Reactive N emissions from cropland and their mitigation in the North China Plain

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**Abstract**

Excessive application of chemical nitrogen (N) fertilizer and inefficient N management are still common in the North China Plain, leading to large reactive N (Nr) losses and pollution, threatening environmental security and public health. Three improved N management practices (33% reduction in N applied (OU), OU combined with partial organic fertilizer substitution (UOM) and the urea in UOM amended with a urease inhibitor (ULOM)) together with no N application (CK) and farmers’ conventional practice (CU) were tested on a maize-wheat rotation at Quzhou, Hebei, North China Plain (NCP). Nr emissions were related to WFPS (Water Filled Pore Space), soil mineral N (NH4+-N and NO3--N) and soil temperature. Nr emissions and yield-scaled Nr emissions were significantly reduced by partial substitution of organic fertilizer for chemical fertilizer: NH3 emissions were reduced by 55.8-62.4%. Using a urease inhibitor (Limus®), further reduced NH3 emissions by 40.2-64.5%. Yield-scaled NH3 emissions were, on average, reduced by 60.0% and 55.2% in the maize and wheat growing season, respectively, relative to the UOM treatment. Long-term application of organic fertilizer had a significant positive effect on N use efficiency (NUE). Overall, the study shows that appropriated N management such as reducing the N application rate, partial substitution of chemical N by organic N and using a urease inhibitor can reduce Nr emissions and promote NUE in the North China Plain. The methods corresponding to the ULOM and UOM treatments were the most and second most effective, respectively, with high net economic benefits.

**Keywords:**

Reactive N emissions mitigation; Organic fertilizer; N stabilizer; maize-wheat rotation

**1. Introduction**

Emissions of reactive N (Nr) gases to the atmosphere from land make a significant contribution to the environmental costs of agricultural production (Jörg et al., 2001; Wintjen et al., 2020). Cropland is one of the main sources of gaseous Nr emissions, contributing 33% of the total ammonia (NH3) and 35% of the total nitrous oxide (N2O) emissions in China (Huang et al., 2012; Luo et al., 2019). This results from the high application rates and low N use efficiency (NUE) (Jung et al., 2016; Zhang et al., 2016). Previous research has reported that the loss of N fertilizer through gaseous emissions accounts for 24.7% of the applied N (Ju et al., 2009). NH3 indirectly contributes to smog pollution, promoting particle formation both in the initial homogeneous nucleation phase and in subsequent particle growth in the creation of haze episodes (Pan et al., 2016; Wang et al., 2016; Sawlani et al., 2021), further influencing human health. As reported, the global deaths and disability-adjusted life years due to air pollution have increased by 102.3% and 67.7% from 1999 to 2019, respectively (Sang et al., 2022). Increasing NH3 emissions also enhance N deposition which leads to soil acidification, loss of biodiversity and eutrophication of water bodies, threatening environmental security (Clark and Tilman, 2008; Liu et al., 2013). In addition, approximately 1% of the NH3 will be converted into N2O after deposition according to the IPCC (Klein et al., 2006), thus indirectly contributing to the greenhouse effect and ozone depletion by producing NO in the stratosphere (Hu et al., 2021). The warming effect of N2O is 273-298 times that of CO2 for a 100-year time horizon, which makes it a major cause of climate change (Masson-Delmotte, 2021; Forster et al., 2007).

To achieve sustainable agricultural production, Nr emissions must be greatly reduced. China has issued a series of action plans, such as the “Zero Increase Action Plan for National Fertilizer Use by 2020” and “Three-year Action Plan on Defending the Blue Sky” to mitigate agricultural non-point source pollution (Liu et al., 2016a; 2020). Some of these have been effective. For example, optimizing the N application rate has improved NUE and decreased Nr losses (Li et al., 2019, Mi et al., 2022) without decreasing crop yields (Cui et al., 2013; Ren et al., 2022). Also replacing some or all of the synthetic fertilizer by organic fertilizer such as recycled manures has been found to be very effective for reducing Nr losses from intensive agricultural production (Guo et al., 2020; Zhang et al., 2018; Zhang et al., 2020; Xu et al., 2022). However, Shi et al. (2017) reported that N2O emissions were increased from summer maize production on saline-alkali soil when organic fertilizer was applied, and Ma et al. (2021) found that organic fertilizer was more likely to increase NH3 emissions. It is therefore vital to clarify the effect of organic fertilizers. In addition, N stabilizers, especially urease inhibitors, can slow urea hydrolysis and have the potential to reduce Nr losses from urea and so improve NUE and yield. The application of urea coated with the urease inhibitor Limus® significantly reduced NH3 volatilization and the amount of N fertilizer required in a maize-wheat rotation in the NCP (Li et al., 2017; Sha et al., 2020a), but its effect on N2O emissions is variable (Feng et al., 2016; Sha et al., 2020b). These uncertainties raise questions as to whether these N management methods can decrease Nr emissions and increase crop yields. The effects of these N practices on environmental effects and crop production need further evaluation.

