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Fate of ^{15}N-labelled urea when applied to long-term fertilized soils of varying fertility

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Abstract: Quantifying the fate of nitrogen (N) fertilizer is essential to develop more sustainable agricultural N management practices. However, our understanding of N losses, particularly in low fertility soils remains incomplete. We evaluated the fate and N use efficiency of N fertilizer under different long-term fertilization regimes, i.e., no N; synthetic N; manure plus synthetic N in a calcareous Cambisol in the North China Plain. A standard rate (160 kg N ha^{-1}) of ^{15}N-labelled urea was applied to the above treatments in summer maize (first crop) and the same amount of unlabelled urea was applied to winter wheat (second crop). We found the manure plus synthetic N treatment had a significantly higher fertilizer N use efficiency (56%) with lower residual fertilizer N in soil (54 kg N ha^{-1}) than the synthetic N treatment (46% and 69 kg N ha^{-1}, respectively), due to the better synchrony of fertilizer N supply and crop demand in the manure plus synthetic N treatment. Surprisingly, compared with the synthetic N treatment, application of N fertilizer to the N-deficient treatment increased fertilizer N use efficiency significantly to 68%, and reduced the residual fertilizer N in soil (37 kg N ha^{-1}). Fertilizer N losses accounted for 7%-12% of applied ^{15}N-labelled urea with no significant differences between treatments. We found that fertilizer N use efficiency was increased in the high fertility soil supplied with manure compared with the low fertility soil supplied with synthetic N fertilizer, which emphasized the importance of recycling the manure or crop residues to soil.

Keywords: long-term fertilization, soil fertility, fate of ^{15}N-labelled urea, N use efficiency

1. Introduction

Nitrogen (N) is of critical importance in boosting crop yields and ensuring food security. To sustain a growing global population, synthetic N fertilizer use has increased from 11 Tg N $year^{-1}$ in 1961 to 109 Tg N $year^{-1}$ in 2018 (FAO 2020). However, current management of N fertilizers for crop production is unsustainable, especially in some developing countries, where it leads to environmental degradation and climate change at the regional and global scale (Bouwman et al. 2013a; Springmann et al. 2018; Yu et al. 2019). Sustainable N management has become one of the major challenges of the 21st century (Zhang et al. 2020). However, to improve N management it is necessary to measure and monitor the fate of N and its impact, so that agricultural practices can be adjusted to achieve better N use efficiency (Zhang et al. 2020).

The fate of fertilizer N in croplands includes crop uptake, residual N remaining in soil and N loss to the environment (Ju and Christie 2011; Sebilo et al. 2013), which is strongly related to field management, climatic conditions and soil properties (Dourado-Neto et al. 2010; Gardner and Drinkwater 2009; Quan et al. 2020). The fate of fertilizer N is known to be affected by N application rates (Ju et al. 2009; Powlson et al. 1986; Stevens et al. 2005a), application methods (Wu et al. 2017; Yao et al. 2018), and timing of application (López-Bellido et al. 2005; Shi et al. 2012; Wang et al. 2016). These studies have shown that applying optimal rates of N fertilizer at the right time and place can achieve high N use efficiency and low fertilizer N losses. Other management activities including the use of straw and irrigation can also directly affect the fate of N fertilizer (Gao et al. 2017; Jia et al. 2011; Quan et al. 2018). However, previous studies have generally focused on the effects of field management and climate on the fate of N fertilizer, but have not taken into account pre-existing gradients in fertility generated by long-term fertilization regimes, especially in regions where soil fertility is low and there is a need to build soil fertility to increase crop productivity.

To achieve high target yields without depleting soil fertility, external resources including fertilizer and manure need to be applied to soil to replenish the soil nutrients removed in agricultural production (Ju and Christie 2011; Yan et al. 2020). Long-term application of N fertilizers or organic amendments (e.g., manure and straw) could increase SOC due to increased root biomass, exudates and shoot biomass returned to the soil (Hirte et al. 2018; Zhu et al. 2016). The application of organic amendments increases SOC to a much greater extent than fertilizer alone, this is because they contain large amounts of organic matter (Maillard and Angers 2014; Powlson et al. 2012). The increased SOC resulting from long-term application of organic amendments generally goes along with the increases in soil total nitrogen (TN) and microbial biomass (Lazcano et al. 2013; Rothamsted Research 2018). Since SOC, TN, microbial biomass and other soil properties play an important role in regulating microbial abundance and activity (Murphy et al. 2011; Serna-Chavez et al. 2013) and therefore soil N transformations (Booth et al. 2005; Li et al. 2019; Li et al. 2020), the long-term application of N fertilizer, especially manure or other organic fertilizers, could greatly increase mineralization-immobilization turnover (MIT) of N in soil. Gross nitrification rates are also stimulated by long-term fertilization of synthetic and organic fertilizers (Dai et al. 2017; Wang et al. 2015).

When N fertilizer is applied to soils with variable background levels of soil fertility caused by different historic fertilization regimes, fertilizer N use efficiency and losses can exhibit very different patterns (Liang et al., 2013). Studying the fate of fertilizer N in soils with variable fertility can provide the scientific basis for improving N use efficiency and mitigating N pollution (Zhang et al. 2020). Soils in the North China Plain (NCP) (mainly calcareous Cambisols) are developed from alluvial sediments of the Yellow River (Li et al. 2017) and are characterized by low fertility due to poor soil structure and low organic matter content (Du et al. 2018). Topsoil SOC concentrations (0-20 cm) of only 0.9% have been reported in the 2010s, which is much lower than that in other intensive agricultural regions of China (Liang et al. 2019; Song et al. 2020). Under these conditions of low fertility, conventional farming practice only achieve a fertilizer N recovery in the crop of 27% with 30% of fertilizer N remaining in the soil and 47% lost to the environment (Ju and Zhang 2017).

Since the 1980s, a growing body of research in the NCP has focused on using organic manure or straw return to increase soil fertility (Huang et al. 2017; Xin et al. 2016). However, few studies have investigated the fate of N fertilizer in calcareous Cambisols with a varying background fertility. We hypothesize that higher soil fertility will increase fertilizer N use efficiency while decreasing fertilizer N losses. We used a long-term field experiment with varying fertilization regimes established in 2006 in the NCP (Huang et al. 2013). The objectives were: 1) to evaluate the effects of long-term fertilization regimes on soil properties; 2) investigate N use efficiency and the fate of fertilizer N under varying levels of background of soil fertility.

2. Materials and methods

2.1 Site description

The study site was located in the Shangzhuang experimental station of the China Agricultural University (40°08.4’N, 116°10.7’E, 50 m above the sea level) in suburban Beijing, China. The site has a soil-climate-crop system that is representative of the NCP, and a temperate monsoon climate, with a mean annual air temperature of 13 °C and mean annual rainfall of 540 mm (1981-2015), in which 60-70% of the rain falls between June and August. The soil in this study was a calcareous Cambisol (fluvo-aquic soil according to Chinese soil genetic classification). The top 20 cm of the soil was sampled when the experiment was established in September 2006 to determine baseline soil properties. At that time, the soil had a particle size distribution of 28% of clay, 32% of silt, and 40% of sand, a bulk density of 1.31 g cm^{-3}, a pH of 8.1 (measured in water, 0.1:2.5 of soil: water), an organic carbon content of 7.1 g kg^{-1}, a total N concentration of 0.8 g kg^{-1}, an ammonium N concentration of 1.2 mg kg^{-1}, a nitrate N concentration of 24.5 mg kg^{-1}, an Olsen-P concentration of 7.8 mg kg^{-1} and an available K concentration of 76.2 mg kg^{-1} (Huang et al. 2013). The crop rotation was a winter wheat-summer maize double rotation, where winter wheat is sown in early October and harvested in the middle of June of the following year, and summer maize is sown immediately after the harvest of winter wheat and harvested in early October of the same year.

