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ORIGINAL ARTICLE

Higher maize yields and lower ammonia emissions by replacing synthetic nitrogen fertiliser with manure in the North China plain

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Abstract Ammonia (NH_3) emitted from synthetic nitrogen (N) fertiliser applications to farmland leads to air pollution and terrestrial acidification. Previous studies have indicated that replacing urea-based N fertilisers by manure may reduce NH_3 emissions and enhance crop yield. However, the long-term effects of replacing urea N fertiliser by manure on crop yield and NH_3 emissions have not been sufficiently quantified. The objective of the field study presented here was to examine the effects of three treatments (i.e., 100% synthetic fertiliser N application (NPK); 50% synthetic fertiliser N +50% manure N application (50%MNS); and 100% manure N application (100%MNS)) on NH_3 emissions and maize grain yields over a 3-year period in a long term (13 years) study site in the North China Plain. Results showed that the NH_3 emissions in the NPK treatment ranged from 9.7 to 11.7 kg ha^{-1} during the three maize growing seasons. Replacement of urea fertiliser by manure significantly decreased the NH_3 emissions by 22–54% in the 50% MNS treatment and by 47–71% in the 100%MNS treatment. Maize grain yields were 14–30% higher in the 50%MNS treatment and 17–45% higher in the 100% MNS treatment, compared to the NPK treatment (8.1–8.8 t ha^{-1}). The NH_3 emission factor for the NPK treatment ranged from 4.1 to 4.8%. Additional ^{15}N labelling work established that 83% of NH_3 emissions originated from the urea fertiliser in the NPK treatment. Manure treatments had a higher maize N uptake and soil organic matter content, and a lower soil pH than the NPK treatment. This study highlights that replacing synthetic N fertiliser with manure on farmland in the North China Plain has long-term beneficial effects on maize yield and N uptake, NH_3 emission mitigation and soil carbon storage.

Keywords Agriculture · Environment · Farmland · Nitrogen · ^{15}N stable isotope

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Introduction

Ammonia (NH_3) emission is one of the dominant pathways of nitrogen (N) loss from farmland in the world. These emissions contribute to air pollution (Gu et al. 2021), soil acidification and water

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eutrophication (Wang et al. 2018; Ti et al. 2019). Several studies have indicated that NH_3 emissions from cropland are notably large in China, because of the large cropland areas and the large use of urea-based N fertilisers. Total NH_3 emissions from farmland in China have been estimated at 3.5–5.8 million t NH_3 yr^{-1}, and NH_3 emission factors for synthetic N fertilisers may range from 4 to 13% (Fu et al. 2020; Zhang et al. 2021). Specifically, three crops (maize, rice and vegetables) account for almost two thirds of the total NH_3 emissions from cropland in China (Wang et al. 2021). The relatively large NH_3 emissions and the relatively high fertiliser N application lead to a low fertiliser N use efficiency and to environmental degradation (Webb et al. 2010; Sha et al. 2021).

The North China Plain produces the food of nearly a half billion people. Winter wheat, summer maize, along with horticulture (vegetables and orchards) are the dominant crops (Zhang et al. 2015). Ammonia emissions and nitrate leaching are the predominant N loss pathways from cropland in the North China Plain (Wan et al. 2021). Emissions of NH_3 from fertilised cropland are affected by many factors, such as crop type, fertiliser type and application rate (Guo et al. 2021), air and soil temperature (Abdo et al. 2021; Anthony and Silver 2021) and soil physical and chemical properties (e.g. bulk density, water moisture content and pH) (Chen et al. 2015). Previous studies have shown that specific management regimes, including the use of controlled-release fertilisers, urease inhibitors, deep placement of fertiliser, and combined applications of organic manure and synthetic N fertiliser can be successful in mitigating NH_3 emissions (Hayashi et al. 2011; Ti et al. 2019; Guo et al. 2021). The success of these specific management regimes strongly depends on soil and environmental (weather) conditions and management (Zhang et al. 2018; Guo et al. 2020; Zhang et al. 2021).

Lowering the N application to the level of the N demand by the crop and replacing urea-based N fertilisers by manure are likely highly effective strategies in the North China Plain. The mechanisms identified in reducing NH_3 emissions following the replacement of urea-based N fertiliser by manure are: (1) a relatively low NH_3 emission potential of composted manure (Xu et al. 2021); (2) an improvement of soil quality, through organic matter and other nutrient additions, which increases the N uptake by the crop (Sha et al. 2019); (3) a reduction of NH_3 emissions during the topdressing period due to a lower urea N fertiliser application (Tang et al. 2021), and (4) a slight decrease of the pH of alkaline soils through manure application (Abdo et al. 2021). However, there are also reports indicating that NH_3 emissions increased and crop yields decreased following the replacement of synthetic N fertiliser by manure; this has been attributed to (1) a high NH_3 emission from animal manure, which led to insufficient N supply, and to a decreased N uptake by the crop (Seufert et al. 2012), and (2) an increase of the soil pH of acidic soils, which may increase NH_3 emissions (Zhang et al. 2021). A recent study by Rahaman et al. (2020) found that NH_3 emissions from manure are less sensitive to air temperature fluctuations than emissions from urea fertiliser.

Evidently, many factors influence the changes in NH_3 emissions following the (partial) substitution of synthetic N fertiliser by animal manure. Most of these factors are well-known, but the long-term effects are less known. Long-term manure application may also affect soil structure and soil micro-biological activity, which may influence the capacity of N retention by microbial communities and thereby indirectly NH_3 emission (Gai et al. 2018; Zhang et al. 2020). It has been suggested also that especially the results of long-term field experiments may convince growers to implement improved management practices (Snyder et al. 2009; Sanz-Cobena et al. 2017).

