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Characterizing Greenhouse Gas Emissions and Global Warming Potential of Wheat-Maize Cropping Systems in Response to Organic Amendments in Eutric Regosols, China

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Abstract: Characterizing greenhouse gas (GHG) emissions and global warming potential (GWP) has become a key step in the estimation of atmospheric GHG concentrations and their potential mitigation by cropland management. However, the impacts of organic amendments on GHG, GWP, and yield-scaled GWP on cropland have not been well documented. Here, we investigate four amendment treatments (no amendment, mineral fertilizers, and pig slurry or crop residue combined with mineral fertilizers) during a two-year field experiment in rain-fed wheat-maize cropping systems. The results show that the average annual cumulative methane (CH₄) flux ranged from −2.60 to −2.97 kg·C·ha^{−1} while nitrous oxide (N₂O) flux ranged from 0.44 to 4.58 kg·N·ha^{−1} across all four treatments. N₂O emissions were significantly correlated with soil inorganic nitrogen (i.e., NH₄⁺-N and NO₃[−]-N), and soil dissolved organic carbon (DOC) during both the winter wheat and summer maize seasons. On average, organic amendments combined with mineral fertilizers increased the annual GWP by 26–74% and yield-scaled GWP by 19–71% compared to those under only mineral fertilizers application. This study indicates that the fertilization strategy for Eutric Regosols can shift from only mineral fertilizers to organic amendments combined with mineral fertilizers, which can help mitigate GHG emissions and GWP while maintaining crop yields.

Keywords: organic fertilizers; CH₄ emissions; N₂O emissions; global warming potential; Eutric Regosols

1. Introduction

Wheat and maize are among the main sources of carbohydrates for human consumption; accordingly, the production of these grains is expected to increase by 1.3% per year by 2025 [1,2]. Along with the growing increase in carbohydrate demand, the production of cereal grains, such as wheat and maize, must be continued, while more land will require agricultural and fertilizer applications to sustain productivity. To reduce mineral nitrogen (N) fertilizers use and subsequent soil degradation, organic soil amendments are being more widely applied [3].

For over two decades reports have shown that the world's food production occurs in 37% of its total land area, and, as a consequence, agricultural practices have become a key source of methane (CH₄) and nitrous oxide (N₂O) emissions, representing 50% of anthropogenic CH₄ and 60% of anthropogenic N₂O emissions globally [4–6]. The extensive application of mineral N fertilizers and organic materials

(i.e., manure and crop residue) to soil for crop production purposes is responsible for the increased CH_4 and N_2O emissions from agricultural practices [7–9]. According to Ravishankara and colleagues [7], CH_4 and N_2O emissions from agriculture contribute approximately 20% to global radiative forcing. N_2O is a strong greenhouse gas with a high potential to deplete the ozone layer [7,10].

Generally, if integrated with crop nutrient demands, organic amendment application to agricultural soil can lead to the creation of small CH_4 sinks [8,11,12]. Conversely, the application of organic amendments over a long period can potentially affect CH_4 and N_2O emissions [9,13,14]. Specifically, an increase in N_2O emissions from organic amendments could occur via nitrification and denitrification processes [12,15,16]. Therefore, soil temperature and soil moisture could both strongly affect N_2O emissions [8,9,13].

The soil locally called “purplish soil” is classified as a Eutric Regosol under the United Nations (UN) Food and Agriculture Organization (FAO) system and a Pup-Orthic-Entisol under the Chinese Soil Taxonomy system [17]. Purplish soil comprises an area of 160,000 km^2 in the upper reaches of the Yangtze River [18], accounting for 68% of the total cropland in Sichuan Province and 7% of China’s total cropland area [19]. Purplish soil is typically fertile, with a relatively high mineral content [19]. Owing to its low organic matter content and high mineral content, purplish soil is liable to generate N_2O emissions, particularly under intensive mineral N fertilizer applications [20]. Additionally, Zhou et al. [12] reported that the combination of organic amendments and mineral N fertilizer application could result in the creation of small CH_4 sinks with annual soil CH_4 emissions of -2.1 to $-1.8 \text{ kg}\cdot\text{C}\cdot\text{ha}^{-1}$. Furthermore, previous studies conducted on N_2O emissions from purplish soil demonstrated that mineral N fertilizer application at a rate of $250 \text{ kg N ha}^{-1}\cdot\text{yr}^{-1}$ in wheat-maize cropping systems will lead to an annual N_2O flux of 1.9 to $6.7 \text{ kg}\cdot\text{N}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$, which corresponds to an emission factor (EF_d) of 0.12 – 1.06% [21].

Given the aim of reducing combined CH_4 and N_2O emissions under the objective of maintaining crop yields, the global warming potential (GWP) values of 28 for CH_4 and 265 for N_2O over a 100-year planning horizon [10] should be taken into account prior to making the shift from mineral fertilizers to organic amendment applications [8]. GWP can alternatively be viewed as a crop yield function, referred to as yield-scaled GWP, with which to evaluate CH_4 and N_2O emission mitigation under the objective of maintaining crop grain yields [2,22]. However, very few studies have concurrently reported on GWP or yield-scaled GWP and the release of CH_4 and N_2O emissions under organic amendment applications in wheat-maize cropping systems. Thus, determining the GWP of CH_4 and N_2O emissions under organic amendment applications in Eutric Regosols could be important for estimating potential CH_4 and N_2O emission effects on regional and global climate systems. Accordingly, the aims of this study were (1) to quantify CH_4 and N_2O emissions under different organic amendments in wheat-maize rotational cropping systems, and (2) to determine the GWP of CH_4 and N_2O emissions and associated yield-scaled GWP in order to find the most suitable management practices for organic amendment applications to Eutric Regosols in the Sichuan Basin, China.

2. Experiments

2.1. Site Conditions

The Yanting Agro-Ecological Station of Purplish Soil ($31^\circ 16' \text{ N}$, $105^\circ 28' \text{ E}$, 420 m a.s.l., Southwest China) was selected as the location of the field experiment. This research station is under the auspice of the Chinese Ecosystem Research Network (CERN). The climate is a moderately subtropical monsoon climate. The annual average air temperature is 17.3°C , and the annual precipitation is 826 mm [14]. The soil is 19.2% clay, 42.1% silt, and 38.7% sand; the pH level is 8.22 (with a water/soil ratio of 2.5:1 w/w); the bulk density (BD) is $1.32 \text{ g}\cdot\text{cm}^{-3}$; the organic carbon (C) content is $8.75 \text{ g}\cdot\text{kg}^{-1}$; and the total N content is $0.62 \text{ g}\cdot\text{kg}^{-1}$ [19].

2.2. Experimental Design

A randomized lysimeter-based block design was used for this experiment (size: $8 \times 4 \text{ m}^2$). Four treatments (including three replicates) with long-term fertilization were established in 2002 using a winter wheat and summer maize rotational system. The four treatments were as follows: (1) no amendment (CK) (i.e., the control), (2) mineral fertilizers (NPK), (3) pig slurry (mineral N application rate = 40%) combined with mineral fertilizers (mineral N application rate = 60%) (OMNPK), and (4) crop residue (mineral N application rate = 20%) combined with mineral fertilizers (mineral N application rate = 80%) (CRNPK). Except for the control (CK), all treatments were amended with $280 \text{ kg} \cdot \text{N} \cdot \text{ha}^{-1}$ annually, which were subdivided into $130 \text{ kg} \cdot \text{N} \cdot \text{ha}^{-1}$ during the winter wheat (*Triticum aestivum* L.) season and $150 \text{ kg} \cdot \text{N} \cdot \text{ha}^{-1}$ during the summer maize (*Zea mays* L.) season. The mineral fertilizers used were urea (NH_4HCO_3) for N, calcium superphosphate ($\text{Ca}(\text{H}_2\text{PO}_4)_2$) at $90 \text{ kg P}_2\text{O}_5 \text{ ha}^{-1}$ for phosphorus (P), and potassium chloride (KCl) at $36 \text{ kg K}_2\text{O ha}^{-1}$ for potassium (K), which were utilized as basal fertilizers for both the winter wheat and summer maize cropping seasons. Pig slurry procured from a single pig farm was utilized for both crops (i.e., wheat and maize). The total N content of the pig slurry used as fertilizer under both crop rotations ranged from 15 to $16 \text{ mg} \cdot \text{kg}^{-1}$, while the total C content ranged from 350 to $338 \text{ mg} \cdot \text{kg}^{-1}$, for an average C:N ratio of 22.0. Both wheat and maize crop residues were used in this experiment, with total N contents of $5.6 \text{ mg} \cdot \text{kg}^{-1}$ and $9.2 \text{ mg} \cdot \text{kg}^{-1}$, total C contents of $429.1 \text{ mg} \cdot \text{kg}^{-1}$ and $415.3 \text{ mg} \cdot \text{kg}^{-1}$, and average C:N ratios of 76.6 and 45.1, respectively. Every year, wheat (variety 01-3570) was sown in early November and harvested in late May for the winter wheat cropping season, while maize (variety Chengdan 60) was planted in early June and harvested late September for the maize cropping season.