2.2 Treatments and managements in the long-term field experiment

The long-term field experiment was initiated in October 2006. Eight treatments were established with four N rates [zero N (N_0), optimum N (N_{opt}), conventional N (N_{con}) and balanced N (N_{bal}+M)] and two straw managements (straw removal and return). The N_0 treatments received no N fertilizer. N rates for crops in the N_{opt} treatments were determined on the basis of soil mineral N tests and the target crop N demand (Cui et al. 2010). The fertilizer N (in the form of urea) application rate in the N_{opt} treatments was 86 to 181 kg N ha^{-1} (average 140 kg N ha^{-1}) for winter wheat and 30 to 212 kg N ha^{-1} (average 124 kg N ha^{-1}) for summer maize from 2006-2017. In the N_{con} treatments, winter wheat received urea at a rate of 300 kg N ha^{-1} and summer maize received 260 kg N ha^{-1}, which is the typical N rate used by local farmers. N rates for N_{bal} treatments were calculated on the basis of a simplified N balance approach and equal to crop N uptake and target soil residual mineral N after harvest minus soil initial mineral N before sowing. N fertilizer used in N_{bal}+M treatments was cattle manure plus urea. Decomposed cattle manure (equivalent to 30 t ha^{-1} of fresh weight) was applied annually before the sowing of winter wheat. We assumed 40% and 20% of total N in manure was available N in the winter wheat and summer maize growing seasons (Qiu et al. 2012), and the gap between available manure N and balanced N rates was filled by synthetic N (urea). The synthetic N application rate in the N_{bal}+M treatments was 0 to 128 kg N ha^{-1} (average 60 kg N ha^{-1}) for winter wheat and 30 to 188 kg N ha^{-1} (average 124 kg N ha^{-1}) for summer maize from 2006-2017. A detailed description of N management in the long-term field experiment can also be found in previous papers (Huang et al.

2013; Huang et al. 2017; Qiu et al. 2012; Song et al. 2019). A randomized complete block design was used with three replications. Each plot was 64 m^2 (8 m × 8 m). Seven of the above treatments were selected for this study, which included treatments with straw removal (N_0, N_{opt}, N_{con}) and straw return (N_0+S, N_{opt}+S, N_{con}+S, N_{bal}+M+S). These treatments can be classified into three categories: (1) no N (NN), (2) synthetic N (SN), (3) manure plus synthetic N (MSN).

All of the seven treatments received the same amount of synthetic phosphorus (P) and potassium (K) fertilizer: 160 kg P_2O_5 ha^{-1} yr^{-1} and 90 kg K_2O ha^{-1} yr^{-1} from 2006 to 2013, 200 kg P_2O_5 ha^{-1} yr^{-1} and 200 kg K_2O ha^{-1} yr^{-1} after 2013. The timing and rates of irrigation for winter wheat were determined on the basis of soil moisture measurements, and irrigation was provided three to five times annually at rates ranging from 28-70 mm on each occasion between 2006-2017. The summer maize was rain-fed and was only irrigated when dry weather occurred at emergence. At the sowing of winter wheat, deep plowing (20 cm) was carried out to incorporate the fertilizer and maize straw (in the straw return treatment). There was no tillage after harvest of the winter wheat, and the wheat straw was shredded and left on the soil surface.

After several years' cultivation, there was a diversity in soil properties in the seven treatments.. The detailed soil properties were measured by Huang et al. (2017) in 2010, Yang et al. (2017) in 2012 and Song et al. (2019) in 2015. These studies generally showed that the MSN treatment had significantly higher SOC and TN than the SN treatments, then followed by the NN treatments. We determined soil properties of the seven treatments in October 2017 (Section 3.1).

2.3 ^{15}N microplots setting up and managements

All the treatments selected for this study (including the zero N treatments) received an application of 160 kg ^{15}N ha^{-1} $season^{-1}$. After the harvest of winter wheat in June 2017, ^{15}N microplots

($1.2 \times 0.8 = 0.96$ m^2) were established in the main plots of the seven treatments. Square tubing made from galvanized sheet iron was inserted into the soil to a depth of 0.40 m, with 0.10 m remaining above the soil surface. The microplots were used for soil and plant sampling of the first crop (summer maize) and the second crop (winter wheat). All ^{15}N microplots received same field management of fertilization, irrigation, straw, tillage, etc.

In the summer maize, ^{15}N labelled urea (5.15 atom%) was band applied at the four-leaf and ten-leaf stages at a rate of 80 kg N ha^{-1} in each application. Four furrows (width, 6 cm) were made by a hoe to a depth of 10 cm over 30 cm within each microplot. ^{15}N labelled urea was dissolved in 0.2 L of pure water and evenly sprayed at the bottom of the four furrows and was then covered by soil. For winter wheat, unlabelled urea was applied as a basal fertilizer at sowing and topdressing at the regreening stage at a rate of 80 kg N ha^{-1} in each application. The basal urea was mixed with 2 kg of fresh sieved soil (2 mm) from each ^{15}N microplot and then spread by hand uniformly. The 0-20 cm soil layer was then distributed using a shovel to achieve a uniform application of ^{15}N fertilizer. A topdressing of urea was dissolved in 1 L of pure water and sprayed uniformly over the soil surface using a hand pressure sprayer, then the ^{15}N microplots were irrigated with 60 mm water (same as the irrigation rate in the main plots) using a watering can.

All plots received 100 kg ha^{-1} of P_2O_5 and 100 kg ha^{-1} of K_2O and applied at the four-leaf stage in the summer maize and sowing in winter wheat together with the N fertilizer. Summer maize received an additional 30 kg ha^{-1} of $ZnSO_4$ at the four-leaf stage when applying the N fertilizer. The P fertilizer was calcium superphosphate, K fertilizer was potassium sulfate, Zn fertilizer was zinc sulfate. No irrigation was provided to the summer maize in 2017. Winter wheat was irrigated with 28 mm at emergence and 60 mm at tillering, regreen, tassel and filling stages, thus the total irrigation rate for winter wheat was 268 mm. Before sowing, all straw in the microplots was removed, no-tillage was used for summer maize and deep plowing (by shovel) was carried out in winter wheat.

2.4 Plant and soil analyses

At maturity, all of the aboveground plants in the ^{15}N microplots were harvested and separated into grain and straw. They were weighed separately for calculating biomass, and straw was manually shredded. Then a subsample of grain and straw was weighed immediately and again after drying at 70 °C to measure water content. They were then ground to pass through a 0.15 mm sieve and the N content and ^{15}N abundance were measured using an isotope ratio mass spectrometer (Europa Scientific Integra, Crewe, UK).

After harvest, six (summer maize) or three (winter wheat) soil cores were collected to a depth of 100 cm using a 3.0 cm diameter soil auger. Each soil core was separated into 20 cm intervals, and then mixed to form a composite sample for each layer. Soil samples were passed through a 2 mm sieve. A sub sample was then extracted with 2 mol L^{-1} KCl using a 5:1 mass ratio of KCl solution to soil. The NH_4^+-N and NO_3^--N concentrations in the extracts were analyzed by an AA3 continuous-flow analyzer (BranCLuebbe GmbH, Norderstedt, Germany). NH_4^+-N and NO_3^--N in the extracts were further separated to determine ^{15}N abundance using the micro-diffusion method (Brooks et al. 1989). The rest of the soil was dried at room temperature and used for measurement of total N content and ^{15}N abundance using an isotope ratio mass spectrometer (Europa Scientific Integra, Crewe, UK).