Maize, wheat and rice have been shown to be the main crops contributing to global NH3 emissions (Liu et al., 2022a) and the maize-wheat rotation is one of the most important production systems for grain production. In China, the area planted to maize and wheat accounts for 36% and 20% of the sown area of grain crops, respectively (Bureau, 2021). The North China Plain (NCP) as China’s granary, produces 31% of national maize production and 73% of national wheat production, contributing greatly to Chinese agricultural production and food security (Xiao et al., 2019). Also the NCP is an area of intensive livestock production, often uncoupled from crop production, reducing resource use efficiency (Hardesty and Tiedeman, 2009). Identifying the effects of organic sources of N on agricultural production (especially the maize-wheat rotation) and designing specific measures to balance the relationship between crop production and livestock production is a win-win method for reducing resource waste and achieving sustainable agricultural production in the NCP.

To this end, we tested three potentially effective N management practices for regional maize production: optimized chemical N (urea) application (OU), OU combined with partial organic fertilizer substitution for the urea (UOM) and the urea in UOM amended with the urease inhibitor Limus® (ULOM). NH3 and N2O emissions, crop yield, NUE and the N balance were measured in order to systematically evaluate the potential for Nr reduction and sustained agricultural productivity. In particular, the best combination of individual N management measures was tested, using the optimized N rate, partly substitution of urea with an organic source of N and stabilized urea as a comprehensive N management technique, with the aim of recommending suitable N management practices for a more efficient maize-wheat rotation system in the NCP, and providing technical support for regional agricultural green development and/or meeting sustainable development goals through higher crop yields and NUE with substantially reduced Nr losses.

**2. Materials and methods**

2.1. Experimental sites

A 2-year field experiment was carried out with three key components (Fig.1) in a maize-wheat rotation field trial at Quzhou Experimental Station, China Agricultural University, Hebei province, China (36°78′01′′ N, 114°94′51′′ E). The common cropping practice is a rotation of summer maize and winter wheat (i.e. two harvests a year). This region has a temperate sub-humid continental monsoon climate with an average air temperature of 13.2℃ and mean annual precipitation of 490 mm; most precipitation occurs from July to September (Fig. 2). The soil is a typical fluvo-aquic soil with 0-20 cm topsoil with: pH 7.95 (2.5:1 water:soil ratio), organic matter 23.1 g kg-1, total N 1.02 g kg-1, available phosphorus (Olsen-P) 6.17 mg kg-1 and available potassium (NH4OAc exchangeable K) 132 mg kg-1.

2.2. Experimental design

The field experiment comprised 5 treatments: (1) CK, no N fertilizer applied either in organic or chemical forms; (2) CU, conventional (farmer’s practice) N fertilizer (urea) applied at a rate of 180 kg N ha-1 to maize and 270 kg N ha-1 to wheat; (3) OU, optimized chemical N application (urea), i.e. a 33% reduction with a rate of 120 kg N ha-1 for maize and 180 kg N ha-1 for wheat; (4) UOM, the same treatment as OU but with 50% of the N supplied by a commercial organic fertilizer (60 kg N ha-1 for maize and 90 kg N ha-1 for wheat) and 50% supplied by urea; (5) ULOM, the same treatment as UOM but with the urea coated with a urease inhibitor (Limus®, 75% N-(n-butyl) thiophosphoric triamide (NBPT) plus 25% N-(n-propyl) thiophosphoric triamide (NPPT)) at a dose rate of 0.1% (w/w). The organic fertilizer was manufactured by Tongliao Lvnong Biochemical Engineering Co. LTD, with N and K2O contents of 12% and 1.5%. During the maize season, the organic fertilizer was applied as a basal application to the surface before sowing and immediately incorporated to 10 cm depth. The urea was evenly broadcast once at the three-expanded leaf stage. The basal fertilizer to topdressing ratio was set at 1:1 in the wheat season (50% chemical N for the CU and OU treatments); all organic N for the UOM and ULOM treatments was applied as basal fertilizer. The remaining urea was top-dressed at jointing stage. All treatments were established in a randomized block design with three replicates. The area of each plot was 140 m2. 90 kg P2O5 ha-1 and 60 kg K2O ha-1 were applied before sowing in both the maize and wheat seasons and uniformly incorporated to depth as above.