2.3. Soil Measurements

Samples used to determine soil ammonia nitrogen ($\text{NH}_4^+\text{-N}$), nitrate nitrogen ($\text{NO}_3^-\text{-N}$), and dissolved organic carbon (DOC) content were collected from the plots at each gas sampling event. Moreover, soil samples were collected randomly from three positions in each plot (at a 20 cm depth) using a small stainless-steel flat-blade shovel. Following sampling, plastic bags were used to promptly seal the soil samples, which were then stored at 4°C until analysis. In the laboratory, 25 mL of a 0.5 M K_2SO_4 solution were used to extract soil samples ($5 \pm 0.5 \text{ g}$), after which $0.45 \mu\text{m}$ membranes were used to filter the supernatant. Following this, an Auto Analyzer-AA3 (Bran + Luebbe, Norderstedt, Germany) was used to analyze the $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$, and DOC contents in the filtrates.

2.4. Crop Yield Measurements

Crop yields in all treatments (0.25 m^2 wheat harvest plots and 1 m^2 maize plots) and replicates were measured during harvest. After biomass harvesting, grains were separated and then oven-dried at 70°C for 48 h to determine their dry weight equivalent grain yield ($\text{Mg} \cdot \text{ha}^{-1}$).

2.5. Soil CH_4 and N_2O Emission Measurements

Emissions of CH_4 and N_2O were measured at the same time over a two-year period (i.e., from November 2016 to October 2018) using static chamber-gas chromatography, which had previously been used in a similar observational study conducted by Zhou and co-workers [8]. After plowing the field plots and prior to planting the crops, we inserted a stainless-steel chamber on a permanent collar ($0.50 \times 0.50 \text{ m}$) into the soil of each plot at a 10 cm depth, which remained in place throughout the entire wheat-maize rotational experimental period. The chambers were encased in a layer of insulating material to minimize exposure to temperature fluctuations when the chambers were in a closed position. The aboveground height of the chamber collar was adjusted according to the specific wheat-maize growth stage, and a circulating fan was used to guarantee uniform mixing within the chamber headspace. We measured CH_4 and N_2O fluxes daily during the first week following fertilization, after which CH_4 and N_2O fluxes were measured at 2-day intervals

during the second week. Thereafter, the sampling frequency was set to twice weekly for the remainder of the measurement period.

Gas samples were collected from the chambers during morning hours (i.e., from 9:00 to 11:00 am) following the procedure recommended by Parkin and Venterea [23]. After closure, five separate gas samples were collected at each flux measurement, using 50 mL plastic syringes that were fitted with three-way stopcocks inserted into Teflon tubes fixed to the chambers. Throughout the gas sampling stage of our experiment, a manual thermoelectric thermometer (model JM624, Tianjin Jinming Instrument Co. Ltd., Tianjin, China) was used to measure both chamber temperature and soil temperature. A portable frequency domain reflectometry probe (RDS Technology Co. Ltd., Nanjing, Jiangsu, China) was used in the same manner to measure soil moisture (soil volumetric water content), which was then converted into water-filled pore space (WFPS in %) as shown in Equation (1).

After sampling, a gas chromatograph (GC) (HP 5890II, Hewlett-Packard, Palo Alto, CA, USA) rigged with a flame ionization detector (FID) was used to analyze CH₄ flux and an electron capture detector (ECD) was used to analyze N₂O flux in gas samples. This study used the inclusive GC configuration procedure by Wang and Wang [24] for CH₄ and N₂O gas analysis. We used high purity dinitrogen (N₂) as a gas carrier and did not directly introduce a high concentration buffering gas such as CO₂ and CH₄ into the detector for analysis. For each measurement, the system was calibrated with known mixed gas concentrations of 0.5 ppm N₂O, 5.1 ppm CH₄, and 386.2 ppm CO₂.

2.6. Calculations

For this study, we expressed soil moisture as WFPS (%). It was calculated following Equation (1):

$$WFPS = SVWC \times \frac{BD}{1 - \left(\frac{BD}{2.65}\right)} \times 100 \quad (1)$$

where SVWC (%) is the soil volumetric water content; BD is the soil bulk density; and 2.65 g·cm³ is the theoretical particle density.

CH₄ and N₂O fluxes from soil were calculated following the method suggested by Wang and Wang [24], while cumulative seasonal and annual CH₄ and N₂O fluxes were calculated using daily flux linear interpolation between gas sampling rates following the method of Gao et al. [25]:

We calculated direct N₂O emission factors (EF_d ; %) generated by the N fertilizer applied in each treatment using Equation (2):

$$EF_d(\%) = 100 \times \frac{E_F - E_O}{N} \quad (2)$$

where E_F denotes cumulative N₂O emissions from the N fertilizer treatments (kg·N·ha⁻¹); E_O denotes cumulative N₂O emissions in the unfertilized treatment (CK, the control) (kg·N·ha⁻¹); and N denotes the N fertilizer application rate (kg·N·ha⁻¹).

To better understand the effects of different organic fertilization practices on N₂O and CH₄ emissions, the corresponding GWP CO₂ equivalents (CO₂-eq kg·ha⁻¹) were calculated based on a 100-year planned horizon. The results showed that the GWP value of CH₄ was 28 and the GWP value of N₂O was 265 [10]. Equation (3) was used to determine these values:

$$GWP = 28 \times CH_4 + 265 \times N_2O \quad (3)$$

where GWP is expressed as kg CO₂-eq·kg⁻¹, and CH₄ and N₂O fluxes are expressed as kg·ha⁻¹·yr⁻¹.

The yield-scaled GWP (kg CO₂-eq·kg⁻¹) of N₂O and CH₄ emission fluxes was calculated by subdividing GWP by the wheat-maize grain yield of each treatment [22].

2.7. Statistical Analysis

Statistical analysis was used to verify the effects of the four treatments on CH₄ and N₂O emission fluxes, grain yields, GWP, and yield-scaled GWP. This was done using one-way analysis of variance (ANOVA) in SPSS version 16.0 (Chicago, IL, USA). The least significant difference (LSD) test was used to verify significant differences in the means of estimated parameters between all four treatments at a 95% confidence level ($p < 0.05$). N₂O emission flux data were log-transformed to meet parametric assumptions associated with normality and homogeneity of variances, while the absolute negative values of CH₄ flux transformed to CH₄ uptake values were used for regression analysis. Data are provided in triplicate as the means $\pm$ standard error (SE). This study used SigmaPlot (version 12.5; Systat, Inc., Chicago, IL, USA) to construct graphs.

3. Results

3.1. Seasonal Variations in Environmental Factors

The obvious differences observed in daily precipitation and daily maximum and minimum air temperatures indicated distinct trends between the winter wheat season (Nov/May) and the summer maize season (June/Oct) (Figure 1a). The annual precipitation from 2016 to 2017 and from 2017 to 2018 was 785 mm and 630 mm, respectively, 70% of which occurred during the summer maize season. The maximum daily air temperature was 38.2 °C from 2016 to 2017 and 37.9 °C from 2017 to 2018, while the minimum daily air temperature was −2.1 °C from 2016 to 2017 and −5.4 °C from 2017 to 2018 (Figure 1a). The annual mean daily temperature was 17.6 °C throughout the two-year experimental period (Figure 1a). The soil temperature of the four treatments during the winter wheat season ranged from 4.1 to 23.1 °C, while the mean soil temperature during the summer maize season ranged from 17.6 to 29.6 °C (Figure 1b) throughout the two-year experimental period. The water-filled pore space (WFPS) of soil throughout the winter wheat growing season varied from 15.7 to 67.1% for all four treatments, while the WFPS of soil throughout the summer maize season varied from 12.1 to 106.6% throughout the two-year experimental period (Figure 1b). Table S1 (Supplementary Materials) provides the mean annual seasonal variation in soil temperature and the WFPS of soil for all four treatments throughout the two-year experimental period.

