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The influences of organic fertilizers produced by different production techniques on rice grain yield and ammonia volatilization in double-cropping rice fields

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ABSTRACT

Ammonia (NH₃) volatilization from rice cultivation contributes to poor air quality and is indicative of low nitrogen use efficiency in farming. Organic fertilizers could meet the nitrogen requirement for rice growth yet, the simultaneous effects of organic fertilizers on ammonia volatilization and yield in rice paddy fields are poorly understood and quantified. To help address this knowledge gap, experimental field plots were constructed in a conventional double-cropping rice paddy fields in the Pearl River Delta, China. Five treatments were used in addition to a no fertilizer control: fresh organic fertilizer, successively composted organic fertilizer, chemical composted organic fertilizer, chemical composted organic fertilizer with inorganic fertilizer, and chemical fertilizer. NH₃ volatilization was measured using a batch-type airflow enclosure method. The results showed no significant differences in grain yield between the different organic fertilizer and conventional chemical fertilizer treatments yet, when compared with the use of chemical fertilizer, the total NH₃ volatilization significantly decreased with the use of a chemical composted organic fertilizer (70% reduction) and a successively composted organic fertilizer (68% reduction). The ammonium (NH₄⁺-N) concentration in the field surface water had a strong positive correlation with NH₃ volatilization for all fertilization treatments ($P < 0.01$). The outcome demonstrates chemical composted organic fertilizer can maintain rice yield and reduce NH₃ volatilization, and an important next step is to up-scale these field-based measurements to similar rice cultivation areas to quantify the regional and national-scale impact on air quality and nitrogen deposition to sensitive areas, and fertilizer best practice.

Key Words: air quality, crop production, manure, NH₃ emission, nitrogen loss

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INTRODUCTION

Rice (*Oryza sativa* L.) is one of the world's most important crops being a dietary staple for more than half of the world's population (Bertora *et al.*, 2018). Rice cultivation covers approximately 1.5×10^8 hectares of the Earth's land surface which is about 10% of all the land cultivated across the globe (Jiang *et al.*, 2018). In China, which produces and consumes the most rice of any nation, the annual rice-planting area is approximately 30 million hectares, which is 23% of the world's rice growing area (Feng *et al.*, 2017; Min *et al.*, 2021b). The application of chemical fertilizers has increased rice production and accelerated economic

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development, yet the long-term excessive fertilizer application has caused multiple issues (Lin *et al.*, 2007). This include worsening soil health, including the deterioration of soil texture and a decreasing organic matter content, and excess nitrogen transfer from the plant-soil system to the atmosphere and freshwater and marine ecosystems contributing to poor air quality, and soil acidification and water eutrophication issues (Kyaw *et al.*, 2005; Hayashi *et al.*, 2008). Improving nitrogen use efficiency in rice production is important to enable food security whilst improving air and water quality. Rice paddy soil is a significant source of ammonia (NH_3) volatilization especially after fertilizer addition (Ma *et al.*, 2021). NH_3 volatilization is the process of ammonia (NH_4^+) transformation to NH_3 in the soil system and subsequent atmospheric diffusion (Wang *et al.*, 2007; He *et al.*, 2018). NH_3 volatilization is not only a nitrogen loss pathway from the soil to the air but enhances atmospheric nitrogen deposition which can adversely impact sensitive ecosystems and cause poor air quality. When NH_3 enters the atmosphere, it can react with atmospheric acidic compounds to produce NH_4HSO_4 and other secondary pollutants. Through atmospheric transport and deposition, these pollutants can enter wetland, forest, lake, grassland and other ecosystems with risk of water eutrophication and soil and water acidification (Chatterjee *et al.*, 2018). NH_3 can also react with NO_x , SO_2 and VOCs (volatile organic compounds) and other substances in the air to generate NH_4NO_3 , $(\text{NH}_4)_2\text{SO}_4$ and other particles, which serve as condensation nucleus to further generate $\text{PM}_{2.5}$, thereby promoting the formation of atmospheric haze (Giltrap *et al.*, 2010; Liang *et al.*, 2017; Lee *et al.*, 2021).

China produces and consumes the most mineral N fertilizer globally, and the application rate of nitrogenous fertilizer accounts for approximately one third of the whole amount of nitrogenous fertilizer used worldwide (Zhao *et al.*, 2020). The nitrogen utilization efficiency of nitrogen is relatively low, with about two thirds of the nitrogen applied by fertilizer lost through multiple ways, and NH_3 volatilization is key transfer pathway from the plant-soil system to the atmosphere (Yin *et al.*, 2020). In 2016, the annual NH_3 emission in China reached 12.11 million tons with a mean emission intensity of 1.28 t km^{-2} and NH_3 emission from fertilizer application contributed 38% (Li *et al.*, 2021). Fertilizer type, dosage and application practice determine NH_3 volatilization in paddy fields with volatilization loss rates of ammonium sulfate and ammonium bicarbonate generally between 30% to 70%, and between 20% to 80% for urea application (Islam *et al.*, 2018). NH_3 volatilization wastage in paddy fields generally increases with nitrogen application rate (Ding *et al.*, 2020) whilst fertilizer injection to depth and irrigation after fertilization have been demonstrated as effective for inhibiting NH_3 volatilization in paddy fields (Liu *et al.*, 2020; Zhong *et al.*, 2021).

Due to the issues caused by mineral fertilizer over-application, organic fertilizer use is receiving more attention in part motivated as a means of using a waste product to replace mineral fertilizer application (Sun *et al.*, 2020). Organic fertilizers, primarily made of plant waste and animal manures sometimes mixed with straw, may also include food waste and solid residue from biogas production (Su *et al.*, 2014). Compared with chemical fertilizer, organic fertilizers have many positive effects on rice paddies including increasing soil carbon and nitrogen, improving the soil physical structure, and enhancing crop yield and quality (Dillon *et al.*, 2012; Zhao *et al.*, 2014).

At the field plot scale, NH_3 volatilization is highly variable and dependent on multiple factors including the raw materials used in the production of the organic fertilizer, application rate and fertilization period (Wu *et al.*, 2019; Sun *et al.*, 2020; Min *et al.*, 2021a). However, there are few reports on the effects of organic fertilizers, produced by different manufacturing techniques, on NH_3 volatilization in rice paddy fields and of those that are reported, they are primarily concentrated in the eastern and central regions in China, with studies in southern China relatively rare (Xu *et al.*, 2021). Ammonia volatilization from paddy fields has different regional emission characteristics and depends on organic fertilizer type, application amount and soil type (Paulot *et al.*, 2014; Li *et al.*, 2021). Therefore, the aim of this study is to quantify the effects of organic fertilizers, with different production processes, on rice grain yield and ammonia volatilization in the paddy

fields in southern China. To achieve the aim, two objectives were defined. The first was to quantify the effects of chemical fertilizer and various organic fertilizers, all with an identical equivalent nitrogen application amount, on rice yield, NH_3 volatilization and surface water mineral N (ammonium, $\text{NH}_4^+\text{-N}$ and nitrate, $\text{NO}_3^-\text{-N}$) concentrations in a double-cropping rice field in the Pearl River Delta. The second objective was to consider the outcomes in terms of soil fertility improvement, N management strategy and development of environment-friendly organic fertilizers.