Maize was sown with row and plant spacings of 60 cm and 22 cm, respectively, while the wheat was sown with a row spacing of 20 cm. The seed rates of maize and wheat were approximately 38 and 225 kg ha-1, respectively. Pesticides and herbicides were applied according to local farmer’s practice. Sowing and harvest dates for maize and wheat are in Table 1.

2.3. Measurements and calculations

2.3.1. NH3 measurement

NH3 volatilization was measured by the Dräger-Tube (Drägerwerk AG, Lübeck, Germany) method (DTM), a dynamic chamber method, calibrated by micrometeorological Integrated Horizontal Flux (IHF) (Li et al., 2015; Pacholski et al., 2006). The measuring system consisted of 4 closed chambers (each covered an area of 100 cm2 and contained 370 cm3 of gas) connected by Teflon tubes through which air was drawn by pumps, each pump having with a volume of 100 cm3. NH3 measurement frequency was determined by the NH3 concentration and adjusted with weather conditions. Details are in Li et al. (2017) and Sha et al. (2020b). Wind speeds at 2.0 m and 0.2 m height, air pressure and temperature were recorded and used to calculate NH3 fluxes (see below).

2.3.2. N2O measurement

A static chamber-gas chromatography technique was used to measure N2O emissions. Each static chamber was 50×60×50 cm, made of stainless steel and covered with 3-cm thick foam to maintain a stable temperature. Three small holes were made on the side of each chamber: one was for the wires of a fan installed inside the chamber for mixing the air; the other two were for a temperature sensor and a Teflon sampling tube. A steel frame 50×60 cm was inserted into the soil to 15 cm depth with a 3-cm wide slot around the top of the frame. Before sampling water was added to the slot to seal the chamber when it was attached. Gas samples were collected continuously for seven days after fertilizer was applied, after which samples were collected every two days for the next two weeks and then twice a week for the next month. After this gas samples were collected once a week for the remainder of the maize season, but with daily sampling for 2 days when rainfall occurred. Gas samples were collected from 8:30 to 10:00 in the morning, a representative time for estimating mean daily fluxes according to previous research (Tian et al., 2012). Four 50 mL gas samples were collected at 10-minute intervals using a 60 mL syringe of which 20 mL were used for N2O analysis.

The N2O concentration was measured using a gas chromatograph (GC; Agilent 7890A, Agilent Technologies, Santa Clara, CA, USA) as in Yue et al. (2018). N2O flux rates were calculated as follows:

(1)

Where () is the N2O flux; is the unit conversion factor (1000); the pressure at the time of sampling; is the absolute temperature during sampling; and are the absolute temperature and pressure of the air under standard conditions, respectively; represents the molecular weight of N per mole of N2O; is the molar volume of gas under standard conditions (22.4 L mol-1); (m) is the height of the chamber; indicates the rate of change in N2O concentration over time.

2.3.3. Soil Mineral N (SMN) and Water Filled Pore Space (WFPS)

Surface soil samples were collected on the day of NH3 or N2O measurement. Three samples were randomly collected from each plot, combined, stored in a portable icebox and brought back to the laboratory. After removal of stones, roots, etc, and sieving, the soil samples were added to 0.01 mol L-1 CaCl2 solution and shaken for 1h at room temperature on a horizontal shaker at 150 rpm. The solution was filtered and soil NO3--N and NH4+-N measured using an Auto Analyzer (AA3, Germany).

Soil water content was determined using the hot air-drying method (20.00 g fresh soil sample was oven-dried at 105℃ for 8 hours), and WFPS calculated as follows:

(2)

in which soil bulk density was determined using a cutting ring; soil particle bulk density was assumed to be 2.65 g cm-3.

2.3.4. Environmental data

Air temperature (AT) was obtained from local weather data. Soil temperature (ST) was measured with a thermometer inserted 5-10 cm into the soil. Precipitation samples were collected in rain gauges (SDM6, Tianjin Weather Equipment Inc., China) and the volume determined with a measuring cylinder.

2.3.5. Yield and plant total N

At maturity maize and wheat plants were harvested from 9 m2 and 4 m2 areas in each plot, respectively. Grain samples were oven-dried (75℃) and weighed to calculate crop yield (adjusted to 14% moisture content).