3.2. CH₄ Fluxes

During both the winter wheat and summer maize seasons, CH₄ fluxes subsequently acted as small sinks (Figure 2). During the winter wheat season, the CH₄ fluxes varied from −0.10 to −0.02 mg·C·m^{−2}·hr^{−1} in the 2016/2017 cropping season and from −0.08 to −0.01 mg·C·m^{−2}·hr^{−1} in the 2017/2018 cropping season for CK (Figure 2a); from −0.09 to −0.01 mg·C·m^{−2}·hr^{−1} in the 2016/2017 cropping season and from −0.17 to −0.01 mg·C·m^{−2}·hr^{−1} in the 2017/2018 cropping season for NPK (Figure 2b); from −0.12 to −0.01 mg·C·m^{−2}·hr^{−1} in the 2016/2017 cropping season and from −0.11 to −0.01 mg·C·m^{−2}·hr^{−1} in the 2017/2018 cropping season for OMNPK (Figure 2c); and from −0.14 to −0.01 mg·C·m^{−2}·hr^{−1} in the 2016/2017 cropping season and from −0.11 to −0.01 mg·C·m^{−2}·hr^{−1} in the 2017/2018 cropping season for CRNPK (Figure 2d). The largest CH₄ flux peak of −0.01 mg·C·m^{−2}·hr^{−1} was observed for all four treatments (except for CK in the 2016/2017 cropping season), while the smallest CH₄ flux peak was observed in the CRNPK treatment in 2016/2017 and in the NPK treatment in the 2017/2018 cropping season.

Conversely, during the summer maize season, the CH₄ fluxes varied from −0.07 to −0.01 mg·C·m^{−2}·hr^{−1} in both the 2017 and 2018 cropping seasons for CK (Figure 2a); from −0.09 to −0.01 mg·C·m^{−2}·hr^{−1} in the 2017 cropping season and from −0.09 to −0.02 mg·C·m^{−2}·hr^{−1} in the 2018 cropping season for NPK (Figure 2b); from −0.27 to −0.01 mg·C·m^{−2}·hr^{−1} in the 2017 cropping season and from −0.05 to 0.00 mg·C·m^{−2}·hr^{−1} in the 2018 cropping season for OMNPK (Figure 2c); and from −0.06 to −0.01 mg·C·m^{−2}·hr^{−1} in the 2017 cropping season and from −0.05 to −0.01 mg·C·m^{−2}·hr^{−1} in the 2018 cropping season for CRNPK (Figure 2d). The largest CH₄ flux peak of −0.01 mg·C·m^{−2}·hr^{−1}

was observed for all four treatments (except for NPK and OMNPK in the 2018 cropping season), while the smallest CH_4 flux peak was observed in the OMNPK treatment in the 2017 cropping season, and in the NPK treatment in the 2018 cropping season. Table S1 (Supplementary Materials) provides the mean seasonal annual CH_4 variations.

Figure 1. (a) Daily precipitation (mm), daily maximum and minimum air temperature (°C) and (b) seasonal variations in mean soil water-filled pore space (WFPS) and mean soil temperature (topsoil at 5 cm) in the wheat-maize rotations for the four treatments over the two-year period from 2016–2018.

For the winter wheat season, the mean cumulative CH_4 fluxes among the four treatments were $-1.54 \text{ kg}\cdot\text{C}\cdot\text{ha}^{-1}$ for CK, $-1.77 \text{ kg}\cdot\text{C}\cdot\text{ha}^{-1}$ for NPK, $-1.75 \text{ kg}\cdot\text{C}\cdot\text{ha}^{-1}$ for OMNPK, and $-2.02 \text{ kg}\cdot\text{C}\cdot\text{ha}^{-1}$ for CRNPK (Table 1). The seasonal mean cumulative CH_4 fluxes for CK were significantly greater than those of the other treatments ($p < 0.05$), while NPK and OMNPK were not significantly different from each other. CRNPK showed the smallest significant mean cumulative CH_4 flux ($p > 0.05$) (Table 1). Comparatively, for the summer maize season, the mean cumulative CH_4 fluxes were $-0.84 \text{ kg}\cdot\text{C}\cdot\text{ha}^{-1}$ for CK, $-0.99 \text{ kg}\cdot\text{C}\cdot\text{ha}^{-1}$ for NPK, $-0.82 \text{ kg}\cdot\text{C}\cdot\text{ha}^{-1}$ for OMNPK, and $-0.78 \text{ kg}\cdot\text{C}\cdot\text{ha}^{-1}$ for CRNPK (Table 1). The mean cumulative CH_4 fluxes for the CK, OMNPK, and CRNPK treatments showed no significant differences from each other ($p < 0.05$), while the NPK treatment had the smallest significant mean cumulative CH_4 flux for the maize season (Table 1). Moreover, the annual mean cumulative CH_4 fluxes were $-2.60 \text{ kg}\cdot\text{C}\cdot\text{ha}^{-1}$ for CK, $-2.97 \text{ kg}\cdot\text{C}\cdot\text{ha}^{-1}$ for NPK, -2.97 for OMNPK, and $-2.93 \text{ kg}\cdot\text{C}\cdot\text{ha}^{-1}$ for CRNPK (Table 1). The annual mean cumulative CH_4 fluxes for CK were significantly greater than those of the other treatments ($p < 0.05$), which were not significantly different from each other ($p > 0.05$). Overall, the annual cumulative CH_4 flux was significantly different among the four treatments in the order of $\text{CK} > \text{NPK} > \text{CRNPK} > \text{OMNPK}$ in the 2016/2017 cropping year (Figure 3a), while different trends were observed in the 2017/2018 cropping year, with the order of $\text{CK} > \text{OMNPK} > \text{CRNPK} > \text{NPK}$ (Figure 3b).

Figure 2. Seasonal variations in methane (CH_4) emissions in the wheat-maize rotations for CK = control (a), NPK = mineral fertilizers (b), OMNPK = pig slurry combined with mineral fertilizers (c) and CRNPK = crop residue combined with mineral fertilizers (d) treatments over the two-year period from 2016–2018. The vertical bars in the panels indicate the standard errors of spatial replicates ($n = 3$).

Table 1. Seasonal and annual cumulative fluxes of methane (CH₄, kg·C·ha^{−1}), nitrous oxide (N₂O, kg·N·ha^{−1}), and direct emission factors (EF_d, %) for the four treatments over the two-year period from 2016–2018.

Year	Treatments	Wheat Season			Maize Season			Annual		
		CH ₄	N ₂ O	EF _d	CH ₄	N ₂ O	EF _d	CH ₄	N ₂ O	EF _d
2016–2017	CK	−1.79 ± 0.15a	0.19 ± 0.01d	-	−0.83 ± 0.02a	0.19 ± 0.06c	-	−2.81 ± 0.15a	0.42 ± 0.09c	-
	NPK	−1.81 ± 0.07a	0.73 ± 0.04b	0.41	−0.93 ± 0.04a	0.69 ± 0.05b	0.34	−2.98 ± 0.05ab	1.48 ± 0.08b	0.38
	OMNPK	−2.05 ± 0.31a	0.62 ± 0.02c	0.33	−0.89 ± 0.03a	3.65 ± 0.02a	2.31	−3.52 ± 0.08c	4.76 ± 0.04a	1.55
	CRNPK	−2.38 ± 0.04b	0.83 ± 0.05a	0.49	−0.79 ± 0.03a	0.81 ± 0.09b	0.42	−3.27 ± 0.06bc	1.77 ± 0.14b	0.48
2017–2018	CK	−1.29 ± 0.10a	0.19 ± 0.02d	-	−0.84 ± 0.02a	0.23 ± 0.02d	-	−2.37 ± 0.08a	0.46 ± 0.03d	-
	NPK	−1.74 ± 0.06b	0.59 ± 0.04b	0.31	−1.05 ± 0.05b	0.64 ± 0.05c	0.27	−2.96 ± 0.09b	1.33 ± 0.09c	0.31
	OMNPK	−1.45 ± 0.07ab	0.50 ± 0.01c	0.24	−0.75 ± 0.01a	2.92 ± 0.01a	1.79	−2.42 ± 0.04a	4.40 ± 0.02a	1.41
	CRNPK	−1.66 ± 0.11b	0.83 ± 0.02a	0.50	−0.78 ± 0.02a	0.87 ± 0.05b	0.42	−2.58 ± 0.13a	1.82 ± 0.02b	0.49
Mean (2016–2018)	CK	−1.54 ± 0.07a	0.19 ± 0.01d	-	−0.84 ± 0.01a	0.21 ± 0.04d	-	−2.60 ± 0.06a	0.44 ± 0.05d	-
	NPK	−1.77 ± 0.01b	0.66 ± 0.04b	0.36	−0.99 ± 0.05b	0.67 ± 0.04c	0.30	−2.97 ± 0.06b	1.40 ± 0.07c	0.34
	OMNPK	−1.75 ± 0.05b	0.56 ± 0.00c	0.28	−0.82 ± 0.02a	3.28 ± 0.02a	2.05	−2.97 ± 0.04b	4.58 ± 0.01a	1.48
	CRNPK	−2.02 ± 0.04c	0.83 ± 0.02a	0.50	−0.78 ± 0.03a	0.84 ± 0.06b	0.42	−2.93 ± 0.06b	1.80 ± 0.07b	0.49

Mean ± SE: Means in columns followed by different lower case letter are significantly different (least significant difference (LSD), $p < 0.05$) and means followed by the same lower case letter are not significantly different (LSD, $p > 0.05$). No amendment as control (CK), mineral fertilizers (NPK), pig slurry combined with mineral fertilizers (OMNPK), and crop residue combined with mineral fertilizers (CRNPK). Note: The CH₄ and N₂O fluxes for winter wheat cropping season (159 days) or summer maize cropping season (104 days) are expressed as kg·C·ha^{−1}·season^{−1} or kg·N·ha^{−1}·season^{−1}, while the annual cumulative is expressed in kg·C·ha^{−1}·yr^{−1} or kg·N·ha^{−1}·yr^{−1}.

