

# Carbon budget and greenhouse gas balance during the initial years after rice paddy conversion to vegetable cultivation



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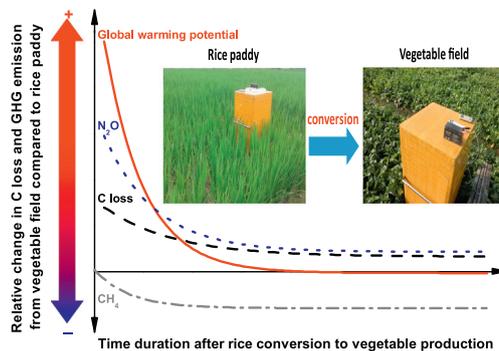
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## HIGHLIGHTS

- N fertilized rice paddy soil sequestered 1.14 Mg C ha<sup>-1</sup> yr<sup>-1</sup>.
- Conversion of rice paddy to vegetable cultivation led to substantial soil C losses.
- Low C input and fast decomposition explained C loss after land-use conversion (LUC).
- The GWP (C loss, CH<sub>4</sub> and N<sub>2</sub>O) strongly increased in the first year after LUC.
- It is especially critical to consider C and GHG balance in the first year after LUC.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Rice paddy conversion to vegetable production is a common agricultural practice driven by economic benefits and shifting diets. However, little is known on the initial effects of this land-use conversion on net ecosystem carbon budget (NECB) and greenhouse gas (GHG) balance. Annual NECB and emissions of CH<sub>4</sub> and N<sub>2</sub>O were measured from a native double rice cropping system (Rice) and a vegetable field recently converted from rice paddy (Veg) under no nitrogen (N) fertilization (Rice-N<sup>0</sup> and Veg-N<sup>0</sup>) and conventional N fertilization (Rice-N<sup>+</sup> and Veg-N<sup>+</sup>) during the initial four years upon conversion in subtropical China. Land-use conversion from rice to vegetable cultivation led to substantial C losses (2.6 to 4.5 Mg C ha<sup>-1</sup> yr<sup>-1</sup>), resulting from strongly reduced C input by 44–52% and increased soil organic matter mineralization by 46–59% relative to Rice. The magnitude of C losses from Veg was highest in the first year upon conversion, and showed a decreasing trend over time. N fertilization shifted rice paddy from a slight C source in Rice-N<sup>0</sup> (−1.0 Mg C ha<sup>-1</sup> yr<sup>-1</sup>) to a significant C sink in Rice-N<sup>+</sup> (1.1 Mg C ha<sup>-1</sup> yr<sup>-1</sup>) and alleviated the impact of land-use conversion on C loss via increased C input from higher crop productivity. Land-use conversion greatly increased the global warming potential (GWP) from Veg by 116–395% relative to Rice in the first year, primarily due to increased C losses and N<sub>2</sub>O emission outweighing the decreased CH<sub>4</sub> emission. However, the GWP did not show obvious difference between Rice and Veg in the

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following years. N fertilization and land-use conversion interactively increased GWP in the first year via increased N<sub>2</sub>O production. Concluding, NECB and GHG emissions in the first year after conversion are crucial and should be considered when evaluating the environmental consequences of land-use conversion.

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## 1. Introduction

Rice is the staple food for over 50% of population on earth. As the world's largest rice producer, China contributed 35% of the world's total rice production in 2012 (FAO, 2013). Given the decline in profitability of traditional rice cultivation while increasing demands and economic benefits from vegetables, a considerable share of rice paddy fields have been and will still be drained for vegetable production in China (Hao et al., 2008; Lu et al., 2010). Previous studies demonstrated that agricultural land-use conversion (LUC) has consequences for soil physico-chemical properties, biodiversity and the associated biogeochemical cycles (Kong et al., 2009; Sheng et al., 2013; Wang et al., 2014). Soil carbon (C) and nitrogen (N) cycling, and associated C balance and greenhouse gas (GHG) emissions are of increasing concern in the context of agricultural productivity and climate change (Lal et al., 2015; Weller et al., 2016; McCalmont et al., 2017).

Submerged conditions for rice cultivation have the potential to sequester C released by plants into the soil (Kögel-Knabner et al., 2010), and is responsible for considerable CH<sub>4</sub> emission (Linguist et al., 2012) while acting as negligible or relevant source and sink of N<sub>2</sub>O (Mejjide et al., 2017). Flooded rice paddy conversion to upland cultivation modifies cropping systems associated with differences in water regimes, cultivation intensification and N fertilization rates. These management practices have profound impacts on soil C and N transformation processes and therefore soil organic carbon (SOC) and GHG balance (Nishimura et al., 2008; Wang et al., 2014; Qin et al., 2016). Soil C dynamics is the predominant determinant of soil fertility and quality, and is closely related to crop productivity and sustainability (Lal, 2004; Nishimura et al., 2008). LUC induces changes in C inputs into the soil through rhizodeposits, dead roots, crop straws and organic manure incorporation, and C outputs by emissions of CO<sub>2</sub> and CH<sub>4</sub> and by dissolved organic carbon (DOC) leaching, leading to a net build-up or depletion of SOC pool (Nishimura et al., 2008). The magnitude of C input and output and resulting change rates of SOC stock following LUC varies widely over time, and is most pronounced during the initial years and become stabilized after decades (West and Post, 2002; Kurganova et al., 2014; Hounkpatin et al., 2018). The existing studies mostly evaluated the impact of LUC on soil C stock by estimating the overall loss or gain of SOC at steady-state conditions, ignoring the temporal dynamics of SOC during the initial years upon conversion (Battlebayer et al., 2010). Soil C change can be directly calculated by determining SOC stock in croplands using soil core sampling technique (Shang et al., 2011). However, significant changes in SOC in response to LUC cannot be detected in a short-term timescale, due to high spatial heterogeneity and huge background of SOC stock (Don et al., 2007; Smith et al., 2010). The net ecosystem carbon budget (NECB) analysis is a superior tool for indirectly determining the short-term C gains/losses relative to SOC stock variation (Smith et al., 2010). The NECB is the balance between C inputs and outputs. These C fluxes (C inputs and outputs) can be well quantified at finer spatial-temporal scale, thereby providing a scientific basis for improved understanding of C fluxes between the soil and atmosphere in response to LUC (Mu et al., 2008). CO<sub>2</sub> efflux derived from soil organic matter (SOM) mineralization is the primary pathway for C losses from soil and is a major component of terrestrial C budget (Fang et al., 1998; Nishimura et al., 2015). Large amounts of organic matter previously stored in paddy field are particularly vulnerable to increased decomposition after conversion to upland cultivation, potentially contributing to significant C losses (Nishimura et al., 2008). Studies on NECB in response to LUC are

therefore required with respect to mitigating CO<sub>2</sub> emissions and helping maintain soil fertility for sustainable crop production.

Conversion of flooded rice paddy to upland vegetable cultivation reduces CH<sub>4</sub> emission via decreased production accompanying with increased CH<sub>4</sub> consumption (Liu et al., 2015, 2017). While this LUC enhances N<sub>2</sub>O fluxes via accelerated N mineralization and increased mineral N supply for nitrification and denitrification (L. Wu et al., 2017). The mitigation benefits of CH<sub>4</sub> emission may be partially offset or even fully counteracted by the accompanying increased CO<sub>2</sub> and N<sub>2</sub>O emissions in response to LUC. The balance among net emissions of CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O constitutes the overall global warming potential (GWP) of a cropping system. Thus, the trade-offs among changes in emission of these GHGs should be taken into account when assessing LUC response. However, most previous investigations on the impacts of LUC focused on SOC dynamics (Nishimura et al., 2008; Huang et al., 2014) and emissions of CH<sub>4</sub> (Eusufzai et al., 2010; Hu et al., 2016) and N<sub>2</sub>O (Nishimura et al., 2005; L. Wu et al., 2017) separately and extensively. Comprehensive studies that simultaneously address NECB and the GWP in response to LUC are lacking (Weller et al., 2016). It also should be noted that the long-term effects of LUC on soil C balance and GHG emissions are quite different to the short-term impacts. Soil conditions immediately following LUC may accelerate C decomposition and nitrification coupled denitrification processes, possibly resulting in considerable changes in soil C balance and GHG emissions (Nikiéma et al., 2012; Kraus et al., 2016). Significant knowledge gaps exist concerning soil C balance and the dynamics of GHG emissions in the initial years following LUC (Weller et al., 2016; X. Wu et al., 2017). Improved understanding the initial impacts of land-use conversion on NECB and GHG emissions is an urgent need to mitigate climate warming by maintaining crop productivity.

Nitrogen fertilizer application strongly influences the NECB in agroecosystems by promoting plant biomass production thereby increasing biomass returns (Pan et al., 2004), and by decreasing (Zang et al., 2016; Li et al., 2017) or increasing (Dossou-Yovo et al., 2016) the decomposition rates of organic residues and SOC. N fertilization can also regulate CH<sub>4</sub> emission through impacts on the activities of methanogens and methanotrophs (Bodelier and Laanbroek, 2004; Liu et al., 2017), and enhance N<sub>2</sub>O emission via increased available substrates for nitrification and denitrification (X. Zhang et al., 2016). However, how LUC and N fertilization interactively affect soil C dynamics and GHG emission remains unclear.