MATERIALS AND METHODS

Experimental sites

The experimental field was located in Huizhou, Guangdong Province, China ($114^\circ 5' \text{ E}$, $23^\circ 1' \text{ N}$) (Fig. S1) (see Supplementary Material for Fig. S1). Huizhou has a subtropical monsoon climate, with an average annual temperature of 22°C and an average annual precipitation of 2 200 mm (Fig. 1). Typhoon weather frequently occurs in July and August. Huizhou is the primary rice cropping area of China's Pearl River Delta, in which the main rice cultivation pattern is double-cropping, mainly comprising rice-rice-vegetable, rice-rice-potato or rice-rice-winter fallow. The soil is an Anthrosol with pH (H_2O) of 5.8, organic matter of 18.6 g kg^{-1} soil, total N, p and K of 1.1, 0.9 and 18.9 g kg^{-1} soil, respectively, for mixed samples within the 0--20 cm depth.

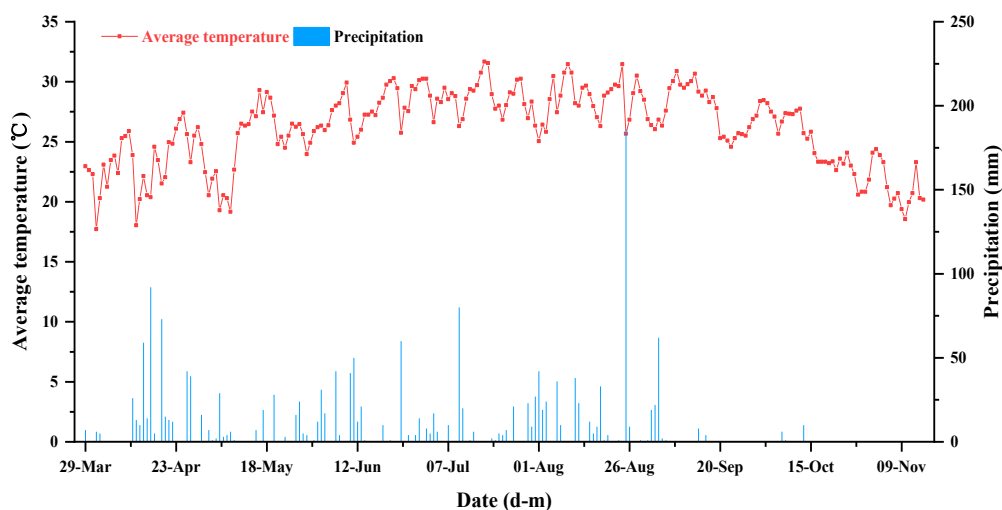


Fig. 1 The dynamic variation of precipitation and average temperature during the rice growth seasons in 2019.

Fertilizer manufacture

Chicken manure was selected as the production raw material for the organic fertilizers. Two traditional, and a recent rapidly composting technology, were selected as the fertilizer manufacture methods. The two traditional organic fertilizer manufacturing techniques contained fresh organic fertilizer (FOF) and successively composted organic fertilizer (SOF). The FOF was fresh chicken manure only, processed with appropriate physical treatments for convenient transportation and application. SOF was produced by a conventional strip pile composting process. Successive composting has been shown to increase mineralization and humification of the organic raw materials, so that the nutrients present can be more easily absorbed and utilized by crops (Wang *et al.*, 2017; Feng *et al.*, 2020). In addition, a late-model rapidly composting technology produced organic fertilizer by the addition of a chemical decomposition agent to the organic raw materials (chemical composted organic fertilizer, COF) to promote physicochemical

decomposition, and then the manure was heat treated at a high temperature. Heat treatment kills the pathogenic microorganisms present, and the decomposition rapidly degrades cellulose, hemicellulose and other substances shortening the ripening time of the organic materials. Finally, a chemical composted organic fertilizer with inorganic fertilizer (COIF) was produced by adding chemical fertilizer to the COF.

Experimental design and field management

In addition to the control plot where no fertilizer (CK) was applied, the five fertilizer treatments were applied to other plots: chemical fertilizer (CF), fresh organic fertilizer (FOF), successively composted organic fertilizer (SOF), chemical composted organic fertilizer (COF), and chemical composted organic fertilizer with inorganic fertilizer (COIF). The treatments of chemical fertilizer and organic fertilizers had the same nitrogen application rates of 105 kg pure N ha⁻¹. The area of each plot was 7.5 m × 5.7 m = 42.75 m², with 4 replicates per treatment. All experimental plots were arranged randomly. To prevent water and fertilizer moving between adjacent plots, cement ridges were constructed between plots. Chemical fertilizer was divided into basal and tillering fertilizer. Organic fertilizers were applied once only as basal fertilizer and no topdressing was applied (Table I).

TABLE I

Fertilization treatments in each experimental field plot

| Treatment | Basal fertilization | Supplemental fertilization |
|--|---|----------------------------|
| No fertilizer (CK) | kg ha ⁻¹ | kg ha ⁻¹ |
| Chemical fertilizer (CF) | Urea (nitrogen content: 46%) 90, 15-15-15 compound fertilizer 240 | urea 60 |
| Fresh organic fertilizer (FOF) | FOF (nitrogen content: 4.43%) 7156 | / |
| Successively composted organic fertilizer (SOF) | SOF (nitrogen content: 1.63%) 7096 | / |
| Chemical composted organic fertilizer (COF) | COF (nitrogen content: 2.35%) 6058 | / |
| Chemical composted organic fertilizer with inorganic fertilizer (COIF) | COIF (nitrogen content: 9%) 1299 | / |

The basal fertilizer of early-season rice was applied on March 29, and the seedlings were transplanted on March 31, 2019. The seedling variety was Meixiangzhan 2 which has a growing period of approximately one month. The seedling row spacing was approximately 20 cm. The early-season rice was harvested on July 15, 2019. The late-season rice received basal fertilizer on August 8, was transplanted on August 10, 2019 and harvested on November 15, 2019. The transplant, harvest and field management methods were the same for both the early- and late-season rice, and field management followed the conventional practice of local farmers. Before the experiment, the experimental plots were flooded for one to two weeks, and then cultivated and harrowed, following the application of basal fertilizer and transplanting of rice seedlings. About one month after transplanting, the field surface was in a state of flood, and then there was one to two weeks of mid-season drainage. Following this, the field surface would again return to a state of flood, and the rice field water was then drained and the plots dried in the last ten days or so of rice yellow maturity stage, which allowed the rice to be harvested (Fig. S2) (see Supplementary Material for Fig. S2).

The batch-type airflow enclosure method

The batch-type airflow enclosure method, which is the national standard method, was used to measure

NH₃ volatilization (Dong *et al.*, 2019) (Fig. S3) (see Supplementary Material for Fig. S3). The method uses boric acid to absorb the NH₃ volatilized from the rice field, and then the absorption solution is titrated with dilute sulfuric acid to obtain the volume of dilute sulfuric acid consumed by titration. The ammonia volatilization flux was calculated according to the sulfuric acid volume as Eq. 1 (Zhong *et al.*, 2021):

$$F = V \times 10^{-3} \times C \times 0.014 \times 10^4 \times \pi^{-1} \times r^{-2} \times 6 \quad (1)$$

where F is the ammonia volatilization flux (kg N ha⁻¹ d⁻¹), V is volume of sulfuric acid for titration (mL), 10^{-3} is volume conversion factor, C is calibration concentration of sulfuric acid for titration (mol L⁻¹), 0.014 is relative atomic mass of nitrogen atom (kg mol⁻¹), 10^4 is area conversion factor, r is radius of air chamber (m), and 6 is the ratio of 24 h to daily ammonia volatilization collection time.