Surface soils (0-20 cm) were sampled from main plots of the seven treatments in October 2017, to analyze physical and chemical properties. For each plot, ten cores were taken from 0-20 cm using a 3.0 cm diameter soil auger, and then mixed to form a composite sample. After collection, soils were immediately sieved (2 mm) and dried at room temperature. SOC was analyzed by wet-digestion with H_2SO_4-$K_2Cr_2O_7$, and TN was determined by semi-micro Kjeldahl digestion followed by distillation and titration. The available P content was determined by sodium bicarbonate extraction followed by colorimetric analysis. Available K was determined by ammonium acetate extraction and flame atomic absorption spectrophotometry. Soil pH was determined in a 1: 2.5 soil/water mixture using a pH meter (S220 Seven Compact, Mettler Toledo, Switzerland). Cation exchange capacity (CEC) was determined by the ammonium acetate saturation method. Bulk density (BD) was determined by the cutting ring method in September 2018. A detailed description of these measurement methods can be found in Lu (2000).

2.5 Calculations and statistical analysis

The percentage of crop N and soil N derived from ^{15}N fertilizer was calculated by:

(1) $Ndff_{crop\ (soil)}$ (%) = $(c-b) / (a-b) \times 100$

Where c, a, b is the ^{15}N abundance of crop (soil), fertilizer (5.15%) and natural abundance (0.3663%), respectively. The ^{15}N abundance of plant and soils in the unfertilized treatment was not measured in the present study. We assumed that the use of atmospheric natural abundance (0.3663%) would not introduce a large bias for the calculation of N recovery and is acceptable when comparing the fate of ^{15}N-labelled fertilizers, which is commonly used in similar studies.

Crop N uptake, residual fertilizer N in soil and fertilizer N losses were calculated as follows:

(2) Total N uptake (kg N ha^{-1}) = biomass (kg ha^{-1}) × crop N content (g N kg^{-1}) / 1000

(3) Fertilizer N uptake (kg N ha^{-1}) = $Ndff_{crop}$ (%) × total N uptake (kg N ha^{-1}) / 100

(4) Soil N uptake (kg N ha^{-1}) = total N uptake (kg N ha^{-1}) − fertilizer N uptake (kg N ha^{-1})

(5) Residual fertilizer N in soil (kg N ha^{-1}) = $Ndff_{soil}$ (%) × bulk density (g cm^{-3}) × thickness (cm) × soil N content (g N kg^{-1})

(6) Fertilizer N losses (kg N ha^{-1}) = N rate (kg N ha^{-1}) − fertilizer N uptake (kg N ha^{-1}) − residual fertilizer N in soil (kg N ha^{-1})

Nitrogen use efficiency, soil N balance, soil surface N surplus and soil nitrate accumulation were calculated as follows:

(7) Fertilizer N use efficiency (%) = fertilizer N uptake (kg N ha^{-1}) / N rate (kg N ha^{-1}) × 100

(8) Partial factor productivity from applied N (kg kg^{-1}) = Grain yield (Mg ha^{-1}) / N rate (kg N ha^{-1}) × 1000

(9) Physiological efficiency of N use (kg kg^{-1}) = total biomass (kg ha^{-1}) / total N uptake (kg ha^{-1})

(10) Soil N balance (kg N ha^{-1}) = Residual fertilizer N in soil (kg N ha^{-1}) + N from atmospheric deposition (kg N ha^{-1}) + biological N fixation − soil N uptake (kg N ha^{-1}) (Ju and Christie 2011)

(11) Soil surface N surplus (Soil apparent N surplus, kg N ha^{-1}) = N input (kg N ha^{-1}) − total N uptake (kg N ha^{-1}), N input include N from fertilizer, atmospheric deposition and biological fixation.

(12) Soil nitrate accumulation (kg N ha^{-1}) = bulk density (g cm^{-3}) × thickness (cm) × soil nitrate concentration (mg N kg^{-1}) / 10 (Yang et al., 2020)

Atmospheric N deposition data was obtained from Yin et al. (2017), who reported that N deposition was 35 kg N ha^{-1} during maize season (from June to September) in the NCP. Estimates of non-symbiotic N fixation for wheat and maize (5 kg N ha^{-1}) was obtained from Bouwman et al. (2013b).

Statistical analyses including stepwise regression and ANOVA were performed by the SPSS 20.0 (IBM Corp., Armonk, NY, USA). The differences between treatments in biomass, grain yield, N uptake, N use efficiency, fertilizer N fates and other parameters were determined by the Duncan test at the 0.05 probability level ($p < 0.05$). Graphs were produced with Sigmaplot 14.0.

3. Results

3.1 Changes in soil properties under the long-term field experiment

Long-term application of manure plus synthetic N (MSN treatment) significantly increased soil organic carbon (SOC), total nitrogen (TN), available P and K concentrations but also significantly reduced soil bulk density (BD), in comparison with long-term application of synthetic N alone (SN treatments) and no N application (NN treatments). The MSN treatment had a significantly lower pH but higher CEC than the NN treatments. Where straw was returned, the SN treatments tended to have a higher SOC and TN than the NN treatments, but significant differences were only found in TN between N_0+S and N_{opt}+S treatment. The SN treatments had significantly lower available P than the NN treatments, and there were no significant differences in available K, pH, CEC and BD between the SN and the NN treatments. Under the same N rates, straw incorporation increased SOC and TN, but significant differences were only observed between N_{opt} and N_{opt}+S treatment. There were no significant differences in the C/N ratio of any of the treatments which were in the range of 7.4-8.7 (Table 1).

3.2 Crop biomass and N uptake of the first crop

In the first crop of summer maize, straw, grain and aboveground biomass in the NN treatments were comparable to those in the MSN treatment. In the NN treatments and MSN treatment, straw biomass was 7.92-9.84 t ha^{-1} (average 8.74 t ha^{-1}) and grain biomass was 9.58-10.19 t ha^{-1} (average 9.87 t ha^{-1}). In the SN treatments, straw biomass was 5.85-7.36 t ha^{-1} (average 6.57 t ha^{-1}) and grain biomass was 6.34-8.45 t ha^{-1} (average 7.47 t ha^{-1}), which were lower than those in the NN treatments and MSN treatment (Fig. 1a).

Crop N uptake by maize in the various treatments followed a similar trend to biomass. Aboveground N uptake in the MSN treatment (229 kg N ha^{-1}) was comparable to that in the N_0+S treatment (198 kg N ha^{-1}), but was significantly higher than that in the N_0 treatment (183 kg N ha^{-1}). Aboveground N uptake in the SN treatments was much lower than that in the NN treatments and MSN treatment, which was in the range of 136-168 kg N ha^{-1}, with an average of 151 kg N ha^{-1} (Fig. 1b).

Fertilizer N uptake by maize in the NN treatments was 100-104 kg N ha^{-1}, with an average of 102 kg N ha^{-1}, which was significantly higher than that in the SN treatments and MSN treatment. The MSN treatment had a fertilizer N uptake of 78 kg N ha^{-1}, which was higher than that in the SN treatments (61-69 kg N ha^{-1}, average 66 kg N ha^{-1}), but significant differences were only found when compared with N_{con}+S treatment (Fig. 1c). The proportion of crop aboveground N derived from fertilizer (NDFF) in the NN treatments were 53-55% (average 54%), but in N_{opt}, N_{opt}+S, N_{con} and N_{con}+S treatment, they were 48, 41, 49 and 40%, respectively. NDFF in the MSN treatment was 34%, which was the lowest among all treatments (Fig. 1d).