The objectives of this study were to 1) quantify NECB and the dynamics of CH<sub>4</sub> and N<sub>2</sub>O emissions, and their contributions to the GWP during the initial years upon rice conversion to vegetable cultivation, and 2) investigate how N fertilization modifies the effects of LUC on NECB and GHG emissions. We hypothesized that LUC from rice to vegetable cultivation will lead to substantial C losses, and increase the GWP. N fertilization will alleviate the effects of LUC on NECB, and enhance the effects of LUC on the GWP.

## 2. Materials and methods

### 2.1. Experimental field

The field experiment was conducted at the Changsha Research Station for Agricultural & Environmental Monitoring of the Chinese Academy of Sciences (28°32'46" N, 113°19'50" E, and 80 m elevation) in Jinjing town, Hunan Province, China. The study region belongs to a subtropical humid monsoon climate with annual mean air temperature of

17.5 °C from 1951 to 2011 (Fig. S1). The annual precipitation averages 1370 mm, with 70% of it occurring between April and June. The daily air temperature and precipitation were recorded by an automatic meteorological monitoring system (Intelimet Advantage, Dynamax Inc., USA) station located nearby the experimental site throughout the study period and are shown in Fig. S2. Annual sequence of rice-rice-fallow (double rice cultivation) is predominant in local agriculture, where parts of paddy fields have been converted to vegetable cultivation in recent years. The paddy soil studied is classified as Stagnic Anthrosols derived from granite-weathered Quaternary red parental material. The topsoil (0–20 cm) was characterized by clay texture (44.2% clay, 29.1% silt, and 26.7% sand), with organic carbon of 18.8 g C kg<sup>-1</sup>, total nitrogen of 2.1 g N kg<sup>-1</sup> and pH of 5.4.

## 2.2. Experimental design

Prior to the onset of the experiment, all the selected field plots had been under permanent cultivation of double rice cropping for >100 years with conventional N fertilization for at least 20 years. The experiment was initiated in July 2012, and all the selected plots were cultivated with late rice under conventional mineral N fertilization. After late rice harvest in October 2012, portions of these selected plots were randomly assigned to convert to vegetable (Veg) cultivation, while the rest remained for rice (Rice) production as a reference. Both Rice and Veg were further subdivided for no N fertilization (Rice-N<sup>0</sup> and Veg-N<sup>0</sup>) and conventional N fertilization (Rice-N<sup>+</sup> and Veg-N<sup>+</sup>) in a randomized design with three replicate plots (15 m × 20 m) per treatment. Field management (including cropping rotations, tillage, fertilization dose and time, irrigation, and growth period) over the study period followed local conventional practices and is summarized in Table S1. For double rice cultivation, local rice cultivars (*Oryza sativa* L.), Xiang 45 and Tyou 207 were used for the early and late rice-cropping seasons, respectively. N fertilizer (urea, 120 and 150 kg N ha<sup>-1</sup> season<sup>-1</sup> for early rice and late rice seasons, respectively) was applied with three splits in Rice-N<sup>+</sup>: 50% as basal fertilizer, 30% as tiller fertilizer and 20% as panicle fertilizer. P (calcium superphosphate, 40 kg P<sub>2</sub>O<sub>5</sub> ha<sup>-1</sup>) and K (potassium chloride, 100 kg K<sub>2</sub>O ha<sup>-1</sup>) were concurrently applied as basal fertilizers before rice transplanting for both rice seasons in Rice-N<sup>0</sup> and Rice-N<sup>+</sup>. Rice seedlings (30 days old) were transplanted at a hill density of 16.7 cm by 20 cm. After transplanting, the paddy field remained flooded for approximately one month. Then, it was drained for two weeks, followed by intermittent irrigations until its final drainage one week before rice harvest. For vegetable cultivation, the fields were cultivated with red cabbage, pepper, radish and water spinach rotations according to local vegetable cropping regimes. Before vegetable transplantation/sow, all vegetable fields were plowed, and compound fertilizer (a mixture of (NH<sub>4</sub>)<sub>2</sub>HPO<sub>4</sub> and KCl, with N: P<sub>2</sub>O<sub>5</sub>: K<sub>2</sub>O = 15%: 15%: 15%) was applied as basal N fertilizer in Veg-N<sup>+</sup>. For the topdressing, urea was dissolved in the water and then evenly applied to the field with irrigation. Veg-N<sup>0</sup> was treated without N fertilization, but additional field management practices were the same as those of Veg-N<sup>+</sup>. Detailed information, including the crops, fertilization dose and time, is provided in Table S1. All the plots were kept weed-free by regularly weeding if necessary. No organic fertilizers were added to rice paddy and vegetable fields during the study period. For rice, grain and parts of the aboveground straw were removed, leaving stubbles (10 to 15 cm in height) and roots in the field at the harvest stage. For vegetables, all the crop residues were removed from the field when harvest.

## 2.3. Measurements of CH<sub>4</sub> and N<sub>2</sub>O fluxes, ecosystem respiration and heterotrophic respiration rates

Fluxes of CH<sub>4</sub> and N<sub>2</sub>O, and ecosystem respiration (R<sub>e</sub>) rates were measured in situ simultaneously in all plots using the static opaque chamber method (Zheng et al., 2008) from July 2012 to July 2016 with a total of 416 sampling events. In each plot, a square stainless steel

chamber base frame (0.5 m × 0.5 m × 0.3 m) with a groove around the top edge was inserted 30 cm into the soil and remained in situ except for tillage. Each of the square chambers (0.5 m × 0.5 m × 1.0 m high) was equipped with a battery-powered circulating fan inside to ensure homogenization of gases in the chamber headspace, and wrapped with plastic foam insulation outside to minimize temperature change inside during gas sampling. A 30 cm long Teflon tube (internal diameter of 0.2 cm) was connected to the backside of each chamber for gas sampling. When gas sampling, the chamber was manually placed over the chamber base frame with a groove that was filled with water to provide an airtight seal. The planting density of rice seedlings or vegetables inside the frames was identical to that outside the frames. For soil heterotrophic respiration measurement (R<sub>h</sub>, the proxy for SOC mineralization), an additional base frame was installed in each Rice-N<sup>+</sup> and Veg-N<sup>+</sup> plot without plants to serve as a root exclusion area for CO<sub>2</sub> fluxes measurement. The root exclusion subplot was 2 × 2 m, providing sufficient distance to prevent adjacent crop root growth into the subplot. All the root exclusion subplots were kept free of plant by regular weeding. A boardwalk was constructed to allow access to each base frame without soil disturbance when gas sampling. For each measurement of CH<sub>4</sub>, N<sub>2</sub>O and CO<sub>2</sub> fluxes, five gas samples were collected from the headspace of each chamber using 30-mL polypropylene syringes via the Teflon tube that was fitted with a three-way stopcock at 0, 10, 20, 30, and 40 min after chamber closure. The collected gas samples were immediately transferred into 12-mL pre-evacuated glass vials sealed with screw-cap septa (Labco Exetainer, Labco Limited, UK), and analyzed within 24 h on a modified gas chromatograph (Agilent 7890A, Agilent Technologies, Palo Alto, California, USA). The gas chromatograph was equipped with a flame ionization detector (FID) for CO<sub>2</sub> and CH<sub>4</sub> concentration analyses at 250 °C and an electron capture detector (ECD) for N<sub>2</sub>O concentration analyses at 350 °C, and the carrier gas was purified N<sub>2</sub>. The gas samples were collected between 9:00 and 11:00 a.m. to approximate average daily fluxes and minimize diurnal variation in flux patterns (Reeves and Wang, 2015). In each plot, CH<sub>4</sub>, N<sub>2</sub>O and CO<sub>2</sub> fluxes were measured twice per week from July 2012 to July 2016, with greater frequency (every two days for 7–10 days) following tillage, fertilization and irrigation events. The air temperature inside the chamber and soil temperature at depth of 5 cm belowground in the vicinity of the base frames were measured using portable digital thermometers (JM624, Liwen Electronics LTD, Tianjin, China) when gas sampling. Surface water depths in the rice paddy plots and soil moisture were also monitored in each plot throughout the study period. CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O fluxes were calculated on the basis of changes in gas concentration in the chamber measured over the closure time and corrected by chamber temperature and atmospheric pressure. Annual cumulative CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O emissions were calculated by sequentially linear interpolation of fluxes between every two adjacent sampling intervals.

## 2.4. Calculation of net ecosystem carbon budget

The NECB, based on mass balance approach, was calculated as the difference between C inputs and outputs to estimate the rate of organic C accumulation (or loss) from each plot (Smith et al., 2010). NECB was calculated using the following equations based on previous studies (Jia et al., 2012; Ma et al., 2013; Wang et al., 2015), with negative values indicating net C loss from ecosystem and positive values net sink of atmospheric CO<sub>2</sub>:

$$\text{NECB} = \sum \text{C input} - \sum \text{C output} = \text{GPP} - (\text{R}_e + \text{Harvest} + \text{CH}_4) \quad (1)$$

$$\text{GPP} = \text{NPP} + \text{R}_a \quad (2)$$

$$\text{R}_e = \text{R}_a + \text{R}_h \quad (3)$$

where, GPP (gross primary production) was inferred from NPP (net

primary production) via the NPP/GPP ratio of 0.52 deduced from the resulting MODIS GPP and NPP products from Zhang et al. (2009).  $R_e$  (the sum of plant respiration ( $R_a$ ) and soil microbial respiration ( $R_h$ )),  $R_h$ , and  $CH_4$  emissions were measured using the above described static chamber method. Harvest included grain and residue C removed from the field at the harvesting stage. Soil C losses through emission of volatile organic compounds, runoff, and leaching were negligible (Smith et al., 2010; Hounkpatin et al., 2018), and therefore not accounted for in the present study.

