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Annual emissions of nitrous oxide and nitric oxide from a wheat–maize cropping system on a silt loam calcareous soil in the North China Plain

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A B S T R A C T

Nitrogen amendment followed by flooding irrigation is a general management practice for a wheat–maize rotation in the North China Plain, which may favor nitrification and denitrification. Consequently, high emissions of nitrous oxide (N_2O) and nitric oxide (NO) are hypothesized to occur. To test this hypothesis, we performed year-round field measurements of N_2O and NO fluxes from irrigated wheat–maize fields on a calcareous soil applied with all crop residues using a static, opaque chamber measuring system. To interpret the field data, laboratory experiments using intact soil cores with added carbon (glucose) and nitrogen (nitrate, ammonium) substrates were performed. Our field measurements showed that pulse emissions after fertilization and irrigation/rainfall contributed to 73% and 88% of the annual N_2O and NO emissions, respectively. Soil moisture and mineral nitrogen contents significantly affected the emissions of both gases. Annual emissions from fields fertilized at the conventional rate (600 kg N ha^{-1} yr^{-1}) totaled 4.0 ± 0.2 and 3.0 ± 0.2 kg N ha^{-1} yr^{-1} for N_2O and NO, respectively, while those from unfertilized fields were much lower (0.5 ± 0.02 kg N ha^{-1} yr^{-1} and 0.4 ± 0.05 kg N ha^{-1} yr^{-1}, respectively). Direct emission factors (EF_ds) of N_2O and NO for the fertilizer nitrogen were estimated to be 0.59 ± 0.04% and 0.44 ± 0.04%, respectively. By summarizing the results of our study and others, we recommended specific EF_ds (N_2O: 0.54 ± 0.09%; NO: 0.45 ± 0.04%) for estimating emissions from irrigated croplands on calcareous soils with organic carbon ranging from 5 to 16 g kg^{-1}. Nitrification dominated the processes driving the emissions of both gases following fertilization. It was evident that insufficient available carbon limited microbial denitrification and thus N_2O emission. This implicates that efforts to enhance carbon sink in calcareous soils likely increase their N_2O emissions.

1. Introduction

Atmospheric nitrous oxide (N_2O) is an important contributor to radiative forcing and a key substance in atmospheric chemistry and is therefore contributing to ongoing global warming as well as to stratospheric ozone destruction (IPCC, 2007; Ravishankara et al., 2009). Due to human activities, the atmospheric abundance of N_2O has been rising dramatically from pre-industrial value of approximately 270 ppbv to at present 319 ppbv (Flückiger et al., 1999; IPCC, 2007; Sowers, 2001). Agricultural soils are recognized as the major source of atmospheric N_2O, globally contributing 1.7–4.8 Tg N yr^{-1} (IPCC, 2007) to the global atmospheric N_2O budget of approximately 14 Tg N yr^{-1} (Fowler et al., 2009).

Nitric oxide (NO), another important player in atmospheric chemistry, participates in regulating the oxidant balance in the troposphere (Fowler et al., 2009). It is a key precursor of tropospheric ozone that is a greenhouse gas and contributes to the atmospheric deposition of nitrogen globally (Galloway et al., 1994; IPCC, 2007). Nitrogen fertilized agricultural soils being the main driver of soil NO emissions (1.6 Tg N yr^{-1}), may account for up to 18% of the global soil source (8.9 Tg N yr^{-1}) (Bouwman et al., 2002b; IPCC, 2007). Agricultural NO emissions play an important role in the tropospheric ozone chemistry of rural regions away from intensive fossil fuel combustion (Bouwman et al., 2002b; Butterbach-Bahl et al., 2009).

In agricultural ecosystems, N_2O and NO are predominantly formed in biological nitrification and denitrification (Davidson, 1991) while the contribution of other processes (e.g., chemical denitrification) is still poorly known (Wrage et al., 2001). In nitrification ammonium is oxidized via nitrite to nitrate. Some N_2O and NO are formed in the first step of nitrification, i.e., in ammonium oxidation (Firestone and Davidson, 1989). The primary regulating factors of nitrification in the majority of soils are the ammonium (NH_4^+) and oxygen (O_2) supplies, though the importance of soil

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temperature and pH are non-ignorable (Robertoson and Groffman, 2007). Denitrification is the stepwise reduction of NO_3^- to N_2 performed by denitrifying microorganisms in the absence of O_2 as electron acceptor. NO and N_2O are obligate intermediate products of the denitrification process. Supplies of carbon, oxidized inorganic nitrogen substrates (NO_3^-, NO_2^-, NO, N_2O) and O_2 concentration in soil are the three most important regulating factors affecting soil denitrification rates. Oxygen is by far regarded as the dominant control on denitrification rates in general, while the importance of carbon and NO_3^- (or other nitrogen oxides) will vary by ecosystem (Robertoson and Groffman, 2007). Due to the complex interactions between these factors, large temporal and spatial variations of N_2O and NO emissions are usually observed in cropland soils.

