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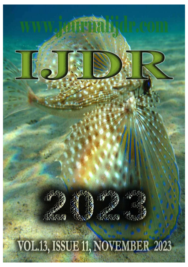

RESEARCH ARTICLE

OPEN ACCESS

INFLUENCE OF ORGANIC AMENDMENTS ON NOx, NH_3 EMISSIONS AND NITROGEN BALANCE FROM CROPLAND OF EUTRIC REGOSOLS

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ABSTRACT

Croplands contribute significantly to the global nitrogen oxide (NOx) emissions, ammonia (NH_3) volatilization, as well as nutrient pollution in water bodies, which is directly related to global warming and affects water and air quality. This study investigates the nitrogen (N) balance of croplands used for wheat-maize rotation farming using field free-drain lysimeters and incubation experiments. Six fertilizer treatments including regular mineral fertilizers (NPK), pig slurry as organic manure (OM), crop residues (CR), organic manure combined with mineral fertilizers (OMNPK), crop residues combined with mineral fertilizers (CRNPK) and a control plot without fertilizer (CK), were used for the field monitoring. N was applied at 280 kg N ha^{-1}, 130 kg N ha^{-1} during the wheat season and maize season was 150 kg N ha^{-1}. The results showed 0.44 to 10.04 kg N ha^{-1} average N_2O flux from the croplands. The total N (TN) loss annually was found to be 8.3 to 27.6 kg N ha^{-1} but organic amendments in croplands significantly reduced TN losses through overland flow by 85% and 117% via interflow compared with NPK treatments. The organic N mineralization rate differed significantly among the organic amendments. Significantly higher N mineralization rates were found for OM (47.51 mg kg^{-1}) and CRNPK (46.93 mg kg^{-1}) as compared to other treatments ($p < 0.05$). The cumulative annual NOx fluxes ranged from 0.12 to 1.27 kg N ha^{-1} across the croplands. Compared to NPK, OMNPK and CRNPK treatments were found to decrease annual NOx fluxes by 45% and 186%, respectively. Similarly, OM, OMNPK, and CRNPK treatments increased the annual NH_3 fluxes by 40%, 26%, and 24%, respectively. The annual NH_3 fluxes varied from 15.90 to 31.03 kg N ha^{-1} while the annual N balance ranged from 103.1 to 164.2 kg N ha^{-1} across all treatments. A significantly higher N balance was found under CRNPK compared to other treatments, suggesting higher N retention in soils under CRNPK compared with NPK treatment ($p < 0.05$). This optimal N supply and higher N balance under CRNPK organic amendments made it recommendable to maintain crop yield in cropland of Eutric Regosols.

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INTRODUCTION

Agricultural activities account for 80 to 90% of the emissions sources (Bouwman *et al*., 1997; Kang *et al*., 2016). Despite, nitrogen supply in agricultural soils is key factor for increasing crop grain production, however, nitrogen such as urea has high concentration and easy to transport into air and water (Cai *et al*., 2012; Ni *et al*., 2014). Consistently, nitrogen applied to agricultural soils need to be hydrolyzed and converted into NH_4^+, then to be further absorbed by crops. The hydrolysis process is very fast under the suitable hydrothermal conditions, because the soil NH_4^+ and pH increase rapidly due to the fertilization and led to an increasing risk of NH_3 volatilization (Sommer *et al*., 2004). Previous studies indicated that the average NH_3 volatilization rate is about 20 to 25%, or higher than 50% of the applied N rate (Rochette *et al*., 2008; Hunter *et al*., 2013). Consequently, nitrogen lost in form of NH_3 volatilization not only cause economic damage, but also affect the environment and significant negative impact of human health (Stokstad, 2014; Gu *et al*., 2014). According to Zhang *et al*. (2015), nitrogen fertilization practices could affect soil properties and climate conditions, while NH_3 volatilization in cropland of Etric Regosolis serious during summer maize season. For example, some measures could be conducted to significantly reduce NH_3 volatilization in cropland such deep application of N fertilizer, irrigation after fertilization (Rochette *et al*., 2013). Zhang *et al.* (2016) have previously compared two NH_3 volatilization measurements methods (Wind tunnel method and continuous air flow enclosure chamber method) under summer maize season in cropland of Eutric Regosol.

The nitrogen oxides (NOx) and ammonia (NH_3) emissions from cropland could negatively influences water and air quality. Improving crop productivity while reducing the environmental impacts of nitrogen gaseous loss from fertilized agricultural soils could be a best practice for sustaining crop production. However, overuse of mineral N fertilizer has led to NOx and NH_3 emissions, as well as an increase in N deposition, consequently led to air pollution worldwide (Erisman *et al*., 2008; Kuang *et al*., 2016). In fertilized agricultural soils, nitrogen oxides (NO and NO_2) results from denitrification, which is the microbial processes that reduces NO_3^- and NO_2^- into NO and NO_2 (Butterbach-Bahl *et al*., 2013). According to Stavrakou *et al.* (2008), NOx indirectly affects earth's radiative balance, but they catalyze the ozone (O_3) production in the troposphere through the photochemical reactions, which affects the greenhouse gases emissions and environmental quality. Over the past two decades, many studies have attempted to determine N balance around the worldwide. For example, the N balance in Belgium, Luxembourg, Japan and Netherlands exceed 160 kg N ha^{-1} (Hoang and Alauddin, 2009), while in Germany N balance was estimated about 100 kg N ha^{-1} from 1992 to 2006 (Panten *et al*., 2009). In contrast, Bechmann *et al.* (2014) reported a significant lower N balance from -12 N ha^{-1} yr^{-1} to 132 kg N ha^{-1} yr^{-1} in Norway. Zhou *et al.* (2016) reported an annual N balance of 28.6 to 72.9 kg N ha^{-1}in wheat-maize rotation systems in cropland of purplish soils in Southern China, without considering N supply from the soil. Moreover, He *et al.* (2018) estimated N balance in China using modelling and concluded that N balance has decreased trends for all regions in China for the period of 2000 to 2010, except in the southeast and southwest of China due to higher increased rate of N input than the lower increased rate of N output. According to He *et al.* (2018) N balance can be characterized as follows: When N balance is equal to value of zero (N balance = 0), it imply that there is no N surplus in the soil; while, when N balance is lower than zero (N balance < 0), there is a lack of N in the soil, so N addition is required for crop growth; and when N balance is higher than zero (N balance > 0), there is N surplus in soil, which can have environmental risks of N gas emissions and hydrological N loss. Purplish soil is an important cropland in the upper reaches of the Yangtze River. The soil occupied 68% total croplands in Sichuan Province and accounted for 7% of total cropland in China. Purple soil is classified as a Eutric Regosol under the United Nations (UN) Food and Agriculture Organization (FAO) system as well as Pup-Orthic-Entisol under the Chinese Soil Taxonomy system (Zhu *et al*., 2012). Eutric Regosoloccupied about 160 000 km^2 in the upper reaches of the Yangtze River (Zhu *et al*., 2009). Therefore, understanding the N balance of EutricRegosol will be crucial for soil fertility and nutrient management practices relevant for crop production and environmental protection (He *et al*., 2018). This study investigates NOx emissions and NH_3 volatilization under wheat-maize cropping systems over one year cropping season. The objective of this study wasto analyze the effects of different organic amendments on gaseous N loss (NOx and NH_3), and to quantify N balance under organic amendments in cropland of Eutric Regosol.