Measurement of NH₃ volatilization and surface water sampling and analysis

The field plot NH₃ volatilization fluxes were measured, on each day after fertilization, until no difference was observed in the measurements between the fertilized plots and the non-fertilized control for more than four consecutive days. During the experiment, NH₃ gas samples were collected, starting at 0700--0900 and 1500--1700 daily, for 4 h, and the daily NH₃ volatilization flux was calculated based on the NH₃ volatilization with the 4 h measured each day. Total NH₃ volatilization was calculated by summing the daily volatilization fluxes for the experiment duration which was 38 d for the early-season rice and 34 d for the late-season rice.

Field surface water samples were collected throughout both the early- and late-season rice and a five-point sampling method was used. Briefly, 100 mL of surface water was collected from five points in each plot to produce a composite sample. The surface water was sampled on days 1, 2, 3, 6, 10, 15, 21, 27 and 38 after the application of basal fertilizer for the early-season rice. For the late-season rice, the sampling frequency of surface water sampling was once a day after the application of basal fertilizer, and 34 samples were taken. On each sampling day, the water samples were transported promptly to the laboratory for measurement of the NH₄⁺-N and NO₃⁻-N concentration and the pH.

Statistical analyses

The least significant difference (LSD) method of one-way analysis of variance (ANOVA), implemented in the IBM SPSS Statistics 21.0 software, was used to analyze the significance of any differences in rice yield, NH₃ volatilization flux, and cumulative NH₃ volatilization among the five different treatments and with the control plot.

RESULTS

Rice yields

The annual rice yield was largest with the FOF, followed by COIF, CF, COF, and SOF, and least in CK (Table II). The fertilization treatments significantly enhanced rice yields ($P < 0.05$), for both the early- and the late-season rice, compared to the non-fertilized control. For the early-season rice, except for the SOF treatment, the FOF, COF and COIF treatments increased early-season rice yields by 4.0%, 3.1% and 2.0% in comparison with the CF treatment ($P > 0.05$). The COIF and FOF treatments increased late-season rice yield by 4.2% and 2.6% compared with the CF treatment ($P > 0.05$). In general, the yield of the late-season rice was higher than the early-season rice, and for the rice annual yield, there was no significant difference between the chemical fertilizer treatment and the four organic fertilizer treatments (Table II).

TABLE II

Rice yields in different fertilization treatments

| Treatment ^{a)} | Rice yield ^{b)} | | |
|-------------------------|--------------------------|---------------------|---------------------|
| | Early-season rice | Late-season rice | Whole year |
| | kg ha ⁻¹ | kg ha ⁻¹ | kg ha ⁻¹ |
| CK | 3176 ± 245b | 3799 ± 169c | 6975 ± 228b |
| CF | 4902 ± 233a | 6355 ± 636a | 11257 ± 401a |
| FOF | 5088 ± 141a | 6529 ± 290a | 11618 ± 323a |
| SOF | 4821 ± 156a | 5546 ± 319b | 10367 ± 276a |
| COF | 5038 ± 198a | 5490 ± 171b | 10527 ± 454a |
| COIF | 4975 ± 304a | 6623 ± 140a | 11598 ± 353a |

^{a)}CK, no fertilizer; CF, chemical fertilizer; FOF, fresh organic fertilizer; SOF, successively composted organic fertilizer; COF, chemical composted organic fertilizer; COIF, chemical composted organic fertilizer with inorganic fertilizer;

^{b)}Different small letters following the value meant significant difference among treatments at 5% level.

NH₃ volatilization flux from double-cropping rice field

NH₃ volatilization fluxes were notably higher for the fertilizer treatments than for the non-fertilized control in the early-season rice, with the NH₃ volatilization occurring mainly within one week of basal fertilization (Fig. 2A). The NH₃ volatilization fluxes peaked on the first day after fertilization and then gradually decreased on the second day and thereafter. A second NH₃ volatilization flux peak appeared on the sixth day after the fertilization, and then gradually decreased to the background level. There was no obvious peak in NH₃ volatilization flux after tillering fertilization with chemical fertilizer on April 16, 2019. Only two small NH₃ volatilization flux peaks occurred 20 and 26 d after fertilization with COIF and SOF. Among all treatments, the peak NH₃ volatilization flux was the highest for the COIF treatment, followed by COF, CF, FOF, and SOF, and least for the control (CK) (Fig. 2A). Late-season rice NH₃ volatilization was also concentrated mainly in the week after basal fertilization, which was consistent with the volatilization emission timing of the early-season rice (Fig. 2B). NH₃ volatilization fluxes peaked on the first day for COIF, on the second day for FOF, COF, and SOF, and on the third day for CF after the fertilization of the late-season rice (Fig. 2B). The NH₃ volatilization fluxes dropped on the fourth day (August 11, 2019) for all treatments due to a heavy rainstorm. After the tillering chemical fertilizer was added on September 1, 2019, the NH₃ volatilization flux increased rapidly, peaked on the second day after the supplemental fertilization, and then declined to a background level. Of the five treatments to the late-season rice, the peak NH₃ volatilization fluxes were highest for COIF, followed by CF, FOF, COF, SOF and then the CK (Fig. 2B).

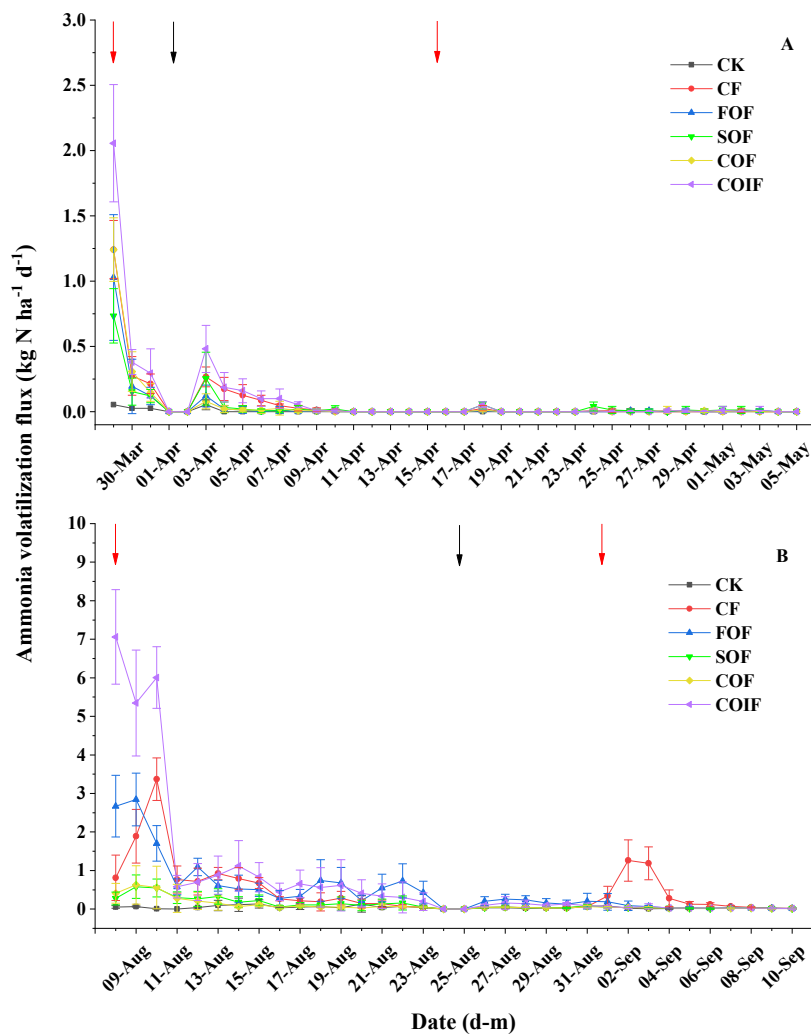


Fig. 2 Ammonia volatilization flux from field plots receiving chemical fertilizer and organic fertilizers in the early-season rice (A) and the late-season rice (B) growing periods in 2019 (CK: no fertilizer; CF: chemical fertilizer; FOF: fresh organic fertilizer; SOF: successively composted organic fertilizer; COF: chemical composted organic fertilizer; COIF: chemical composted organic fertilizer with inorganic fertilizer). The red arrow indicates fertilization, and the black arrow indicates typhoon that caused the experiment to be suspended.