The North China Plain (NCP), mostly containing calcareous soils, is an intensive agricultural region. It covers approximately 300,000 km^2 and provides about one-fourth of China's total grain yield (Liu et al., 2001). A winter wheat–summer maize rotation is a general cropping system, which is characterized by high addition synthetic nitrogen fertilizers but low (less than 30%) nitrogen-use efficiency (Jia et al., 2004; Zhao et al., 2006). A number of studies have found fertilizer application rates of up to 600 kg N ha^{-1} yr^{-1} or even higher (Ju et al., 2007; Zhao et al., 2006). Nitrogen amendment followed by flooding irrigation is a general management practice for the wheat–maize cropping systems of the NCP (Liu et al., 2001). This practice is likely to result in favorable conditions for nitrification and denitrification and associated N_2O/NO emissions. The published direct emission factors (EF_ds) of N_2O for the wheat–maize cropping systems in NCP range from 0.02 to 1.93% (Ding et al., 2007; Dong et al., 2000; Ju et al., 2011; Meng et al., 2005; Zhang et al., 2011) and nitrification has been the main process for the N_2O emission because low availability of easily degradable carbon substrates evidently limits denitrification in this cropping system (Ju et al., 2011). However, it remains unclear whether their conclusions can be applied more generically to other areas of the NCP region with calcareous soils. There is only one earlier field study on soil NO emissions in the NCP (Zhang et al., 2011). The large variation in the measured EF_ds of N_2O and the rare reports concerning NO emissions from croplands in the NCP demand further field investigations on the fluxes of both gases and their regulating factors.

Therefore, the aims of our study were to: a) investigate the temporal variation in N_2O and NO emissions in situ; b) quantify annual emissions and direct emission factors (EF_ds) of both gases by measuring their fluxes from unfertilized and conventionally fertilized fields; c) to test the hypothesis that nitrogen fertilization immediately followed by flooding irrigation stimulates intensive emissions of N_2O and NO; and d) test if availability of carbon limits denitrification and thus N_2O emissions.

2. Materials and methods

2.1. Field site

Our field measurements were conducted from October 2008 to September 2009 at the experimental station (36°58′ N, 117°59′ E, approx. 17 m above sea level) founded by China Agriculture University. The field site is located at a suburban arable field site approximately 10 km away from the downtown of Huantai County, a key grain production area in the center of Shandong province. It is representative of the intensive agricultural areas of the NCP where the winter wheat–summer maize rotation is the typical cropping system. The field site has a warm temperate continental monsoon climate. From 2000 to 2008, the Weifang climate station had a mean annual air temperature of 13.0 °C and average annual precipitation of 586 mm (National Climate Data Center, ftp://ftp.ncdc.noaa.gov/pub/data/gsod/). The area has calcaric cambisols (WRB, 2007), with a silt loam texture. Soil properties (0–20 cm) are listed in Table 1. The ground water table depth is 8–12 m (L. Chen, personal communication). The winter wheat–summer maize rotation system has been in use here for at least 50 years.

2.2. Field treatments

In our experiments, we used the conventional double cropping system, with winter wheat (*Triticum aestivum* L.: early October to mid-June) and summer maize (*Zea mays* L., late June to late September) in rotation. In an uniform wheat–maize field, three replicate plots were randomly selected for both the control and fertilized treatment. Each plot covered an area of 51 m^2. Fertilizer applications, totaling 600 kg N ha^{-1} (270 and 330 kg N ha^{-1} for wheat and maize, respectively), were conducted following the local conventional regimes. The fertilizers were amended with four split applications. Each crop was fertilized with nitrogen twice: a basal fertilization before or immediately after sowing (wheat: 60%; maize: 50%) and a top-dressing at wheat tillering (40%) or when the maize had 11–12 leaves (50%). All phosphorous and potassium fertilizers (wheat: 105 kg P_2O_5 ha^{-1} and 60 kg K_2O ha^{-1}; maize: 110 kg P_2O_5 ha^{-1} and 110 kg K_2O ha^{-1}) were basally and equally applied for both treatments. Four irrigation events for wheat (totally 250 mm water) and one for maize (60 mm water) were adopted. The additional field management of both field treatments, such as tillage, wheat and maize species, time for crop sowing and harvest, and residues incorporation, also followed the local conventional practices. Immediately before wheat sowing, harvested maize straw was fully cut into small pieces and plowed into soil (0–20 cm) at a rate of approximately 1664 kg C ha^{-1} (with a C:N ratio of 65). At wheat harvest, standing stubbles (approx. 15 cm high) remained in the field, and the harvested straw (approx. 1686 kg dry C ha^{-1}; with a C:N ratio of 93) was applied back to the field by covering the ground. One week later, maize seeds were sown into prepared rows, with the field itself remaining untilled. Details of field management practices are shown in Table 2.

2.3. Field measurements of N_2O and NO fluxes

In the center of each plot, one subplot was permanently set for measurements of N_2O and NO emissions by inserting a stainless steel base frame (covering an area of 0.5 m × 0.5 m) to a depth of 10 cm. Fluxes of both gases were simultaneously measured with static, opaque chambers (made of stainless steel sheet and coated with heat-isolation material), which were mounted onto the permanently installed base frames. A gas-tight seal was ensured by filling the groove with water. Negative pressure in the chamber during sampling was prevented with a Teflon tube (inner diameter 1/4 inches, 10 m long) between the chamber headspace and the atmosphere. An active carbon filter, which was used to remove NO from the air flowing into the chamber, was connected to the external end of the pressure balance tube. The chamber heights were adapted to plant height (varying from 0.5 m, to 1.0 m and

Table 1
Soil properties.

Soil depth (cm)	pH	Bulk density (g cm^{-3})	Organic matter (g kg^{-1})	Total N (g kg^{-1})	Clay[a] (%)	Silt[b] (%)	Sand[c] (%)
0–20	8.29	1.42	17.73	1.05	17.1	66.1	16.8

[a] <0.002 mm.
[b] 0.002–0.05 mm.
[c] 0.05–2 mm.