MATERIAL AND METHODS

Site description: This study was conducted at YantingAgro-Ecological Station of Purplish Soil (31°16′ N, 105°28′ E, elevation 400-600 m above sea level), one of the research stations under the Chinese Ecosystem Research Network (CERN) in Sichuan Province, Southwest China (Figure 1). The soil is clayed loam texture (0-10 cm) with 19.2% clay, 42.1% silt, and 38.7% sand (Zhu *et al*., 2009; Zhou *et al*., 2013). The soil has a pH of 8.3 ± 0.1, 6.35 ± 0.93 g kg^{-1} organic carbon, 0.55 ± 0.15 g kg^{-1} total N and C/N ratio of 11.5 ± 0.6. On the other hand, it has a bulk density of 1.32 ± 0.12 g cm^{-3} and saturated hydraulic conductivity of 18.6 ± 1.2 mm h^{-1} (Zhou *et al*., 2014). The experimental wheat-maize croplands were laid out in a completely randomised block design consisting of six fertiliser treatments replicate three times. These includes cropland without amendment (CK, as control), conventional mineral fertilizers (NPK) as a control, fresh pig slurry as organic manure at N application rate equivalent to mineral N in NPK (OM), crop residues only with N equivalent to 20% of applied N in treatment of NPK (CR), fresh pig slurry with N equivalent to 40% mineral N plus 60% mineral N at total N rate equivalent to NPK treatment (OMNPK), and crop residues at 20% N equivalent plus 80% mineral N with total N same as in the NPK treatment (CRNPK). Each plot was connected to a lysimeter (Size: 8 × 4 m^2, slope: 6.5°) to collect water samples. The annual N application rate was 280 kg N ha^{-1}, except for the CR treatment, the rate changed to130 kg N ha^{-1} during wheat season (*Triticum aestivum* L.) and 150 kg N ha^{-1} during maize season (*Zea mays* L.). Urea $CO(NH_2)_2$ was mineral N fertilizers source, while calcium superphosphate and potassium chloride were source of P (90 kg P_2O_5 ha^{-1}) and K (36 kg K_2O ha^{-1}), respectively (Zhu *et al*., 2009; Zhou *et al*., 2014). Crop residues chopped to < 5 cm in length were incorporated into the CR and CRNPK treated plots before planting. All fertiliser treatments were applied uniformly into the soil to a depth of 10 cm before sowing. Winter wheat (variety 01-3570) was planted early November and harvested in May, followed by maize (variety Chengdan 60) cropping season with planting June and harvested September.

Soil sampling and incubation of soil: Using a flat-bladed stainless-steel shovel, three cores of field moist soil samples (0–15 cm) were collected in late summer from each treatment plot following the harvest of maize. In order to have four replicates for each treatment in the incubation experiment, three 300 g soil samples and three 100 g samples from each of the three replicates were taken from each plot. To get rid of roots and fragments, field-moist soil samples were air dried and sieved at a mesh size of 2 mm. The soil samples were quickly sealed with plastic bags and stored at 4 °C until the incubation experiments began. For organic N mineralization, soil samples (30.0 g on an oven-dried basis) were put into 250 mL glass jars. As per the protocol of Raiesi and Kabiri (2017), glass jar bottles were incubated in the dark at a temperature of 25 ± 1°C and a water holding capacity of 70%. In order to compensate for the soil temperature and water content lost during soil sampling and air-drying time (Broos *et al*., 2007), reduce labile soil organic matter concentration, and activate the soil microbial population (Raiesi and Kabiri, 2017), soil samples were pre-incubated for one week under WHC of 70% prior to measuring N mineralization.

Measurements of soil N_2O emissions: Static chamber-gas chromatography was used to measure N_2O emissions at the same time in 2017–2018. Zhou *et al.* (2017) had previously employed this method for a related observational study. We inserted a stainless-steel chamber on a permanent collar (0.50 x 0.50 m) into each plot's soil at a depth of 10 cm after ploughing the field plots and before planting the crops. This chamber remained in place for the duration of the wheat-maize rotational experimental period. To reduce exposure to temperature changes when the chambers were closed, a layer of insulating material was placed over them. A circulating fan was employed to ensure even mixing within the chamber headspace, and the aboveground height of the chamber collar was adjusted in accordance with the particular wheat-maize growth stage. During the first week after fertilization, we measured N_2O fluxes every day. During the second week, we measured N_2O fluxes every two days. For the remainder of the measurement period, the sampling frequency was then set to twice weekly. As directed by Parkin and Ventera (2010), morning hours (i.e., 9:00 to 11:00 am) were used to gather gas samples from the chambers. Five distinct gas samples were obtained at each flux measurement following closure, utilizing 50 ml plastic syringes equipped with three-way stopcocks that were placed into Teflon tubes that were fastened to the chambers. Following sampling, N_2O flux in gas samples from both treatments was examined using a gas chromatograph (GC) (HP 5890II, Hewlett-Packard, Palo Alto, California, USA) equipped with an electron capture detector (ECD). For the analysis of N_2O gas, this study employed Wang *et al.* (2003)'s inclusive GC configuration procedure. Rather than directly introducing high concentration buffering gases like CO_2 and CH_4 into the detector for analysis, we employed high purity dinitrogen (N_2) as a gas carrier. The system was calibrated using known mixed gas concentrations of 386.2 ppm CO_2, 5.1 ppm CH_4, and 0.5 ppm N_2O for each measurement.

Measurements of interflow and overland flow: Cement partition walls were used to hydrologically isolate each free-drain lysimeter plot for the purpose of measuring overland flow and interflow water. In order to prevent unanticipated seepage into adjacent plots, the partition walls were inserted at least 60 cm into the bedrock (Zhu *et al.*, 2009). To measure overland flow and interflow following each rainfall event, a conflux trough was incorporated into the design of the free-drain lysimeter, both on the topsoil and the bedrock surface. Under each conflux trough, distinct ponds were built to collect water from both interflow and overland flow (Wang and Zhu, 2011; Zhou *et al.*, 2012). Each pond's water level was measured four times prior to the collection of water samples. Clean 500 mL polythene bottles were used to collect water from both the overland flow and the interflow. Every time it rained and when there was no more water flowing, water was taken from various ponds. To guarantee homogeneity, water samples from overland flows were thoroughly mixed prior to sampling.

Measuring nitrogen in runoff water: Following water sampling, nitrogen compounds were examined in both interflow water and runoff water obtained from overland flow. Whatman No. 5 filter paper was used to filter water samples, with the exception of the total nitrogen (TN) measurement. A continuous flow auto analyzer (AA3, Bran + Luebbe, Norderstedt, Germany) was used to measure total dissolved nitrogen (TDN), ammonia nitrogen (NH_4^+-N), and nitrate nitrogen (NO_3^--N). TDN minus NH_4^+-N and NO_3^--N was used to calculate dissolved organic nitrogen, or DON. TDN minus TN was used to compute the particulates N, or PN (Jiao *et al.*, 2009; Gao *et al.*, 2014).