The main objective of our study was to increase the understanding of the long-term effects of replacing synthetic fertilisers by animal manure on NH_3 emissions and crop yields. The specific objectives were (1) to estimate the NH_3 emission factors and crop yield effects of single applications of synthetic fertiliser and animal manure to a maize cropping system in the North China Plain; (2) to measure the NH_3 emissions and crop yield effects following a partial replacement (by 50%) of synthetic fertiliser by manure, and (3) to estimate how changes in soil parameters following long-term manure application affect NH_3 emissions and crop yields. The measurements were conducted over a period of 3 years in a long-term field experiment (during the 11th, 12th and 13th experimental year). Also, a ^{15}N labelling technique was used to better understand the fate and transformation of N fertiliser in soil and the source of NH_3 emissions following long-term N applications.

Our main hypothesis is that replacing synthetic fertiliser by manure decreases NH_3 emissions because of (i) omitting a topdressing; (ii) increased maize N uptake due to an improved soil fertility over time, and (iii) a slight decrease in soil pH following long-term manure application (Guo et al. 2020).

Materials and methods

Site description

The study site (40.22°N, 116.23°E, altitude 43.3 m) was located in the Beijing Changping Soil Quality National Field Science Observation Research Station, in the North China Plain. This location has a typical sub-humid temperate continental monsoon climate with warm and wet summers and cold and dry winters. The average annual temperature is 11.5 °C and the annual rainfall is 625 mm. More than 80% of the rainfall occurs from June to October. The soil is a Haplic Luvisol with a light loam texture (FAO classification), which is widely distributed in the North China Plain.

The long-term field experiment was set up in 2007. The soil physical and chemical properties of the 0-200 mm soil layer at the beginning of the experiment in 2007 were: soil bulk density = 1320 kg m^{-3}, pH (H_2O) = 8.3, soil organic carbon (SOC) = 7.9 g kg^{-1}, total nitrogen (TN) = 0.76 g kg^{-1}, extractable nitrate (NO_3^--N) = 0.89 mg kg^{-1}, and extractable ammonia (NH_4^+-N) = 0.15 mg kg^{-1}. The mean daily air temperature during the three maize growing seasons (May to October) of 2017–2019 were 30.2, 31.5 and 29.7 °C, respectively (Fig. 1a), and the total rainfall plus irrigation water input were 526, 259 and 320 mm, respectively (Fig. 1a).

Experimental design and field management

Three treatments with equal total N application rates (240 kg N ha^{-1}) were laid out in a randomized block design and replicated three times. In each plot, there were micro-plots. The bottom and sides of the micro-plots were made of concrete and there was an isolated drainage outlet (Fig S1). The size of each micro-plot covered an area of 2.4 m^2 (1.2 m × 2 m). Treatments were: (1) fertilisation with synthetic nitrogen,

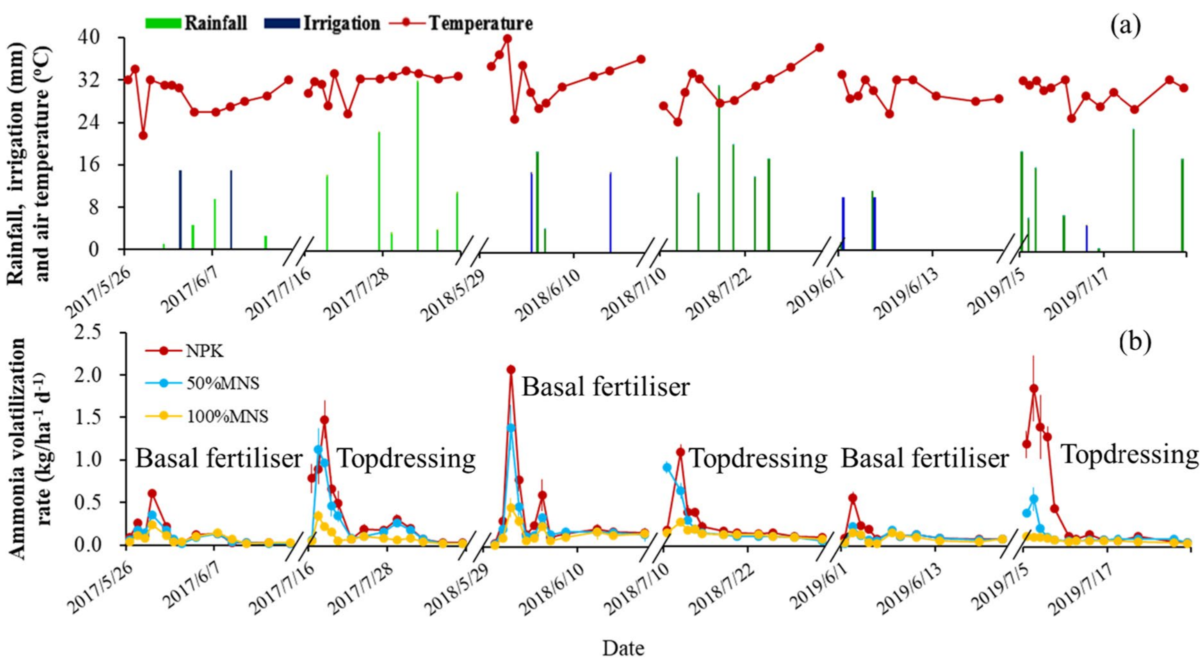

Fig. 1 Temporal variations of **(a)** rainfall, irrigation and air temperature and **(b)** ammonia emission rates during the three maize growing seasons of 2017–2019. Data were collected from day 1 to day 24 after basal fertiliser or topdressing fertiliser application. The error bars are standard deviations of ammonia emission (n = 3)

phosphorus and potassium (NPK); (2) 50% synthetic N + 50% manure N application (50%MNS) and (3) 100% manure N application (100%MNS). The synthetic N, P and K fertilisers were applied as urea (46% N), superphosphate and potassium chloride, respectively. For the NPK treatment, superphosphate and potassium chloride were applied at 52 kg P ha^{-1} and 149 kg K ha^{-1} on the same day of sowing. For the 50%MNS and 100%MNS treatments, the superphosphate and potassium chloride application were corrected for the amounts of P and K applied via manure. Thus, the total N, P, and K inputs were equal for all treatments. The manure was a partially dried and composted swine manure; it contained on average 51.7 g kg^{-1} of organic matter, 24.3 g kg^{-1} of total N, 15.7 g kg^{-1} of total P, 17.9 g kg^{-1} of total K, and 25% moisture, on a dry weight basis. The pH of the manure was 6.7.