$R_h$  was not determined in Rice- $N^0$  and Veg- $N^0$ , and was estimated indirectly by Eq. (4):

$$R_h = R_e - R_a = R_e - (GPP - NPP) \quad (4)$$

NPP in cropland was estimated by Eq. (5) (Smith et al., 2010):

$$NPP = NPP_{\text{aboveground}} + NPP_{\text{root}} + NPP_{\text{litter}} + NPP_{\text{rhizodeposits}} \quad (5)$$

The NPP of aboveground and root were calculated using the dry biomass weighed and corresponding C content at harvest.  $NPP_{\text{litter}}$  and  $NPP_{\text{rhizodeposit}}$  were estimated using allometric functions. Litter was estimated to account for 5% and 3% of the total biomass of rice and vegetable, respectively (Kimura et al., 2004). Rhizodeposits (including exudates, root hairs and fine roots sloughed off) were estimated to account for 15% (Mandal et al., 2008) and 7% (Gregory, 2006) of the total biomass for rice and vegetables, respectively. The default value of 0.70 Mg C  $ha^{-1}$  for C load of aquatic algal mass in rice paddy was used for C input (Mandal et al., 2008). Shifts in C allocation to aboveground and belowground in response to fertilization or management were not accounted for in the present study.

At the physiological maturity stage of each crop, aboveground and belowground biomass were measured from three randomly sampling quadrats (1 m  $\times$  1 m) in each plot, and the average biomass of the three harvested areas was treated as a replicate for each plot. The aboveground biomass was collected by cutting the stems close to the soil surface. The belowground biomass was determined by collecting two soil cores at the center of each quadrat using an 8-cm-diameter soil auger to a depth of 30 cm. The soil core samples were carefully washed with rinsing water to remove the soil so as to measure root biomass. Collected plant biomass was separated into harvested grain, shoot, leaves and root. All biomass samples were oven-dried at 65 °C to a constant mass and weighted, and subsamples were ground for C content analysis using a CN analyzer (Vario Max CN, Elementar, Hanau, Germany). The C inputs via root and aboveground biomass were calculated by multiplying dry biomass of crop residues remaining in soil and corresponding C contents. The amounts of C removed as the harvested crop were also determined.

For annual NECB calculation, an entire year is considered nominally from harvest to harvest.

### 2.5. Net global warming potential

The net GWP (Mg  $CO_2$ -equivalent  $ha^{-1}$ ) of the cropping system was calculated as the  $CO_2$  equivalent of  $CH_4$  and  $N_2O$  emissions minus NECB using Eq. (6) based on 100-year time horizon (IPCC, 2013; Ma et al., 2013; Hwang et al., 2017):

$$\text{Net GWP} = 28 \times CH_4 + 265 \times N_2O - 44/12 \times \text{NECB} \quad (6)$$

### 2.6. Statistical analysis

Analysis of variance was used to evaluate the effects of LUC, N fertilization, year, and their interactions on NECB,  $CH_4$  and  $N_2O$  emissions, and GWP. To examine the temperature sensitivity ( $Q_{10}$ ) of  $R_h$ , exponential regression models were used to determine the relationships

between  $R_h$  and soil temperature (T):  $R_h \text{ flux} = \alpha e^{\beta T}$ ,  $\alpha$  and  $\beta$  were constants.  $Q_{10}$  of  $R_h$  flux was calculated as  $Q_{10} = e^{10\beta}$ . Linear regression analyses were used to determine whether significant relationships existed between annual emissions of  $CH_4$  and  $N_2O$  versus both NPP and NECB. The significant level of all statistical tests was set at the value of  $p < 0.05$ . All data statistical analysis was conducted using the SPSS software package (SPSS 20.0, SPSS Inc., IL, Chicago, USA).

## 3. Results

### 3.1. Environmental conditions

Daily precipitation and air temperature ( $T_a$ ) varied seasonally across the four-year study period (Fig. S2). Annual rainfall ranged from 1230 to 1385 mm, with mean value (1317 mm) close to the 60-year average (1370 mm, Fig. S1). Daily  $T_a$  ranged from  $-1.5$  to 34.3 °C, and the four-year mean value (17.8 °C) was slightly higher than the preceding long-term average (17.5 °C, Fig. S1).

Soil temperature ( $T_s$ ) followed the same seasonal pattern of  $T_a$ , with minimum values in January and maximums in July (Fig. S2). Conversion from rice to vegetable cultivation significantly reduced soil volumetric water content from 58% to 39% on average over the four years (Fig. S2).

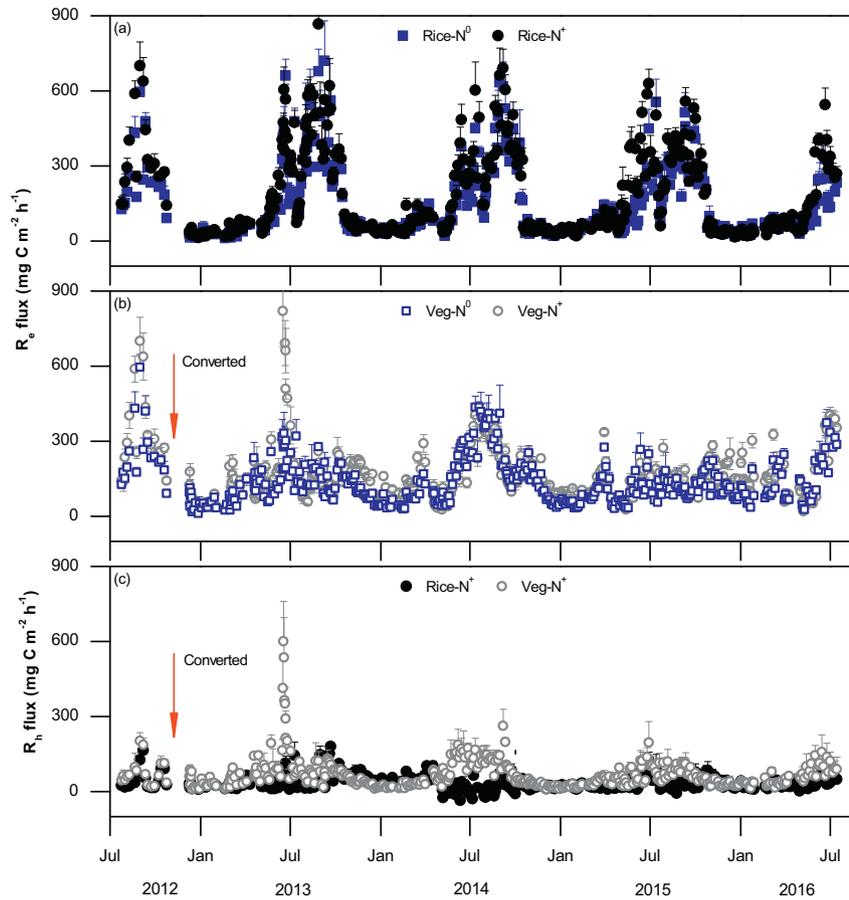
### 3.2. Components of NECB

$R_e$  fluxes ranged from 10.8 to 865.8 mg C  $m^{-2} h^{-1}$  in Rice and 11.6 to 1025.5 mg C  $m^{-2} h^{-1}$  in Veg over the study period, following the seasonal patterns of plant growth and  $T_a$  (Fig. 1a and b). LUC decreased the annual cumulative  $R_e$  from 12.9 Mg C  $ha^{-1}$  in Rice- $N^0$  to 10.5 Mg C  $ha^{-1}$  in Veg- $N^0$  and from 15.8 Mg C  $ha^{-1}$  in Rice- $N^+$  to 12.5 Mg C  $ha^{-1}$  in Veg- $N^+$  averaged over the four years ( $p < 0.05$ , Table 1). N fertilization significantly increased  $R_e$  in Rice and Veg, while it did not interact with LUC to affect  $R_e$  (Table 2).

$R_h$  fluxes from Rice- $N^+$  ranged from  $-34.5$  to 180.9 mg C  $m^{-2} h^{-1}$ , with some negative values during rice growing seasons (Fig. 1c). For Veg- $N^+$ ,  $R_h$  fluxes ranged from 9.0 to 599.1 mg C  $m^{-2} h^{-1}$ , following the same seasonal pattern of  $R_e$ . Annual C loss via  $R_h$  was greatly influenced by LUC, year, and their interaction (Tables 1, 2). Annual cumulative  $R_h$  from Veg- $N^+$  increased ( $p < 0.05$ ) by 52–99% relative to Rice- $N^+$  during the first three years, but no difference in  $R_h$  existed in the fourth year (Table 1).  $R_h$  of Veg- $N^+$  decreased from 6.30 to 3.41 Mg C  $ha^{-1} yr^{-1}$  over time. There existed positive exponential relationships between  $R_h$  fluxes and  $T_s$  in Rice- $N^+$  and Veg- $N^+$  across the four years except for Rice- $N^+$  in 2013/2014 and 2015/2016 (Fig. S3). The  $R_h$  fluxes from Veg- $N^+$  responded more strongly to increasing  $T_s$  than that from Rice- $N^+$ , owing to lower soil moisture in Veg- $N^+$  relative to Rice- $N^+$ . This result suggested that temperature plays a key role in  $R_h$  flux when soil moisture is optimal. The temperature sensitivity ( $Q_{10}$ ) of  $R_h$  in Rice- $N^+$  was 1.2, smaller than that in Veg- $N^+$  (1.9) over the four years, indicating that LUC strengthened the impact of  $T_s$  on  $R_h$ .