Uptake of Crop N : After being oven dried for 48 hours at 70 °C to calculate their dry weight equivalent, the grains, shoots, and roots were ground and put through a 0.5 mm sieve to measure the TN content. An elemental analyzer (Vario EL/micro cube, Germany) was used to calculate TN. N uptake in wheat and maize was calculated using Malhi *et al.* (2010).

Measurements of soil NOx emissions: The two components of nitrogen oxides (NOx) are nitrogen dioxide (NO_2) and nitrogen oxide (NO). A unique gas bag with valves connected to a pump (12V, KNF, Germany) in the outlet was used to collect samples of NOx gas from the installed chambers at a predetermined 7-minute interval. Plastic tubes were used to connect the inlet to the chambers. During the first week following the application of organic fertilizers, NOx gas samples were taken every morning between 9:00 and 11:00. In the second week of the experiment, however, NOx gas samples were taken every two days. The sampling frequency was changed to twice weekly after the second week and remained that way until the gas collection period ended. A Thermo-Model 42i (Fischer Scientific, USA) was used to analyze the obtained gas samples for NOx content right away.

Measurements of soil ammonia (NH_3) volatilization: Using a continuous air flow enclosure chamber method, the NH_3 volatilization was measured for three weeks during the wheat and maize seasons, adhering to the protocols of Cao *et al.* (2013). As advised by Hayashi *et al.* (2006), the NH_3 volatilization was collected over the course of two hours, in the morning (between 8:00 and 10:00 am) and in the evening (between 3:00 and 5:00 pm), using an air flow rate of 15 L min^{-1}. The materials for the NH_3 volatilization sampling consist of an acid trap (80 ml 0.05 M H_2SO_4) to collect volatilized NH_3, a vacuum pump, and a cylindrical chamber constructed of methyl methacrylate (15 cm high and 20 cm inner diameter). In order to collect NH_3, a cylindrical chamber was embedded in the ground and pumped with ambient air to mix the interior air. This allowed the H_2SO_4 acid to trap the NH_3 emissions inside two glass bottles. Additionally, NH_3 from the surrounding air was extracted using a chamber that is not submerged in the ground. Afterward, the ammonium (NH_4^+) concentration was measured using an Auto Analyzer-AA3 (Bran + Luebbe, Norderstedt, Germany).

Analyzing data: The method proposed by Wang *et al.* (2003) was used to calculate N_2O fluxes from soil, whereas Gao *et al.* (2014) proposed using daily flux linear interpolation between gas sampling rates to calculate cumulative seasonal and annual N_2O fluxes. The equations (1) and (2) were used to determine the N loss loadings in a single flow (Qi) for every individual runoff event (Wang and Zhu, 2011; Wang *et al.*, 2012).

$$Qi = Ci \times \frac{qi}{100} \tag{1}$$

where Ci is the overland flow or interflow's N content (mg L^{-1}), *qi* is the runoff depth per unit area (mm), and *Qi* is the loss flux of the overland flow or interflow (kg ha^{-1}). The following formula was used to determine annual N loss loadings:

$$Q = \sum Qi \tag{2}$$

where i is the number of runoff events in a given year and Q is the N loss load (kg ha^{-1}).

Equation (3) was used to calculate the NH3 volatilization (Hayashi *et al.*, 2006).

$$F = \frac{(Cs - Cb) * V * 10^{-2}}{\pi * r^2} * \frac{24}{t} \tag{3}$$

where *F* is the daily NH_3 volatilization (kg N ha^{-1} d^{-1}), *V* is the acid (H_2SO_4) volume (L), *r* is the cylindrical chamber radius (m), t is the duration of NH_3 volatilization sampling (h), Cs is the NH_4^+-N concentration trapped in acid (H_2SO_4) solution for soil (mg L-1), and *Cb* is the NH_3 in the ambient air (mg L^{-1}). By adding up the NH_3 from the wheat and maize seasons, the cumulative NH_3 volatilization was determined. Through linear interpolation of the daily flux between the sampling days, the cumulative NH_3 fluxes for the wheat and maize seasons were determined.

Equation (4) was used to estimate the nitrogen balance (Ju *et al.*, 2009).

$$\text{N balance} = N_{inputs} - N_{outputs} \tag{4}$$

where $N_{outputs}$= Crop N uptake + Soil erosion + Hydrological N loss + Gaseous N loss, and N_{inputs} = N deposition + N fertilizer + N mineralization.

Statistical analyses: Using one-way analysis of variance (ANOVA) in IBM SPSS Statistics 20.0 (IBM Corp., Armonk, NY, USA), the effects of the organic amendments were assessed on simultaneous overland flow, interflow of N loss loadings, N mineralization, nitrous oxides (N_2O) fluxes, nitrogen oxides (NOx) emissions, and ammonia (NH_3) volatilization. The least significant difference test (LSD) at $P < 0.05$ was used to confirm significant differences in the mean of estimated parameters among treatments. The data are presented as mean values with mean standard error (±SE) in triplicate. Sigma Plot software (version 12.5, Systat Inc., USA) was used to visualize the data.

RESULTS

Seasonal patterns of soil NOx emissions: Nitrogen oxides (NOx) emissions showed a decreasing trend in the wheat season and an increasing trend in the maize season for the all treatments over the one-year observation period of 2017-2018 (Fig. 2). During the wheat season, the NOx emissions ranged from -8.04 to 1.31 μg N m^{-2} hr^{-1}

for CK (Fig. 2a), from 0.08 to 89.83 µg N m^{-2} hr^{-1} for OM (Fig. 2b), from 0.18 to 3.63 µg N m^{-2} hr^{-1} for CR (Fig. 2c), from 0.45 to 193.18 µg N m^{-2} hr^{-1} for NPK (Fig. 2d), from 0.13 to 97.23 µg N m^{-2} hr^{-1} for OMNPK (Fig. 2e), from 0.09 to 33.49 µg N m^{-2} hr^{-1} for CRNPK (Fig. 2f).