Superphosphate, potassium chloride, and manure were applied once per year, as basal application, uniformly spread onto the soil surface and immediately incorporated into the soil (0–200 mm depth) by ploughing. Sowing of the maize was conducted on the same day. Urea fertiliser was applied for 35.5% as base application and for 64.5% as topdressing. The topdressing was applied when the maize had 6 leaves, i.e., on 15th July 2017; 9th July 2018 and 4th July 2019. The topdressings were spread evenly over the experimental fields.

Soil and plant sampling and measurement

At each harvest, three soil samples (0-200 mm) were collected from each plot by an auger and a composite soil sample was obtained per plot by mixing the soil samples thoroughly. In total, 27 composite soil samples were collected during 2017–2019 (3 years × 3 treatments × 3 replicates). Soil samples were gently ground and sieved over a 2-mm mesh sieve. A subsample of 150 g was used for the determination of exchangeable NH_4^+-N, NO_3^--N, microbial biomass carbon (MBC), microbial biomass nitrogen (MBN), activities of β-D-glucosidase (BG), N-acetylglucosaminidase (NAG) and the genes abundance of ammonia-Oxidizing Archaea (AOA) and Ammonia-Oxidizing Bacteria (AOB). Another subsample of 200 g was air-dried, ground and sieved over a 150-µm mesh sieve for the determination of TN, SOM and pH. Soil NO_3^--N and NH_4^+-N was determined in 1 mol L^{-1} KCl extracts with a continuous flow analyser (AA3, Seal, Germany). Soil pH was measured using a 1:2.5 ratio of soil to deionised water. Soil organic matter was determined using the potassium dichromate oxidation method (Skjemstad and Baldock 2007). Soil TN was ascertained using the Kjeldahl method (Thomas et al. 1967).

The MBC and MBN were analysed with the chloroform fumigation-extraction method (Vance et al. 1987), followed by analysis with a CN analyser. The activities of BG and NAG were measured by the method of Zhu et al. (2014). Total DNA was extracted from 0.5 g fresh soil using the Fast DNA® SPIN Kit for Soil following manufacturer's instructions. The AOA genes and transcripts were quantified by using the Arch-amo AF (5′-STAATGGTCTGGCTTAGACG-3′) and Arch-amo AR (5′-GCGGCCATCCATCTGTATGT-3′) primers for amplification. The AOB genes and transcripts were quantified by using the amoA-1 F (5′-GGGGTTTCTACTGGTGGT-3′) and amoA-2R (5′-CCCCTCKGSAAAGCCTTCTTC-3′) primers for amplification.

Above ground crop samples (separated into cornstalks and grains) from the three treatments were collected in the field and dried at the laboratory (at 105 °C for 30 min, and then at 75 °C until they reached a constant weight). The dry matter (DM) content of grains and cornstalks was determined; grain yield is evaluated at 16% water content in China. Maize grain yields were calculated as follows:

$$\text{Maize grain yields} = \text{dried maize grain yields}\,(\%)$$

In addition, N uptake by the crop was estimated from the N concentration in the dry matter and the mass of dry matter. Partial productivity of applied N (PFP_N, kg kg^{-1}) was estimated as follows:

$$PFP_N = \text{grain yield/total N input}$$

Measurement of NH_3 emissions

Fluxes of NH_3 were measured from each plot using a battery operated chamber system (Zhang et al. 2018). The chamber had a cylindrical shape and consisted of a plastic frame (250 mm inner diameter, 350 mm height) and a top chamber (260 mm inner diameter, 150 mm height) (Fig S2). The chamber was placed between maize rows before basal fertilisation. The bottom frame was pushed into the ground to a depth of 200 mm. During measurement days, water was added to the groove of the bottom frame. Then the top chamber was put on the bottom frame, in the groove with water, which sealed to top chamber to the frame. The top chamber had an air inlet and an air outlet. The inlet was connected to an impinger containing dilute sulfuric acid solution to eliminate traces of NH_3 from the air before being drawn into the chamber at a flow rate of 1.5 L min^{-1} for 2 h (Shang et al. 2014). The outlet of the chamber was also connected to an impinger, which contained 40 mL of 0.05 mol L^{-1} sulfuric acid solution. The trapped NH_3 was measured using an XY-2 sampler connected to a continuous-flow analyser, and the NH_3 fluxes were calculated based on the flow rate and collection time per unit area of soil. Ammonia emissions were recorded for 24 days following the basal fertilisation and following the topdressing. Ammonia emissions were measured once a day, between 9:00 and 11:00 am (Zhang et al. 2018). The average NH_3 flux observed during the 2 h measurement period was assumed to be representative for the mean daily flux. The chambers were covered by insulation materials during the sampling process to minimise temperature effects. All chamber systems were periodically calibrated before use in the field, by measuring the recovery of a spike of NH_3 produced by reacting to a known concentration of NH_4OH with NaOH under complete enclosure for 2 h. A 95% recovery of the NH_3 spike was considered acceptable.