Cumulative NH₃ volatilization and NH₃ volatilization intensity in paddy fields

In the early-season rice growing period, the cumulative NH₃ volatilization was highest for the COIF treatment, followed by CF, COF, FOF, SOF and CK (Fig. 3A). The four organic fertilizer treatments and the chemical fertilizer treatment significantly increased cumulative NH₃ volatilization compared with the non-fertilized control ($P < 0.01$), however, compared with the CF treatment, the cumulative NH₃ volatilization decreased by 24.9%, 38.5% and 38.7% in the COF, FOF and SOF treatments respectively ($P < 0.05$), and increased by 54.9% ($P < 0.05$) in COIF treatment (Fig. 3A). The cumulative NH₃ volatilization was greatest in the COIF treatment, followed by FOF, CF, SOF and COF, and least in the non-fertilized control in the late-rice growing season. The cumulative NH₃ volatilization significantly increased after application of the COIF, FOF and CF compared with the non-fertilized control ($P < 0.01$). The cumulative NH₃ volatilization in the SOF and COF treatments significantly decreased by 72.7% ($P < 0.01$) and 77.6% ($P < 0.01$) compared with the CF treatment. In contrast, the cumulative NH₃ volatilization in COIF treatment

significantly increased by 79.8% ($P < 0.01$) (Fig. 3B).

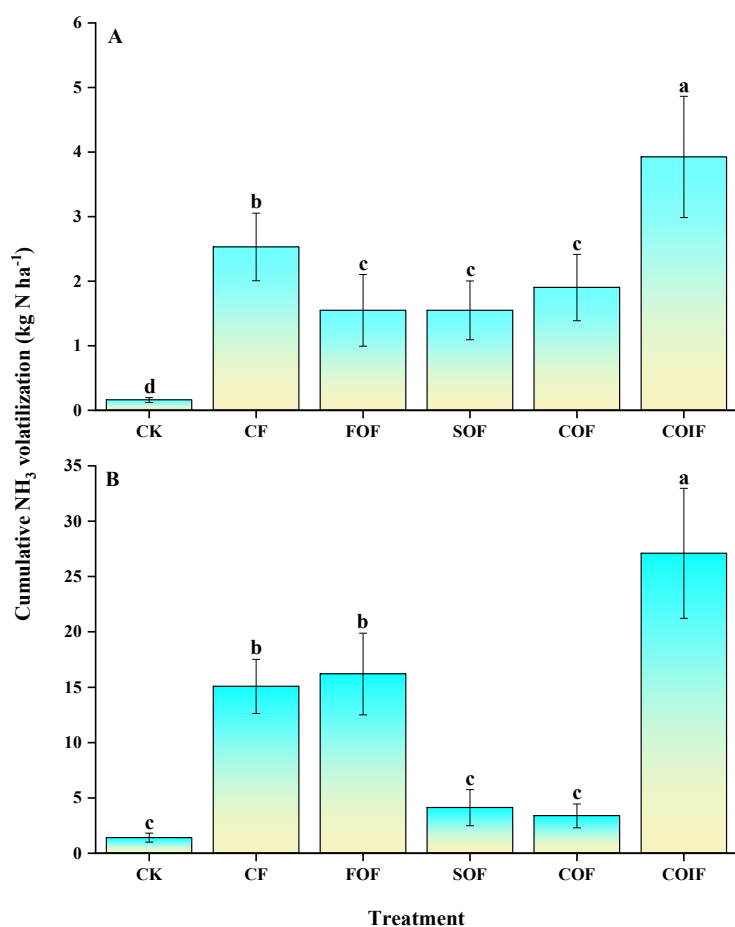


Fig. 3 Cumulative ammonia volatilization from field plots receiving chemical fertilizer and organic fertilizers in the early-season rice (A) and the late-season rice (B) growing periods in 2019 (CK: no fertilizer; CF: chemical fertilizer; FOF: fresh organic fertilizer; SOF: successively composted organic fertilizer; COF: chemical composted organic fertilizer; COIF: chemical composted organic fertilizer with inorganic fertilizer). Different small letters meant significant difference among treatments at 5% level.

The combined effect of different fertilizers on rice yield and NH₃ volatilization was evaluated by calculating the NH₃ volatilization flux per unit rice yield (Table III). For all the late-season rice treatments, the volatilization flux per unit rice yield was much higher than that for the early-season rice. The total NH₃ volatilization intensity of the early- and the late-season rice was highest for the COIF treatment, followed by CF, FOF, SOF, and COF, and least in the non-fertilized control. The NH₃ volatilization intensity for the early- and the late-season rice decreased by 1.9% ($P > 0.05$), 64.7% ($P < 0.01$) and 67.9% ($P < 0.01$) for the FOF, SOF and COF treatments respectively compared with the CF treatment.

TABLE III

| Cumulative ammonia volatilization and volatilization intensity of ammonia in different fertilization treatments | | | | | | |
|---|---|-----|------------|----------------------------------|-----|------------|
| Treatment ^{a)} | Cumulative ammonia volatilization ^{b)} | | | Ammonia volatilization intensity | | |
| | ESR ^{c)} | LSR | Whole year | ESR | LSR | Whole year |
| | kg N ha ⁻¹ | | | | | |

| | | | | | | |
|------|--------------|---------------|---------------|---------------|--------------|--------------|
| CK | 0.16 ± 0.03d | 1.41 ± 0.24c | 1.57 ± 0.29d | 0.05 ± 0.01c | 0.37 ± 0.05c | 0.23 ± 0.06d |
| CF | 2.53 ± 0.32b | 15.07 ± 3.46b | 17.60 ± 3.51b | 0.52 ± 0.05ab | 2.37 ± 0.31b | 1.56 ± 0.28b |
| FOF | 1.55 ± 0.19c | 16.20 ± 3.15b | 17.75 ± 3.68b | 0.30 ± 0.02b | 2.48 ± 0.38b | 1.53 ± 0.35b |
| SOF | 1.55 ± 0.27c | 4.11 ± 0.62c | 5.66 ± 0.63c | 0.32 ± 0.04b | 0.74 ± 0.06c | 0.55 ± 0.06c |
| COF | 1.90 ± 0.22c | 3.38 ± 0.49c | 5.28 ± 0.52c | 0.38 ± 0.03b | 0.62 ± 0.04c | 0.50 ± 0.05c |
| COIF | 3.92 ± 0.47a | 27.09 ± 5.95a | 31.01 ± 6.24a | 0.79 ± 0.12a | 4.09 ± 0.81a | 2.67 ± 0.75a |

^{a)}CK, no fertilizer; CF, chemical fertilizer; FOF, fresh organic fertilizer; SOF, successively composted organic fertilizer; COF, chemical composted organic fertilizer; COIF, chemical composted organic fertilizer with inorganic fertilizer; ^{b)}Different small letters following the value meant significant difference among treatments at 5% level; ^{c)}ESR, the early-season rice; LSR, the late-season rice.