Furthermore, during the maize season, the NOx emissions ranged from -0.23 to 3.99 µg N m^{-2} hr^{-1} for CK (Fig 2a), from 0.15 to 398.20 µg N m^{-2} hr^{-1} for OM (Fig. 2b), from 0.05 to 23.84 µg N m^{-2} hr^{-1} for CR (Fig. 2c), from 0.06 to 190.17 µg N m^{-2} hr^{-1} for NPK (Fig. 2d),

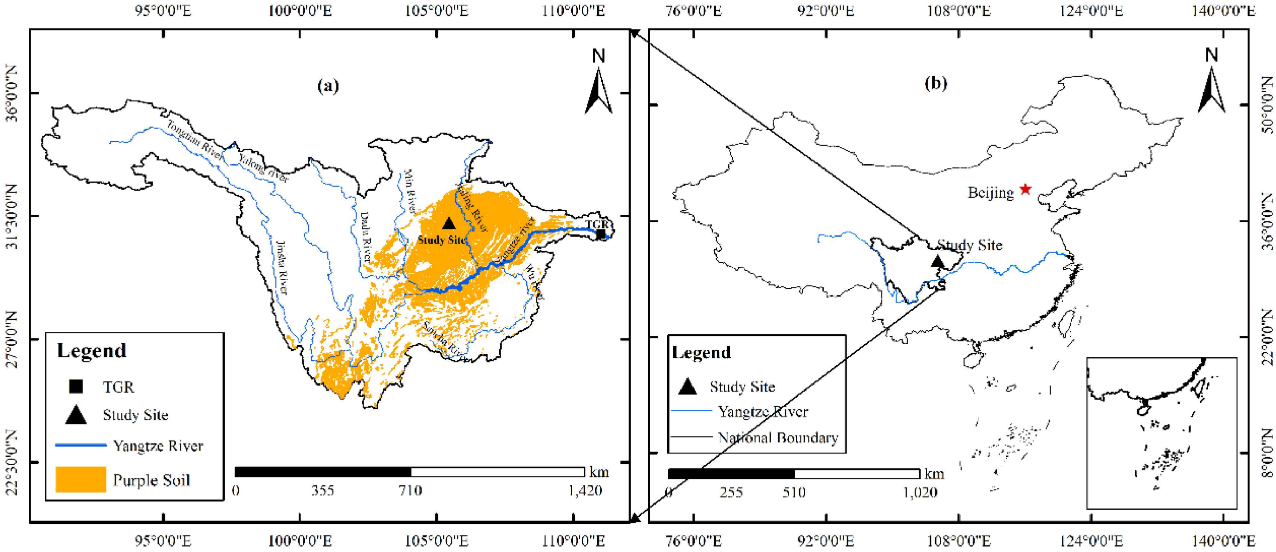

Figure 1. Sketch map showing the study site: (a) distribution of purple soil (Eutric Regosol) in the upper Yangtze River; and (b) its location on the China map from Bah et al. (2020)

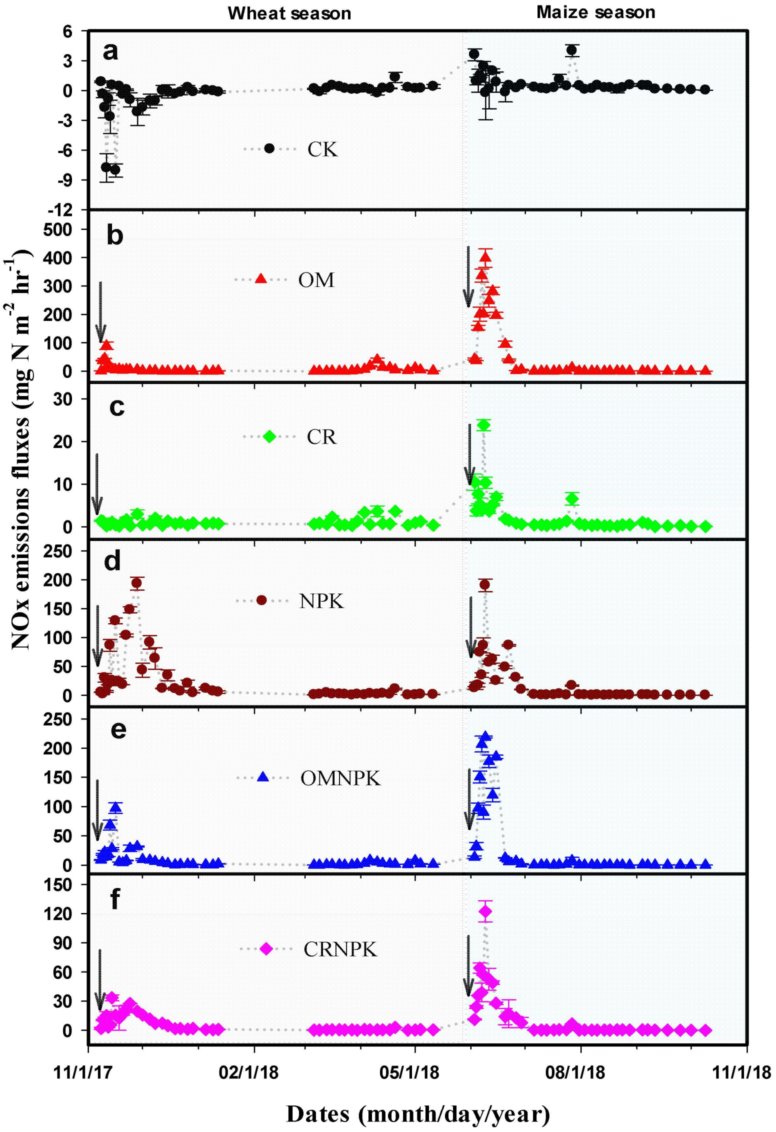

Figure 2. Seasonal changes in NOxemissions for the all treatments during 2017-2018. The vertical bars indicate the standard error of the three spatial replicates (n=3). The downward arrows indicate the date of different organic amendments applications

from 0.08 to 219.15 µg N m^{-2} hr^{-1} for OMNPK (Fig. 2e), from 0.02 to 122.31 µg N m^{-2} hr^{-1} for CRNPK (Fig. 2f). Our results showed that the highest peaks of NOx emissions fluxes were observed in the NPK treatment in the wheat cropping season and OM treatment in the maize cropping season. The mean cumulative NOx fluxes over the one-year period between all the treatments were significantly different for the wheat and the maize seasons ($p < 0.05$), varying from -0.01 to 0.80 kg N ha^{-1}, and from 0.02 to 0.94 kg N ha^{-1}, respectively (Table 1). On average, NOx emissions fluxes for the maize season were two times higher than those for the wheat season. Annual cumulative CH_4 fluxes of the organic amendments differed significantly, with highest significant mean annual NOx fluxes observed in OM treatments ($p < 0.05$) (Table 1). Furthermore, annual cumulative NOxfluxes showed significant difference between the all treatments and followed the order of; OM>NPK> OMNPK>CRNPK>CR>CK (Fig. 3).