Air and soil (0–100 mm) temperatures were recorded using temperature probes, and soil moisture levels were determined using a TDR (Spectrum Technologies, Inc., Item # 6440FS). Reading of the TDR were converted to soil moisture content following calibration.

The rate of NH_3 emission (R, kg d^{-1} ha^{-1}) was calculated by averaging the measured fluxes (Eq. 1) (Zhang et al. 2021):

$$R = \frac{M \times 10^{-2}}{\pi \times r^2} \times \frac{24}{t} \tag{1}$$

where M is the NH_3 emissions amounts during the collection (mg), t is the duration of collection (h), and r is the chamber radius (m). The cumulative NH_3 emission (kg N ha^{-1}) is the sum of the daily emission rates over the measurement period. The NH_3 emission factor was calculated as follows:

$$NH_3 \text{ emission factor} = \text{cumulative } NH_3 \text{emission/sum of N inputs.}$$

Analysis of ^{15}N enrichment

Labelled urea with a ^{15}N abundance of 20% (Shanghai Research Institute of Chemical Industry, Shanghai, China) was applied at the start of the growing seasons of 2017, in order to understand the fate and transformation of NH_3 from urea N application. The isotopic enrichments of NH_3 emitted from the plot with ^{15}N labelled fertiliser application, was compared with the isotopic enrichments of NH_3 emitted from the plot without ^{15}N labelled fertiliser application.

The atom% ^{15} N of the trapped NH_3 was measured using a Stable Isotope Ratio Mass Spectrometer (Isoprime100, UK). All operation procedures were conducted in sequence from lower to higher atom% ^{15}N to avoid cross-contamination.

The NH_3 emissions (kg N ha^{-1}) from the applied urea-N fertiliser was calculated using the following formula (Eq. 2):

$$N_{NH_3} = N_x \times \frac{(B - C)}{A} \tag{2}$$

where N_x represents the measured NH_3 emissions (kg N ha^{-1}); A represents the atom percent excess of the applied labelled fertiliser N, which is the difference between the ^{15}N abundance of urea (20%) and the natural ^{15}N abundance (0.3722%); B represents the ^{15}N abundance of NH_3-N treated with labelled fertiliser N; and C represents the ^{15}N abundance of NH_3-N that was treated without labelled fertiliser N.

Statistical analysis

Microsoft Excel 2016 and R software (ggplot2 package, R 3.6.3) were used to analyse the data and to create figures. Analysis of variance (ANOVA) was used to determine the statistical significance of the treatment effects in maize grain yields, NH_3 emissions and soil parameters. Multiple comparisons of mean values of maize grain yields, NH_3 emissions and soil parameters were performed using the Fisher's least significant difference (LSD, $P < 0.05$) among the different treatments. IBM SPSS 19.0 software was used for all statistical tests. With the aim to reveal the relationships among environmental factors (e.g., soil pH, SOM and crop N uptake) and NH_3 emissions in the NPK, 50%MNS and 100%MNS treatments, redundancy analysis (RDA) was performed using the vegan package in R 3.6.3. ANOVA was used to test the RDA model and adjusted R square was calculated to identify parameters that significantly explained the variance of NH_3 emissions.

Results

Soil properties

The differences in soil properties among treatments are the result of the cumulative impact of the treatment during the period 2007 to 2019. During the measurement period 2017–2019, the mean SOM content of the topsoil of the 50%MNS treatment was 8% higher and that of the 100%MNS treatment 9% higher than the SOM content of the NPK treatment (Fig. 2). In addition, soil TN, NO_3^--N and NH_4^+-N contents of the 50%MNS and 100%MNS treatments were also higher than those of the NPK treatment. It was noteworthy that soil NO_3^--N and NH_4^+-N in the 100%MNS treatment were statistically lower than those in the 50%MNS treatment (Fig. 2). Soil MBC was 34% higher and MBN 13% higher in the 50%MNS treatment compared to the NPK treatment. Soil MBC was 59% higher and MBN 29% higher in the 100%MNS treatment compared to the NPK treatment (Table S1). Correspondingly, the β-D-Glucosidase and N-acetylglucosidase were also significantly enhanced in the treatments with manure (Table S1). The 50%MNS and 100% MNS treatments decreased AOA gene abundance and increased AOB gene abundance (Table S1).

Ammonia emissions

Days 1 to 7 after N fertiliser application are commonly considered the period with relatively high NH_3 emissions, but these being influenced by temperature and rainfall. Indeed, days 1 to 7 after N fertiliser application accounted for 52 to 65% of the NH_3 emissions following basal fertilisation and for