Figure 3. Seasonal and annual cumulative CH_4 fluxes (a,b) and cumulative N_2O fluxes (c,d) in the wheat-maize rotations for the four treatments over the two-year period from 2016–2018. Each colored circle point in the panels indicates the mean for measured data for three spatial replicates ($n = 3$) with error bars not shown.

3.3. N_2O Emissions

This study found that N_2O flux during the winter wheat season was lower than that during the summer maize season (Figure 4). Higher N_2O emissions were measured in the initial two weeks after the various amendments were applied in both the winter wheat and summer maize seasons. During the winter wheat season, the N_2O emissions ranged from 1.6 to 6.6 $\mu\text{g}\cdot\text{N}\cdot\text{m}^{-2}\cdot\text{hr}^{-1}$ in the 2016/2017 cropping season and from 1.4 to 7.2 $\mu\text{g}\cdot\text{N}\cdot\text{m}^{-2}\cdot\text{hr}^{-1}$ in the 2017/2018 cropping season for CK (Figure 4a); from 2.7 to 50.7 $\mu\text{g}\cdot\text{N}\cdot\text{m}^{-2}\cdot\text{hr}^{-1}$ in the 2016/2017 cropping season and from 3.4 to 45.0 $\mu\text{g}\cdot\text{N}\cdot\text{m}^{-2}\cdot\text{hr}^{-1}$ in the 2017/2018 cropping season for NPK (Figure 4b); from 2.3 to 104.3 $\mu\text{g}\cdot\text{N}\cdot\text{m}^{-2}\cdot\text{hr}^{-1}$ in the 2016/2017 cropping season and from 1.8 to 47.8 $\mu\text{g}\cdot\text{N}\cdot\text{m}^{-2}\cdot\text{hr}^{-1}$ in the 2017/2018 cropping season for OMNPK (Figure 4c); and from 3.1 to 62.7 $\mu\text{g}\cdot\text{N}\cdot\text{m}^{-2}\cdot\text{hr}^{-1}$ in the 2016/2017 cropping season and from 2.8 to 96.6 $\mu\text{g}\cdot\text{N}\cdot\text{m}^{-2}\cdot\text{hr}^{-1}$ in the 2017/2018 cropping season for CRNPK (Figure 4d). The highest N_2O flux pulse event during the winter wheat season occurred in the OMNPK treatment in the 2016/2017 cropping season and in CRNPK treatment in the 2017/2018 cropping season.

Conversely, the N_2O emissions for the summer maize season ranged from 1.0 to 25.0 $\mu\text{g}\cdot\text{N}\cdot\text{m}^{-2}\cdot\text{hr}^{-1}$ in the 2017 cropping season and from 2.2 to 22.6 $\mu\text{g}\cdot\text{N}\cdot\text{m}^{-2}\cdot\text{hr}^{-1}$ in the 2018 cropping season for CK (Figure 4a); from 1.0 to 175.4 $\mu\text{g}\cdot\text{N}\cdot\text{m}^{-2}\cdot\text{hr}^{-1}$ in the 2017 cropping season and from 3.4 to 106.7 $\mu\text{g}\cdot\text{N}\cdot\text{m}^{-2}\cdot\text{hr}^{-1}$ in the 2018 cropping season for NPK (Figure 4b); from 1.9 to 1492.1 $\mu\text{g}\cdot\text{N}\cdot\text{m}^{-2}\cdot\text{hr}^{-1}$ in the 2017 cropping season and from 2.6 to 1545.4 $\mu\text{g}\cdot\text{N}\cdot\text{m}^{-2}\cdot\text{hr}^{-1}$ in the 2018 cropping season for OMNPK (Figure 4c); and from 1.5 to 195.7 $\mu\text{g}\cdot\text{N}\cdot\text{m}^{-2}\cdot\text{hr}^{-1}$ in the 2017 cropping season and from 7.5 to 138.9 $\mu\text{g}\cdot\text{N}\cdot\text{m}^{-2}\cdot\text{hr}^{-1}$ in the 2018 cropping season for CRNPK (Figure 4d). For the amendment

treatments, the highest N₂O emission pulse events during the summer maize season both occurred in the OMNPK treatment, while the lowest significant N₂O emissions occurred in the NPK treatment in the 2017 and 2018 cropping seasons. Table S1 (Supplementary Materials) provides the mean seasonal and annual N₂O emission variations.

Figure 4. Seasonal variations in N₂O emissions in the wheat-maize rotations for rotations for CK = control (a), NPK = mineral fertilizers (b), OMNPK = pig slurry combined with mineral fertilizers (c) and CRNPK = crop residue combined with mineral fertilizers (d) treatments over the two-year period from 2016–2018. The vertical bars in the panels indicate the standard errors of spatial replicates ($n = 3$). The downward arrows indicate the date of different amendment applications.

Moreover, the mean cumulative N₂O fluxes from all four treatments during the winter wheat season were 0.19 kg·N·ha^{−1} for CK, 0.66 kg·N·ha^{−1} for NPK, 0.56 kg·N·ha^{−1} for OMNPK, and 0.83 kg·N·ha^{−1} for CRNPK (Table 1). During the winter wheat season, the mean cumulative N₂O flux for CRNPK was significantly higher than that in the other treatments, which significantly differed from each other ($p < 0.05$). The mean EF_d % (i.e., the emission factor) values ranged from 0.28–0.50% (i.e., below 1.0%) (Table 1) during the winter wheat season.

Comparatively, the mean cumulative N_2O fluxes from all four treatments during the summer maize season were $0.21 \text{ kg}\cdot\text{N}\cdot\text{ha}^{-1}$ for CK, $0.67 \text{ kg}\cdot\text{N}\cdot\text{ha}^{-1}$ for NPK, $3.28 \text{ kg}\cdot\text{N}\cdot\text{ha}^{-1}$ for OMNPK, and $0.84 \text{ kg}\cdot\text{N}\cdot\text{ha}^{-1}$ for CRNPK (Table 1). The mean cumulative N_2O flux for the summer maize season differed significantly among the four treatments ($p < 0.05$). The OMNPK treatment had a significantly higher N_2O flux (i.e., approximately two times higher) than the other treatments in both 2017 and 2018 (Table 1). Moreover, the mean EF_d (%) values, ranging from 0.42–2.05%, were higher during the summer maize season (Table 1).

Furthermore, the annual mean cumulative N_2O fluxes throughout the two-year experimental period for all four treatments were $0.44 \text{ kg}\cdot\text{N}\cdot\text{ha}^{-1}$ for CK, $1.40 \text{ kg}\cdot\text{N}\cdot\text{ha}^{-1}$ for NPK, $4.58 \text{ kg}\cdot\text{N}\cdot\text{ha}^{-1}$ for OMNPK, and $1.80 \text{ kg}\cdot\text{N}\cdot\text{ha}^{-1}$ for CRNPK (Table 1). The annual mean cumulative N_2O flux for OMNPK was significantly greater than that in the other treatments, followed by the CRNPK and NPK treatments, which differed significantly from each other ($p < 0.05$). The mean annual EF_d (%) values exhibited trends similar to those of the mean seasonal EF_d (%), ranging from 0.49–1.48%, with slight differences observed between the two-year experimental periods. Overall, the annual cumulative N_2O flux was significantly different among the four treatments in the order of OMNPK > CRNPK > NPK > CK in the 2016/2017 cropping year (Figure 3c), with similar trends observed in the 2017/2018 cropping year (Figure 3d). The OMNPK treatment emitted more N_2O than the CRNPK and NPK treatments, contributing approximately 56% of the total mean annual N_2O flux. Thus, the OMNPK treatment was the greatest N_2O source.