LUC decreased ( $p < 0.05$ ) NPP from 9.3 Mg C  $ha^{-1} yr^{-1}$  in Rice- $N^0$  and 13.3 Mg C  $ha^{-1} yr^{-1}$  in Rice- $N^+$  to 4.4 Mg C  $ha^{-1} yr^{-1}$  in Veg- $N^0$  and 7.4 Mg C  $ha^{-1} yr^{-1}$  in Veg- $N^+$  averaged over the four years (Fig. 2). N fertilization increased NPP in Rice and Veg, and it also interacted with LUC to affect NPP ( $p < 0.05$ , Table 2).

For Rice- $N^0$ , C inputs via NPP ranged from 8.2 to 10.4 Mg C  $ha^{-1} yr^{-1}$ , being insufficient to compensate for the C outputs (ranging from 9.3 to 11.7 Mg C  $ha^{-1} yr^{-1}$ ) via harvest and decomposed C losses ( $R_h$  and  $CH_4$  emission). The balance between C inputs and outputs resulted in Rice- $N^0$  acting as a net C loss with NECB ranging from  $-1.7$  to  $-0.03$  Mg C  $ha^{-1} yr^{-1}$  (Fig. 2). N fertilization increased ( $p < 0.05$ ) C input in Rice- $N^+$  by 39–49% relative to Rice- $N^0$ , shifting rice field from a slight C source in Rice- $N^0$  to a significant C sink in Rice- $N^+$  (0.6 to 1.8 Mg C  $ha^{-1} yr^{-1}$ ). LUC decreased C input in Veg by 44–52% relative to Rice over the four years ( $p < 0.05$ ). The C losses via decomposition increased significantly in response to LUC. Furthermore, the contribution of



**Fig. 1.** Dynamics of (a) ecosystem respiration rates ( $R_e$ ) from rice paddy without nitrogen fertilization (Rice-N<sup>0</sup>) and rice paddy with conventional nitrogen fertilization (Rice-N<sup>+</sup>), (b)  $R_e$  from vegetable field without nitrogen fertilization (Veg-N<sup>0</sup>) and vegetable field with conventional nitrogen fertilization (Veg-N<sup>+</sup>), and (c) soil heterotrophic respiration rates ( $R_h$ ) from Rice-N<sup>+</sup> and Veg-N<sup>+</sup> during the 4-year study period. Values are means  $\pm$  standard errors of three replicates.

decomposed C losses to total C output increased greatly from 33 to 46% in Rice to 56–71% in Veg. However, LUC had no impact on C output, due to the increase of C decomposition offsetting the decreased harvest

removal in Veg. As a result, LUC reduced ( $p < 0.05$ ) NECB of Veg by 3.59–3.70 Mg C ha<sup>-1</sup> yr<sup>-1</sup> compared to Rice, leading to substantial C losses. N fertilization significantly increased C input in Veg-N<sup>+</sup> by 67%,

**Table 1**  
Annual ecosystem respiration ( $R_e$ ), heterotrophic respiration ( $R_h$ ), net ecosystem carbon budget (NECB), CH<sub>4</sub> and N<sub>2</sub>O emissions and net global warming potential (GWP) in the first four years upon land-use conversion from rice to vegetable cultivation.

Year	Treatment	$R_e$ Mg C ha <sup>-1</sup> yr <sup>-1</sup>	$R_h$ Mg C ha <sup>-1</sup> yr <sup>-1</sup>	NECB Mg C ha <sup>-1</sup> yr <sup>-1</sup>	CH <sub>4</sub> kg C ha <sup>-1</sup> y <sup>-1</sup>	N <sub>2</sub> O kg N ha <sup>-1</sup> y <sup>-1</sup>	Net GWP <sup>a</sup> Mg CO <sub>2</sub> -eq ha <sup>-1</sup> y <sup>-1</sup>
2012/2013	Rice-N <sup>0</sup>	11.7 $\pm$ 0.7 <sup>ab</sup>		-1.1 $\pm$ 0.7 <sup>a</sup>	309.7 $\pm$ 11.8 <sup>a</sup>	1.3 $\pm$ 0.2 <sup>c</sup>	16.2 $\pm$ 2.7 <sup>c</sup>
	Rice-N <sup>+</sup>	15.0 $\pm$ 0.3 <sup>a</sup>	3.2 $\pm$ 0.1 <sup>b</sup>	0.6 $\pm$ 0.2 <sup>a</sup>	385.2 $\pm$ 75.0 <sup>a</sup>	1.8 $\pm$ 0.1 <sup>c</sup>	13.1 $\pm$ 2.0 <sup>c</sup>
	Veg-N <sup>0</sup>	10.5 $\pm$ 1.1 <sup>b</sup>		-6.1 $\pm$ 1.0 <sup>b</sup>	25.1 $\pm$ 2.2 <sup>b</sup>	28.0 $\pm$ 3.2 <sup>b</sup>	35.1 $\pm$ 4.2 <sup>b</sup>
2013/2014	Veg-N <sup>+</sup>	12.6 $\pm$ 1.5 <sup>ab</sup>	6.3 $\pm$ 0.7 <sup>a</sup>	-4.5 $\pm$ 1.4 <sup>b</sup>	51.7 $\pm$ 13.8 <sup>b</sup>	111.8 $\pm$ 13.3 <sup>a</sup>	65.0 $\pm$ 5.9 <sup>a</sup>
	Rice-N <sup>0</sup>	14.5 $\pm$ 1.2 <sup>ab</sup>		-1.7 $\pm$ 0.8 <sup>b</sup>	431.7 $\pm$ 60.9 <sup>a</sup>	0.1 $\pm$ 0.1 <sup>c</sup>	22.3 $\pm$ 1.4 <sup>a</sup>
	Rice-N <sup>+</sup>	17.0 $\pm$ 1.3 <sup>a</sup>	4.0 $\pm$ 0.1 <sup>b</sup>	0.7 $\pm$ 0.4 <sup>a</sup>	411.6 $\pm$ 70.0 <sup>a</sup>	0.4 $\pm$ 0.3 <sup>c</sup>	12.9 $\pm$ 3.7 <sup>b</sup>
2014/2015	Veg-N <sup>0</sup>	13.7 $\pm$ 0.5 <sup>b</sup>		-5.5 $\pm$ 0.7 <sup>c</sup>	3.8 $\pm$ 0.3 <sup>b</sup>	5.9 $\pm$ 0.9 <sup>b</sup>	22.7 $\pm$ 2.3 <sup>a</sup>
	Veg-N <sup>+</sup>	15.3 $\pm$ 0.3 <sup>ab</sup>	6.1 $\pm$ 0.5 <sup>a</sup>	-3.0 $\pm$ 0.5 <sup>b</sup>	5.3 $\pm$ 1.0 <sup>b</sup>	14.6 $\pm$ 2.6 <sup>a</sup>	17.2 $\pm$ 2.5 <sup>ab</sup>
	Rice-N <sup>0</sup>	13.3 $\pm$ 0.8 <sup>b</sup>		-0.03 $\pm$ 0.06 <sup>b</sup>	460.3 $\pm$ 89.2 <sup>b</sup>	-0.2 $\pm$ 0.1 <sup>b</sup>	17.2 $\pm$ 3.5 <sup>a</sup>
2015/2016	Rice-N <sup>+</sup>	16.4 $\pm$ 1.4 <sup>a</sup>	2.7 $\pm$ 0.3 <sup>b</sup>	1.8 $\pm$ 0.8 <sup>a</sup>	761.1 $\pm$ 140.8 <sup>a</sup>	-0.06 $\pm$ 0.04 <sup>b</sup>	21.8 $\pm$ 7.7 <sup>a</sup>
	Veg-N <sup>0</sup>	9.1 $\pm$ 0.3 <sup>c</sup>		-4.2 $\pm$ 0.3 <sup>d</sup>	8.5 $\pm$ 2.9 <sup>c</sup>	21.0 $\pm$ 5.0 <sup>a</sup>	24.5 $\pm$ 2.2 <sup>a</sup>
	Veg-N <sup>+</sup>	10.7 $\pm$ 0.3 <sup>bc</sup>	4.6 $\pm$ 0.2 <sup>a</sup>	-1.9 $\pm$ 0.6 <sup>c</sup>	13.9 $\pm$ 0.7 <sup>c</sup>	30.9 $\pm$ 5.9 <sup>a</sup>	20.4 $\pm$ 4.7 <sup>a</sup>
2012/2016	Rice-N <sup>0</sup>	12.2 $\pm$ 0.6 <sup>b</sup>		-1.0 $\pm$ 0.3 <sup>b</sup>	297.5 $\pm$ 57.7 <sup>a</sup>	0.1 $\pm$ 0.0 <sup>c</sup>	14.9 $\pm$ 2.8 <sup>a</sup>
	Rice-N <sup>+</sup>	14.7 $\pm$ 0.8 <sup>a</sup>	2.8 $\pm$ 0.1 <sup>a</sup>	1.5 $\pm$ 0.6 <sup>a</sup>	416.0 $\pm$ 54.4 <sup>a</sup>	0.1 $\pm$ 0.2 <sup>c</sup>	10.0 $\pm$ 4.4 <sup>a</sup>
	Veg-N <sup>0</sup>	8.6 $\pm$ 0.1 <sup>c</sup>		-2.4 $\pm$ 0.1 <sup>c</sup>	1.6 $\pm$ 0.5 <sup>b</sup>	12.6 $\pm$ 3.0 <sup>b</sup>	14.0 $\pm$ 1.6 <sup>a</sup>
2012/2016	Veg-N <sup>+</sup>	11.2 $\pm$ 0.5 <sup>b</sup>	3.4 $\pm$ 0.5 <sup>a</sup>	-0.8 $\pm$ 0.3 <sup>b</sup>	3.2 $\pm$ 0.2 <sup>b</sup>	21.2 $\pm$ 2.4 <sup>a</sup>	12.0 $\pm$ 1.8 <sup>a</sup>
	Rice-N <sup>0</sup>	12.9 $\pm$ 0.3 <sup>b</sup>		-1.0 $\pm$ 0.3 <sup>b</sup>	374.8 $\pm$ 42.7 <sup>a</sup>	0.3 $\pm$ 0.0 <sup>c</sup>	17.6 $\pm$ 0.6 <sup>bc</sup>
	Rice-N <sup>+</sup>	15.8 $\pm$ 0.7 <sup>a</sup>	3.2 $\pm$ 0.1 <sup>b</sup>	1.1 $\pm$ 0.3 <sup>a</sup>	493.5 $\pm$ 81.7 <sup>a</sup>	0.6 $\pm$ 0.1 <sup>c</sup>	14.5 $\pm$ 4.1 <sup>c</sup>
2012/2016	Veg-N <sup>0</sup>	10.5 $\pm$ 0.4 <sup>c</sup>		-4.5 $\pm$ 0.4 <sup>d</sup>	9.8 $\pm$ 0.7 <sup>b</sup>	16.9 $\pm$ 1.6 <sup>b</sup>	24.1 $\pm$ 2.1 <sup>ab</sup>
	Veg-N <sup>+</sup>	12.5 $\pm$ 0.4 <sup>b</sup>	5.1 $\pm$ 0.1 <sup>a</sup>	-2.6 $\pm$ 0.3 <sup>c</sup>	18.5 $\pm$ 3.9 <sup>b</sup>	44.6 $\pm$ 5.8 <sup>a</sup>	28.7 $\pm$ 2.8 <sup>a</sup>