3.3 The fate and use efficiency of ^{15}N-labelled urea in the first crop

As all the treatments received same amount of N fertilizer, fertilizer use efficiency (RE_N) showed a similar trend to fertilizer N uptake. RE_N in the NN treatments were 63%-65% (average 64%) and was significantly higher than that in the SN treatments and MSN treatment. The MSN treatment had a RE_N of 49%, which was higher than that in the SN treatments (38%-43%, average 41%), but significant differences were only found in the N_{con}+S treatment (Fig. 2a). Partial factor productivity from applied N (PFP_N) in the NN treatments and MSN treatment was comparable and in the range of

60-64 kg kg^{-1} (average 62 kg kg^{-1}), PFP_N in the N_{opt}+S treatment was 53 kg kg^{-1} and there were no significant differences between the N_{opt}+S, NN and MSN treatments. PFP_N in the N_{opt}, N_{con} and N_{con}+S treatments was 40-48 kg kg^{-1} (average 45 kg kg^{-1}) and significantly lower than that in the NN and MSN treatments (Fig. 2b). No significant differences in physiological efficiency of N use (PE_N) were observed between the NN and SN treatments (89-97 kg kg^{-1}, average 94 kg kg^{-1}). The N_{opt} treatment had a significantly higher PE_N than that in the MSN treatment (85 kg kg^{-1}) (Fig. 2c).

There was 46-47 kg N ha^{-1} residual fertilizer N in the NN treatments, and this accounted for 29% of applied N fertilizer, which was significantly lower than in the SN and MSN treatments. Residual fertilizer N in the SN treatments was 78-92 kg N ha^{-1} (average 85 kg N ha^{-1}) and accounted for 49-58% (average 53%) of applied ^{15}N fertilizer. Residual fertilizer N in the MSN treatment was 69 kg N ha^{-1} and accounted for 43% of applied ^{15}N fertilizer (Fig. 3). Fertilizer N losses across all treatments ranged from 7-14 kg N ha^{-1} (average 10 kg N ha^{-1}) and accounted for 4%-9% (average 6%) of applied fertilizer N. There were no significant differences in fertilizer N loss between treatments (Fig. 3).

3.4 Crop biomass, N uptake and the fate of ^{15}N-labelled urea in the second crop

In the second crop of winter wheat, we found that wheat biomass in different soils exhibited a similar trend to summer maize (first crop). In the NN and MSN treatments, straw biomass was 4.07-4.50 t ha^{-1} (average 4.29 t ha^{-1}) and grain biomass was 5.49-5.71 t ha^{-1} (average 5.62 t ha^{-1}). In the SN treatments, straw biomass was 2.50-3.16 t ha^{-1} (average 2.82 t ha^{-1}) and grain biomass was 3.77-4.22 t ha^{-1} (average 3.94 t ha^{-1}), which was significantly lower than those in the NN treatments and MSN treatment (Fig. 4a). Aboveground N uptake in the NN treatments and MSN treatment was comparable and in the range of 134-143 kg N ha^{-1}, and significantly higher than the aboveground N uptake in the SN treatments, which was 94-105 kg N ha^{-1} (average 100 kg N ha^{-1}) (Fig. 4b).

In the NN and SN treatments, 6 to 9 kg N ha^{-1} (average 7 kg N ha^{-1}) of the applied ^{15}N fertilizer was taken up by winter wheat. Winter wheat in the MSN treatment recovered more ^{15}N fertilizer (11 kg N ha^{-1}) than the NN and SN treatments. Taking into account the first crop, fertilizer N use efficiency in the NN, SN and MSN treatments was 67%-69% (average 68%), 43%-49% (average 46%) and 56%, respectively. After the harvest of winter wheat, residual fertilizer N in the NN treatments was 36-39 kg N ha^{-1} (average 37 kg N ha^{-1}) and accounted for 23%-24% (average 23%) of applied ^{15}N fertilizer, which was significantly lower than that in the SN treatments and MSN treatment. Residual fertilizer N in the SN treatments was 64-73 kg N ha^{-1} (average 69 kg N ha^{-1}) and accounted for 40%-46% (average 43%) of applied ^{15}N fertilizer. Residual fertilizer N in the MSN treatment was 54 kg N ha^{-1} and accounted for 34% of applied N fertilizer. Fertilizer N losses across all treatments were 3-11 kg N ha^{-1} (average 6 kg N ha^{-1}) and accounted for 2%-7% (average 4%) of applied fertilizer N. There were no significant differences in fertilizer N losses between treatments (Table 2).

3.5 Distribution of residual ^{15}N-labelled urea and nitrate in the soil profile

After the harvest of the summer maize, residual ^{15}N-labelled urea in the soil decreased with increasing soil depth. Almost all residual ^{15}N-labelled urea was in the 0-60 cm soil layer which accounted for 93%-96% of residual ^{15}N (Fig. S1a). After the harvest of winter wheat, ^{15}N in the 0-40 cm soil layer had decreased while ^{15}N below 40 cm soil layer was increased, which indicated that the residual ^{15}N-labelled urea in the first crop had moved down into the deeper soil layers during the second crop season (Fig. S1b). The details of distribution of nitrate in the soil profile can be found in the supporting text and Figure S1.

The form of residual ^{15}N-labelled urea in soil after harvest was analyzed. We found that the proportion of residual soil ^{15}N in the organic or inorganic form in winter wheat exhibited a similar trend to summer maize (Fig. 5). In the NN treatments, organic ^{15}N account for 76-91% (average 85%) of the total residual ^{15}N. Organic ^{15}N in the MSN treatment accounted for 53-56% (average 55%) of the total residual ^{15}N. ^{15}N in the SN treatments was mainly in the inorganic form, which accounted for 57-79% (average 68%) of the total residual ^{15}N (Fig. 5).

The amount of residual ^{15}N in the inorganic form increased with the total residual ^{15}N in soil ($R^2>0.93$, $p<0.01$) (Fig. S2a). There was a positive linear regression between the proportion of residual ^{15}N in the inorganic form and the amount of total residual ^{15}N in the soil ($R^2>0.85$, $p<0.01$). As there is strong nitrification in these calcareous Cambisols (Ju and Zhang 2017), the soil inorganic N was mainly in the NO_3^--N form, which accounted for 93-97% (average 95%) of inorganic N in the SN treatments and MSN treatment, and 70-90% (average 80%) of inorganic N in the NN treatments. The above results indicate that the more residual ^{15}N that was in the soil, the more ^{15}N (both in terms of the amount and proportion) will enter the soil NO_3^--N pool (Fig. S2b).

3.6 N budgets in the fertilizer-soil-crop continuum

The soil N balance in the NN treatments was close to zero (-7 to 3 kg N ha^{-1}) however, the SN treatments had a higher soil N balance than the NN treatments and MSN treatment, which was 25-54 kg N ha^{-1}. The soil N balance in the MSN treatment (-42 kg N ha^{-1}) was the lowest of all the treatments. The soil surface N balance in the NN treatments was 2-17 kg N ha^{-1} in the SN treatments it was 32-64 kg N ha^{-1}, and the MSN treatments had the lowest soil N balance (-29 kg N ha^{-1}; Table 3).

4. Discussion

4.1 Soil properties

4.1 Soil fertility alter fertilizer N use efficiency

When same amount of external synthetic N fertilizer was applied to the three categories of historic fertility management (no nitrogen, synthetic nitrogen and manure plus synthetic nitrogen), grain yield in the MSN treatment with high soil fertility was higher than that in the SN treatments, with the MSN treatment providing a far higher soil nitrogen supply (Fig. 6). These results are likely to be related to higher mineralization-immobilization turnover (MIT) in the MSN treatment, and the better synchrony of N supply and crop demand. Thus, the fertilizer N use efficiency in the MSN treatment was higher (49%) than that in the SN treatments (41%). Surprisingly, we found grain yields in the NN treatments were substantially increased when they received the standard amount of synthetic N fertilizer and were comparable with that in the MSN treatment and higher than those in the SN treatments. This led to a much higher N use efficiency in the NN treatments (64%, Fig. 6). A similar study was conducted in a 19-year field experiment to investigate the fate of ^{15}N fertilizer (165 kg urea-N ha^{-1}) in three contrasting fertilizer treatments (no fertilizer; inorganic NPK fertilizers and manure plus inorganic NPK fertilizers) but showed different results. Wheat grain yield in the manure plus inorganic NPK fertilizers treatment was significantly higher than the inorganic NPK fertilizers treatment, and both were higher than the no fertilizer treatment (Liang et al. 2013).