At the same time 6 maize plants and 1 m2 wheat were harvested from each plot, the grain and straw separated and used for total N determination. Plant samples were digested with H2SO4-H2O2 and total N determined by the Kjeldahl method. Crop N uptake was calculated from N content and dry matter weight.

2.3.6. Calculation of Yield-scaled Nr emissions, NUE and N budgets

Yield-scaled Nr emissions, NUE and N balance were calculated using the following formulae:

(3)

(4)

here is the N uptake of treatment with N fertilizer application; is the N uptake of CK in which no N fertilizer was applied; is the N application rate.

(5)

(6)

(7)

in which represents the amount of N mineralization from soil, estimated by the balance of inputs and outputs in the CK treatment. Outputs consisted of plant N uptake, residual NO3--N in the 0-1m soil layer in the CK treatment after harvest, and input was initial NO3--N in the 0-1m soil layer in the CK treatment before planting; represents the N fertilizer application rate; represents the initial soil NO3--N (NH4+-N was negligible and so ignored) in the 0-1 m soil layer; represents the residual accumulation of NO3--N in the 0-1 m soil layer after harvest. For analysis, 0-1 m soil samples were collected in 5 x 20 cm layers before sowing and after harvest and analyzed using the method given earlier.

2.4. Data analysis

Differences in NH3 emissions, N2O losses, yields, etc., between treatments were computed using a one-way analysis of variance (ANOVA). NH3 and N2O emissions and soil properties (e.g. WFPS, NO3--N, NH4+-N) during the measurement period were used for correlation analysis or data fitting.

Statistical analyses and mapping were performed using the SPSS 26.0, Origin 2016, R-studio 5.0 analytical software package.

**3. Results**

3.1. NH3 emissions

Over the 2-year period, NH3 flux peaks were observed 1-3 days after N fertilizer application during the maize growing season both in 2019 and 2020 (Fig. 3). The peak fluxes from the urea-only treatments CU (conventional) and OU (optimized) were 0.6 and 0.4 kg N ha-1 h-1 in 2019 and 2.4 and 2.3 kg N ha-1 h-1 in 2020, respectively. Thus N optimization significantly reduced NH3 volatilization by 27.7% in 2019, but with no significant effect in 2020. The peak fluxes from UOM (urea and organic N, optimized rate) and ULOM (as UOM with the Limus® inhibitor) were 0.4 and 0.2 kg N ha-1 h-1 in 2019 and 0.6 and 0.2 kg N ha-1 h-1 in 2020, respectively. Thus compared to OU, substituting 50% of the N with N in organic fertilizer decreased NH3 flux peaks from maize by 17.5 and 75.3% in 2019 and 2020, respectively, and applying the Limus® inhibitor reduced flux peaks significantly by another 42.4% and 71.0% in 2019 and 2020, respectively.

During the wheat growing season, NH3 fluxes were relatively low and there were no significant differences between the four treatments after basal fertilizer application (Fig. 3). As in the maize growing season, NH3 fluxes peaked 1-3 days after fertilizer was applied at topdressing. The maximum fluxes from OU were 41.7 and 17.1% lower than those from CU in 2020 and 2021, respectively. The peak flux from the UOM treatment was 0.18 kg N ha-1 h-1, which was 52.2% less than that from CU (0.38 kg N ha-1 h-1) in 2020. Again the use of the inhibitor in ULOM reduced NH3 volatilization compared to the other three treatments with fluxes ranging from 0-80.3 g N ha-1 h-1 in 2020, and from 0-70.8 g N ha-1 h-1 in 2021; i.e. peak fluxes were significantly decreased by 78.6 and 65.0% compared to CU in 2020 and 2021, and by 55.3-63.3% and 57.7-66.5% compared to OU (the same level of N application as CU) in 2020 and 2021, respectively.

Further analysis shows that the NH4+-N content of the soil and the WFPS had significant positive impacts on NH3 fluxes in the maize growing season (Fig. 4a), while there was a significant negative correlation between soil NO3--N content and NH3 emissions (P<0.01). During the wheat growing season (Fig. 4b), as in the maize season, high WFPS and NH4+-N content increased the NH3 emission potential; increasing air temperature (AT) also increased NH3 emissions (P<0.05).