Table 1. Seasonal and annual soil N supply, soil erosion, soil N gaseous losses from the organic amendments the wheat-maize for the period of 2017-2018

Treatments	Soil N supply (kg N ha^{-1})	Soilerosion (kg N ha^{-1})	N_2O fluxes (kg N ha^{-1})	NOx fluxes (kg N ha^{-1})	NH_3 fluxes (kg N ha^{-1})	Total gaseous N (kg N ha^{-1})
Wheat season						
CK	42.47 ± 1.43e	-	0.19 ± 0.02f	-0.01 ± 0.01d	3.90 ± 0.03d	4.08 ± 0.03d
OM	97.41 ± 2.59a	-	0.68 ± 0.03b	0.20 ± 0.01bc	10.53 ± 0.01a	11.40 ± 0.02a
CR	87.26 ± 2.64b	-	0.30 ± 0.01e	0.04 ± 0.00d	-	-
NPK	61.91 ± 0.81d	-	0.59 ± 0.04c	0.80 ± 0.04a	5.20 ± 0.03c	6.58 ± 0.04c
OMNPK	73.48 ± 2.02b	-	0.50 ± 0.01d	0.24 ± 0.00b	5.85 ± 0.03c	6.58 ± 0.01c
CRNPK	93.47 ± 0.78a	-	0.83 ± 0.02a	0.16 ± 0.01c	6.50 ± 0.28b	7.49 ± 0.02b
Maize season						
CK	42.47 ± 1.43e	2.96 ± 0.14a	0.23 ± 0.02e	0.02 ± 0.00f	12.00 ± 0.10c	12.25 ± 0.02e
OM	97.41 ± 2.59a	0.96 ± 0.19b	6.72 ± 0.08a	0.94 ± 0.01a	20.50 ± 0.76a	28.16 ± 0.08a
CR	87.26 ± 2.64b	0.23 ± 0.02c	0.34 ± 0.01e	0.05 ± 0.00e	-	-
NPK	61.91 ± 0.81d	1.08 ± 0.11b	0.64 ± 0.05d	0.37 ± 0.01c	13.50 ± 1.26c	14.51 ± 0.05d
OMNPK	73.48 ± 2.02b	1.18 ± 0.04b	2.92 ± 0.01b	0.55 ± 0.01b	19.50 ± 1.00a	22.97 ± 0.02b
CRNPK	93.47 ± 0.78a	0.54 ± 0.09c	0.87 ± 0.05c	0.23 ± 0.01d	18.00 ± 1.80b	19.10 ± 0.04c
Annual						
CK	42.47 ± 1.43e	2.96 ± 0.14a	0.46 ± 0.03f	0.02 ± 0.02f	15.90 ± 0.13d	16.37 ± 0.02e
OM	97.41 ± 2.59a	0.96 ± 0.19b	10.44 ± 0.13a	1.27 ± 0.02a	31.03 ± 0.89a	42.73 ± 0.12a
CR	87.26 ± 2.64b	0.23 ± 0.02c	0.71 ± 0.02e	0.12 ± 0.01e	-	-
NPK	61.91 ± 0.81d	1.08 ± 0.11b	1.33 ± 0.09d	1.21 ± 0.04b	18.70 ± 1.26c	21.23 ± 0.07d
OMNPK	73.48 ± 2.02b	1.18 ± 0.04b	4.40 ± 0.02b	0.83 ± 0.02c	25.35 ± 0.98b	30.58 ± 0.03b
CRNPK	93.47 ± 0.78a	0.54 ± 0.09c	1.82 ± 0.02c	0.42 ± 0.00d	24.50 ± 1.56b	26.74 ± 0.03c

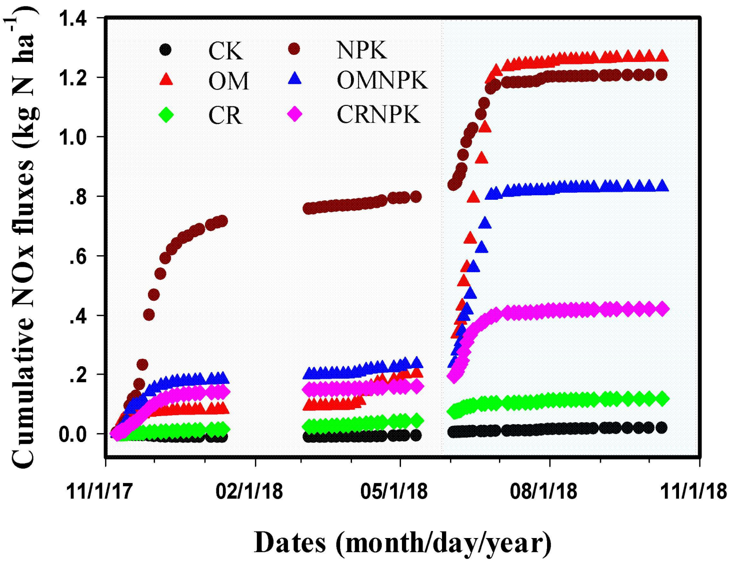

Figure 3. Seasonal and annual cumulative NOx fluxes for organic amendments during the year of 2017-2018

Seasonal pattern of soil NH_3 emissions fluxes: The daily flux of NH_3 emissions showed a decreasing trend in the wheat season and an increasing trend in the maize season for the five treatments over the three weeks measurements for each season (Fig. 4). During the wheat cropping season, the daily flux of NH_3 emissions ranges from 0.00 to 0.12 kg N ha^{-1} d^{-1} for CK (Fig. 4a), from 0.02 to 0.35 kg N ha^{-1} d^{-1} for OM (Fig. 4b), from 0.01 to 0.51 kg N ha^{-1} d^{-1} for NPK (Fig. 4c), from 0.02 to 0.17 kg N ha^{-1} d^{-1} for OMNPK (Fig. 4d), and from 0.03 to 0.52 kg N ha^{-1} d^{-1} for CRNPK (Fig. 4e). Furthermore, during the maize cropping season, the daily flux of NH_3 emissions ranges from 0.00 to 0.16 kg N ha^{-1} d^{-1} for CK (Fig. 4a), from 0.02 to 0.91 kg N ha^{-1} d^{-1} for OM (Fig. 4b), from 0.03 to 1.23 kg N ha^{-1} d^{-1} for NPK (Fig. 4c), from 0.03 to 0.28 kg N ha^{-1} d^{-1} for OMNPK (Fig. 4d), and from 0.03 to 1.00 kg N ha^{-1} d^{-1} for CRNPK (Fig. 4e). The highest peaks of daily flux of NH_3 volatilization were observed in the OM, NPK and CRNPK treatments in both the wheat and the maize seasons. Nonetheless the NH_3 flux for only CR was not determined. There were significant differences in the seasonal and annual cumulative NH_3 fluxes among the five treatments ($p < 0.05$), as shown in Figure 8.4. Across the five treatments, the wheat season cumulative NH_3 flux ranges from 3.90 to 10.53 kg N ha^{-1}, and the maize season cumulative NH_3 flux range from 12.00 to 20.50 kg N ha^{-1}, while the annual cumulative NH_3 fluxes ranged from 15.90 to 31.03 kg N ha^{-1} (Table1, Fig. 5). Furthermore, annual cumulative NH_3 fluxes showed significant difference between the all treatments and were in the order; OM>OMNPK~CRNPK>NPK>CK (Table 1, Fig. 5).