There might be several reasons for the high grain yield and N use efficiency after the addition of N fertilizer to the N-deficient soil (NN treatments). It has been shown that a balanced supply of N, P, and K can increase crop yield (Wang et al. 2010; Yousaf et al. 2017). The N-deficient soil in the present study had a relatively high available P and K supply (Table 1) due to the annual application of P and K fertilizer and low P and K uptake by crops caused by an unbalanced nutrient supply. When synthetic N was applied to the N-deficient soil, the soil N, P and K were balanced thus increasing the grain yield. In the study of Liang et al. (2013), the N-deficient soil (long-term no fertilizer treatment) received no P and K and the Olsen-P in the no fertilizer soil was only 1.4 mg P kg^{-1}. This imbalance in N, P and K supply led to the low grain yield. In the present study, the available P and K were sufficient for crop growth, although the available P concentrations in the NN treatments (46-52 mg kg^{-1}) were significantly higher than those in the SN (24-35 mg kg^{-1}). The available P levels in the SN treatments were high enough to achieve the target yield, since the critical levels of soil Olsen-P required for high crop yields range from 11-21 mg kg^{-1} across different agro-ecological regions in China (Bai et al. 2013). We assume another reason for the high grain yield in the NN treatments was related to the substantial growth of lateral roots which increased the crop's ability to capture fertilizer N. In a split-root experiment in which half of the plant root was grown in uniform low N soil (equivalent to NN in this study) and half of the root was grown in uniform high N soil (equivalent to fertilization zone in NN in this study), it was found that the lateral root growth is suppressed in the low-N compartment but enhanced in the high-N compartment (Oldroyd and Leyser 2020). This root response is thought to be regulated by the shoot and a number of signaling processes.

Taken together, crops in the MSN treatment with high soil fertility had a higher fertilizer N uptake and use efficiency, and also had a larger soil N pool and N availability, thus increased crop N uptake from both fertilizer and soil and achieved the highest grain yield. Although the NN treatments had a small soil N pool and low N availability, the addition of fertilizer N largely increased crop N uptake from fertilizer. Crop N uptake from soil and fertilizer in the SN treatments was relatively low thus led a lower grain yield than the NN and MSN treatments. Another reason for the lower grain yield in the SN treatments may have resulted from the lack of nutrients such as P and K (Fig. 6).

4.2 Influence of soil fertility on fate of ^{15}N fertilizer

Residual fertilizer N in the soil after crop harvest was most abundant in the SN treatments, followed by the MSN treatment and then the NN treatments. This was opposite to the order of fertilizer N uptake. Fertilizer N losses in all the treatments were very low and no significant differences were found. It is generally accepted that ammonia volatilization and nitrate leaching are the main pathways of N loss in these calcareous Cambisols (Ju et al. 2009; Ju and Zhang 2017). However, in the present study, urea was band applied at a depth of 10 cm which is enough to achieve a negligible ammonia volatilization (Rochette et al. 2013). Since almost all of the residual N occurred in the 0-60 cm soil layer (Fig. S1a), we deduced that fertilizer N losses via nitrate leaching in this study were very low. This was due to fertilizer N being applied at the optimized rate which is a key strategy for reducing N leaching loss in these calcareous Cambisols (Ju and Zhang 2017).

It has been reported that the form of residual ^{15}N in soil is highly related to the fertilizer N rate. Stevens et al. (2005b) found that residual ^{15}N in the soil was mainly in the organic form when N was applied at no more than the optimum rate. Also the proportion of organic residual ^{15}N was reduced from 97% to 64% when N applications increased from 67 to 268 kg N ha^{-1} (Stevens et al. 2005b). However, in the present study, even under the optimum N rate, the proportion of organic residual ^{15}N in soil was only 32% in the SN treatments. This is probably because the results of Stevens et al. (2005b) were obtained from a high carbon soil, and it is generally accepted that inorganic N is easily converted to organic N where concentrations of SOC are high (Dai et al. 2017). The calcareous Cambisols used in the present study are characterized by low SOC which has a weak N immobilization potential (Ju and Zhang 2017), thus leading to a low proportion of organic residual ^{15}N. In contrast to the SN treatments, the long-term application of manure significantly increased

SOC in the MSN treatment and further increased the proportion of organic ^{15}N in the soil to 55%. We found that the high total residual ^{15}N in the soil led to a large amount and proportion of residual ^{15}N in the inorganic form (Fig. 5), which increased the risk of nitrate accumulation and leaching (Huang et al. 2017; Ju and Zhang 2017). These results indicate the importance of recycling manure and optimizing N rates in low SOC soils to reduce fertilizer N losses.

Surprisingly, we found the NN treatments with lowest SOC had the highest proportion of organic ^{15}N (85%) across all treatments. This was probably due to the high fertilizer N uptake by the crop which led to much lower residual ^{15}N-labelled urea in soil, which could be largely immobilized by the SOC pool. Therefore, soil properties, the amount of residual ^{15}N in the soil and the crop's ability to capture fertilizer N are all important factors for affecting the form and amount of residual fertilizer N in soil.

4.3 Effect of soil fertility on soil N balance and soil surface N surplus

The soil N balance is a proxy for the soil N surplus or deficit, which indicates whether soil N uptake by crops can be replenished by the external N supply (e.g., residual fertilizer N, atmospheric N deposition and biological N fixation) (Ju and Christie 2011). A negative value for the soil N balance indicates a risk of depleting soil fertility while a positive value for the soil N balance indicates a risk to the environment. The NN treatments roughly maintained the N balance but had the potential to mine soil N (soil N balance of -7 to 3 kg N ha^{-1}). The SN treatments had a high soil N balance (25-54 kg N ha^{-1}) due to the low crop N uptake from soil. If crop yield and N balance in the SN treatments were maintained at the current level, over the long run, the surplus N will eventually result in losses to the environment. The soil N balance in the MSN treatment was negative (-42 kg N ha^{-1}) which was probably due to the high crop yield and N uptake from soil. The fertilizer N application rate in the present study was uniformly set to 160 kg N ha^{-1} per crop season for all treatments, but to maintain the high target yield and achieve a balanced soil N supply, an additional 42 kg N ha^{-1} of N should be applied to the MSN treatment.

The soil surface N surplus allows an evaluation of N management strategies in cropping systems without the need for laborious ^{15}N field experiments (Zhang et al. 2019). The soil surface N surplus is generally regarded as an indicator of N losses. N surplus benchmarks could be regarded as the allowable maximum N loss within a given cropping system (Ju and Gu 2017; Zhang et al. 2019). If the N surplus is higher than a benchmark, N release to the environment will be expected to be unacceptable. On the contrary, there will be a risk of soil N mining where the balance falls below zero (EU Nitrogen Expert Panel 2015). At the regional scale, it has been proposed that the N surplus benchmark for summer maize in the North China Plain should be 80 kg N ha^{-1} (Ju and Gu 2017; Zhang et al. 2019). The N surpluses in the NN, SN and MSN in this study were 10, 49 and -29 kg N ha^{-1}, respectively; all lower than the proposed N surplus benchmark. But this does not mean all treatments were mining soil N. The 80 kg N ha^{-1} of the N benchmark is a realistic target to improve the N management of current conventional practices with a high N surplus, and could be further reduced by the improvements in fertilization techniques and agronomic management in China (Zhang et al. 2019). All the treatments in the present study received the optimum N management, for example, 160 kg N ha^{-1} of fertilizer N was a reasonable N rate for summer maize and winter wheat in the study region (Ju and Gu 2014). Fertilizer was applied at the right time and deeply incorporated by ploughing or irrigation (Huang et al. 2013; Huang et al. 2017; Qiu et al. 2012; Song et al. 2019), and thus the N losses were very low (Fig. 3). Overall, the NN treatment had the potential of mining soil N, while SN had risks of high N losses and MSN was likely to mine soil N when 160 kg N ha^{-1} of fertilizer N was applied with current yields. To maintain the high target yields, an additional 53 kg N ha^{-1} of N should be applied to the MSN treatment, which is close to the additional N required for the high yield and soil N balance calculated by soil N balance approach.