3.4. Changes in Soil Substrates

The ammonia nitrogen ($\text{NH}_4^+\text{-N}$), nitrate nitrogen ($\text{NO}_3^-\text{-N}$), and dissolved organic carbon (DOC) in the soil substrates under four treatments in both the winter wheat and summer maize seasons are shown in Figure 5. During the winter wheat season, the $\text{NH}_4^+\text{-N}$ ranged from 0.1 to $2.9 \text{ mg}\cdot\text{N}\cdot\text{kg}^{-1}$ in the 2016/2017 cropping season and from 0.3 to $3.3 \text{ mg}\cdot\text{N}\cdot\text{kg}^{-1}$ in the 2017/2018 cropping season for CK (Figure 5a); from 0.6 to $13.0 \text{ mg}\cdot\text{N}\cdot\text{kg}^{-1}$ in the 2016/2017 cropping season and from 0.7 to $10.1 \text{ mg}\cdot\text{N}\cdot\text{kg}^{-1}$ in the 2017/2018 cropping season for NPK (Figure 5a); from 0.5 to $12.6 \text{ mg}\cdot\text{N}\cdot\text{kg}^{-1}$ in the 2016/2017 cropping season and from 0.8 to $8.8 \text{ mg}\cdot\text{N}\cdot\text{kg}^{-1}$ in the 2017/2018 cropping season for OMNPK (Figure 5a); and from 0.8 to $11.5 \text{ mg}\cdot\text{N}\cdot\text{kg}^{-1}$ in the 2016/2017 cropping season and from 0.9 to $10.5 \text{ mg}\cdot\text{N}\cdot\text{kg}^{-1}$ in the 2017/2018 cropping season for CRNPK (Figure 5a). We observed a significantly higher soil $\text{NH}_4^+\text{-N}$ content in the NPK treatment in 2016/2017, while the NPK and CRNPK treatments showed similar soil $\text{NH}_4^+\text{-N}$ contents in the 2017/2018 wheat cropping season (Figure 5a).

During the summer maize season, a much lower range of $\text{NH}_4^+\text{-N}$ contents was observed, varying from 0.1 to $2.3 \text{ mg}\cdot\text{N}\cdot\text{kg}^{-1}$ in the 2017 cropping season and from 0.2 to $3.8 \text{ mg}\cdot\text{N}\cdot\text{kg}^{-1}$ in the 2018 cropping season for CK (Figure 5a); from 0.1 to $2.8 \text{ mg}\cdot\text{N}\cdot\text{kg}^{-1}$ in the 2017 cropping season and from 0.1 to $4.4 \text{ mg}\cdot\text{N}\cdot\text{kg}^{-1}$ in the 2018 cropping season for NPK (Figure 5a); from 0.1 to $2.5 \text{ mg}\cdot\text{N}\cdot\text{kg}^{-1}$ in the 2017 cropping season and from 0.2 to $4.9 \text{ mg}\cdot\text{N}\cdot\text{kg}^{-1}$ in the 2018 cropping season for OMNPK (Figure 5a); and from 0.6 to $2.8 \text{ mg}\cdot\text{N}\cdot\text{kg}^{-1}$ in the 2017 cropping season and from 0.1 to $6.2 \text{ mg}\cdot\text{N}\cdot\text{kg}^{-1}$ in the 2018 cropping season for CRNPK (Figure 5a). We observed the highest $\text{NH}_4^+\text{-N}$ content in the NPK treatment in the 2017 cropping season and in the CRNPK treatment in the 2018 cropping season. Compared to that in the summer maize season, the soil $\text{NH}_4^+\text{-N}$ content in the winter wheat season was higher.

Similarly, the soil $\text{NO}_3^-\text{-N}$ content during the winter wheat season ranged from 0.7 to $18.3 \text{ mg}\cdot\text{N}\cdot\text{kg}^{-1}$ in the 2016/2017 cropping season and from 3.5 to $41.8 \text{ mg}\cdot\text{N}\cdot\text{kg}^{-1}$ in the 2017/2018 cropping season for CK (Figure 5b); from 3.9 to $140.4 \text{ mg}\cdot\text{N}\cdot\text{kg}^{-1}$ in the 2016/2017 cropping season and from 12.4 to $122.3 \text{ mg}\cdot\text{N}\cdot\text{kg}^{-1}$ in the 2017/2018 cropping season for NPK (Figure 5b); from 2.6 to $113.0 \text{ mg}\cdot\text{N}\cdot\text{kg}^{-1}$ in the 2016/2017 cropping season and from 1.8 to $89.5 \text{ mg}\cdot\text{N}\cdot\text{kg}^{-1}$ in the 2017/2018 cropping season for OMNPK (Figure 5b); and from 2.5 to $86.5 \text{ mg}\cdot\text{N}\cdot\text{kg}^{-1}$ in the 2016/2017 cropping season and from 1.8 to $155.9 \text{ mg}\cdot\text{N}\cdot\text{kg}^{-1}$ in the 2017/2018 cropping season for CRNPK (Figure 5b). The NPK and CRNPK treatments exhibited significantly higher soil $\text{NO}_3^-\text{-N}$ contents in 2016/2017 and 2017/2018 cropping seasons, respectively.

Figure 5. Seasonal variations of soil ammonia nitrogen ($\text{NH}_4^+\text{-N}$) (a), soil nitrate nitrogen ($\text{NO}_3^-\text{-N}$) (b), and dissolved organic carbon (DOC) (c) contents of topsoil (0–15 cm) of wheat-maize rotations for the four treatments over the two-year period from 2016–2018. The vertical bars indicate the standard errors of spatial replicates ($n = 3$).

Conversely, the soil $\text{NO}_3^-\text{-N}$ content during the summer maize season ranged from 2.2 to 15.5 mg·N·kg⁻¹ in the 2017 cropping season and from 1.0 to 16.1 mg·N·kg⁻¹ in the 2018 cropping season for CK (Figure 5b); from 2.3 to 68.6 mg·N·kg⁻¹ in the 2017 cropping season and from 0.6 to 34.1 mg·N·kg⁻¹ in the 2018 cropping season for NPK (Figure 5b); from 2.8 to 79.0 mg·N·kg⁻¹ in the 2017 season and from 4.2 to 69.1 mg·N·kg⁻¹ in the 2018 cropping season for OMNPK (Figure 5b); and from 4.1 to 38.9 mg·N·kg⁻¹ in the 2017 cropping season and from 1.1 to 67.2 mg·N·kg⁻¹ in the 2018 cropping season for CRNPK (Figure 5b). We observed the highest $\text{NO}_3^-\text{-N}$ content in the OMNPK treatment during the summer maize season in both 2017 and 2018. Compared to that in the summer maize season, the soil $\text{NO}_3^-\text{-N}$ content in the winter wheat season was higher.

Furthermore, the soil dissolved organic carbon (DOC) content during the winter wheat season ranged from 12.1 to 98.7 mg·C·kg⁻¹ in the 2016/2017 cropping season and from 25.3 to 92.1 mg·C·kg⁻¹ in the 2017/2018 cropping season for CK (Figure 5c); from 40.2 to 152.8 mg·C·kg⁻¹ in the 2016/2017 cropping season and from 55.8 to 108.3 mg·C·kg⁻¹ in the 2017/2018 cropping season for NPK (Figure 5c);

from 35.3 to 152.5 mg·C·kg⁻¹ in the 2016/2017 cropping season and from 63.5 to 137.9 mg·C·kg⁻¹ in the 2017/2018 cropping season for OMNPK (Figure 5c); and from 57.4 to 178.9 mg·C·kg⁻¹ in the 2016/2017 cropping season and from 64.9 to 137.9 mg·C·kg⁻¹ in the 2017/2018 cropping season for CRNPK (Figure 5c). The DOC content exhibited a similar pattern across all four treatments, and the highest soil DOC content was observed in the CRNPK treatment in 2016/2017, while the OMNPK and CRNPK treatments showed the highest soil DOC contents in the 2017/2018 cropping season.

Meanwhile, the soil DOC during the summer maize season ranged from 29.3 to 82.9 mg·C·kg⁻¹ in the 2017 cropping season and from 14.6 to 82.7 mg·C·kg⁻¹ in the 2018 cropping season for CK (Figure 5c); from 49.6 to 106.0 mg·C·kg⁻¹ in the 2017 cropping season and from 25.4 to 100.1 mg·C·kg⁻¹ in the 2018 cropping season for NPK (Figure 5c); from 48.0 to 115.5 mg·C·kg⁻¹ in the 2017 cropping season and from 54.5 to 138.8 mg·C·kg⁻¹ in the 2018 cropping season for OMNPK (Figure 5c); and from 63.8 to 125.8 mg·C·kg⁻¹ in the 2017 cropping season and from 41.2 to 194.3 mg·C·kg⁻¹ in the 2018 cropping season for CRNPK (Figure 5c). The CRNPK treatment had the highest soil DOC content during the summer maize season in both the 2017 and 2018 cropping seasons. Table S2 (Supplementary Materials) provides the variation in mean seasonal and annual NH₄⁺-N, NO₃⁻-N, and DOC contents.