Rice-N<sup>0</sup> and Rice-N<sup>+</sup> indicate no nitrogen fertilization and nitrogen fertilization in rice paddy, respectively. Veg-N<sup>0</sup> and Veg-N<sup>+</sup> indicate no nitrogen fertilization and nitrogen fertilization in vegetable field, respectively. Different lowercase letters indicate significant differences between treatments within specific year at  $p < 0.05$ .

<sup>a</sup> Net GWP (Mg CO<sub>2</sub>-eq ha<sup>-1</sup> y<sup>-1</sup>) = 28  $\times$  CH<sub>4</sub> + 265  $\times$  N<sub>2</sub>O - 44/12  $\times$  NECB.

**Table 2**

Statistical analysis for the effects of land-use conversion (LUC), nitrogen fertilization (F), year (Y) and their interactions on soil moisture, the components of net ecosystem carbon budget (NECB) and greenhouse gas emissions.

	Soil moisture	NPP	Harvest	R <sub>c</sub>	R <sub>h</sub>	NECB	CH <sub>4</sub>	N <sub>2</sub> O	GWP
LUC	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001
F	0.256	<0.001	<0.001	<0.001	<0.001	<0.001	0.027	<0.001	0.708
Y	0.158	<0.001	<0.001	<0.001	<0.001	<0.001	0.008	<0.001	<0.001
LUC × F	0.388	0.006	0.001	0.326		0.861	0.055	<0.001	0.045
LUC × Y	0.080	<0.001	<0.001	0.014	0.035	0.014	0.004	<0.001	<0.001
F × Y	0.741	0.978	0.898	0.951		0.874	0.239	<0.001	0.003
LUC × F × Y	0.928	0.941	0.929	0.916		0.883	0.228	<0.001	0.003

Bold indicates significance at  $p < 0.05$  level.

increasing NECB of Veg-N<sup>+</sup> by 2.0 Mg C ha<sup>-1</sup> yr<sup>-1</sup> relative to Veg-N<sup>0</sup> on average over the four years (Fig. 2). NECB of Veg exhibited large interannual variation. Across the four years, NECB increased from -6.1 to -2.4 Mg C ha<sup>-1</sup> yr<sup>-1</sup> in Veg-N<sup>0</sup> and from -4.5 to -0.8 Mg C ha<sup>-1</sup> yr<sup>-1</sup> in Veg-N<sup>+</sup> over time, primarily resulting from decreased R<sub>h</sub>, indicating the reduction in C loss from Veg yearly after LUC. This study revealed that rice paddy conversion to vegetable cultivation caused substantial soil C loss, particularly in the first year.

### 3.3. CH<sub>4</sub> and N<sub>2</sub>O emissions

CH<sub>4</sub> fluxes from Rice varied seasonally, ranging from -0.04 to 70.9 Mg C m<sup>-2</sup> h<sup>-1</sup>, with 94–99% of the annual emission occurring during rice growing seasons over the four years (Fig. 3a). N fertilization had no impact on CH<sub>4</sub> emission from Rice except for 2014/2015, in which CH<sub>4</sub> emission was significantly higher in Rice-N<sup>+</sup> compared to Rice-N<sup>0</sup> (Table 1). Annual CH<sub>4</sub> emissions from Rice were positively related to NPP and NECB (only for Rice-N<sup>+</sup>) across the four years (Fig. S4). LUC greatly reduced CH<sub>4</sub> emission from Veg by 96–97% relative to Rice (Table 1, Fig. 3). CH<sub>4</sub> fluxes from Veg varied from -0.3 to 7.5 Mg C m<sup>-2</sup> h<sup>-1</sup> over the four years (Fig. 3b). Distinct pulses of CH<sub>4</sub> fluxes from Veg were sustained during the initial few months after conversion, contributing to considerably larger cumulative emission in the first year as compared to any later years (Table 1, Fig. 3b).

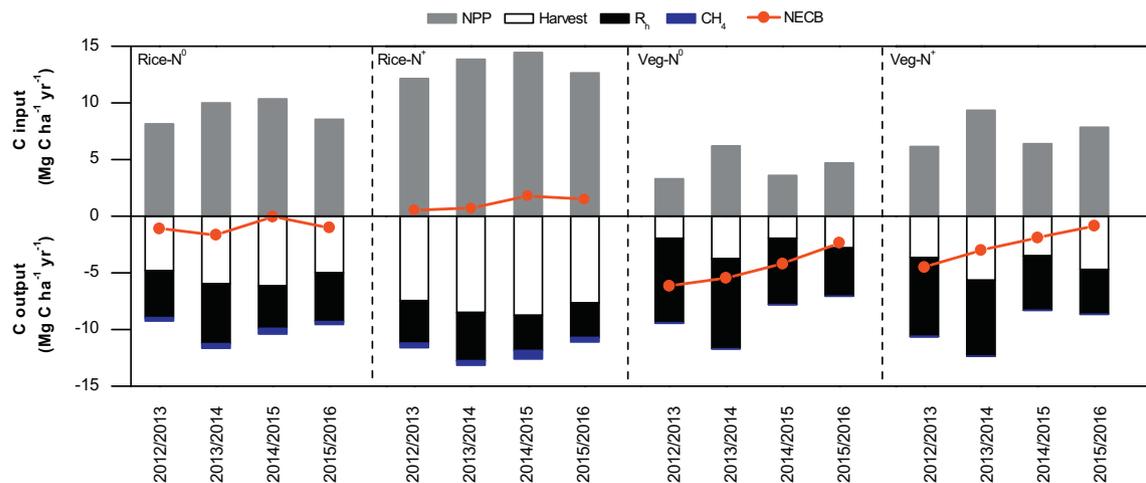
N<sub>2</sub>O fluxes from Rice were at low levels except for several peaks occurring when drainage during rice growing seasons (Fig. 3c). The N<sub>2</sub>O emission ranged from -0.2 to 1.3 kg N ha<sup>-1</sup> yr<sup>-1</sup> in Rice-N<sup>0</sup> and -0.06 to 1.8 kg N ha<sup>-1</sup> yr<sup>-1</sup> in Rice-N<sup>+</sup>. LUC from Rice to Veg greatly increased N<sub>2</sub>O emission (Table 1, Fig. 3c and d). N<sub>2</sub>O fluxes from Veg showed distinct seasonal variation, with higher fluxes during the summer seasons especially following N fertilization. N<sub>2</sub>O emission ranged

from 5.9 to 28.0 kg N ha<sup>-1</sup> yr<sup>-1</sup> in Veg-N<sup>0</sup> and 14.6 to 111.8 kg N ha<sup>-1</sup> yr<sup>-1</sup> in Veg-N<sup>+</sup>, varying greatly across the four years (Table 1). The variability in annual N<sub>2</sub>O emission from Veg was negatively linked to NPP variation (Fig. S4). N fertilization substantially increased N<sub>2</sub>O emission from Veg-N<sup>+</sup> by 164% relative to Veg-N<sup>0</sup> over the four years, and enhanced the effects of LUC on N<sub>2</sub>O emission, leading to the highest annual emission from Veg-N<sup>+</sup> in the first year (Tables 1, 2).