5. Conclusions

This study was able to clearly demonstrate that the combined application of manures and synthetic N fertilizer was able to deliver higher soil fertility, grain yield and crop N uptake, and achieved a higher fertilizer N use efficiency than N fertilizer applied alone. The combination of manure and fertilizer also increased the proportion of soil residual N in the organic form, thus decreased fertilizer N losses. These results highlight the importance of recycling manure or crop residues in soil, especially in the regions with low SOC. Surprisingly, when applying N fertilizer to soils with a history of no N application, the grain yield and aboveground N uptake were increased more significantly and to higher levels than those in soils that had a history of synthetic N application. Fertilizer N use efficiency in these previously unfertilized soils was the highest of all treatments. The proportion of residual N in the organic form in previously unfertilized soils was far higher than that in soils that had received annual inputs of synthetic N. Further research from the perspective of plant physiology and root biology is needed to explore the mechanism responsible for the high fertilizer use efficiency in the N-deficient soil.

Declaration of Competing Interest

The authors report no declarations of interest.

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Table captions

Table 1 Soil physical and chemical properties in 2017 after 11-year fertilizations [a]

Table 2 Fate of residual fertilizer N in winter wheat (kg N ha^{-1}) [a]

Table 3 Soil N balances and soil surface N surpluses in summer maize (kg N ha^{-1})

Table 1 Soil physical and chemical properties in 2017 after 11-year fertilizations [a]

Treatments	pH	Organic C ($g\ kg^{-1}$)	Total N ($g\ kg^{-1}$)	C: N	Available P ($mg\ kg^{-1}$)	Available K ($mg\ kg^{-1}$)	CEC [b] ($cmol\ kg^{-1}$)	Bulk density ($g\ cm^{-3}$)
N_0	8.1±0.1 a	6.8±0.3 b	0.84±0.07 d	8.1±0.9 a	46±4 b	87±5 b	12.7±0.3 b	1.48±0.01 a
N_0+S	8.1±0.1 a	7.3±0.5 b	0.87±0.05 cd	8.4±0.4 a	52±7 b	88±6 b	12.6±0.2 b	1.48±0.01 a
N_{opt}	8.0±0.0 a	6.9±0.4 b	0.83±0.03 d	8.3±0.4 a	27±6 c	82±7 b	12.9±0.2 b	1.48±0.00 a
N_{opt}+S	7.9±0.1 ab	8.2±0.7 b	1.12±0.04 ab	7.4±1.1 a	35±3 bc	84±0 b	13.0±0.2 ab	1.44±0.01 ab
N_{con}	7.9±0.1 ab	7.4±0.4 b	0.88±0.10 cd	8.7±1.1 a	29±2 c	81±1 b	13.0±0.1 ab	1.49±0.01 a
N_{con}+S	7.7±0.1 b	8.4±0.6 b	1.07±0.03 bc	7.9±0.7 a	24±2 c	89±5 ab	13.2±0.4 ab	1.46±0.04 ab
N_{bal}+M+S	7.7±0.1 b	10.9±0.7 a	1.28±0.09 a	8.6±0.2 a	72±8 a	105±7 a	13.7±0.3 a	1.41±0.02 c

[a] Numbers were expressed as mean ± standard error (n=3), means followed by the same letter are not significantly different at $p<0.05$). [b] CEC represents cation exchange capacity

Table 2 Fate of residual fertilizer N in winter wheat (kg N ha^{-1}) [a]

Treatments	Residual fertilizer N in soil following the first crop	Crop uptake in the second crop	Residual fertilizer N in soil following the second crop	Losses in the second crop
N_0	46 ± 7 c	7 ± 2 b	36 ± 4 c	3 ± 3 a
N_0+S	47 ± 3 c	6 ± 1 b	39 ± 1 c	3 ± 2 a
N_{opt}	85 ± 6 ab	8 ± 1 b	71 ± 6 a	7 ± 0 a
N_{opt}+S	84 ± 3 ab	9 ± 1 ab	69 ± 2 ab	6 ± 4 a
N_{con}	78 ± 6 ab	7 ± 0 b	64 ± 7 ab	7 ± 5 a
N_{con}+S	92 ± 6 a	8 ± 1 ab	73 ± 5 a	11 ± 2 a
N_{bal}+M+S	69 ± 8 b	11 ± 1 a	54 ± 6 b	4 ± 3 a

[a] Number represents mean ± standard error (n=3), means followed by the same letter are not significantly different ($p<0.05$).

Table 3 Soil N balances and soil surface N surpluses in summer maize (kg N ha^{-1})

Soil	Fertilizer N	N deposition	Biological N fixation	Crop aboveground N uptake	Crop aboveground N uptake from soil	Residual fertilizer N in soil	Soil N balance	Soil surface N surplus
N_0	160	35	5	183	83	46	3	17
N_0+S	160	35	5	198	94	47	-7	2
N_{opt}	160	35	5	136	71	85	54	64
N_{opt}+S	160	35	5	168	99	84	25	32
N_{con}	160	35	5	144	74	78	44	56
N_{con}+S	160	35	5	154	92	92	40	46
N_{bal}+M+S	160	35	5	229	151	69	-42	-29

Figure captions

Fig. 1 Grain and straw biomass (a), grain and straw N uptake (b), aboveground N derived from fertilizer and soil (c) and the proportion of aboveground N uptake derived from fertilizer and soil (d) in summer maize. Error bars represent standard errors (n=3), lowercase letters compared the parameters of the corresponding legend among treatments, uppercase letters compared the parameters of the corresponding whole column among treatments, the different lowercase or uppercase letters indicate significant differences ($p<0.05$) between treatments.

Fig. 2 Fertilizer N use efficiency (a), partial factor productivity from applied N (b) and physiological efficiency of N use (c) in summer maize. Error bars represent standard errors (n=3), the different lowercase letters indicate significant differences ($p<0.05$) between treatments.

Fig. 3 Fate of fertilizer N in summer maize. Error bars represent standard errors (n=3), the different lowercase letters indicate significant differences ($p<0.05$) between treatments.

Fig. 4 Grain and straw biomass (a), grain and straw N uptake (b) in winter wheat. Error bars represent standard errors (n=3), lowercase letters compared the parameters of the corresponding legend among treatments, uppercase letters compared the parameters of the corresponding whole column among treatments, the different lowercase or uppercase letters indicate significant differences ($p<0.05$) between treatments.

Fig. 5 Proportion of soil residual ^{15}N in the organic and inorganic form after the harvest of summer maize (a) and winter wheat (b). Error bars represent standard errors (n=3). Different lowercase letters indicate significant differences ($p<0.05$) between treatments.

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Fig. 6 N flows in the fertilizer-soil-crop-environment continuum under the three clustered-background fertility of soils, Data were derived from the maize season in 2017, the data of the NN treatments is the average value of the N_0 and N_0+S treatment, the SN treatments is the average value of N_{opt}, N_{opt}+S, N_{con} and N_{con}+S treatment, the MSN treatment is the value of N_{bal}+M+S treatment. Except for grain yield (Y in the Fig. 6), soil organic carbon (SOC in the Fig. 6) and soil total nitrogen (TN in the Fig. 6), the unit for the numbers in the Fig. 6 are kg N ha^{-1}. N_{fer}、N_{fer1}、N_{fer2}、N_{loss} and N_{soil} represent fertilizer N rate, fertilizer N uptake by crop, residual fertilizer N in soil, fertilizer N losses to the environment, crop N uptake from soil. N_{other} represent N from atmospheric deposition and biological N fixation.