Crop N uptake: The results of crop N uptake are summarized in Table 7.2. The crop N uptake across all treatments was in the range of 12.24 to 88.24 kg N ha^{-1} for the wheat season and 27.09 to 150.05 kg N ha^{-1} for the maize season, while the annual crop N uptake ranged

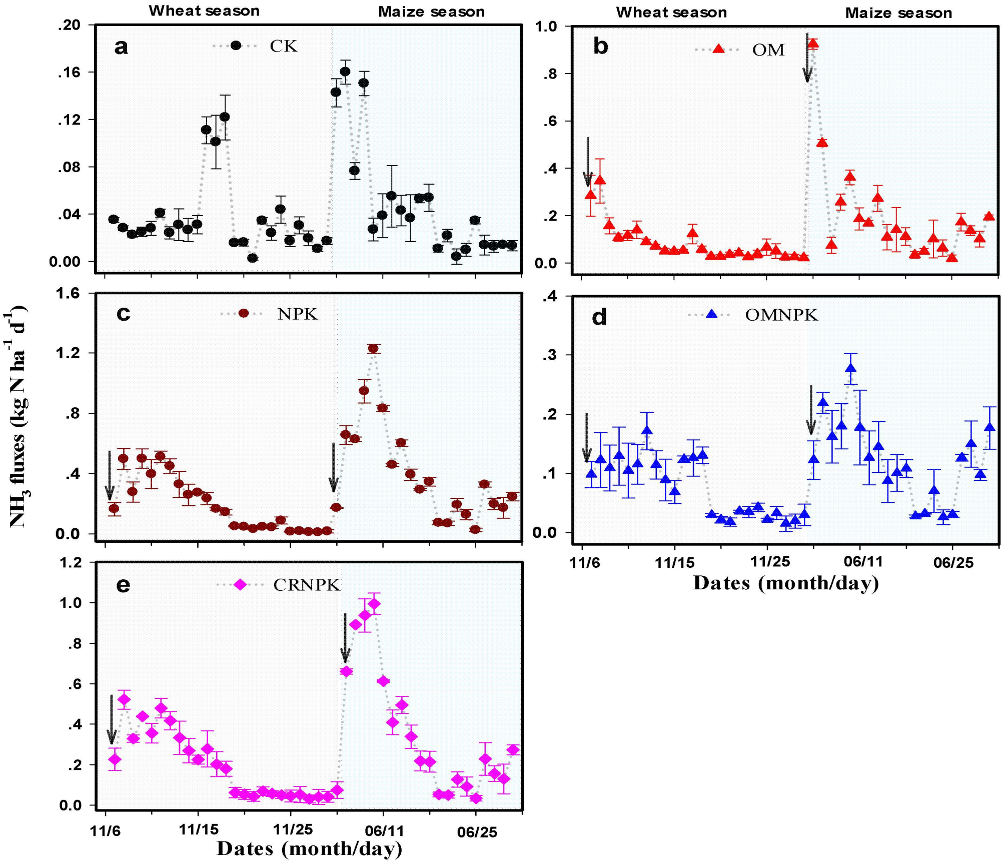

Figure 4. Seasonal changesin NH_3 volatilization of organic amendments during the year of 2017-2018. The vertical bars indicate the standard error of the three spatial replicates (n=3)

Table 2. Seasonal and annual crop grain yield and biomass, crop N uptake from the organic amendmentsin the wheat-maize seasons for 2017-2018

Treatments	Grain yield (Mg ha^{-1})	Shoot biomass (Mg ha^{-1})	Root biomass (Mg ha^{-1})	Grain N uptake (kg N ha^{-1})	Shoot N uptake (kg N ha^{-1})	Root N uptake (kg N ha^{-1})	Crop N uptake (kg N ha^{-1})
Wheat season							
CK	0.55 ± 0.06d	0.57 ± 0.05d	0.26 ± 0.03c	7.89 ± 0.63	2.88 ± 0.34c	1.48 ± 0.08c	12.24 ± 0.98d
OM	4.46 ± 0.06a	3.37 ± 0.13a	0.86 ± 0.03a	63.72 ± 0.96	18.81 ± 1.53a	5.72 ± 0.60a	88.24 ± 2.05a
CR	1.80 ± 0.17c	1.59 ± 0.09c	0.49 ± 0.06b	25.10 ± 2.09	7.09 ± 1.60bc	2.56 ± 0.35bc	34.76 ± 3.51c
NPK	3.56 ± 0.21b	1.81 ± 0.05c	0.43 ± 0.01b	60.41 ± 5.50	12.30 ± 1.45ab	3.00 ± 0.21bc	75.71 ± 6.58ab
OMNPK	3.87 ± 0.15b	2.55 ± 0.36b	0.70 ± 0.04a	58.22 ± 5.75	16.14 ± 2.89a	4.83 ± 0.21ab	79.18 ± 8.68ab
CRNPK	3.68 ± 0.06b	2.52 ± 0.09b	0.70 ± 0.09a	49.52 ± 2.17	13.38 ± 2.57ab	4.97 ± 1.33ab	67.87 ± 5.68b
Maize season							
CK	0.56 ± 0.16c	2.14 ± 2.14c	0.29 ± 0.04c	7.06 ± 2.53c	17.95 ± 1.46c	2.08 ± 0.14c	27.09 ± 3.47c
OM	5.78 ± 0.47a	4.66 ± 4.66a	1.56 ± 0.02a	87.98 ± 4.27a	45.87 ± 7.11a	16.20 ± 1.00a	150.05 ± 8.97a
CR	3.98 ± 0.17b	3.08 ± 3.08bc	0.77 ± 0.04b	58.29 ± 4.36b	23.57 ± 2.32bc	5.50 ± 0.88b	87.37 ± 4.89b
NPK	5.14 ± 0.49a	3.29 ± 3.29bc	0.71 ± 0.12b	81.19 ± 5.43a	33.41 ± 2.86ab	5.82 ± 0.56b	120.43 ± 7.95a
OMNPK	5.27 ± 0.25a	4.04 ± 4.04ab	0.93 ± 0.06b	86.72 ± 7.18a	41.32 ± 5.11a	8.83 ± 1.33b	136.88 ± 10.86a
CRNPK	5.26 ± 0.46a	4.48 ± 4.48a	0.88 ± 0.07b	84.21 ± 6.94a	40.25 ± 6.22a	7.81 ± 0.87b	132.26 ± 13.78a
Annual							
CK	1.11 ± 0.16d	2.72 ± 0.17d	0.55 ± 0.05d	14.95 ± 2.65c	20.83 ± 1.12c	3.56 ± 0.20d	39.33 ± 3.10d
OM	10.24 ± 0.42a	8.03 ± 0.41a	2.42 ± 0.05a	151.70 ± 4.89a	64.67 ± 8.29a	21.92 ± 1.54a	238.29 ± 10.65a
CR	5.78 ± 0.16c	4.67 ± 0.56c	1.26 ± 0.04c	83.39 ± 2.72b	30.67 ± 2.50bc	8.07 ± 1.05c	122.13 ± 2.02c
NPK	8.70 ± 0.69b	5.10 ± 0.31c	1.14 ± 0.11c	141.60 ± 9.94a	45.71 ± 2.94a	8.82 ± 0.77c	196.14 ± 12.74b
OMNPK	9.14 ± 0.32ab	6.59 ± 0.17b	1.64 ± 0.02b	144.94 ± 9.75a	57.46 ± 4.97a	13.66 ± 1.25b	216.06 ± 14.13ab
CRNPK	8.94 ± 0.41ab	7.00 ± 0.40ab	1.58 ± 0.10b	133.73 ± 5.08a	53.63 ± 5.80a	12.78 ± 0.95b	200.13 ± 11.44b

from 39.33 to 238.29 kg N ha^{-1} (Table 2). There were significant differences ($p < 0.05$) in the crop N uptake in both the wheat and the maize seasons. The OM treatments exhibited the highest significant crop N uptake in both the wheat and the maize seasons. Conversely, seasonal and annual grain N uptake, shoot N uptake, and root N uptake over the experimental year are shown in Table 2.