3.2. N2O emissions

Significant N2O emission peaks appeared after fertilizer application during the maize growing season but not the wheat growing season (Fig. 5). In the maize season the highest fluxes were measured 3-5 days after application, and peak fluxes from the OU treatment were 440.3 µg N m-2 h-1 in 2019 and 530.1 µg N m-2 h-1 in 2020; there were no significant differences compared to CU. N2O peak fluxes from UOM and ULOM were lower than those from CU and OU, with 80.3-88.8% reductions in 2019 and 59.1-87.1% reductions in 2020.

During the wheat growing season N2O fluxes from the UOM and ULOM treatments were consistently low with very small peaks in 2019-2020, no more than 42.6 and 42.8 µg N m-2 h-1 (Fig. 5).Even so these were significantly less, by 53.4-53.7% and 42.2-42.6%, compared to those from the CU and OU treatments, respectively. In 2020-2021, N2O fluxes rapidly increased at the late growth period of wheat, reaching 658.9 and 598.9 µg N m-2 h-1 for the CU and OU treatments, respectively. Fluxes from UOM and ULOM were also high at more than 118.5 µg N m-2 h-1, but still significantly lower than those from the urea-only treatments.

Correlation analysis showed that WFPS and soil NH4+-N content had a significant positive effect on N2O emissions during the maize growing season, but an increase in AT significantly inhibited N2O emissions. In contrast ST, AT and soil NO3--N content had significant positive relationships with N2O emissions in the wheat growing season. N2O emissions increased, but not significantly, with WFPS and soil NH4+-N content.

3.3. Cumulative NH3 and N2O emissions

Optimizing the N applications decreased cumulative NH3 emissions significantly in 2019-2020 for wheat and in 2020 for maize by 21.8 and 17.6% compared to the CU treatment, respectively (Fig. 7a). At the same N application rate, using 50% organic N significantly reduced NH3 emissions over the 2 years by 21.8-34.0% and 31.6-37.0% in the maize and wheat growing seasons, respectively. Compared to the UOM treatment, using the urease inhibitor (ULOM) had an additional effect in reducing NH3 emissions, with cumulative emissions significantly reduced by 55.8-62.4% and 40.2-64.5% from maize and wheat at the 0.05 level, respectively.

Optimizing the N rate was also effective at reducing N2O emissions during the maize growing season, with cumulative emissions reduced by 29.9% and 31.3% in 2019 and 2020, respectively (Fig. 7b). In addition, compared to the OU treatment, the partial substitution of urea by organic N (UOM) or using the inhibitor (ULOM) significantly reduced N2O emissions by 40.5-65.0% and 48.2-68.7% in the maize and wheat growing seasons, respectively, but there was no significant difference between the UOM and ULOM treatments.

3.4. Crop yield, yield-scaled Nr emissions and NUE

The control treatment without N fertilizer had the significantly lowest grain yield as expected in 2019-2021 (Table 2). Optimizing the N application rate did not impact grain yield and NUE significantly, but significantly reduced yield-scaled NH3 emissions by 29.3% and 20.5% during the 2019-2020 wheat and 2020 maize growing seasons, respectively. Compared to the CU treatment, yield-scaled N2O emissions from the OU treatment were significantly lower by 27.2% and 29.1% during the maize growing season in 2019 and 2020, respectively. There was a significant effect from the partial substitution of chemical fertilizer by organic N on increasing yield in the second rotation, and the yields of the UOM treatment were on average 14.4% higher than those from the OU treatment in the whole maize-wheat rotation in 2020-2021. Compared to the OU treatment, yield-scaled NH3 emissions from the UOM treatment were significantly reduced by 18.3-46.3% over the 2 years. Similarly, yield-scaled N2O emissions from the UOM treatment were effectively mitigated, with a 55.7-73.6% reduction over the whole maize-wheat rotation except in the 2019-2020 wheat season. Partial substitution of urea by organic N increased NUE in the wheat growing season by 25.0% and 29.1% in 2020 and 2021, respectively. Compared to the UOM treatment, the use of the inhibitor showed significant advantages in decreasing yield-scaled NH3 emissions, with average 60.0% and 55.2% reductions in the maize and wheat growing seasons, respectively. However, there were no statistically significant differences in yield, yield-scaled N2O emissions and NUE between the UOM and ULOM treatments.