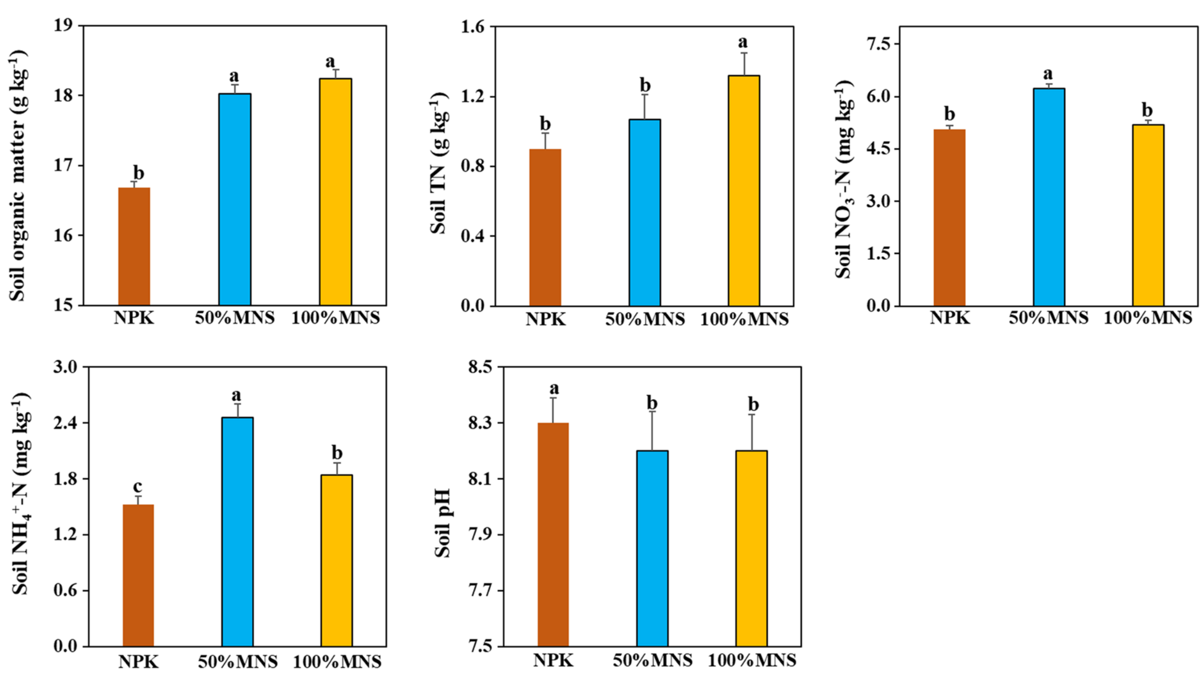

Fig. 2 Effect of manure substitution on the topsoil (0-200 mm) attributes. Data are the average values of three years (n = 9, 3 replicate plots × 3 years). Different small letter above the deviation bar represents significant difference among the treatments. Abbreviations: TN: total nitrogen

43–83% following topdressing in the NPK treatment (Table 1). As manure was applied as basal fertilisation, no significant NH_3 emissions occurred from the 100%MNS treatment during the topdressing period (Fig. 1b). The NH_3 emissions were larger during the topdressing periods than during the basal fertilisation period in 2017 and 2019. However, the opposite was true in 2018 (Fig. 1b), likely because of the differences in temperature and rainfall (Table 1).

Replacing urea N fertiliser by manure N decrease NH_3 emissions, especially in the 100%MNS treatment. Compared with the NPK treatment, the NH_3 emissions during the maize growing period of 2017–2019 decreased by 22 to 54% in the 50%MNS treatment and by 47 to 71% in the 100%MNS treatment (Table 2).

Results from the ^{15}N labelling work conducted in 2017 showed that the total NH_3 emission from the NPK treatment was 9.7 kg ha^{-1}. About 83% of the NH_3 emissions (8.1 kg ha^{-1}) originated from urea (Fig. 3). In the 50%MNS treatment, 3.4 kg ha^{-1} of the NH_3 emissions stemmed from the applied urea and 4.0 kg ha^{-1} from other N sources (i.e., manure N and soil/crop system) (Fig. 3). The N loss via NH_3 emissions accounted for 3.4% and 2.8% of the applied urea in the NPK and 50%MNS treatments, respectively.

Maize grain yields and N surplus

Maize grain yields in the NPK treatment ranged from 8.1 to 8.7 t ha^{-1} during the measurement period. Compared with the NPK treatment, maize grain yields were 14–30% higher in the 50%MNS treatment, and 17 to 46% higher in the 100%MNS treatments (Table 3). The partial productivity of applied N (PFP_N) was 17 to 50% higher in the 100%MNS treatment, and 14 to 30% higher in the 50%MNS treatment compared to the NPK treatment (Table 3). Two-way ANOVA analyses indicated that the maize grain yields and PFP_N were significantly affected by treatments but did not significantly depend on the year and on the interaction between treatment and year (Table 3).

The N surpluses of the 50%MNS treatment (−1 to −14 kg N ha^{-1}) and of the 100%MNS treatment (−25 to −48 kg N ha^{-1}) were lower than the N surpluses (6–42 kg N ha^{-1}) of the NPK treatment, because of the increased N uptake in the manure application treatments (Table 4).

Table 1 The NH_3 emissions (kg ha^{-1}) during days 1–7 after N fertiliser application and the corresponding mean air temperatures, soil water-filled pore space and soil temperatures during days 1–7

Year	Treatment	Basal fertilisation period					Topdressing period				
		NH_3 emissions during days 1–7 (kg N ha^{-1})	Proportion[‡] (%)	AT[§] (°C)	ST (°C)	SMC (%)	NH_3 emissions during days 1–7 (kg N ha^{-1})	Proportion (%)	AT (°C)	ST (°C)	SMC (%)
2017	NPK[†]	1.71	64.5%	32.1	24.1	16.4	4.65	65.6%	28.3	26.1	39.3
	50%MNS	1.23	60.0%				3.25	61.8%			
	100%MNS	0.77	43.0%				0.98	45.2%			
2018	NPK	3.88	62.0%	34.4	24.3	26.1	2.30	42.5%	27.5	24.8	41.9
	50%MNS	2.53	53.5%				2.26	51.7%			
	100%MNS	1.15	39.4%				0.99	30.5%			
2019	NPK	1.36	52.1%	29.6	22.8	12.4	6.49	83.3%	28.1	24.2	27.1
	50%MNS	0.72	36.7%				1.46	52.1%			
	100%MNS	0.55	37.4%				0.62	40.3%			

[†]N: nitrogen; NPK: 100% synthetic N application; 50%MNS: 50% synthetic N + 50% manure N application; 100%MNS: 100% manure N application

[‡]The NH_3 emission proportion was calculated by dividing the NH_3 emissions in basal fertilisation periods or topdressing periods with NH_3 emissions during day1-7 after N fertiliser application