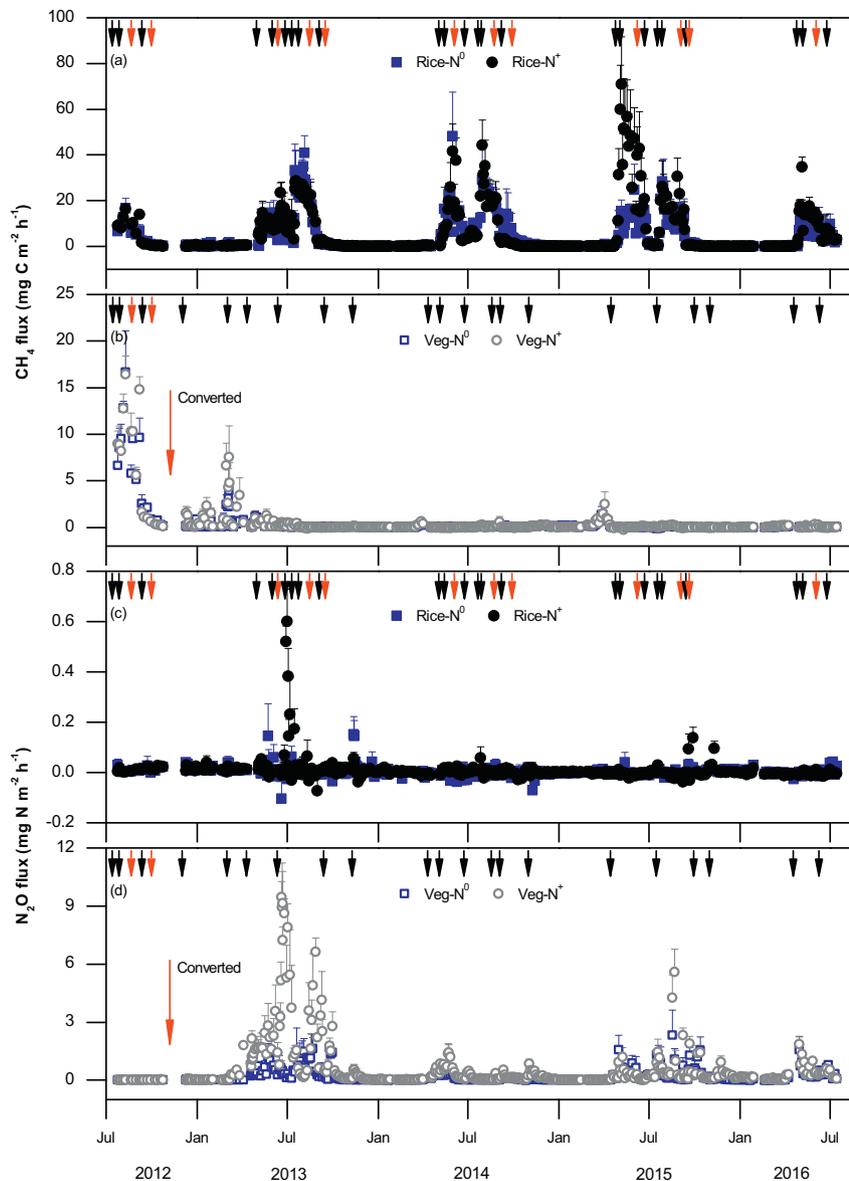
### 3.4. GWP

The GWP of NECB and emissions of CH<sub>4</sub> and N<sub>2</sub>O ranged from 14.9 to 22.3 Mg CO<sub>2</sub>-eq ha<sup>-1</sup> yr<sup>-1</sup> in Rice-N<sup>0</sup> and 10.0 to 21.8 Mg CO<sub>2</sub>-eq ha<sup>-1</sup> yr<sup>-1</sup> in Rice-N<sup>+</sup> (Table 1). N fertilization had no impact on GWP in Rice over the four years. LUC greatly increased GWP in Veg by 116–395% relative to Rice in the first year, resulting from the increased C loss and N<sub>2</sub>O emission far outweighing the decreased CH<sub>4</sub> emission (Fig. 4a and b). While in the following years, difference in annual GWP was not obvious between Rice and Veg, irrespective of N fertilization. N fertilization significantly increased GWP in Veg-N<sup>+</sup> by 85% compared to Veg-N<sup>0</sup> in the first year, resulting from increased N<sub>2</sub>O emission overriding the decreased C loss (Fig. 4d). The interactive effects of LUC and N fertilization on GWP in the first year were higher than their individual effects, suggesting a synergistic effect on increased N<sub>2</sub>O emission. When averaged over the four years, N fertilization enhanced the effects of LUC on GWP.

The relative contribution of individual GHG emission to the GWP changed significantly in response to LUC and N fertilization (Table 1). The annual GWP in Rice was mainly attributed to CH<sub>4</sub> emission across the four years, irrespective of N fertilization. LUC greatly decreased the contribution of CH<sub>4</sub> to the annual GWP from 73 to 151% in Rice to 0.4–3% in Veg, while increased the contribution of N<sub>2</sub>O to GWP from minus 1 to 6% in Rice to 11–75% in Veg. For Veg-N<sup>0</sup>, C loss (NECB)



**Fig. 2.** Annual carbon inputs (NPP: net primary production), carbon outputs (Harvest, R<sub>h</sub>: heterotrophic respiration, CH<sub>4</sub> emission), and net ecosystem carbon budget (NECB, Mg C ha<sup>-1</sup> yr<sup>-1</sup>) in the first four years upon land-use conversion from rice to vegetable cultivation. Rice-N<sup>0</sup> and Rice-N<sup>+</sup> indicate no nitrogen fertilization and nitrogen fertilization in rice paddy, respectively. Veg-N<sup>0</sup> and Veg-N<sup>+</sup> indicate no nitrogen fertilization and nitrogen fertilization in vegetable field, respectively.



**Fig. 3.** Dynamics of (a) CH<sub>4</sub> fluxes from rice paddy without nitrogen fertilization (Rice-N<sup>0</sup>) and rice paddy with conventional nitrogen fertilization (Rice-N<sup>+</sup>), (b) CH<sub>4</sub> fluxes from vegetable field without nitrogen fertilization (Veg-N<sup>0</sup>) and vegetable field with conventional nitrogen fertilization (Veg-N<sup>+</sup>), (c) N<sub>2</sub>O fluxes from Rice-N<sup>0</sup> and Rice-N<sup>+</sup>, and (d) N<sub>2</sub>O fluxes from Veg-N<sup>0</sup> and Veg-N<sup>+</sup> during the 4-year study period. Black vertical arrows indicate N fertilization events, and red vertical arrows indicate mid-season N drainage events. Values are means ± standard errors of three replicates. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

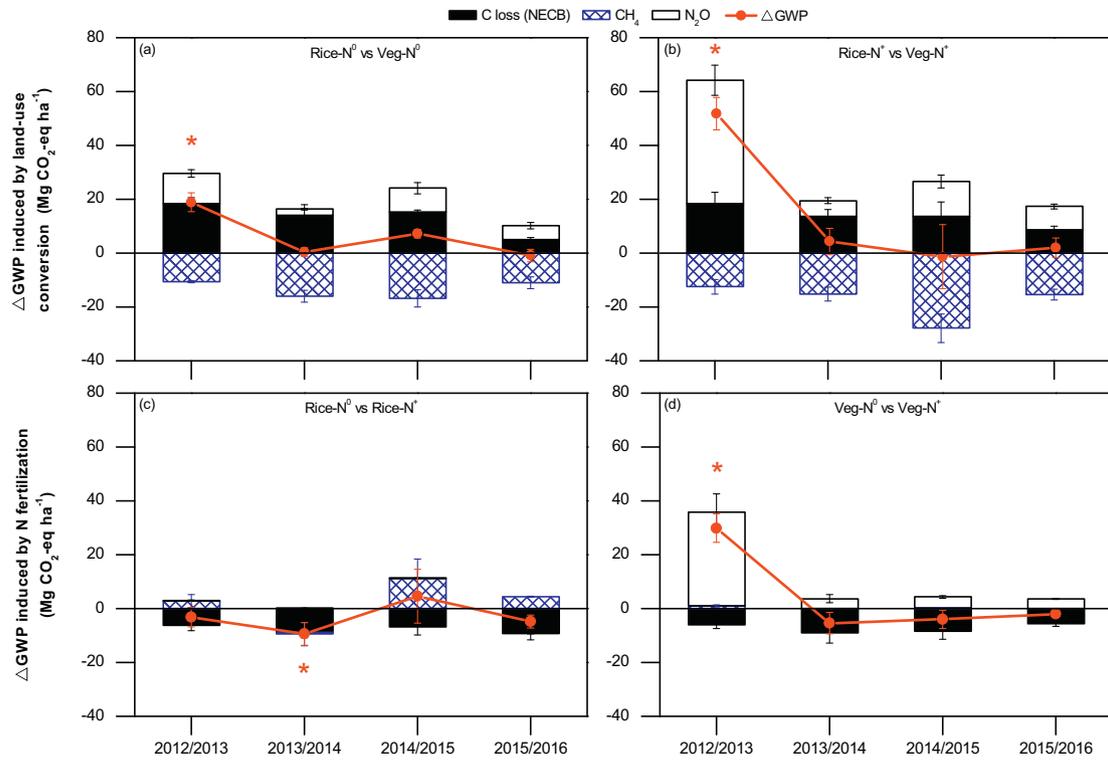
contributed the most (63–88%) of GWP over the four years, followed by N<sub>2</sub>O (11–37%). N fertilization shifted from C loss in Veg-N<sup>0</sup> to N<sub>2</sub>O in Veg-N<sup>+</sup> as the dominant GHG contributing to the annual GWP over the four years, with the exception of the second year when C loss contributed most (64%) in Veg-N<sup>+</sup>.