3.5. Nitrogen budgets

The initial NO3--N content of the soil had a significant effect on N budgets especially in the CU treatment (Table 3). The N uptakes from the optimized N (OU) treatment were higher than those from the conventional N application treatment (CU) in both the maize and wheat growing seasons, and those from the treatment that used 50% organic N and the inhibitor exceeded those from the urea treatments. N surpluses increased with increasing N applications. When applying urea-N, N surpluses ranged from 240-411 kg N ha-1 for CU and 132-193 kg N ha-1 for OU. Partial substitution of urea by organic N and using the N inhibitor reduced N surpluses, with the ULOM treatment having the largest effect in reducing N losses, increasing N utilized and thus agricultural sustainability.

3.6. Cost-benefit analysis

In 2019-2021, input costs were reduced by 5.6 and 14.7% in the maize and wheat seasons, respectively, when the optimized N rate was used (OU; Table 4) but both the use of organic N (UOM) and the inhibitor (ULOM) increased input costs, especially the use of the inhibitor. As a result of this, and yield variation (Table 2) caused by the weather and disease, effects on net economic benefit (profit from increased yield minus extra costs) were variable. In the 2019 maize season, all the treatments reduced profit and net economic benefit, but in the next three cropping seasons, results were better, although not always beneficial, especially for OU, the simple reduction in urea N application. In these three seasons, UOM and ULOM improved profit, ranging from 6.7% (UOM treatment) to 21.1% (ULOM treatment) and net economic benefit from 8.8% (UOM treatment) to 34.6% (ULOM treatment).

**4. Discussion**

4.1. Nr emissions

Alkaline soils such as those in this experiment are at risk of NH3 loss. Other soil conditions such as moisture content influencethe dynamic equilibrium between the aqueous and gaseous phases of NH3 (Duan and Xiao, 2000; Sha et al., 2019). We found that soil NH4+-N content had a significant positive correlation with NH3 fluxes, so NH3 emissions increased rapidly after fertilizer application due to the increasing NH4+-N content in the surface soil, especially in the CU and OU treatments. However, NH3 volatilization was low after basal fertilizer was applied in the wheat season because of the cool AT and the deep placement of fertilizer that led to a low content of NH4+-N in the surface soil. Increasing WFPS after 9.9 mm precipitation promoted NH3 volatilization in the 2019 maize season because of accelerated urea hydrolysis (Li et al., 2015). Soil NO3--N content had a negative correlation with NH3 fluxes because increasing soil NO3--N indicates that the substrate (NH4+) of NH3 volatilization is quickly consumed by nitrification (Zhu et al., 2020).

The production pathway of N2O is complex and is dependent on a number of variables. Wang et al. (2021) divided these into three categories: environmental, management and measurement factors, which interact with each other. Soil moisture is one of the major factors affecting N2O emissions and our results showed a positive effect of WFPS on N2O emissions. Some studies have reported maximum N2O emissions when WFPS is >60% (Bateman and Baggs, 2005; Rabot et al., 2014; Wang et al., 2021), and this was confirmed by the rapidly increasing N2O fluxes at the late stage in the 2020-21 wheat season in this experiment (Fig. 5). We also found a positive correlation between NH4+-N content and N2O fluxes, especially in the maize season when it was significant at the 0.001 level, possibly because, as a precursor of soil nitrification, increasing NH4+-N is rapidly nitrified (Sha et al., 2020b). In the wheat season, N2O emissions were significantly related to soil NO3--N content at the 0.05 level. ST had contrasting effects on N2O emissions in the maize (negative effect) and wheat seasons (positive effect), perhaps because the relationship between N2O emissions and ST is not linear (Yin et al., 2015): the optimum temperature for nitrification is 25-35℃, and we found that 62% of ST measurements exceeded 25℃, ranging from 25.1-33.4℃, in the maize seasons, which may enhance the conversion of NH4+-N to NO3--N and decrease the formation of intermediate N2O (Wang et al., 2010).