3.5. Global Warming Potential (GWP)

Table 2 provides the seasonal and annual GWP of N₂O and CH₄ emissions from all four treatments, including the mean GWP throughout the two-year wheat-maize rotational experimental period. Significant differences were observed between all four treatments in both cropping seasons and years ($p < 0.05$) as shown in Table 2. The mean GWP in the winter wheat season was 0.02 Mg CO₂-eq·ha⁻¹ for CK, 0.21 Mg CO₂-eq·ha⁻¹ for NPK, 0.17 Mg CO₂-eq·ha⁻¹ for OMNPK, and 0.27 Mg CO₂-eq·ha⁻¹ for CRNPK (Table 2). The mean GWP in the winter wheat season for the CRNPK treatment was significantly greater than those for the other treatments, which were significantly different from each other ($p < 0.05$). Moreover, the mean GWP in the summer maize season was 0.06 Mg CO₂-eq·ha⁻¹ for CK, 0.24 Mg CO₂-eq·ha⁻¹ for NPK, 1.34 Mg CO₂-eq·ha⁻¹ for OMNPK, and 0.32 Mg CO₂-eq·ha⁻¹ for CRNPK. In contrast to the pattern observed in the winter wheat season, the mean GWP in the summer maize season for the OMNPK treatment was significantly higher than that in the other treatments, which also significantly differed from each other ($p < 0.05$) (Table 2). During the summer maize season, the OMNPK treatment had a higher GWP in 2017 (compared to 2018), accounting for 71% and 65% of the total GWP, respectively.

Furthermore, the mean annual GWP was 0.08 Mg CO₂-eq·ha⁻¹ for CK, 0.47 Mg CO₂-eq·ha⁻¹ for NPK, 1.80 Mg CO₂-eq·ha⁻¹ for OMNPK, and 0.64 Mg CO₂-eq·ha⁻¹ for CRNPK (Table 2). The OMNPK treatment showed a significantly higher mean annual GWP than the other treatments, which was attributable to the high N₂O emissions during the summer maize season in both years. The CRNPK treatment had the second highest annual GWP throughout the two-year experimental period, which differed significantly from those in the NPK and CK treatments (Table 2). Compared to that in the OMNPK and CRNPK treatments, the annual GWP in NPK was significantly lower in both years, which was attributed to the low N₂O emissions and the smallest CH₄ fluxes.

3.6. Grain Yield

Table 2 provides grain yields from the wheat-maize rotational system throughout the two-year experimental period. During the winter wheat season, the mean grain yields were 0.64 Mg·ha⁻¹ for CK, 2.93 Mg·ha⁻¹ for NPK, 3.50 Mg·ha⁻¹ for OMNPK, and 3.09 Mg·ha⁻¹ for CRNPK (Table 2). The mean grain yields for wheat cropping seasons for OMNPK were significantly greater than those for the other treatments ($p < 0.05$), while the NPK and CRNPK treatments exhibited no significant differences in grain yield.

Table 2. Seasonal and annual global warming potential of CH₄ and N₂O emissions (GWP, Mg CO₂-eq·ha^{−1}), grain yield (Mg·ha^{−1}), and yield-scaled global warming potential of CH₄ and N₂O emissions (yield-scaled GWP, kg CO₂-eq·Mg^{−1} gain) for the four treatments over the two-year period from 2016–2018.

Year	Treatments	Wheat Season			Maize Season			Annual	
		GWP	Yield	Yield-Scaled GWP	GWP	Yield	Yield-Scaled GWP	GWP	Yield-Scaled GWP
2016–2017	CK	0.01 ± 0.01c	0.73 ± 0.13c	22.24 ± 16.22b	0.05 ± 0.02c	1.35 ± 0.10b	34.07 ± 16.86b	0.07 ± 0.03c	33.20 ± 14.48c
	NPK	0.24 ± 0.01a	2.29 ± 0.28b	107.82 ± 21.65a	0.25 ± 0.02b	4.22 ± 0.33a	60.26 ± 1.09b	0.50 ± 0.03b	77.31 ± 4.41bc
	OMNPK	0.18 ± 0.01b	3.14 ± 0.12a	58.11 ± 3.48ab	1.49 ± 0.01a	4.38 ± 0.50a	347.20 ± 36.94a	1.85 ± 0.01a	249.41 ± 21.66a
	CRNPK	0.26 ± 0.02a	2.51 ± 0.12b	103.13 ± 12.36a	0.31 ± 0.04b	4.88 ± 0.09a	63.64 ± 8.85b	0.62 ± 0.06b	83.86 ± 9.80b
2017–2018	CK	0.03 ± 0.01c	0.55 ± 0.06b	54.66 ± 10.75b	0.07 ± 0.01d	0.56 ± 0.16b	134.20 ± 32.72b	0.10 ± 0.01d	98.04 ± 24.85b
	NPK	0.18 ± 0.02b	3.56 ± 0.21a	51.08 ± 5.98b	0.23 ± 0.02c	5.14 ± 0.49a	44.82 ± 4.51c	0.44 ± 0.04c	51.42 ± 5.55c
	OMNPK	0.15 ± 0.00b	3.87 ± 0.15a	39.55 ± 2.47b	1.19 ± 0.01a	5.27 ± 0.25a	226.28 ± 11.72a	1.74 ± 0.01a	190.98 ± 7.00a
	CRNPK	0.29 ± 0.01a	3.68 ± 0.06a	77.65 ± 2.50a	0.33 ± 0.02b	5.26 ± 0.46a	63.88 ± 3.44c	0.66 ± 0.01b	74.29 ± 4.08bc
Mean (2016–2018)	CK	0.02 ± 0.01d	0.64 ± 0.05c	38.45 ± 12.04c	0.06 ± 0.02d	0.96 ± 0.13b	84.14 ± 22.43b	0.08 ± 0.02d	65.62 ± 18.99b
	NPK	0.21 ± 0.02b	2.93 ± 0.16b	79.45 ± 13.08ab	0.24 ± 0.02c	4.68 ± 0.39a	52.54 ± 1.72b	0.47 ± 0.03c	64.37 ± 4.44b
	OMNPK	0.17 ± 0.00c	3.50 ± 0.10a	48.83 ± 1.39bc	1.34 ± 0.01a	4.83 ± 0.13a	286.74 ± 12.62a	1.80 ± 0.00a	220.19 ± 8.19a
	CRNPK	0.27 ± 0.01a	3.09 ± 0.09b	90.39 ± 6.16a	0.32 ± 0.02b	5.07 ± 0.19a	63.76 ± 2.89b	0.64 ± 0.03b	79.08 ± 3.54b

Mean ± SE: Means in columns followed by different lower case letter are significantly different (least significant difference (LSD), $p < 0.05$) and means followed by the same lower case letter are not significantly different (LSD, $p > 0.05$). No amendment as control (CK), mineral fertilizers (NPK), pig slurry combined with mineral fertilizers (OMNPK), and crop residues combined with mineral fertilizers (CRNPK).

Conversely, throughout the summer maize season, the mean grain yields were $0.96 \text{ Mg}\cdot\text{ha}^{-1}$ for CK, $4.68 \text{ Mg}\cdot\text{ha}^{-1}$ for NPK, $4.83 \text{ Mg}\cdot\text{ha}^{-1}$ for OMNPK, and $5.07 \text{ Mg}\cdot\text{ha}^{-1}$ for CRNPK (Table 2). The mean maize grain yields for the NPK, OMNPK, and CRNPK treatments did not differ significantly ($p > 0.05$) in the summer maize cropping seasons. Moreover, over the two-year experimental period, the CRNPK treatment produced the highest annual maize grain yield, but it was not significantly different from the other amendment treatments ($p = 0.58$), indicating the higher productivity of summer maize compared to winter wheat. Furthermore, the maize grain yields for the OMNPK and CRNPK treatments increased by 5 and 16%, respectively, in the winter wheat season and by 3–8% in the summer maize season compared to those in the NPK treatment. These results indicated that combining organic amendments with mineral fertilizers can sustain wheat and maize grain yields.

3.7. Yield-Scaled GWP

Table 2 provides the seasonal and annual yield-scaled GWP during the winter wheat and summer maize seasons. The mean yield-scaled GWP during the winter wheat season for the CK, NPK, OMNPK, and CRNPK treatments was 38.45 , 79.45 , 48.83 , and $90.38 \text{ kg}\cdot\text{CO}_2\text{-eq}\cdot\text{ha}^{-1}\cdot\text{Mg}^{-1}$ grain, respectively (Table 2). The NPK and CRNPK treatments exhibited the highest mean yield-scaled GWP values, but these values did not significantly differ in 2016/2017 (Table 2). With the low N_2O emissions during the winter wheat season, we observed even lower and significantly different yield-scaled GWP values. During the summer maize season, the mean yield-scaled GWP for CK, NPK, OMNPK, and CRNPK was 84.14 , 52.54 , 286.74 , and $63.76 \text{ kg}\cdot\text{CO}_2\text{-eq}\cdot\text{ha}^{-1}\cdot\text{Mg}^{-1}$ grain, respectively (Table 2). The mean yield-scaled GWP for OMNPK was significantly higher than that in the other three treatments ($p < 0.05$), while those in the CK, NPK, and CRNPK treatments did not significantly differ from each other ($p > 0.05$). On the other hand, the N_2O content of the OMNPK treatment was high, which resulted in higher yield-scaled GWP values in both the 2017 and 2018 cropping seasons. Interestingly, the control (CK, no amendment) exhibited similar annual yield-scaled GWP values as NPK and CRNPK in the 2017 cropping season, but significantly higher values than those in NPK and CRNPK in the 2018 cropping season (Table 2). This could be due to the decrease in grain yield and an increase in CH_4 uptake in the CK treatment.