Relationship between NH₃ volatilization flux and NH₄⁺-N, NO₃⁻-N concentration and pH in field surface water

The NH₃ volatilization flux increased with higher NH₄⁺-N concentrations in the field surface water. For both rice growing seasons, the changes in the surface water NH₄⁺-N concentrations for the different treatments had a linear relationship with the NH₃ volatilization flux changes (Fig. 4). The maximum NH₄⁺-N concentration in field surface water appeared within one week of basal fertilization. Similarly, the field surface water NH₄⁺-N concentration increased significantly after the application of tillering fertilizer in the CF treatment, and then decreased gradually (Fig. 5A and 5B). All chemical and organic fertilizer applications significantly increased the surface water NH₄⁺-N concentration compared with the non-fertilized control for both the early- and the late-season rice. A linear regression analysis showed a strong correlation between ammonia volatilization flux and surface water NH₄⁺-N concentration for both chemical and organic fertilizer treatments. Except for the control plot for late-season rice, the *P* values of all other treatments were less than 0.01, and the correlation coefficient *r* was between 0.72 and 0.99 (Fig. 4).

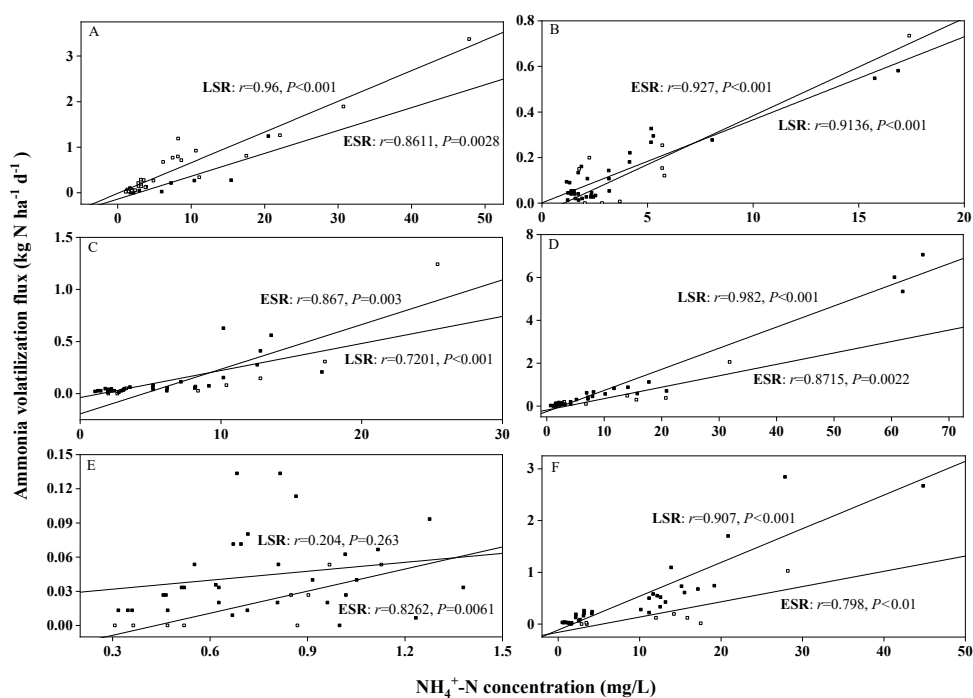


Fig. 4 The relationships between ammonia volatilization flux and NH₄⁺-N concentration in field surface water as determined by linear regression analysis (A, chemical fertilizer; B, successively composted organic fertilizer; C,

chemical composted organic fertilizer; D, chemical composted organic fertilizer with inorganic fertilizer; E, no fertilizer; F, fresh organic fertilizer). Hollow symbols denote the early-season rice (ESR) and solid symbols denote the late-season rice (LSR).

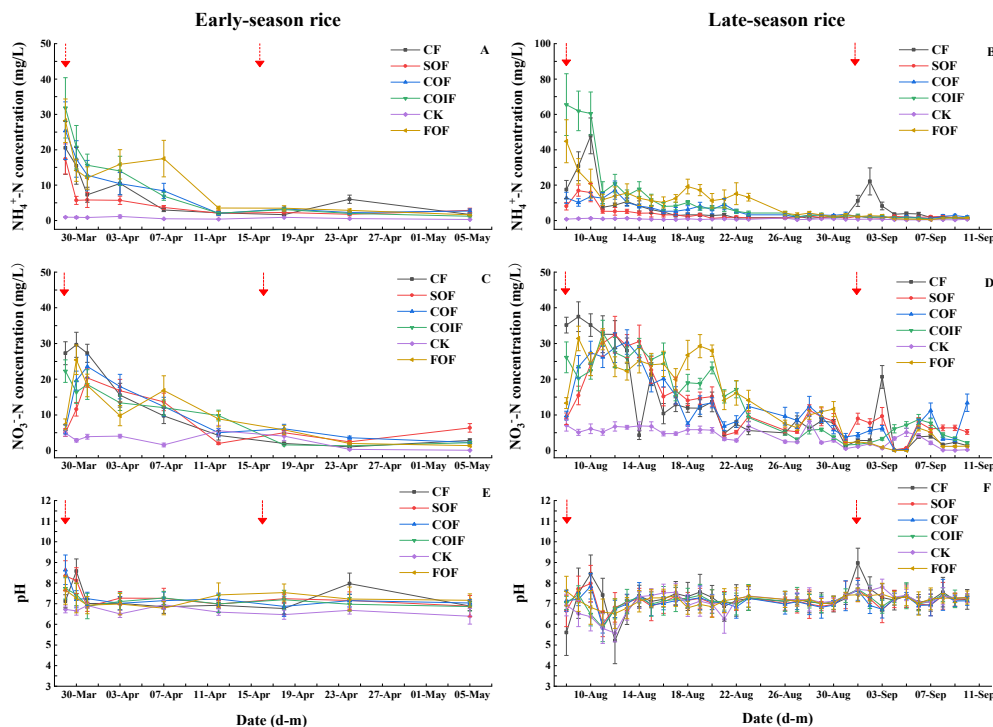


Fig. 5 $\text{NH}_4^+\text{-N}$ concentration, $\text{NO}_3^-\text{-N}$ concentration and pH values in field surface water for the early-season rice and the late-season rice growing periods in 2019 (CK: no fertilizer; CF: chemical fertilizer; FOF: fresh organic fertilizer; SOF: successively composted organic fertilizer; COF: chemical composted organic fertilizer; COIF: chemical composted organic fertilizer with inorganic fertilizer). The red arrow indicates fertilization.

The field surface water $\text{NO}_3^-\text{-N}$ concentration for both early- and late-season rice increased rapidly after basal fertilization, with a peak that occurred within one week of fertilization, and then decreased gradually (Fig. 5C and 5D). Compared with the non-fertilized control, the application of chemical fertilizer and organic fertilizers significantly increased the surface water $\text{NO}_3^-\text{-N}$ concentration. In contrast to the rapid decrease of surface water $\text{NH}_4^+\text{-N}$ concentration, the $\text{NO}_3^-\text{-N}$ concentration remained high until two weeks after fertilization. Correlation analysis indicated that ammonia volatilization and $\text{NO}_3^-\text{-N}$ concentration in field surface water were significantly positively correlated ($P < 0.05$) in all treatments for the late-season rice, and significantly positively correlated only for the COIF treatment ($P < 0.05$) for the early-season rice (Table IV).