Globally the application of chemical fertilizers is the main source of NH3 and N2O emissions from cropland (Davidson and David, 2014; Kang et al., 2016; Xu et al., 2015), and the N application rate has been found to be positively correlated with Nr emissions (Maaz et al., 2021; Sha et al., 2021). Our results are consistent with this: with the 33% reduction in urea-N applied there were 11.0-22.4% and 17.6-21.8% reductions in NH3 emissions and 24.5-26.6% and 15.7-17.9% reductions in N2O emissions for maize and wheat, respectively. Large Nr losses from the CU treatment are probably due to the excessive N supplied compared with crop demand. The emission reductions from the UOM treatment relative to the CU treatment can be explained by the reduced N application rate but also the replacement of urea-N by organic N. This slowed the speed of N mineralization, improved the fixation of N by microorganisms and so soil NH4+-N content was reduced (Guo et al., 2020; Xia et al., 2017a). This also caused the observed NH3 and N2O emissions reduction from the UOM treatment compared to the OU treatment. Cumulative NH3 emissions of the ULOM treatment were significantly decreased by 55.8-62.4% and 40.2-64.5% from maize and wheat, respectively, compared to the UOM treatment, owing to the use of Limus®, slowing the rate of urea hydrolysis and controlling NH4+-N release (Sha et al., 2020b). However, there was no significant difference in N2O emissions between the UOM and ULOM treatments, perhaps because there was no significant difference in the utilization of NH4+-N by crops on the UOM and ULOM treatments and a similar amount of NH4+-N was therefore nitrified (Akiyama et al., 2009). In contrast, Sha et al. (2020b) reported that applying urea coated with Limus® significantly reduced N2O emissions due to the reduction of soil NH4+-N content in an indoor controlled environment experiment (Sha et al., 2020a). The effect of Limus® on N2O emissions clearly needs further research.

4.2. Crop productivity and N budgets

Optimizing the N application rate should not significantly affect crop yields (Lu et al., 2021), and we achieved this using urea as the only N source (Table 2). NUE was increased, but not statistically significantly, by reducing the N application rate. Although 33% less N was applied, a large amount of soil N was present, and so N mineralization could have satisfied crop N requirements. The application of 50% urea-N and 50% organic N also sustained crop yields relative to urea alone. An organic fertilizer will increase soil organic matter and so slowly provide mineral N to crops (Chen et al., 2018). According to previous research, the use of organic N increases soil organic carbon content, improving soil N storage and crop productivity (Guo et al., 2020; Lazcano et al., 2021; Singh et al., 2009). This can also improve NUE: the long-term application of organic fertilizer significantly increased NUE on average by 27.0% in the wheat growing seasons in our 2-year experiment relative to the OU treatment. However, compared to the UOM treatment, crop yields were not significantly increased by using Limus®, consistent with the conclusions of (Li et al., 2017; Li et al., 2015). Thus, although NH3 emissions were reduced in the UOM treatment, this was not large enough to increase available N and significantly affect crop yields.

Yield-scaled NH3 emissions were significantly increased by 280.5-453.7% by using urea compared to no N, and the increase was greater at higher N application rates (Huang et al., 2016). Yield-scaled N2O emissions varied from 0.06-0.11 kg N t-1 grain on the CU treatment, which is within the range (0.06-0.35 kg N t-1) found across the NCP (Lv et al., 2020). Yield-scaled emissions of NH3 and N2O from the OU treatment were reduced compared to the CU treatment, indicating that optimizing N applications mitigate yield-scaled Nr emissions (Sha et al., 2021). In addition, the combination of organic N and urea (but without Limus®) significantly reduced yield-scaled NH3 and N2O emissions by 24.5-54.9% and 67.7-78.5% relative to the CU treatment because of (1) the reduction of Nr emissions and (2) the increase in crop yields. Limus® reduced the yield-scaled NH3 emissions relative to the UOM treatment.

Nitrogen budgets were calculated to assess the most efficient measures for mitigating gaseous Nr losses and ensuring crop productivity (Eerdt and Fong, 1998). Fertilizer N is usually considered to be the main source of N to crops (Wang et al., 2017). In our experiment the contributions of fertilizer to N inputs were 34.8% and 49.6% in maize and wheat seasons, respectively (Table 3). Zheng et al. (2015) reported that approximately half of applied N fertilizer was taken up by crops. In our experiment, residual soil mineral NO3--N available at the beginning of the season (or released later in the season due to further mineralization) was also a large source of N, especially in the maize season, indicating that N applications can often be reduced while still meeting crop needs. Generally, soil residual NO3--N was often affected by precipitation (soil NO3--N decreased by 10 mm with precipitation of 2-3 mm) (Ma et al., 2021; Zhang et al., 1998). During the 2020 maize season, the amount of precipitation reached 398.1 mm and one rainfall event, in excess of 100 mm before harvest, could have increased N leaching to depth, consistent with the finding that soil NO3--N leached to about 1 m when precipitation was 394 mm in the wheat season (Lv et al., 1998). This could also explain why soil residual NO3--N content changed so much during the 2020 maize season, compared to previous seasons.