Moreover, the annual yield-scaled GWP values for CK, NPK, OMNPK, and CRNPK were 65.62 , 64.37 , 220.19 , and $79.08 \text{ kg}\cdot\text{CO}_2\text{-eq}\cdot\text{ha}^{-1}\cdot\text{Mg}^{-1}$ grain, respectively (Table 2). The mean annual yield-scaled GWP for the OMNPK treatment was significantly higher than that for the other treatments throughout the two-year experimental period. This was due to the high N_2O flux during the summer maize season. However, no significant differences were observed between the CK, NPK, and CRNPK treatments in terms of the mean annual yield-scaled GWP ($p < 0.05$).

3.8. Relationships between CH_4 Uptake and N_2O Emissions, Environmental Factors, and Soil Substrates

During the winter wheat season, we found a significant correlation between CH_4 uptake and soil temperature (Figure 6a); however, no corresponding significant relationships were observed during the summer maize season (Figure 6c). Moreover, no significant correlations were observed between CH_4 uptake and soil water-filled pore space (WFPS) during the winter wheat and summer maize seasons (Figure 6b,d). Additionally, no significant correlations were observed between CH_4 uptake and $\text{NH}_4^+\text{-N}$ content during the winter wheat season (Figure 7a), while CH_4 uptake and $\text{NH}_4^+\text{-N}$ content were negatively correlated during the summer maize season (Figure 7d). Conversely, CH_4 uptake and $\text{NO}_3^-\text{-N}$ content were negatively correlated during both the winter wheat and summer maize seasons (Figure 7b,e). Moreover, no significant correlations were observed between CH_4 uptake and DOC content during the winter wheat season (Figure 7c), while we found that CH_4 uptake and DOC content were negatively correlated during the summer maize season (Figure 7f). Our study revealed significant correlations between N_2O emissions and soil temperature during the winter wheat and summer maize seasons ($p < 0.0001$) (Figure 8a). Conversely, no significant correlations were observed between N_2O emissions and the WFPS of soil during the winter wheat season (Figure 8b); however,

N_2O emissions and the WFPS of soil were negatively correlated during the summer maize season (Figure 8d). Last, significant linear relationships were observed between N_2O emissions and $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$, and DOC contents during both the winter wheat season and the summer maize season throughout the two-year experimental period ($p < 0.0001$) (Figure 9).

Figure 6. Relationships between CH_4 uptake, soil temperature, and soil water-filled pore space (WFPS) in the wheat season (a,b) and in maize season (c,d) for the wheat and the maize seasons across the four treatments over the two-year period from 2016–2018.

Figure 7. Relationships between CH_4 uptake and $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$, and DOC content and soil in the wheat season (a–c) and in the maize season (d–f) for the wheat and the maize seasons across the four treatments over the two-year period from 2016–2018.

Figure 8. Relationships between N_2O emissions, soil temperature, and soil water-filled pore space (WFPS) in the wheat season (a,b) and in the maize season (c,d) for the wheat and the maize seasons across the four treatments over the two-year period from 2016–2018.

Figure 9. Relationships between N_2O emissions and $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$, and DOC content and soil in the wheat season (a–c) and in the maize season (d–f) for the wheat and the maize seasons across the four treatments over the two-year period from 2016–2018.

4. Discussion

4.1. On CH₄ Fluxes

The average annual CH₄ fluxes of the four treatments ranged from -2.6 to -3.0 kg·C·ha⁻¹·yr⁻¹ (Table 1) throughout the two-year experimental period. Comparatively, average annual CH₄ fluxes from the control (CK, no amendment) significantly differed from those from the amendment treatments, which did not differ significantly from each other (Table 1). The three amendment treatments reduced CH₄ fluxes by approximately 12% compared to those in the control. Similarly, Zhou et al. [12] reported similar CH₄ fluxes, ranging from -1.8 to -2.1 kg·C·ha⁻¹·yr⁻¹, while Liu and colleagues [26] reported corresponding CH₄ fluxes from -1.5 to -1.8 kg·C·ha⁻¹·yr⁻¹ in a wheat-maize rotational system under the application of similar organic amendments. The results from these studies support the assumption that cropland with wheat-maize rotational systems had the smallest CH₄ fluxes and support the hypothesis that rain-fed agricultural soil generally acts as a CH₄ sink [11,13]. The smallest CH₄ fluxes in our experimental site resulted from the relatively high CH₄ oxidation rates associated with light sandy soil, which promotes CH₄ production diffusion within the subsoil during methanogenesis [12]. CH₄ uptake and soil temperature exhibited significant linear relationships during the winter wheat season throughout the two-year experimental period (Figure 6a), indicating that CH₄ uptake increased linearly with an increase in soil temperature. Similar significant relationships between CH₄ uptake and soil temperature during the wheat season were detected by Zhou and co-workers [8] via long-term CH₄ emission measurements. Nevertheless, our study did not find a correlation between CH₄ uptake and soil temperature during the summer maize season (Figure 6b) or the WFPS of soil during either the winter wheat or the summer maize season (Figure 6c,d).

Moreover, although CH₄ uptake was negatively correlated with inorganic NO₃⁻-N content in the soil (Figure 7b), CH₄ uptake did not correlate with soil inorganic N (i.e., NH₄⁺-N and DOC) content during the winter wheat season (Figure 7a,c). Conversely, we found that CH₄ uptake was significantly correlated with soil NH₄⁺-N, NO₃⁻-N, and DOC contents during the summer maize season (Figure 7d–f). This indicated that NH₄⁺-N, NO₃⁻-N, and DOC contents in soil are regulatory factors that influence CH₄ fluxes during the maize growing season in Eutric Regosols. Given that CH₄ uptake from cropland is both the main C and energy sources of methanotrophs, CH₄ uptake decreases when significant changes (i.e., conversion from CH₄ to NH₄⁺-N) occur in soil substrates, as reported by Hu [27]. Additionally, Aronson and Helliher [28] demonstrated that this mechanism is more critical for cropping systems in cropland Eutric Regosols. Moreover, previous studies confirmed the existence of similar structures between CH₄ and NH₄⁺-N, while the preference of most methanotrophs is to oxidize NH₄⁺-N (rather than CH₄) under a higher NH₄⁺-N to CH₄ ratio [29,30]. More studies are necessary to better understand the interactions between soil temperature and soil NH₄⁺-N, NO₃⁻-N, and DOC contents in regulating CH₄ uptake in cropland Eutric Regosols.

4.2. On N₂O Emissions

In this study, both seasonal and annual N₂O emissions differed significantly among the four treatments (Table 1). For the four treatments, the average annual N₂O emissions ranged from 0.4 to 4.6 kg·N·ha⁻¹·yr⁻¹ throughout the two-year experimental period. Similar ranges have been reported, such as from 0.14 to 1.88 kg·N·ha⁻¹·yr⁻¹ [12], from 0.56 to 3.01 kg·N·ha⁻¹·yr⁻¹ [14], and from 0.5 to 2.2 kg·N·ha⁻¹ [31], for wheat-maize cropping systems under different organic fertilization treatments. Compared to the control (CK, no amendment), the NPK, CRNPK, and OMNPK treatments all exhibited a significant increase in average annual N₂O emissions, specifically by 69%, 76%, and 90%, respectively (Table 1). In this study, the mean EF_d varied from 0.28–0.50% during the winter wheat season and from 0.30–2.05% during the summer maize season; these values are significantly higher than the results from wheat-maize cropping systems reported by Hu et al. [32] (i.e., from 0.45–0.59%) and Gao et al. [25] (i.e., from 0.17–0.53%) in the North China Plain. The slight differences reported in OMNPK treatments

for N_2O emissions by different studies can be explained by the long-term incorporation of CRNPK and OMNPK as organic fertilizers, resulting in an increase in N_2O emissions [33–35].