[§]AT: Air temperature; ST: Soil temperature; SMC: Soil moisture content

Table 2 Total NH_3 emissions (kg ha^{-1}) during the maize growing periods of 2017~2019 (mean ± standard deviation, n = 3). Small letters following the standard deviation represent significant difference among the treatments ($p < 0.05$). Two-way ANOVA for the effects of fertilisation (F) and year (Y) on NH_3 emissions (p value)

Year	Treatment	NH_3 emissions			NH_3/N inputs	Reduction rate (%)‡
		Basal fertiliser	Topdressing	Total		
2017	NPK†	2.65 ± 0.11a	7.09 ± 0.39a	9.74 ± 0.50a	4.1%	–
	50%MNS	2.05 ± 0.06b	5.26 ± 0.29b	7.32 ± 0.34b	3.1%	24.9
	100%MNS	1.79 ± 0.06c	2.17 ± 0.14c	3.96 ± 0.15c	1.7%	59.3
2018	NPK	6.26 ± 0.23a	5.41 ± 0.25a	11.7 ± 0.30a	4.8%	–
	50%MNS	4.73 ± 0.35b	4.37 ± 0.09b	9.10 ± 0.43b	3.8%	22.1
	100%MNS	2.92 ± 0.07c	3.25 ± 0.12c	6.17 ± 0.13c	2.6%	47.2
2019	NPK	2.61 ± 0.25a	7.79 ± 0.39a	10.4 ± 0.34a	4.3%	–
	50%MNS	1.96 ± 0.10b	2.80 ± 0.15b	4.76 ± 0.06b	2.0%	54.3
	100%MNS	1.47 ± 0.15c	1.54 ± 0.11c	3.01 ± 0.26c	1.3%	71.1
Model effect	df	p values				
F	2	< 0.001	< 0.001	< 0.001		
Y	2	< 0.001	< 0.001	< 0.001		
F * Y	4	< 0.001	< 0.001	< 0.001		

†N: nitrogen; NPK: 100% synthetic N application; 50%MNS: 50% synthetic N + 50% manure N application; 100%MNS: 100% manure N application

‡Reduction rate of NH_3 emissions in the 50%MNS and 100%MNS treatments were compared with the NPK treatment

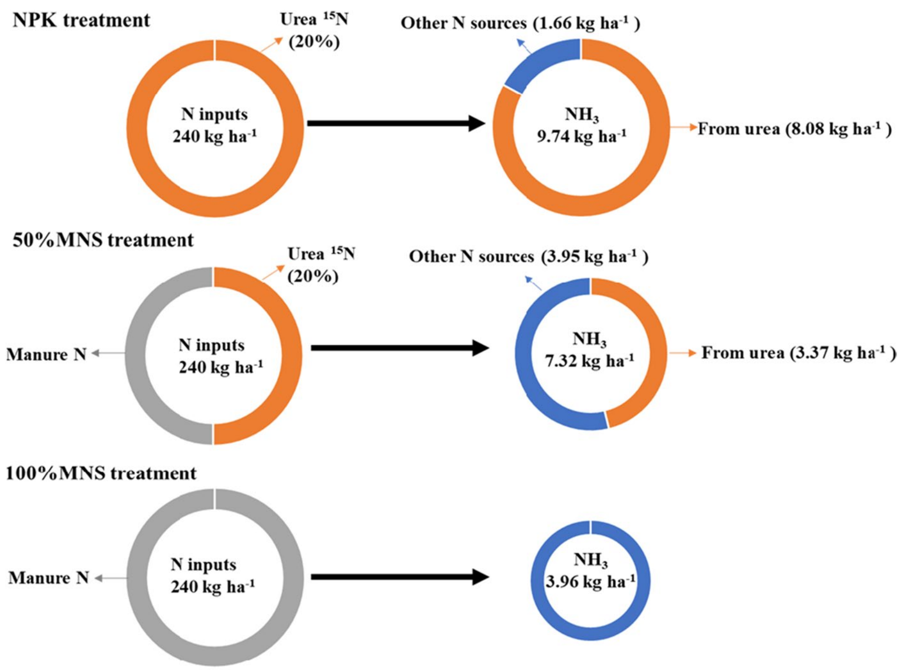

Fig. 3 NH_3 emissions from urea-N and other N source in the NPK, 50% manure N substitution (50% MNS) and 100% manure N substitution (100% MNS) treatments in the maize growing period of 2017

Table 3 Maize grain yields and partial productivity of applied N (PFP_N) during the experimental periods. Small letters following the standard deviation represent significant difference among the treatments ($p<0.05$). Two-way ANOVA for the effects of fertilisation (F) and year (Y) on maize grain yields and PFP_N (p value)

Year	Treatment	Maize grain yields ($t\ ha^{-1}$)	PFP_N[‡] ($kg\ kg^{-1}$)
2017	NPK[†]	8.7 ± 1.54c	36.3c
	50%MNS	10.4 ± 1.57b	43.3b
	100%MNS	12.7 ± 0.24a	52.9a
2018	NPK	8.8 ± 1.62b	36.7b
	50%MNS	10.0 ± 0.22a	41.7a
	100%MNS	10.3 ± 0.50a	42.9a
2019	NPK	8.1 ± 0.30b	33.8b
	50%MNS	10.5 ± 1.02a	43.8a
	100%MNS	10.5 ± 1.27a	43.8a
Model effect	df	p values	
F	2	< 0.001	< 0.001
Y	2	0.176	0.174
F * Y	4	0.206	0.206

[†]N: nitrogen; NPK: 100% synthetic N application; 50%MNS: 50% synthetic N + 50% manure N application; 100%MNS: 100% manure N application

[‡]PFP_N was calculated by dividing the N application rate with maize grain yields

Relationship between soil properties and ammonia emissions

To further explain the long-term impacts of manure substitution, relationships between SOM, soil TN, pH, and maize N uptake on the one hand and NH_3 emissions on the other hand were investigated (Fig. 4). Redundancy analysis (RDA) showed that soil pH had a positive effect on NH_3 emissions while the impacts of SOM, soil TN and N uptake were negative (Fig. 4). The first and second axes of RDA explained 62% (adjusted RDA value) of the total variation of NH_3 emissions. In addition, it was found that SOM ($F=13$, $p=0.009$) was the main factor explaining the differences in NH_3 emissions among the NPK and manure substitution treatments.

