted at each experimental plot on a weekly basis during the winter but were performed once every 3 to 4 days during the other seasons. Sampling was carried out daily following the events that were likely to stimulate intensive N_2O and NO emissions like fertilizer application, irrigation, rainfall, and tillage. Such intensified observation lasted for more than 10 to 15 days after fertilizer application and 3 to 7 days following the other events, depending on variation in the gas fluxes. The sampling was completed between 08:00 a.m. and 11:00 a.m. at the local standard time to obtain a single flux of a gas at each plot for the day.

All air samples that were stored inside the plastic syringes were analyzed within 10 h after sampling using an Agilent 6890 Gas Chromatograph (Agilent Technologies Inc., USA) that was equipped with an Electron Capture Detector (ECD). Molecular nitrogen (N_2) with a purity of 99.999% was used as the carrier gas for N_2O analysis. To eliminate the interference by CO_2 in the air samples with the ECD signal of N_2O, we introduced 10% CO_2 in N_2 directly into the detector cell at a flow rate of 1–3 mL min^{-1}. More details on the principles, techniques, instrumental configurations and operation procedures were described by Zheng et al. (2008) and Wang et al. (2010).

The air samples that were stored in the gas bags were analyzed within 2 h after collection using a 42C NO–NO_2–NO_x analyzer (Thermo Environment Instruments Inc., USA), which was calibrated once every two months using the calibration system from the same manufacture and the standard gas from the National Center of Standard Matters (Beijing, China).

Each N_2O flux was determined by using the initial change rate of the concentrations within the enclosure, the headspace volume, the soil surface area that was covered by the chamber, and the gas density (Zheng et al., 2008). The gas density at 273 K and 1013 h Pa was corrected with the actual air pressure and headspace air temperature. Because the altitude of the field site is very near the average sea level, we directly used 1013 h Pa to represent the actual air pressure. Using this constant value could cause flux errors of 2%, at most, as a consequence of diurnal or seasonal variation in ambient air pressure. The initial change rate of the gas concentrations within the chamber headspace of each enclosure was determined using the significant nonlinear (exponential) or linear fitting of the five concentration observations against the enclosure time (Kroon et al., 2008; Valente et al., 1995; Mei et al., 2009, 2011). If the nonlinear or linear correlation between the gas concentrations and the enclosure time was not significant at the 95% confidence interval (i.e., $P > 0.05$), the