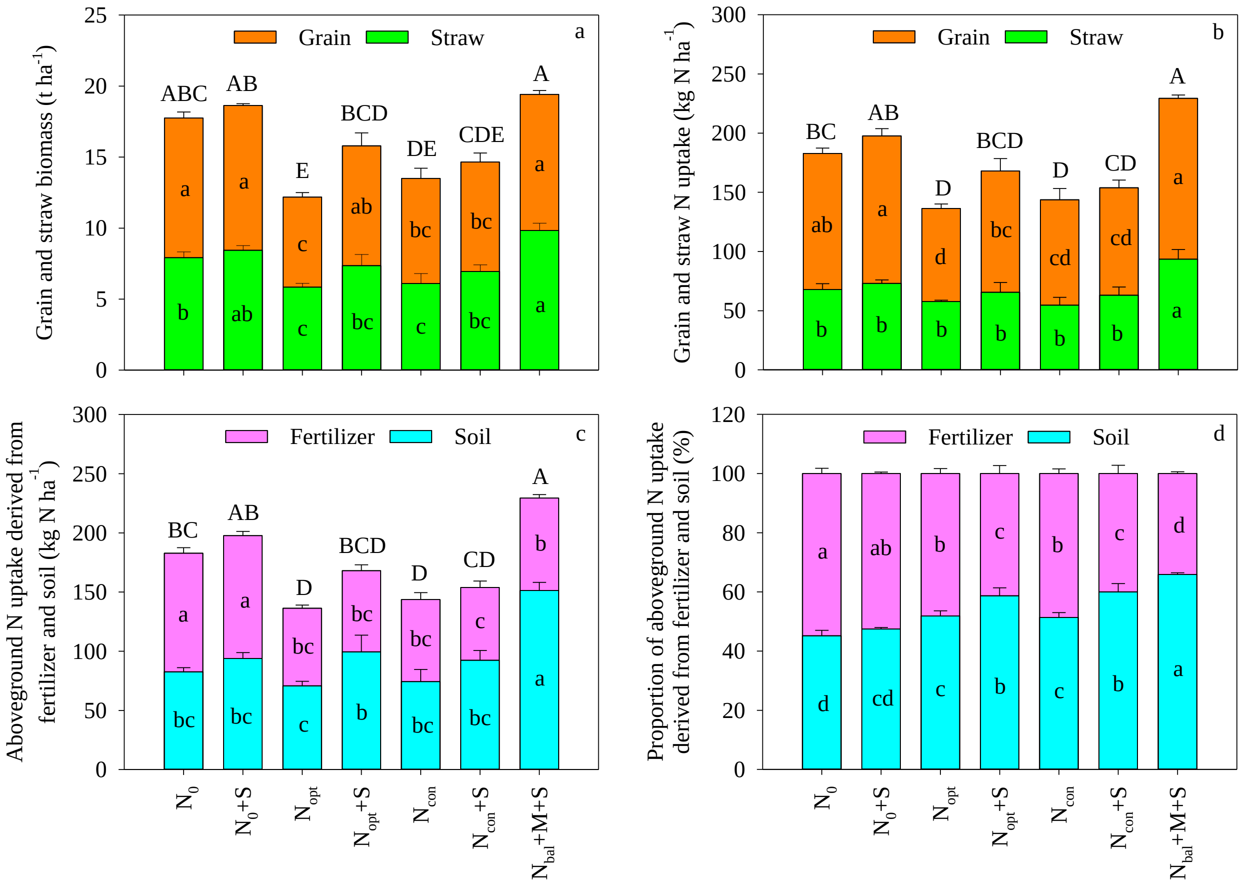

Fig. 1 Grain and straw biomass (a), grain and straw N uptake (b), aboveground N derived from fertilizer and soil (c) and the proportion of aboveground N uptake derived from fertilizer and soil (d) in summer maize. Error bars represent standard errors (n=3), lowercase letters compared the parameters of the corresponding legend among treatments, uppercase letters compared the parameters of the corresponding whole column among treatments, the different lowercase or uppercase letters indicate significant differences ($p<0.05$) between treatments.

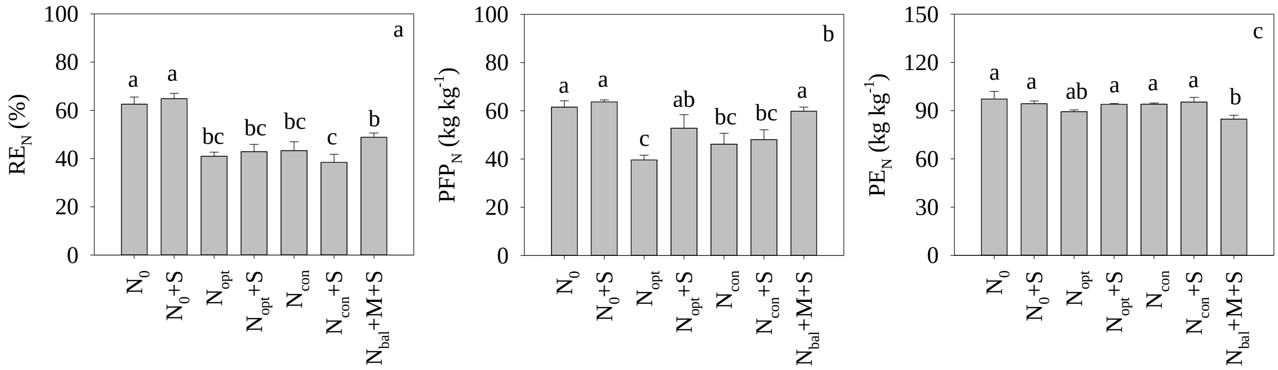

Fig. 2 Fertilizer N use efficiency (a), partial factor productivity from applied N (b) and physiological efficiency of N use (c) in summer maize. Error bars represent standard errors (n=3), the different lowercase letters indicate significant differences ($p<0.05$) between treatments.

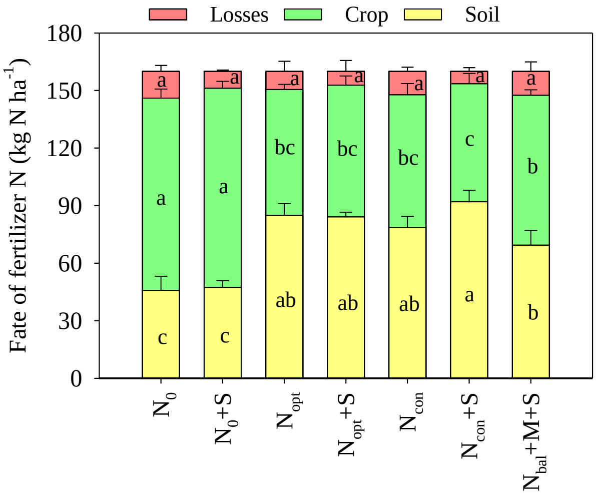

Fig. 3 Fate of fertilizer N in summer maize. Error bars represent standard errors (n=3), the different lowercase letters indicate significant differences (p<0.05) between treatments.