Quantification of nitrogen balance: The N balance components of soil N supply, soil erosion and gaseous N loss are shown in Table 1. While the crop N uptake are presented in Table 2, and the total hydrological N loss through both overland flow and interflow in Table 3. Overall, the N balance inputs and outputs are presented in Table 4 for all treatments except CR treatments. From Table 4, the N balance for the wheat season ranges from 38.9 to 160.8 kg N ha^{-1} across all treatments. Wheat season N balance for CRNPK was significantly higher than those other treatments, which were also significantly different from each other ($p < 0.05$). The maize season N balance ranges of 14.2 to 97.6 kg N ha^{-1} for all treatments (Table 4). Similarly, N balance for CRNPK was significantly higher compared to the other treatments in the maize season. On average, the N balance in the wheat season is two times higher than that in the maize season.

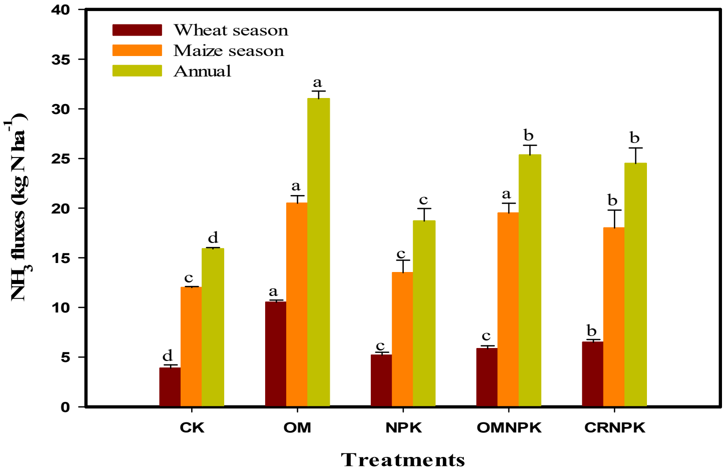

Figure 5. Seasonal and annual cumulative NH$_3$ fluxes of organic amendments during the period of 2017-2018. The vertical bars indicate the standard error of the three spatial replicates (n=3)

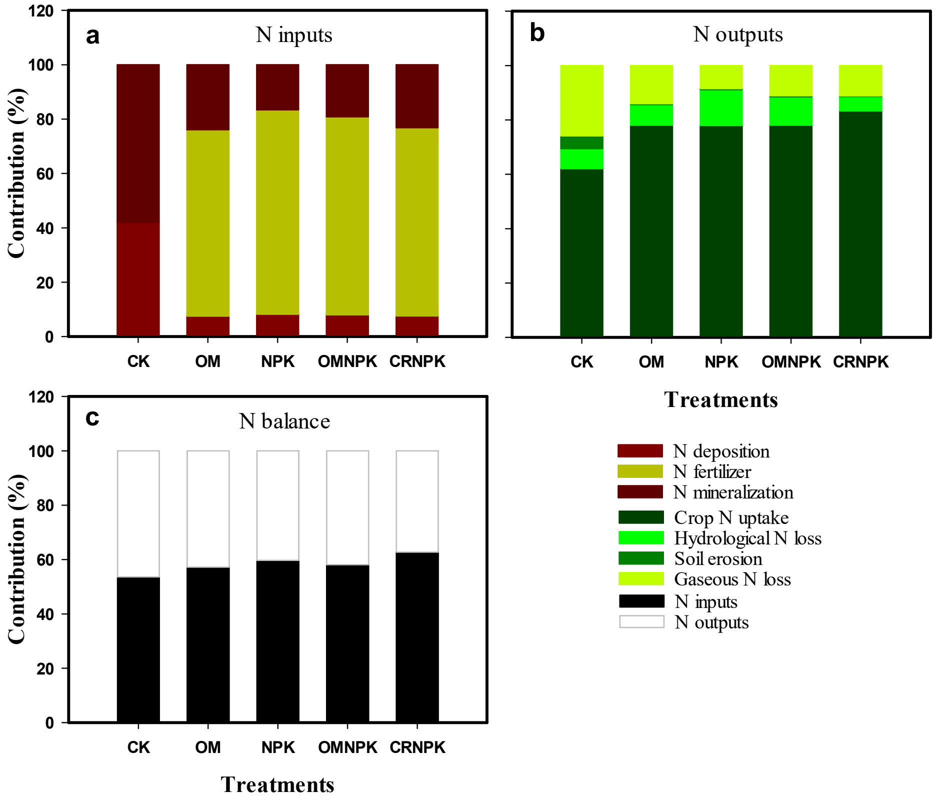

Figure 6. Annual contribution of N balance components to N balance inputs (a) and outputs (b) and the contribution of inputs and outputs (c) for organic amendments

Table 3. Annual hydrological N loss from organic amendments for period of 2017-2018

Treatments	Hydrological N losses				
	PN (kgN ha^{-1})	NH$_4^+$-N (kg N ha^{-1})	NO$_3^-$-N (kg N ha^{-1})	DON (kg N ha^{-1})	Total (kg N ha^{-1})
Overland flow					
CK	0.68 ± 0.02a	0.03 ± 0.01a	0.13 ± 0.01d	0.12 ± 0.00d	0.97 ± 0.05b
OM	0.26 ± 0.02e	0.02 ± 0.00ab	0.22 ± 0.01c	0.22 ± 0.00a	0.71 ± 0.01d
CR	0.18 ± 0.01f	0.02 ± 0.00ab	0.16 ± 0.03d	0.10 ± 0.00e	0.46 ± 0.02e
NPK	0.55 ± 0.01b	0.03 ± 0.00a	0.36 ± 0.00a	0.14 ± 0.00c	1.08 ± 0.01a
OMNPK	0.46 ± 0.00c	0.02 ± 0.00ab	0.23 ± 0.00c	0.18 ± 0.00b	0.89 ± 0.00c
CRNPK	0.33 ± 0.02d	0.01 ± 0.00c	0.31 ± 0.01b	0.11 ± 0.01e	0.77 ± 0.01d
Interflow					
CK	0.24 ± 0.01e	0.09 ± 0.00b	3.11 ± 0.23e	0.41 ± 0.10e	3.85 ± 0.30f
OM	3.25 ± 0.01c	0.08 ± 0.01b	12.92 ± 0.29c	6.41 ± 0.13c	22.67 ± 0.20c
CR	0.35 ± 0.01e	0.08 ± 0.01b	9.22 ± 0.54d	0.59 ± 0.12e	10.24 ± 0.44e
NPK	6.39 ± 0.09a	0.11 ± 0.00a	18.99 ± 0.27a	6.78 ± 0.08a	32.27 ± 0.38a
OMNPK	5.10 ± 0.01b	0.08 ± 0.01b	15.50 ± 0.28b	7.44 ± 0.01b	28.12 ± 0.28b
CRNPK	0.52 ± 0.02d	0.09 ± 0.00b	8.78 ± 0.10d	2.60 ± 0.02d	11.98 ± 0.13d

PN, particulate nitrogen, NH$_4^+$-N, ammonia nitrogen, NO$_3^-$-N, nitrate nitrogen and DON, dissolved organic nitrogen.