Discussion

Replacing fertiliser by manure increased maize grain yields

Compared to the NPK treatment, the maize grain yields in the 50%MNS and 100%MNS treatments significantly increased during the three maize growing seasons (Table 3). This is in line with the main

Table 4 Nitrogen inputs, outputs and N surplus during the three maize growing seasons of 2017–2019 ($kg\ N\ ha^{-1}$)

Year	Treatments	Basal urea N inputs[‡]	Top-dressing urea N inputs	Manure N inputs	Total N inputs	Crop N uptake[§]	N surplus[a]
2017	NPK[†]	90	150	0	240	198 ± 39b	42
	50%MNS	45	75	120	240	254 ± 53a	−14
	100%MNS	0	0	240	240	265 ± 45a	−25
2018	NPK	90	150	0	240	234 ± 51b	6
	50%MNS	45	75	120	240	241 ± 20b	−1
	100%MNS	0	0	240	240	278 ± 36a	−38
2019	NPK	90	150	0	240	200 ± 6c	40
	50%MNS	45	75	120	240	248 ± 24b	−8
	100%MNS	0	0	240	240	285 ± 26a	−45

[†]N: nitrogen; NPK: 100% synthetic N application; 50%MNS: 50% synthetic N + 50% manure N application; 100%MNS: 100% manure N application

[‡]The urea with ^{15}N labelled was only used in 2017 while the N fertiliser applied in 2018 and 2019 were the urea without ^{15}N labelled

[§]N uptake is the sum of the N assimilated by maize grain and straw

[a]N surplus is calculated by subtracting N uptake (including grain N and straw N) from the N inputs

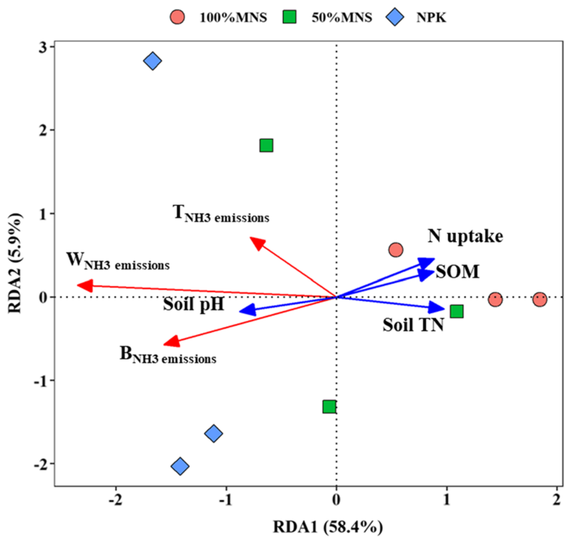

Fig. 4 RDA analysis shows the relationships among soil properties, maize N uptake and ammonia emissions during the basal fertilisation period ($B_{NH_3 emissions}$), top-dressing fertilisation period ($T_{NH_3 emissions}$) and whole period of maize growing seasons ($W_{NH_3 emissions}$) in the NPK, 50%MNS and 100%MNS treatments

finding of Zhang et al. (2021), i.e., substituting synthetic fertiliser with manure increased wheat yields in northern China. However, the yield effect depends on the substitution rate (Seufert et al. 2012; Zhang et al. 2020; Liu et al. 2021). The meta-analysis by Zhang et al. (2020) indicated that a partial substitution of synthetic fertilisers by manure increased the yields of upland crop by on average 6% while full substitution decreased yields by 9.6%. Liu et al. (2021) obtained comparable results; they arrived at an optimal substitution of 70% of synthetic fertiliser N by manure N.

The increased maize grain yields in our study may be ascribed to the following two reasons. First, the long-term manure application has improved soil quality and thereby increased plant growth (Guo et al. 2020). Soil TN, NO_3^--N, NH_4^+-N, MBC and MBN contents were significantly higher in the 50%MNS and 100% MNS treatments than in the NPK treatment (Fig. 2 and Table S1). The increase of MBC and MBN reflects an increased capacity of N retention by microbial communities in the manure treatments. This was backed up by significantly enhanced enzymes activities of β-D Glucosidase and N-acetylglucosidase and the AOB gene abundance (Table S1). Further, manure applications may have increased the availability of micronutrients (e.g. Zn, Cu, Mn) in soil, as well as soil structure. Long-term beneficial effects of manure applications were also observed in the meta-analysis by Zhang et al. (2020); the yield effect of a full substitution of fertiliser by manure was larger when the duration of the experiments exceeded 10 years.

Second, the N application rate of 240 kg N ha^{-1} was relatively high compared with the average N application rate of 150–200 kg N ha^{-1} in the North China Plain (Du et al. 2020). This means that the N supply to maize was sufficient, also when taking into account the lag time for mineralisation of manure N to inorganic N (Xu et al. 2021). Zhang et al. (2020) also found that full replacement of synthetic N by manure would not decrease crop yields, when N application rates for cereals were ≥ 250 kg N ha^{-1}.