TABLE IV

Correlation between ammonia volatilization flux and $\text{NH}_4^+\text{-N}$ concentration, $\text{NO}_3^-\text{-N}$ concentration and pH in field surface water, air temperature, soil temperature (5 cm), and precipitation during the early- and late-season rice growing periods

| Treat- ment ^{a)} | $\text{NH}_4^+\text{-N}$ | | $\text{NO}_3^-\text{-N}$ | | pH | | Precipitation | | Air temperature | | Soil temperature | |
|------------------------------|--------------------------|----------|--------------------------|--------|-------|-------|---------------|--------|-----------------|-------|------------------|---------|
| | ESR ^{b)} | LSR | ESR | LSR | ESR | LSR | ESR | LSR | ESR | LSR | ESR | LSR |
| CK | 0.826** | 0.204 | 0.474 | 0.482* | 0.078 | 0.071 | -0.109 | -0.119 | 0.004 | 0.052 | 0.236 | 0.225 |
| CF | 0.861** | 0.960*** | 0.637 | 0.692* | 0.036 | 0.223 | -0.072 | -0.191 | 0.003 | 0.203 | 0.237 | 0.451** |

| | | | | | | | | | | | | |
|------|----------|----------|--------|--------|---------|-------|--------|--------|-------|---------|-------|---------|
| FOF | 0.798** | 0.907*** | 0.012 | 0.533* | 0.345 | 0.107 | -0.014 | -0.089 | 0.01 | 0.452** | 0.162 | 0.570** |
| SOF | 0.914*** | 0.927*** | 0.109 | 0.624* | 0.752* | 0.177 | -0.104 | -0.013 | 0.104 | 0.417* | 0.118 | 0.649** |
| COF | 0.867** | 0.720*** | 0.043 | 0.560* | 0.964** | 0.097 | -0.025 | -0.024 | 0.015 | 0.433* | 0.167 | 0.627** |
| COIF | 0.872** | 0.982*** | 0.677* | 0.455* | 0.802** | 0.186 | -0.071 | -0.155 | 0.005 | 0.479** | 0.218 | 0.590** |

*, ** and *** indicate $P < 0.05$, $P < 0.01$ and $P < 0.001$.

^a)CK, no fertilizer; CF, chemical fertilizer; FOF, fresh organic fertilizer; SOF, successively composted organic fertilizer; COF, chemical composted organic fertilizer; COIF, chemical composted organic fertilizer with inorganic fertilizer;

^b)ESR, the early-season rice; LSR, the late-season rice.

The paddy surface water pH increased first and decreased later after basal fertilization in the early- and late-rice growing seasons (Fig. 5E and 5F). The pH increased from 5.6 on the first day, to 7.45 on the second day, and then to 8.45 on the third day, then gradually decreased after the basal application of chemical fertilizer for late-season rice (Fig. 5F). Higher pH for all organic fertilizer treatments occurred mostly in the first three days after the basal fertilization, and the NH_3 volatilization peak flux coincided with this stage for all the organic fertilizer treatments. The pH range of the field surface water was smaller for the organic fertilizer treatments than that of the chemical fertilizer treatment (Fig. 5E and 5F). The correlation between the NH_3 volatilization flux and pH in field surface water in early-season rice was higher than that in late-season rice, which was significantly correlated for SOF ($P < 0.05$), COF ($P < 0.01$) and COIF ($P < 0.01$) treatments (Table IV).

Effects of meteorological factors on NH_3 volatilization

The mean air temperature of ammonia volatilization experimental period in the early-season rice was 22.9 °C, which was lower compared to that for the late-season rice with 28.8 °C (Fig. 1). The soil temperature (5 cm depth) in the early-season rice experimental period was lower than that of late-season rice and the precipitation during the early-season rice experimental period was more continuous and frequent than that during the late-season rice. There were 29 rainy days of the 38 days for the early-season rice experimental period with a mean precipitation of 16.5 mm, and 18 rainy days of 34 with a mean precipitation of 14.1 mm during the late-season rice (Fig. S4) (see Supplementary Material for Fig. S4). The correlation analysis showed that there was a negative correlation between precipitation and NH_3 volatilization flux in the field experiments ($P > 0.05$). In contrast, there was a positive correlation between air temperature and NH_3 volatilization flux, which was significantly correlated for the SOF ($P < 0.05$), COF ($P < 0.05$), FOF ($P < 0.01$), and COIF ($P < 0.01$) treatments for late-season rice. Soil temperature and NH_3 volatilization flux also showed a positive correlation for the chemical fertilizer and organic fertilizer treatments ($P < 0.01$) for late-season rice (Table IV).

DISCUSSION

Effects of organic fertilizers on rice yields

The rice yields for the organic fertilizer treatments FOF, SOF and COF had no significant difference in rice yield when compared to the CF treatment ($P > 0.05$). This is a significant outcome, indicating that these three organic fertilizers have the potential to replace chemical fertilizer. In this study, FOF had a positive effect in maintaining rice yield, and this form of organic fertilizer has advantages of simplicity, convenience, and low cost in the production process (Dong *et al.*, 2019). However, the direct application of untreated manure may lead to the emission of harmful gases such as H_2S and CH_4 , the latter a greenhouse gas (Feng *et*

al., 2020). Moreover, untreated manure may introduce pathogenic bacteria and weed seeds to the paddy ecosystem (Amin, 2020), and also cause high NH_3 volatilization (Fig. 3). For these reasons, the direct application of fresh manure is not recommended for large-scale application (Rajwade *et al.*, 2018). The use of SOF maintained rice yield relative to the use of chemical fertilizer, but the production time was longer than COF. In contrast, COF not only maintained rice yield and reduced ammonia volatilization relative to the application of chemical fertilizer but has the benefits of pathogen sterilization and weed seed treatment, and a short production cycle. Thus, the COF has good prospects for widespread application in the double-cropping rice fields with a similar climate and soil type.

Effects of organic fertilizers on NH_3 volatilization from double-cropping rice field

The application of both chemical fertilizer and organic fertilizer increased NH_3 volatilization loss from the double-cropping rice field, indicating that the addition of exogenous inorganic and organic nitrogen increased NH_3 volatilization, which was consistent with a previous studies (Zhao *et al.*, 2015; Zhao *et al.*, 2021). The COIF treatment significantly increased NH_3 volatilization for both the early-season rice (54.9%) and the late-season rice (79.8%) compared with the CF (Fig. 3). A possible reason for the increased volatilization may be that the COIF consisted of two parts, inorganic nitrogen fertilizer with urea and compound fertilizer and organic nitrogen fertilizer with COF, and the urea addition increased the COIF mineral nitrogen content. The mineral nitrogen then rapidly increased the field surface water NH_4^+ -N concentration, which in turn stimulated higher NH_3 volatilization (Sun *et al.*, 2019; Lee *et al.*, 2021). The NH_3 volatilization was also higher after the application of FOF for both the early- and the late-season rice (Fig. 2). As the FOF was made with fresh chicken manure that was untreated, then the number of microorganisms would be enormous (Feng *et al.*, 2017). The organic nitrogen present in the FOF would likely be converted to mineral nitrogen through microbial decomposition, thereby increasing the FOF NH_4^+ -N content and therefore the NH_3 volatilization (Liu *et al.*, 2015; Lee *et al.*, 2021). The NH_3 volatilization period in FOF treatment case was longer than for the other organic fertilizer treatments during the late-season rice because the field surface water NH_4^+ -N concentration also remained at a higher concentration for a long time (Fig. 5B).

The main reason for the reduced NH_3 volatilization by applying COF is that the organic materials were completely fermented and decomposed in the process of fertilizer production. With most of readily decomposed organic nitrogen consumed during fermentation, then it is thought that the remaining organic nitrogen was difficult to degrade, and this accounted for the lower NH_3 volatilization flux. Moreover, as most of microorganisms in the chicken manure were killed in the COF production process by high temperature sterilization, then the generation of mineral nitrogen after the application of COF may have taken longer. Similarly, most of the degradable organic nitrogen in organic materials was consumed by composting and fermentation, thus the content of mineral nitrogen in SOF was low, thereby reducing the volatilization of NH_3 from rice (Lee *et al.*, 2021).