This could be explained by the higher crop N uptake, hydrological N loss and gaseous N loss during the summer maize season. Moreover, the annual N balance ranges from 9.9 to 164.2 kg N ha^{-1} across all treatments. Furthermore, for the N fertilizer contribution to the N balance inputs accounted for 0 to 75% across all treatments (Fig 6a), while the crop N uptake contribution to the N balance outputs accounted for 62 to 83% for all treatments (Fig. 6b). Meanwhile, the inputs and outputs contribution to N balance accounted for 54 to 63% and 37 to 46% respectively across all treatments (Fig. 6c). While gaseous N loss contributed about 8 to 26% and hydrological N loss about 5 to 13% across all treatments. Furthermore, the results showed that N fertilizer, soil N supply, and crop N uptake were the major contributors to the N balance.

DISCUSSION

Effects of organic amendments on NOx emissions: The NOx emissions fluxes are significantlyinfluenced by different organic amendments (Cui *et al*., 2012; Hu *et al*., 2017). Consistent with the report of Hu *et al.* (2017), average cumulative NOx fluxes over the yearly experiment were significantly lower during the wheat season, ranging from -0.01 to 0.8 kg N ha^{-1}, andhigher during the maize season, ranging from 0.02 to 0.94 kg N ha^{-1} for all experimental treatments and differed significantly from each other (Table 1). Results from this study showed that organic amendments reduced NOx fluxes in the wheat growing season but not in the maize season. Annual cumulative NOx fluxes in this study were 1.21 kg N ha^{-1} for NPK, 1.27 kg N ha^{-1} for OM, 0.83 kg N ha^{-1} for OMNPK and 0.42 kg N ha^{-1} for CRNPK were higher than the annual cumulative NOx fluxes of 0.11 kg N ha^{-1} for NPK, 0.38 kg N ha^{-1} for OM, 0.27 kg N ha^{-1} for OMNPK and 0.10 kg N ha^{-1} for CRNPK, as those reported by Hu *et al.* (2017) in a similar study site conditions over two-year experiment. In contrast, the annual mean NOx fluxes in this study were lower than the annual NOx emissions of 3.0 kg N ha^{-1}reported by Cui *et al*., 2012) for wheat-maize rotation systems under calcareous soil in the North China Plain. The difference between these results could be attributed to the difference in environmental factors, nitrogen application rate and soil properties as reported by several previous studies (Bouwman *et al*., 2002; Bateman and Baggs, 2005; Stavrakou *et al*., 2008). Interestingly, CRNPK treatment significantly reduce NOx flux by 38 and 80% in the wheat and the maize seasons compared to NPK treatment. This could be explained by the crop residues water holding capacity and poor soil aeration, which stimulated the soil microbial respiration and growth (Zhang *et al*., 2020).

Effects of organic-instead fertilization on NH_3 volatilization: The NH_3 volatilization is influenced by organic amendments (Table 1, Fig. 5).Consistent with the report of Zhang et al. (2015); average cumulative NH_3 fluxes over one year experiment were significantly lower for the wheat season, ranging from 3.9 to 10.53 kg N ha^{-1}, andhigher for the maize season, ranging from 12.0 to 20.5 kg N ha^{-1} for all treatments,and differed significantly from each other (Table 1). Zhang et al. (2020) reported that NH_3 volatilization in cropland of EutricRegosol is serious during summer maize season.Results from this study showed that CRNPK treatment significantly increased NH_3 fluxes during the wheat and the maize seasons compared to other organic amendments treatments (Table 1). The urea applied in our study had significant N loss during maize season, this is in agreement with Cao et al. (2013), who reported that urea could be lost as NH_3 volatilization under higher temperature and watered soil conditions. Moreover, fresh pig slurry application showed higher NH_3 flux than those other treatments. In contrast to our results, Huijsmans et al. (2003) previously reported that NH_3 flux from organic manure can be lower due to a certain amount of NH_3 lost during manure storage before field application. Furthermore, annual cumulative NH_3 flux was significantly differed between the organic amendments. Further study on different crop residue combination with mineral fertilizers are needed for understanding the full mechanisms involved. However, recent studies have reported contradictory effects of crop residues returning on NH_3 flux in agricultural soils (Xia et al., 2018; Li et al., 2019).

Effects of organic amendments on N balance: Based on the N balance components, we determined the N balance by computing the difference between N inputs and outputs as suggested by Ju et al. (2009). Seasonal and annual N balance for CRNPK was significantly greater than those other treatments ($p< 0.05$), with no significant difference observed between OM, NPK and OMNPK treatments ($p> 0.05$) (Table 4). Our study annual N balance ranged from 103.1 to 164.2 kg N ha^{-1}, and were higher than those of 28.6 to 72.9 kg N ha^{-1}reported by Zhou et al., (2016) in a similar condition. Our results showed that organic amendments of CRNPK waspromising measures for N retention in cropland of Eutric Regosols. While, mineral fertilizers application resulted to a low N balance both in the wheat and the maize seasons leading to significant N loss in the environment.In agreement with Zhou et al., (2016), N deposition, N fertilizer and N supply were contributed to the annual N balance inputs about 8%, 69% and 23%, respectively, across all organic amendments except CR treatment on average (Fig. 6). While crop N uptake, hydrological N loss and gaseous N loss contribution to the annual N balance outputs were, about 79%, 9%, and 11%, respectively, on average. Moreover, the annual N balance inputs and outputs contribution to the annual N balance was about 59% and 41%, respectively on average (Fig. 6). Overall, determining N balance can be useful for understanding the mechanisms of organic amendments on coupling N supply and retention in cropland of EutricRegosol, and could have significant implication in N loss mitigation control in cropland of Eutric Regosols ecosystems (Blesh and Drinkwater, 2013; Wang et al., 2015; Zhou et al., 2016).Further multiyear monitoring of total gaseous N loss and total hydrological is need for temporal and spatial heterogeneity N balance analysis to support the present results.

CONCLUSION

This study quantified the N balance, which were in the range of 38.9 to 160.8 kg N ha^{-1} for the wheat season, 14.2 to 97.6 kg N ha^{-1} for the maize season, and 9.9 to 164.2 kg N ha^{-1} annually across the different organic amendments. The N balance results indicated that reactive gaseous N loss and hydrologicalN loss mitigation measures could be more focus during summer maize season in cropland of Eutric Regosol. Overall, regular mineral N fertilization regime resulted in significant reactive N loss leading to a low N balance. The results showed that both gaseous N loss exhibited higher daily flux during maize growing season than the wheat season indicating that mitigation measures could be more focus during summer maize season for reducing both N loss in cropland of EutricRegosol. The results indicated that mineral fertilizer applications resulted to a low N balance leading to significant N loss into the environment. While the organic amendmentscould be recommended for N supply and sustaining crop yields in cropland of Eutric Regosols.

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