Replacing urea-N fertiliser by manure decreased NH_3 emissions

Emissions of NH_3 were significantly reduced in treatments (50%MNS and 100%MNS) with replacement of urea-based N fertiliser with manure compared to the NPK control treatment (Fig. 1b; Table 2). This finding is in accordance with our hypothesis and also with results reported by Zhang et al. (2020). The NH_3 emission factor ranged from 4.1 to 4.8% in the NPK treatment (Table 2). The ^{15}N labelling experiment conducted in 2017 supported these results; the NH_3 emission factor of the labelled urea applied in the NPK treatment was 3.4%. This value decreased to 2.8% in the 50%MNS treatment, indicating that NH_3 emissions were lower from manure N than from urea N fertiliser. The NH_3 emission factors derived in our study are at the lower end of the NH_3 emission reported for the North China Plain; most of the NH_3 emission factors for urea fertiliser are in the range of 5–15% (Fu et al. 2020; Guo et al. 2020; Zhang et al. 2021). Commonly, NH_3 emissions are lower from urea applied to summer maize than from urea applied to winter wheat, because maize is grown during the wet season (which washes the urea quickly into the soil) and winter wheat during the dry season (Meade et al. 2011; Martins et al. 2015). Further, the annual mean air temperature is lower in the North China Plain compared to South China, which may lower the potential of NH_3 emissions in the North China Plain compared to South China (Fu et al. 2020).

In 2017 and 2019, NH_3 emissions rates were higher during the topdressing period than during the basal fertilisation period (Fig. 1b; Table 2), likely because the N fertiliser application was 1.67-folds higher as topdressing than as basal fertilisation. The relatively high NH_3 emissions during the basal fertilisation period compared to the topdressing period in 2018 was likely related to the relatively high temperature during the basal fertilisation period (Fig. 1; Table 2). Indeed, air temperature, rainfall and soil moisture content are main factors affecting NH_3 emissions (Singurindy et al. 2009; Yan et al. 2016; Xu et al. 2019). Moist and warm conditions also facilitate the mineralisation of organically bound N in manure to ammonium (NH_4^+), which is a source of NH_3 emissions from manure. This process may have contributed to the relatively modest NH_3 emission mitigation in the 100%MNS treatment compared to the NPK treatment in 2018 (Tables 1 and 2). This suggests that substituting synthetic N fertiliser with manure N may be not a robust NH_3 emission abatement strategy in warm areas with frequent precipitation (Meade et al. 2011; Zhang et al. 2021).

Replacing urea-N fertiliser by manure affected soil properties

Soil NO_3^--N and NH_4^+-N contents were lower in the 100% MNS treatment than in the 50% MNS treatment (Fig. 2). Also, the contents of SOM and total N, MBN and enzyme activities of β-D-Glucosidase and N-acetyl-glucosidase were significantly higher in the 100%MNS treatment than in the NPK treatment. These suggest that the capacity of N retention by microbial communities was enhanced in the manure treatments, which may decrease the potential for NH_3 emissions and nitrate leaching (Abdo et al. 2021; Zhang et al. 2021). Redundancy analysis showed that SOM ($R^2=0.53$, $p<0.05$) and soil pH ($R^2=0.52$) were positive associated with the mitigation of NH_3 emissions (Fig. 4). Thus, increased SOM contents in the manure treatments facilitates the retention of soil N through microbial processes, and may decrease NH_3 emissions losses (Liu et al. 2021; Quan et al. 2021). The significantly positive correlation between SOM and TN emphasised that an increase in soil organic C storage and soil TN build up go hand in hand (Fig. 4). Further, soil pH decreased slightly from 8.3 in the NPK treatment to 8.2 in the manure treatments, which may decrease NH_3 emissions (Guo et al. 2021).

Crop yield and N uptake were higher in the manure treatments than in the NPK treatment (Table 4). As a result, total N losses were likely reduced, including NH_3 emissions (Fig. 2). The decrease in NH_3 emissions occurred especially during the topdressing period (Table 4), in accordance with the findings of Liu et al. (2021).

Recommendations for improved management

Currently, manure-sourced N inputs account for 10% of the total N input into cereal crops, which account for about 70% of all croplands in China (Chen et al. 2017). This suggests a vast potential for replacing synthetic N with manure in cereal crops. Recent policies of the Chinese government also aim at replacing synthetic fertilisers with manure in cereal cropping system, so as to lower the environmental cost of fertiliser and manure use and to improve soil fertility (Jin et al. 2020). Results of our study support this policy initiative; full substitution of urea N fertiliser by manure N gave the highest crop yield and the lowest NH_3 emissions. The strength of our study is that measurements was conducted in a long-term field experiment. However, the N application rate (240 kg ha^{-1}) was relatively high, and may have masked that organically bound N in manure was slowly mineralised in the manure treatment. The long-term manure application has been fundamental to maintain crop yields in our study, because of the build-up of soil N. Thus, the duration of manure use, the N application rate, and the manure type and quality should be taken into account when transferring the results of this study and those of other studies to practices. We speculate that a partial substitution of synthetic fertiliser N by manure N might be suitable in the short term, while full substitution of synthetic fertiliser N by manure should be considered as long-term objective, also to achieve the dual goals of high crop yields and low environmental costs.

Conclusion

In a long-term field experiment, we found that NH_3 emissions were significantly decreased and maize grain yields were significantly increased following a partial or full replacement of urea N fertiliser by manure. The relatively large beneficial effect of a full replacement was likely the result of the long-term manure use, and the relatively high N application rate in the experiment. Replacement of urea-N fertiliser by manure also increased total N uptake by the maize crop, and the contents of soil organic matter (SOM) and total soil nitrogen (TN). Replacement of urea-N fertiliser by manure turned out to be more effective in reducing NH_3 emissions when soil moisture contents were low. This hints at low NH_3 emissions from manure N when the manure and soil were relatively dry, likely because of the slow mineralisation of organically bound N in manure. Future studies should focus on the impacts of different manure types (fresh slurries/liquid versus composted manure), different substitution ratios, and different durations of manure use (short and long-term).

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Data availability The data that support the findings of this study are available from the corresponding author Hongyuan Wang and Hongbin Liu upon reasonable request.

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