Influencing factors of NH_3 volatilization from double-cropping rice field

The field surface water NH_4^+ -N concentration was highly correlated with NH_3 volatilization (Fig. 4). This finding further confirms those from other studies which also indicate that NH_4^+ -N concentration was the main factor affecting the daily dynamics of NH_3 volatilization fluxes under different fertilizer treatments, with higher NH_4^+ -N concentration causing higher volatilization (Deng *et al.*, 2012; Zhou *et al.*, 2016; Uddin *et al.*, 2021). As the higher NH_4^+ -N concentration in the field surface water within one week after basal fertilization led to the greater NH_3 volatilization loss from the rice field during the same period (Figs. 2 and 5), then it is recommended that the further research aimed at the reduction of NH_3 volatilization loss from

double-cropping rice fields in China's Pearl River Delta should focus on the one week period after fertilization because this when NH_3 volatilization occurs particularly for the late-season rice. The field surface water pH was another significant factor affecting NH_3 volatilization from the double-cropping of rice (Ma *et al.*, 2021). Consistent with the findings of Zhong *et al.* (2021) and Sun *et al.* (2019), this study also showed that there was a positive correlation between the field surface water pH and NH_3 volatilization (Table IV).

Meteorological elements, such as temperature, precipitation, wind speed and incident solar radiation, may also affect NH_3 volatilization in paddy field but this appears to be a second order effect when compared to fertilizer addition. In general, sunny weather conditions with high temperature, less precipitation, high wind speed and strong light intensity promote NH_3 volatilization loss in paddy field (Kumagai *et al.*, 2015; Xu *et al.*, 2021). Lower temperature will reduce the hydrolysis rate of urea and organic nitrogen, inhibit the formation of $\text{NH}_4^+\text{-N}$, slow down the reaction of $\text{NH}_4^+\text{-N}$ to NH_3 , reduce the transport and diffusion of NH_3 at the water-air interface, resulting in the reduction of NH_3 volatilization in paddy field (Xia *et al.*, 2019). Precipitation could decrease the $\text{NH}_4^+\text{-N}$ concentration in field surface water, and increase the $\text{NH}_4^+\text{-N}$ loss through runoff and leaching, accounting for the decrease of NH_3 volatilization in paddy field (He *et al.*, 2018). Among all treatments for the early-season rice, almost no NH_3 volatilization occurred after April 9 due to the low temperature and rainy weather conditions (Figs. 1 and 2A). Furthermore, the distinctive climatic conditions with typhoon, wet and rainy, high temperature and heat in China's Pearl River Delta also interfered with the normal operation of field experiment (Chen *et al.*, 2015). In the late-season rice growing period, the experimental field was completely flooded after a heavy rainfall on August 11, 2019, resulting in a sharp decline of NH_3 volatilization fluxes. About two weeks later, the NH_3 volatilization experiment was forced to stop due to the rainstorm when 186 mm of rainfall fell within 24 hours caused by typhoon "Bailu". In addition, the typhoon "Lingling" caused the NH_3 volatilization flux in chemical fertilizer treatment to decrease rapidly back to the background level on the fourth day after the application of tillering fertilizer (Fig. 2B). Therefore, the cumulative NH_3 volatilization from rice field may be lower than the actual value.

It should be pointed out that due to the adverse factors such as COVID-19, the study on the mechanism of ammonia volatilization in rice fields is not deep enough. This study did not analyze the relationship between soil properties (such as ammonium nitrogen, pH and nitrate nitrogen) and ammonia volatilization in paddy fields. At the same time, the activity and gene abundance of soil microorganisms closely related to ammonia volatilization in rice fields and the activity of important enzymes (such as urease) were not analyzed. We hope that in the follow-up study, we could further study the mechanism of ammonia volatilization in paddy field and better explain the effect of organic fertilizer on ammonia volatilization in paddy field.

Overall, in this study, both conventional organic fertilizers produced by successive composting and the late-model rapidly composting organic fertilizer showed promising effects in ensuring rice yields and mitigating NH_3 volatilization from the rice field (Fig. 3, Table II). Compared with COF, SOF has shown almost consistent effects in ensuring rice yield and reducing ammonia volatilization in paddy fields. However, given the shorter fertilizer COF production cycle (about one month shorter than SOF), and pathogen sterilization, the combined advantages of COF seem worthy of further investigation on a larger scale of rice production.

CONCLUSIONS

The research results showed that there was no significant difference in rice yield between chemical fertilizer and four organic fertilizers ($P > 0.05$). COF and SOF produced the same rice yield as that when

chemical fertilizer was used in a double-cropping rice paddy field, whilst reducing NH₃ volatilization. The NH₃ volatilization loss from the late-season rice field was much higher than that from the early-season rice field. The total NH₃ volatilization from the early- and the late-season rice field significantly decreased after the application of COF (70%) and SOF (68%) compared with the chemical fertilizer. The NH₄⁺-N concentration in field surface water was a considerable factor determining the NH₃ volatilization from paddy field ($P < 0.01$). Overall, COF was found to maintain rice yield, mitigating NH₃ volatilization in paddy fields, and to have the additional benefits of pathogen sterilization and weed suppression and short production time, and utilizes chicken manure thus helping to develop a nitrogen circular economy. For these reasons, it is recommended that the more widespread use of COF over larger regions be investigated as a next step to promote efficient nitrogen use in agriculture and to help minimize N losses to the environment.

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Supplementary Material

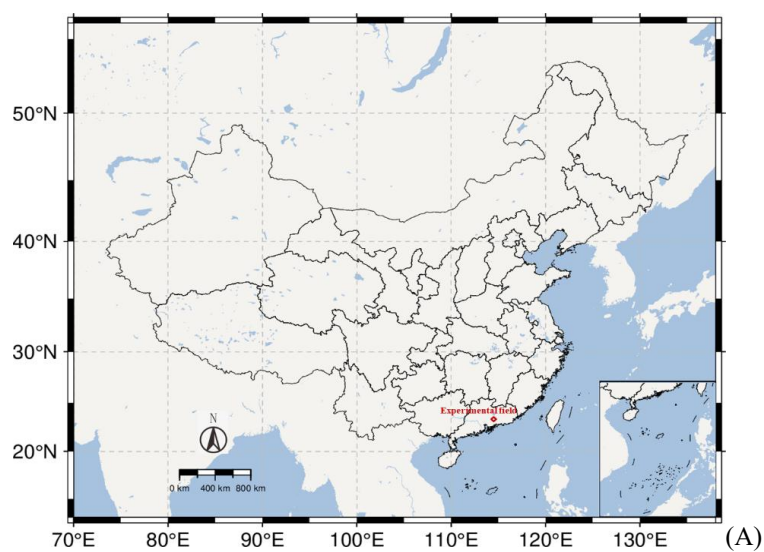


Figure S1 Geographic location and pictures of the experimental field. (A) Geographic location of the experimental field; (B) Aerial photograph of unmanned aerial vehicle in the experimental field; (C) Experiment field plot of chemical composted organic fertilizer treatment; (D) Ammonia absorption device.