Compared to the findings for the winter wheat season, the highest average annual N_2O emission observed in the OMNPK treatment during the summer maize season was three-fold greater than those in the NPK and CRNPK treatments (Table 1, Figure 3). The mean EF_d values for OMNPK during the winter wheat and summer maize seasons were 0.28% and 2.05%, respectively, while these values were lower than 1% in the winter wheat season and higher than 1% in the summer maize season in a report by the Intergovernmental Panel on Climate Change (IPCC) [36]. The highest N_2O emissions observed in the OMNPK treatment resulted from the energy provided by organic amendments to soil microorganisms being used to increase microbial biomass and N availability during soil N_2O production via nitrification and denitrification processes [12,16]. It is interesting to note that the average annual N_2O emission from the CRNPK treatment was $1.80 \text{ kg}\cdot\text{N}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$, which was within the range of 0.20 to $4.54 \text{ kg}\cdot\text{N}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ reported by Huang and colleagues [13], but higher than the range of 0.29 to $1.25 \text{ kg}\cdot\text{N}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ reported by Htun and co-workers [9] in wheat-maize systems under crop residue returning agricultural practices. Moreover, Huang et al. [37] demonstrated that the application of crop residue with a high C/N ratio can induce soil microbes to stimulate N immobilization under mineral fertilizers application. The decrease in N_2O emissions observed in the CRNPK treatment was caused by the organic C added, resulting in the full stimulation of denitrification through the reduction of N_2O to N_2 . Moreover, the organic C added did not heighten the microbial competition driven inorganic N immobilization of $\text{NH}_4^+\text{-N}$ for nitrification or $\text{NO}_3^-\text{-N}$ for denitrification [38–40]. The average EF_d in the CRNPK treatment during both the winter wheat season and the summer maize season was 0.50% and 0.42%, respectively, which was 1% lower than that reported by the IPCC [36].

Regarding the temporal and spatial heterogeneity in environmental factors, we found strong linear relationships between N_2O emissions and soil temperature (Figure 8a), but these relationships were not associated with the WFPS of soil during the winter wheat season (Figure 8b). Additionally, Zhou and colleagues [8] found significant relationships between N_2O emissions and soil temperature during the winter wheat season in their study with long-term N_2O emission monitoring in wheat-rice cropping systems. During the summer maize season, we found correlations for N_2O emissions with soil temperature and the soil WFPS (Figure 8c,d), which confirmed that soil temperature and soil WFPS controlled intra and inter seasonal variation in N_2O emissions.

Significant linear relationships were also detected between N_2O emissions and soil inorganic N (i.e., $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$) and DOC contents during both the winter wheat season (Figure 9a–c) and the summer maize season (Figure 9d–f) across the four treatments throughout the two-year experimental period. This finding showed that variations in soil $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$, and DOC contents throughout the two-year experimental period did not affect intra or inter seasonal N_2O emissions in this study. Thus, increases in soil $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$, and DOC contents resulted in an increase in Eutric Regosol N_2O emissions. Moreover, our results also showed that both soil temperature and WFPS as well as soil $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$, and DOC contents regulate cropland N_2O emissions. Other studies have also shown that soil temperature, WFPS, $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$, and DOC are all key controllers of N_2O emissions from cropland [41–43]. Conversely, Yao et al. [44] showed that N_2O emissions are controlled not only by individual environmental factors, such as soil temperature, WFPS, inorganic N, and dissolved organic C availability, but also by concurrent interactions between these factors. Thus, the aim of our study was to summarize the combined effects of environmental factors on Eutric Regosols.

4.3. GWP and Yield-Scaled GWP

The GWP was lower during the winter wheat season than during the summer maize season across all four treatments and throughout the two-year experimental period (Table 2). On average, the GWP across all four treatments throughout the two-year experimental period during the winter wheat season was $0.17 \text{ Mg}\cdot\text{CO}_2\text{-eq}\cdot\text{ha}^{-1}$, which was significantly lower than that reported

by [12] (i.e., $0.3 \text{ Mg}\cdot\text{CO}_2\text{-eq}\cdot\text{ha}^{-1}$) and [2] (i.e., $0.66 \text{ Mg}\cdot\text{CO}_2\text{-eq}\cdot\text{ha}^{-1}$). Moreover, the average GWP across all four treatments throughout the two-year experimental period during the summer maize season was $0.49 \text{ Mg}\cdot\text{CO}_2\text{-eq}\cdot\text{ha}^{-1}$, which was significantly higher than that reported by Zhou et al. [12] (i.e., $0.38 \text{ Mg}\cdot\text{CO}_2\text{-eq}\cdot\text{ha}^{-1}$) and in a meta-analysis by Linquist and colleagues [2] (i.e., $1.40 \text{ Mg}\cdot\text{CO}_2\text{-eq}\cdot\text{ha}^{-1}$). The GWP during the summer maize season was approximately three-fold greater than that during the winter wheat season, which was similar to findings from Linquist and colleagues [2]. Throughout the two-year experimental period, the OMNPK treatment exhibited a significantly higher GWP than the other treatments (Table 2). The mean annual GWP across all four treatments was $0.75 \text{ Mg}\cdot\text{CO}_2\text{-eq}\cdot\text{ha}^{-1}$, in the range reported by Linquist and colleagues [2] (i.e., 0.003 to $4.40 \text{ Mg}\cdot\text{CO}_2\text{-eq}\cdot\text{ha}^{-1}$) from a meta-analysis of 62 studies and 328 global observations. The highest N_2O emission values observed during the summer maize season contributed to the significantly higher GWP in the OMNPK treatment than in the other treatments. The main difference in GWP between the winter wheat season and the summer maize season was due to emissions of N_2O , the dominant greenhouse gas emitted under rain-fed agricultural conditions. Nonetheless, this study did not include the GWP for fertilizer production or other components for net GWP determination, as previously reported by Huang and colleagues [13]. Further studies are needed to integrate GWP for fertilizer production, power and fuel, soil organic carbon changes, and pesticides to fully characterize net GWP in Eutric Regosols.

The yield-scaled GWP was consistently lower during the winter wheat season than during the summer maize season. For all four treatments, the average yield-scaled GWP throughout the two-year experimental period during the winter wheat season was $64 \text{ kg}\cdot\text{CO}_2\text{-eq}\cdot\text{Mg}^{-1}$ grain, which was lower than that reported by Zhou et al. [12] (i.e., $82 \text{ kg}\cdot\text{CO}_2\text{-eq}\cdot\text{Mg}^{-1}$ grain) and significantly lower than that reported by Linquist and colleagues [2] (i.e., $166 \text{ kg}\cdot\text{CO}_2\text{-eq}\cdot\text{Mg}^{-1}$ grain). Moreover, the average yield-scaled GWP value for all four treatments throughout the two-year experimental period during the summer maize season was $122 \text{ kg}\cdot\text{CO}_2\text{-eq}\cdot\text{Mg}^{-1}$ grain, which was higher than that reported by Zhou et al. [12] (i.e., $84 \text{ kg}\cdot\text{CO}_2\text{-eq}\cdot\text{Mg}^{-1}$ grain) and lower than that reported by Linquist and colleagues [2] (i.e., $185 \text{ kg}\cdot\text{CO}_2\text{-eq}\cdot\text{Mg}^{-1}$ grain). In our study, average yield-scaled GWP values varied significantly among the four treatments, demonstrating the importance of conducting further yield-scaled studies applying a greater quality and quantity of organic amendments with which to identify the optimum greenhouse gas (GHG) and grain yield values when selecting mitigation strategies. Our study showed that the average annual yield-scaled GWP value accounted for 87% of grain yield (Table 2). Other studies have further shown that lower yield-scaled GWP values lead to approximately 90% of the maximum potential grain yield [2,45]. Several studies have recommended the use of yield-scaled GWP when evaluating CH_4 and N_2O emission mitigation without compromising grain yields from cropland [2,11,22].

5. Conclusions

The four treatments evaluated in this study had various effects on seasonal and annual CH_4 and N_2O emissions, GWP, and yield-scaled GWP. N_2O emissions made the greatest contribution to GWP in the OMNPK treatment (i.e., the pig slurry combined with mineral fertilizer treatment) during the summer maize season. Moreover, both the winter wheat and summer maize seasons exhibited distinct patterns of spatiotemporal variability in N_2O emissions. The results revealed significant correlations between N_2O emissions and soil temperature, the WFPS of soil, soil inorganic N (NH_4^+ -N and NO_3^- -N), and soil DOC during both the winter wheat and summer maize seasons. Given the short time span of this study (two years), long-term observations of soil organic carbon stock changes associated with greenhouse gas emissions under organic amendment applications to croplands of Eutric Regosols are necessary to determine the C inputs and mineralization equilibrium.

Supplementary Materials: The following are available online at <http://www.mdpi.com/2073-4433/11/6/614/s1>, Table S1: Mean seasonal and annual variation of soil temperature ($^{\circ}\text{C}$), soil water-filled pore space (WFPS, %), methane emissions (CH_4), and nitrous oxide (N_2O) emissions for the four treatments over the two-year period

from 2016–2018. Table S2: Mean seasonal and annual variation of soil ammonia nitrogen ($\text{NH}_4^+\text{-N}$), nitrate nitrogen ($\text{NO}_3^-\text{-N}$), and dissolved organic (DOC) contents for the four treatments over the two-year period from 2016–2018.

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