## 4. Discussion

### 4.1. Effects of LUC and N fertilization on NECB

The NECB provides a scientific basis to determine the short-term soil C gains and losses via net C exchange between the terrestrial ecosystem and atmosphere (Smith et al., 2010). The NECB of Rice-N<sup>0</sup> averaged  $-1.0 \text{ Mg C ha}^{-1} \text{ yr}^{-1}$  over the four-year study period, indicating net C losses from rice paddy without N fertilization (Fig. 2). N fertilization shifted the rice paddy from a slight C source to a significant C sink, with averaged NECB of  $1.1 \text{ Mg C ha}^{-1} \text{ yr}^{-1}$  in Rice-N<sup>+</sup>. The C sequestration potential in Rice-N<sup>+</sup> in this study is supported by previous findings

(Pan et al., 2004; Ma et al., 2013; Kim et al., 2017) that N fertilization facilitates SOM storage in rice cropping systems by regulating C input and output. NPP was the main component of C input in our rice paddy (Fig. 2). Harvest removal and decomposed C loss ( $R_h$  and CH<sub>4</sub> emission) contributed to 55–71% and 29–45% of C output, respectively. Although CH<sub>4</sub> emission was high in rice paddy, it averagely contributed 4% of the total C output, indicating negligible impact on NECB. N fertilization significantly increased NPP and harvest removal, but had no impact on the decomposed C losses via  $R_h$  and CH<sub>4</sub> emission in rice paddy (Fig. 2). The increased NPP outweighed increased harvest C removal, resulting in considerably increased NECB in Rice-N<sup>+</sup> relative to in Rice-N<sup>0</sup>. These results are partially supported by Kim et al. (2017) who reported that optimal level of N fertilization increased NECB via increased NPP overriding increased harvest C removal and C losses by decomposition. The positive NECB of Rice-N<sup>+</sup> exhibited no significant interannual variation, indicating longer-term C sequestration for double-rice cropping system with conventional N fertilization.



**Fig. 4.** Changes in the global warming potential (GWP) of the combined net ecosystem carbon budget (NECB), CH<sub>4</sub> and N<sub>2</sub>O induced by land-use conversion from rice to vegetable cultivation (a) without (Rice-N<sup>0</sup> and Veg-N<sup>0</sup>) and (b) with (Rice-N<sup>+</sup> and Veg-N<sup>+</sup>) nitrogen fertilization, and by nitrogen fertilization in (c) rice paddy and (d) vegetable field across the four years. \* denotes statistically significant differences at  $p < 0.05$ . Values are means  $\pm$  standard errors of three replicates.

LUC has profound impacts on NECB and soil quality (Nishimura et al., 2008; Batllebayer et al., 2010; Kraus et al., 2016). However, considerable uncertainties remain regarding the direction and magnitude of NECB especially during the initial years upon LUC. Rice paddy conversion to vegetable cultivation led to substantial C losses (2.6 to 4.5 Mg C ha<sup>-1</sup> yr<sup>-1</sup>) because of decreased NPP in the converted vegetable field relative to rice paddy. For rice paddy, the stubbles and roots of rice were remained in the field when harvest. Whereas in the converted vegetable field, the entire vegetables were removed, returning minor residues to the soil. The harvest C removal was decreased in parallel with lower NPP in Veg relative to Rice. Rice paddy conversion to vegetable cultivation greatly increased R<sub>h</sub>, due to better soil aeration enhancing SOM mineralization (L. Wu et al., 2017). The contribution of R<sub>h</sub> to total C output significantly increased from 33 to 46% in Rice to 56–71% in Veg. As a result, the lower NECB in Veg relative to Rice was mainly attributed to much lower NPP as C input and higher R<sub>h</sub> as C output in Veg. N fertilization considerably increased NECB of Veg-N<sup>+</sup> by 2.0 Mg C ha<sup>-1</sup> yr<sup>-1</sup> as compared to Veg-N<sup>0</sup>, owing to more C inputs via increased NPP (M. Zhang et al., 2016). These results indicated that N fertilization alleviated the effect of rice paddy conversion to vegetable cultivation on C loss.

The NECB of vegetable field exhibited a considerable interannual variation with the highest C loss (4.5 to 6.1 Mg C ha<sup>-1</sup> yr<sup>-1</sup>) in the first year (Fig. 2). This phenomenon could be mainly attributed to the following two reasons. Firstly, the previous high level of SOM stored in rice paddy was particularly vulnerable to increased mineralization after initial drainage and tillage for upland cultivation (Nishimura et al., 2008; Palmer et al., 2014). This was demonstrated by decreasing R<sub>h</sub> from 6.3 Mg C ha<sup>-1</sup> yr<sup>-1</sup> in the first year to 3.4 Mg C ha<sup>-1</sup> yr<sup>-1</sup> in the fourth year in Veg-N<sup>+</sup> (Table 1). Secondly, the lower NPP of vegetables during the first year resulted in much lower C input than that in other years (Fig. 2). Above mentioned reasons in combination with the absence of organic manure addition contributed to lower annual NECB of our newly established vegetable field relative to older vegetable fields (Jia et al., 2012; M. Zhang et al., 2016). Consequently, vegetable

cultivation on rice paddy triggered substantial C losses especially in the first year (Fig. 5). This was demonstrated by previous study that conversion of rice paddy to upland crop cultivation induces rapid decomposition of organic matter from cropland soil (Nishimura et al., 2008). It is necessary to implement proper management practices through crop residues and manure amendment to compensate soil C losses induced by LUC (Qiu et al., 2009). Maintenance and accrual of soil C stocks is critical to fertility and sustainable crop production (Lal, 2004).

#### 4.2. Effects of LUC and N fertilization on CH<sub>4</sub> and N<sub>2</sub>O emissions

Flooded paddy soils generally emit higher CH<sub>4</sub> and lower N<sub>2</sub>O relative to upland soils (Linguist et al., 2012; Weller et al., 2016). However, most previous studies regarding evaluation of rice conversion to upland cultivation on GHG emissions mainly focused on long-term effects, with fewer studies considering emissions at the initial stage upon conversion (Kraus et al., 2016). This study is among the few to measure CH<sub>4</sub> and N<sub>2</sub>O fluxes during the initial years upon land-use conversion (Yuan et al., 2015; X. Wu et al., 2017).

In the present study, rice paddy emitted substantial CH<sub>4</sub> (ranging from 297.5 to 761.1 kg C ha<sup>-1</sup> yr<sup>-1</sup>), comparable to the results reported by Shang et al. (2011) in a double rice-cropping system in subtropical China. CH<sub>4</sub> emission from our rice paddy exhibited distinct interannual variation, and it was positively related to NPP across the four years (Fig. S4). These results suggested that NPP could, at least in part, explain the interannual variation in CH<sub>4</sub> emission from rice paddy. The increased plant growth and associated NPP facilitated additional C input for CH<sub>4</sub> production and the pathway for CH<sub>4</sub> emission (Shang et al., 2011; Linguist et al., 2012). N fertilization generally affects CH<sub>4</sub> emission by regulating CH<sub>4</sub> production and consumption processes (Bodelier and Laanbroek, 2004; Wu et al., 2009). Previous studies revealed that N fertilization decreases (X. Wu et al., 2017; Zhou et al., 2017) or increases (Shang et al., 2011) CH<sub>4</sub> emission in rice paddies. However, N fertilizer