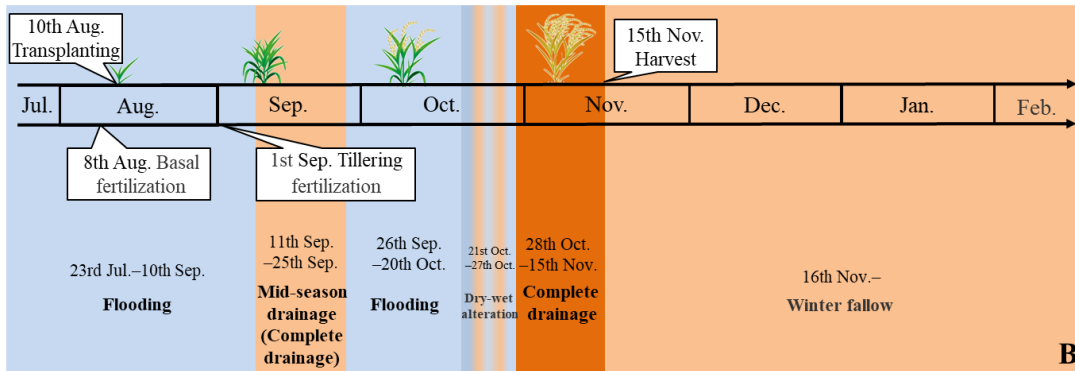
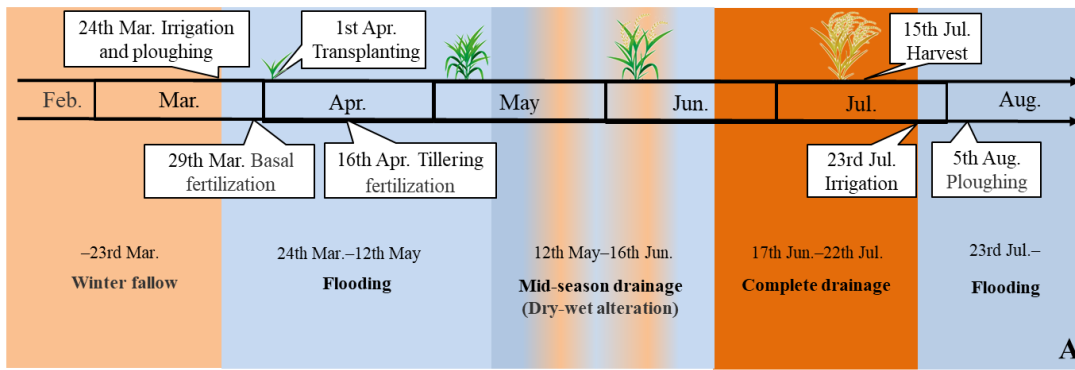


Figure S2 Field management of paddy during the early-season rice (A) and the late-season rice (B) growth period.

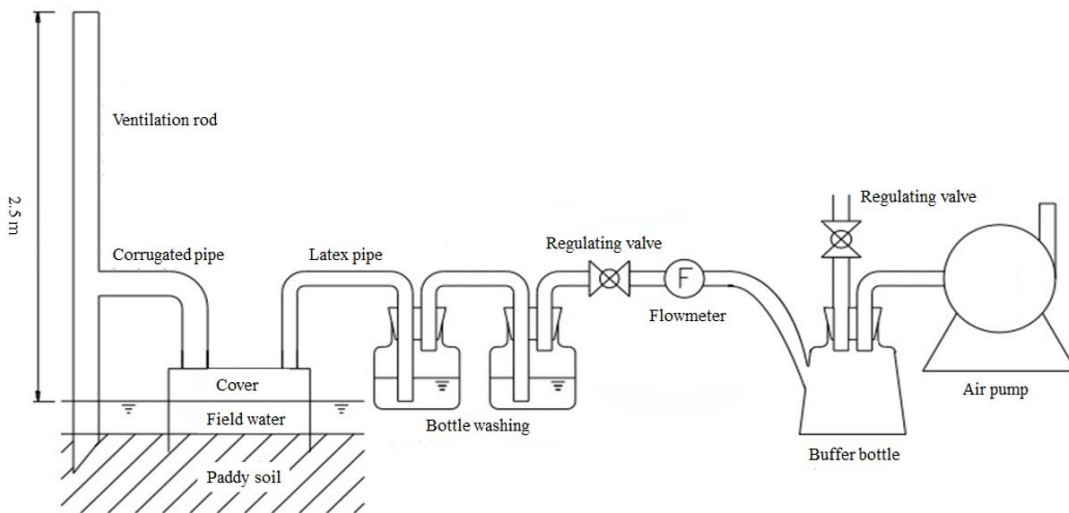


Figure S3 Schematic diagram of field installation for ammonia volatilization test.

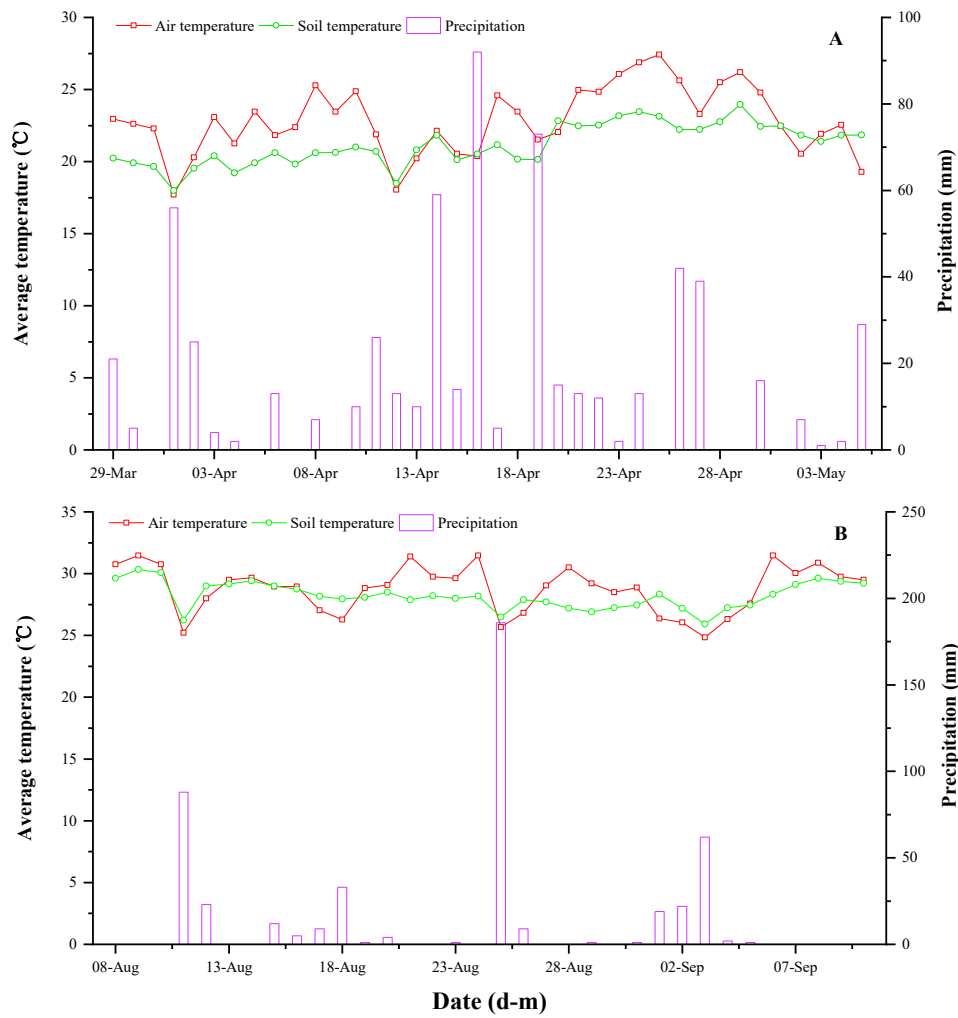


Figure S4 The dynamic change of precipitation and average temperature during ammonia volatilization experiment of the early-season rice (A) and the late-season rice (B) growing seasons in 2019.