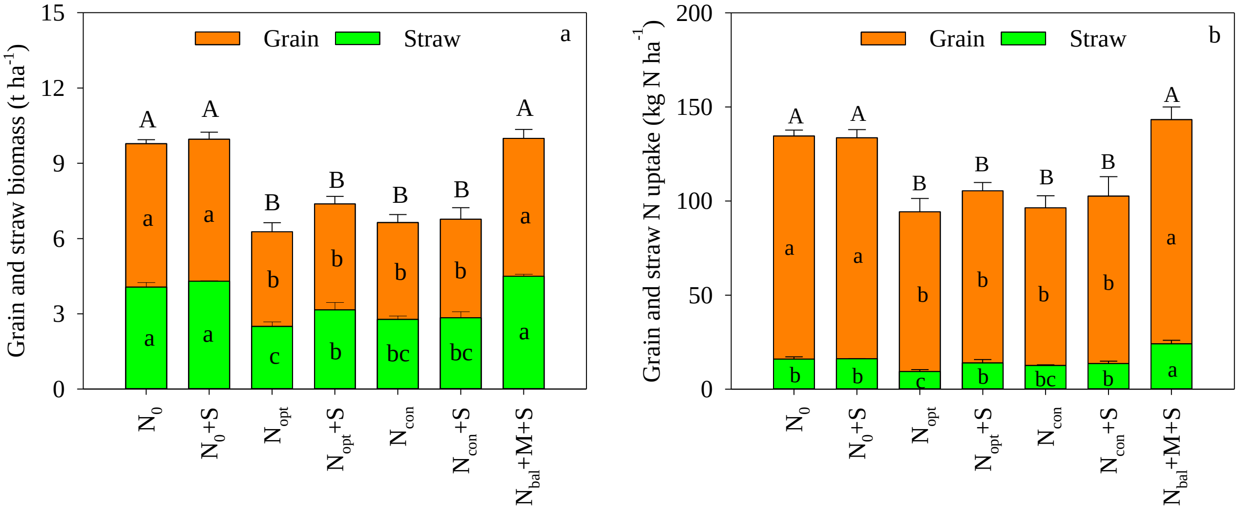

Fig. 4 Grain and straw biomass (a), grain and straw N uptake (b) in winter wheat. Error bars represent standard errors (n=3), lowercase letters compared the parameters of the corresponding legend among treatments, uppercase letters compared the parameters of the corresponding whole column among treatments, the different lowercase or uppercase letters indicate significant differences ($p<0.05$) between treatments.

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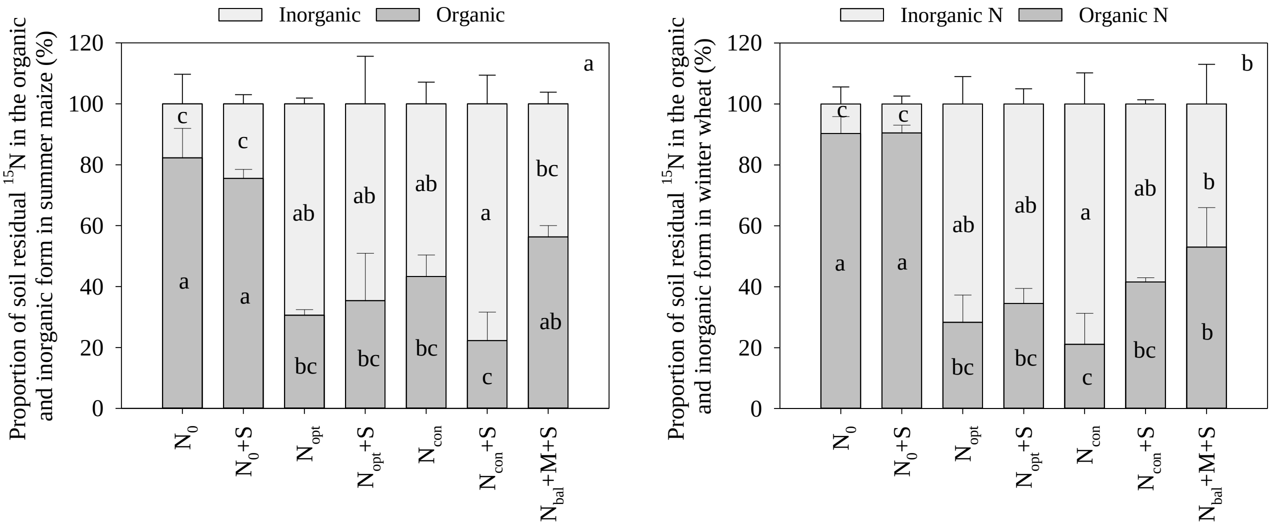

741 **Fig. 5** Proportion of soil residual ^{15}N in the organic and inorganic form after the harvest of summer maize (a) and
742 winter wheat (b). Error bars represent standard errors (n=3). Different lowercase letters indicate significant
743 differences (p<0.05) between treatments.

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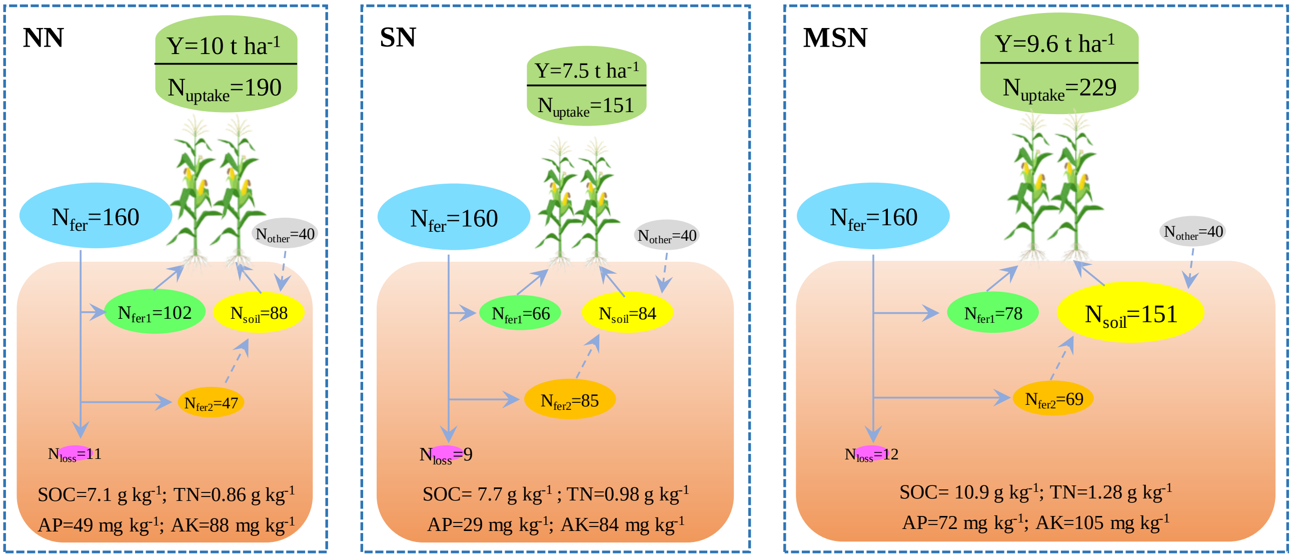

Fig. 6 N flows in the fertilizer-soil-crop-environment continuum under the three clustered-background fertility of soils, Data were derived from the maize season in 2017, the data of the NN treatments is the average value of the N_0 and N_0+S treatment, the SN treatments is the average value of N_{opt}, N_{opt}+S, N_{con} and N_{con}+S treatment, the MSN treatment is the value of N_{bal}+M+S treatment. Except for grain yield (Y in the Fig.6), soil organic carbon (SOC in the Fig.6), soil total nitrogen (TN in the Fig.6), available phosphorus (AP in the Fig. 6) and available potassium (AK in the Fig. 6), the unit for the numbers in the Fig.6 are kg N ha^{-1}. N_{fer}、N_{fer1}、N_{fer2}、N_{loss} and N_{soil} represent fertilizer N rate, fertilizer N uptake by crop, residual fertilizer N in soil, fertilizer N losses to the environment, crop N uptake from soil. N_{other} represent N from atmospheric deposition and biological N fixation.