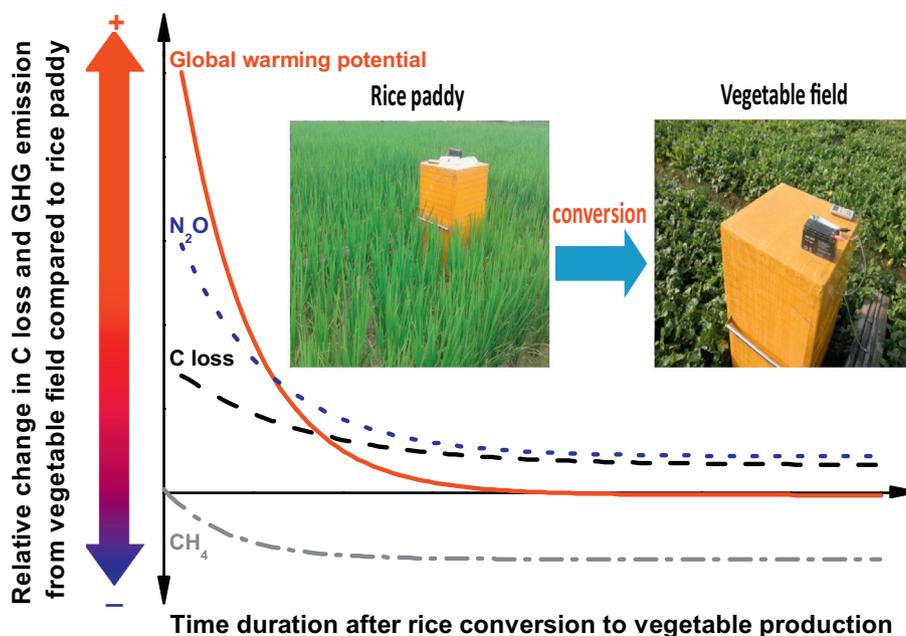


Fig. 5. Conceptual diagram of time duration effects of land-use conversion from rice to vegetable production on C loss, CH<sub>4</sub> and N<sub>2</sub>O emissions, and global warming potential.

application had no impact on CH<sub>4</sub> emission from our rice paddy. This result was supported by Dan et al. (2001) that N fertilization did not influence CH<sub>4</sub> fluxes from paddy soil due to the counterbalance between the stimulation of both CH<sub>4</sub> production and CH<sub>4</sub> oxidation after fertilization. N fertilization increases NPP which promotes CH<sub>4</sub> emissions (Ma et al., 2013). As an essential nutrient for methanotrophs, N fertilizer addition can stimulate the activities of methanotrophs, promoting CH<sub>4</sub> oxidation in soils (Bodelier and Laanbroek, 2004). Thus, no effect of N fertilization on CH<sub>4</sub> emission in our rice paddy was probably the result of the increased NPP being offset by the increase of methanotrophic activities.

LUC substantially reduced CH<sub>4</sub> emission from Veg by 97% compared to Rice over the study period, which is supported by the finding of Yuan et al. (2015). Distinct pulses of CH<sub>4</sub> fluxes were observed during the initial months after vegetable established on rice paddy, probably due to the physical release of entrapped CH<sub>4</sub> when the soil changed from saturated to unsaturated conditions during drainage. These CH<sub>4</sub> pulses from Veg contributed to significantly higher emission in the first year relative to any later years, suggesting that land-use legacy has strong effect on CH<sub>4</sub> emission and should be considered when estimating GHG emissions from agroecosystem (Fig. 5). The aerated soil condition inhibited CH<sub>4</sub> production while facilitating CH<sub>4</sub> oxidation, contributing to negligible CH<sub>4</sub> emission from our newly established vegetable field (Weller et al., 2016; Liu et al., 2017). Consistent with rice paddies, N fertilization did not affect CH<sub>4</sub> fluxes from the converted vegetable field, probably resulting from the increase of CH<sub>4</sub> oxidation from N fertilization being compensated by N stimulation on the substrates (e.g. root exudates) for CH<sub>4</sub> production. Our finding is supported by M. Zhang et al. (2016) who reported that chemical N fertilization had no impact on CH<sub>4</sub> emission from vegetable field in southeastern China. Further studies are required to clarify the underlying mechanisms of CH<sub>4</sub> emission from rice paddy and vegetable soils in response to N fertilization.

Rice paddy emitted negligible N<sub>2</sub>O and sometimes acted as a sink of atmospheric N<sub>2</sub>O (ranging from  $-0.2$  to  $1.8$  kg N ha<sup>-1</sup> yr<sup>-1</sup>) across the four-year study period, in line with previous studies (Shang et al., 2011; Linquist et al., 2012). This phenomenon was mainly caused by the submerged conditions limiting soil nitrification, thus inhibiting the supply of NO<sub>3</sub><sup>-</sup> for subsequent denitrification, and simultaneously favoring the complete reduction of N<sub>2</sub>O to N<sub>2</sub> (L. Wu et al., 2017). N fertilization had no impact on N<sub>2</sub>O emission from our rice paddy, which was in contrast to previous findings that N fertilization increased N<sub>2</sub>O emission

(Shang et al., 2011; X. Wu et al., 2017). This result indicated that soil moisture rather than N availability was likely the primary factor regulating N<sub>2</sub>O emission from rice paddy in the present study.

Rice paddy conversion to vegetable cultivation led to considerable N<sub>2</sub>O emission, primarily due to increased soil aeration and higher N fertilization rates providing substrates for N<sub>2</sub>O production (Weller et al., 2016; L. Wu et al., 2017). The extreme high N<sub>2</sub>O emission from our converted vegetable field in the first year was explained by the following reasons: Firstly, the drainage, tillage, and resulting increased soil aeration conditions promoted mineralization of SOM. This was demonstrated by the increased R<sub>n</sub> in Veg-N<sup>+</sup> by 99% relative to Rice-N<sup>+</sup> in the first year (Table 1, Fig. 1c). Given the tightly coupling of soil C and N cycles, SOM mineralization could serve as a considerable source of C as energy and mineral N as substrate suitable for N<sub>2</sub>O production (L. Wu et al., 2017). Secondly, the increased soil aeration upon LUC initiated nitrification and subsequent denitrification, resulting in tightly coupled nitrification-denitrification processes and thus higher N<sub>2</sub>O production (Penton et al., 2013). Thirdly, the annual N<sub>2</sub>O emission from our converted vegetable field was negatively linked to the variability in NPP (Fig. S4). This highest N<sub>2</sub>O emission from our vegetable field in the first year after conversion was partially explained by decreased N uptake by the lowest NPP, providing sufficient available N for microbial N<sub>2</sub>O production (Goldberg et al., 2010; Gelfand et al., 2011; L. Wu et al., 2017). Taken together, the complex interactions of soil aeration conditions, nutritional status, and additional labile C and N sources led to substantial N<sub>2</sub>O emission especially in the first year after rice conversion to vegetable cultivation (Fig. 5).

#### 4.3. Effects of LUC and N fertilization on the GWP

The impact of LUC on GWP was largely year-dependent. The GWP increased significantly in the first year upon rice paddy conversion to vegetable cultivation, primarily due to the increased C loss and N<sub>2</sub>O emission far outweighing the decreased CH<sub>4</sub> emission. In contrast, there was no distinct difference in the GWP between Rice and Veg in the following years, because the decreased CH<sub>4</sub> emission was counterbalanced by the increased C loss and N<sub>2</sub>O emission. These results indicated immediate and short-term increased GWP in response to LUC, partially confirming our hypothesis that rice paddy conversion to vegetable cultivation increased the GWP (Fig. 5). However, Weller et al.

(2016) stated that double rice cropping emitted larger GHG emissions compared to other cropping systems converted from rice paddy. The divergent results between these studies were largely attributed to substantial C losses upon conversion in the present study (Table 1). C losses contributed 24–88% of the GWP in our converted vegetable field, suggesting that sustainable NECB management in agroecosystem is crucial for mitigating GHG emissions especially in the initial years after land-use conversion. N fertilization enhanced the effects of LUC on the GWP in the first year via increased N<sub>2</sub>O emission, contributing to the highest GWP in Veg-N<sup>+</sup> (Table 1). Our study highlighted the importance of considering GHG emissions during the first year after LUC, and of developing effective mitigation options (Fig. 5). Manure/compost fertilizers, as an alternative N source, could partially substitute chemical N fertilizers, decreasing N<sub>2</sub>O emission, while maintaining crop yields and increasing C sequestration (Qiu et al., 2009). Applying soil amendments (such as biochar) and increasing crop residue incorporation rates are also promising options for SOC conservation and GHG mitigation (Roberts et al., 2010; Wei et al., 2013).

The present study evaluated the impact of LUC on direct GHG emissions. The indirect emissions arising from ammonia volatilization, C and N runoff and leaching, farm operation and agricultural inputs, may also contribute to the total GHG balance (Xia et al., 2016; M. Zhang et al., 2016). Direct and indirect GHG emission should be included when evaluating GHG balance in response to LUC in further studies.

## 5. Conclusions

Land-use conversion from rice to vegetable cultivation led to substantial C losses (2.6 to 4.5 Mg C ha<sup>-1</sup> yr<sup>-1</sup>), due to reduced C input by 44–52% and increased R<sub>n</sub> by 46–59% compared to Rice. The magnitude of C losses from the converted vegetable field was highest in the first year, and showed a decreasing trend in the following years. N fertilization shifted rice paddy from a slight C source in Rice-N<sup>0</sup> to a significant C sink in Rice-N<sup>+</sup> and alleviated the impact of rice conversion to vegetable cultivation on C loss via increased C inputs from higher crop productivity.

Land-use conversion significantly increased the GWP from Veg by 116–395% relative to Rice in the first year, primarily due to the increased C loss and N<sub>2</sub>O emissions far outweighing the decreased CH<sub>4</sub> emission. In contrast, the GWP from Rice were similar to those from Veg in the following years after conversion, because of the decreased CH<sub>4</sub> emission fully offsetting the increased C loss and N<sub>2</sub>O emission. N fertilization and land-use conversion interactively increased the GWP in the first year via increased N<sub>2</sub>O emission, contributing to largest GHG emission in Veg-N<sup>+</sup>. We conclude that C balance and GHG emissions in the first year after conversion are the most important and therefore, should be considered when evaluating the environmental consequences of land-use conversion.

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