Large N applications caused a large N surplus, especially in conventional practice (CU), ranging from 240-333 and 250-411 kg N ha-1 in the maize and wheat seasons, respectively. This is within the range (-18~505 kg N ha-1 and -134~538 kg N ha-1) found in China’s smallholder-dominated cereal production systems (Zhang et al., 2021a). Besides much of this surplus N being lost through NH3 emissions, much of it was unaccounted for, possibly lost by N leaching of the large amounts of residual NO3--N in soil after harvest. Reducing the N application rate, coupled with the replacement of urea-N by organic-N and using Limus® will increase NUE, reduce Nr losses and so ensure sustainable N management and environmental protection.

4.3. Sustainability analysis of N management practices

Considering the sustainability and acceptability of the three tested N management improvements, reducing the application rate reduced Nr emissions and improved NUE (see also Halvorson et al., 2005; Liu et al., 2022b). It will also save input costs and so might be expected to increase net economic income as in Xia et al. (2017b). However, in our experiment, the often small reductions in yield compared to traditional farmer’s practice sometimes reduced net economic benefit, outweighing the cost saving, as also found by Zhang et al. (2021b). This was probably due to a limited N supply (120 and 180 kg N ha-1 applied to maize and wheat, respectively) since the optimal N amounts for these crops have been shown to be 146-180 and 208-230 kg N ha-1 for maize and wheat, respectively (Liu et al., 2021; Liu et al., 2016b). Therefore, reducing N applications is not necessarily acceptable to farmers and so ineffective for agricultural development. The partial replacement of urea by an organic N source (UOM) has been found to be effective in previous research (Cai et al., 2019) and was again in our experiment, reducing Nr emissions and increasing yield and NUE, offsetting the negative effect of simple N-reduction. Although the commercial organic fertilizer used here increased N input costs, it was more likely to increase yield and net economic benefit. The third approach tested, the use of the urease inhibitor Limus®, was confirmed to be effective for maintaining crop yield, economic benefit and so promoting the ‘green development of agriculture’ (Wang et al., 2022), It also showed the best potential for reducing Nr losses and effective N utilization. Compared to the UOM treatment, N input costs were increased by using Limus® but yield and net economic benefit also improved. This is consistent with other results showing that using Limus® was beneficial in a wheat-maize rotation (Sha et al., 2020b). Furthermore, the environmental benefits of reducing agricultural NH3 emissions in China far exceed the abatement costs (Zhang et al., 2020), and the mitigation of Nr emissions using optimized N practices could deliver large environmental benefits (Cheng et al., 2021; Gu et al., 2021).

**5. Conclusions**

Partly substituting urea with organic fertilizer and calculating the optimal N application rate was effective in sustaining yields, reducing Nr emissions (and yield-scaled emissions) and improving NUE and net economic benefit in this wheat-maize rotation in the NCP. NH3 emissions and yield-scaled NH3 emissions were further significantly reduced, and NUE and net economic benefit increased by using the urease inhibitor Limus®, indicating that these practices can ensure crop yields and minimize gaseous Nr emissions and the environmental impacts of maize-wheat rotations in the NCP. However, applying urea with Limus® did not reduce N2O emissions, so further research is needed to explore the influence of Limus® on N2O emission mitigation. We used a commercial organic fertilizer, but the NCP is an area of poorly managed, intense livestock production, resulting in large Nr emissions. It would be preferable and cheaper to recycle manures and other ‘wastes’ from livestock production than to purchase commercial organic fertilizer. Therefore a combination of farming with optimized and enhanced (with an inhibitor) N fertilizer and the recycling of manures and ‘wastes’ from animal husbandry should be the focus of the future research; i.e. linked and balanced crop and livestock production. In the wider sense of achieving “carbon neutral” production, and with agriculture systems being a significant source of greenhouse gas emissions, it is necessary to explore the effects of the above on CO2 and CH4 emissions in future research.

**Credit author statement**

**Xuejun Liu:** Methodology, Supervision, Writing - Review & Editing, Founding acquisition. **Jingxia Wang:** Measurement, Formal analysis, Writing – Original Draft. **Zhipeng Sha:** Validation, Writing - Review & Editing. **Jinrui Zhang:** Measurement. **Jiahui Kang:** Validation. **Wen Xu:** Writing - Review & Editing. **Keith Goulding:** Writing - Review & Editing.

**Acknowledgments**

This work was funded by the National Natural Science Foundation of China (41425007), the Xinlianxin Company supported project “Effect of Xinlianxin efficient nitrogen fertilizer on yield and efficiency increase and ammonia emission reduction in cropland of Northern China” and the High-level Team Project of China Agricultural University.

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