

1 Exponential relationship between N<sub>2</sub>O emission and fertilizer nitrogen  
2 input and mechanisms for improving fertilizer nitrogen efficiency under  
3 intensive plastic-shed vegetable production in China: a systematic  
4 analysis

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**Abstract:** Currently, China has approximately four million hectares of intensively cultivated plastic-shed vegetable production, i.e., with excessive nitrogen (N) fertilization and high irrigation. Plastic-shed vegetable production has helped meet the rapidly increasing consumer demand for fresh vegetables while improving heat, light, and land utilization efficiencies, resulting in very high vegetable yield. We collected all studies from the 1980s to 2020 on N<sub>2</sub>O emissions and N fertilization associated with plastic-shed vegetable production at 40 field sites in China. Fertilizer N utilization efficiencies and N<sub>2</sub>O emissions that were affected by fertilizer N rate and type, irrigation, growth duration, nitrification inhibitors, and soil properties were systematically examined. The results revealed that fertilizer N efficiencies in plastic-shed vegetable production significantly decreased with increasing N fertilization rate. The average N recovery efficiency (RE<sub>N</sub>) and apparent N use efficiency (ANUE) were 6.8% and 33%, respectively; much lower than those of cereal crop production in the same region. In fruit and leafy vegetable production, N<sub>2</sub>O emissions exhibited an exponential and linear relationship with the fertilizer N rate, respectively, and the average contributions of fertilizer N to N<sub>2</sub>O emissions were 68% and 70%, respectively. Compared with synthetic N fertilizer or manure alone, combination of synthetic fertilizer with animal manure significantly increased the N<sub>2</sub>O emissions and emission factors (EFs) at high N fertilization rates (>800 kg N ha<sup>-1</sup> season<sup>-1</sup>), but there were no significant differences among fertilizer types at N rate <800 kg N ha<sup>-1</sup> season<sup>-1</sup>. Nitrification inhibitors reduced N<sub>2</sub>O emissions by 24.0% (95% confidence intervals [CI]: 19.2%–28.9%), and water-saving irrigation is the other effective measure to reduce emissions. Plastic-shed soils with neutral pH, high organic carbon content (> 30 g kg<sup>-1</sup>), growth period of >100 d, and higher irrigation increased the responses of N<sub>2</sub>O emission to N fertilization. As plastic-shed soils are continuously and intensively farmed, soil properties will be negatively

affected, and should be considered together with fertilization and irrigation to maintain high vegetable yield and low N<sub>2</sub>O emissions. Our study highlighted that the exponential relationship was more appropriate to predict the N<sub>2</sub>O emissions in plastic-shed vegetable production, and our findings help to optimize fertilizer N input with consideration of crop yield and greenhouse gas emission.

**Keywords:** fertilizer nitrogen efficiency; exponential relationship; nitrification inhibitor; animal manure; pH

## 1. Introduction

Driven by rapid economic development and increasing demand for fresh vegetable products in China, plastic-shed vegetable production was initiated in the 1990s, and has since been developed at a large scale (CSB, 2019). In 2016, the plastic-shed vegetable production planting acreage in China reached 3.92 million hectares, which is approximately 20% of the total vegetable acreage in the country (CSB, 2017; Li et al., 2018a). This enabled the average annual supply of fresh vegetables to reach 378 kg per capita, which is much greater than the global average of 143 kg per capita (FAO, 2020).

Plastic-shed vegetable production is characterized by an extremely high input of irrigation water and fertilizers (Yao et al., 2019). Typical irrigation rates in plastic-shed vegetable production range from 620 to 1180 mm per vegetable season (Lv et al., 2019; Yao et al., 2019), with an average of 50–120 mm per irrigation event (He et al., 2009; Lv et al., 2019). This is approximately 2–7 times higher than irrigation rates for cereal crops in the same region of China (Xin and Tao, 2020), where most solar plastic-shed vegetable production is located. Similarly, annual fertilizer nitrogen (N) input in typical plastic-shed vegetable fields (two crop seasons per year) reached 3338 kg ha<sup>-1</sup> in

Shouguang County, China, approximately 6–14 times that in local wheat-maize rotations (Liu et al., 2014). Compared with cereals, vegetables have lower nutrient uptake because of their shallow root system and low quantity of harvested dry matter (Min et al., 2012; Wang et al., 2018). Min et al. (2012) and Liang et al. (2015) found that N use efficiency (NUE) was only 18% and 9.5% in a plastic-shed tomato system, respectively, which was significantly lower than the NUE of the main grain systems (29%–45%) in Northern China (Chuan et al., 2010; Li et al., 2015a; Yu and Shi, 2015). Although it is not always appropriate to compare vegetable and cereal crops, these findings indicate there is room to improve the NUE for vegetable production to an acceptable level (e.g., 40%; Zhu and Chen, 2002).

With the intensification and popularization of plastic-shed vegetable production in China, the negative environmental impacts of conventional planting methods have gradually become prominent. Surplus nutrients either remain in the soil (Bai et al., 2020), or are lost to the environment via various pathways, including  $\text{N}_2\text{O}$ ,  $\text{NO}$ ,  $\text{N}_2$  (Yao et al., 2019),  $\text{NH}_3$  (Zhang et al., 2019a), and N leaching (Lv et al., 2019).  $\text{N}_2\text{O}$  emissions from plastic-shed fields account for approximately 25% of total farmland  $\text{N}_2\text{O}$  emissions in China (Yao et al., 2019). Plastic-sheds are characterized by a close inner environment with high soil temperature and moisture (Han et al., 2017), which significantly affects soil properties (Ding et al., 2019; Lv et al., 2020), promotes soil microbial activity, and increases  $\text{N}_2\text{O}$  emissions (Duan et al., 2019a; Ye et al., 2018).

Previous studies found that  $\text{N}_2\text{O}$  emissions for plastic-shed production increase with increasing fertilizer N rates (Chen et al., 2014; Cui et al., 2014; Wang et al., 2018), and their relationships vary among crops (Chen et al., 2014; Wang et al., 2018) and regions (Liyanage et al., 2020; Wang et al., 2018) because of variations in climate (Aliyu et al., 2018), soil (Gu et al., 2020), vegetable species

(Yang et al., 2019), and farming measures (Wu et al., 2020). Compared with leafy vegetables, fruit vegetables with a longer growth period require higher N levels and more irrigation, and therefore take up more N (Luo et al., 2015; Min et al., 2012; Yao et al., 2019). Fertilizer type significantly influences N<sub>2</sub>O emissions (Shcherbak et al., 2014; Zhou et al., 2017). Farmers commonly apply synthetic fertilizer in combination with composted animal manure in Chinese vegetable production (Wang et al., 2018), to meet the crop N demand (Tang et al., 2019), and simultaneously improve the soil organic matter (SOM) and ameliorate negative soil properties (Chen et al., 2020). Recent studies indicated that partial replacement of synthetic N with manure was optimal for mitigating N<sub>2</sub>O emissions from farmland (Chang et al., 2020; Tang et al., 2019; Wu et al., 2018a), either because of slow N release from manure (Ding et al., 2013; Zhou et al., 2019a) or stimulated denitrification (Meijide et al., 2007; Duan et al., 2019b). However, some studies have shown that a combination of synthetic N fertilizer and manure emitted more N<sub>2</sub>O at the same N application rate compared with synthetic fertilizer alone (Dong et al., 2007; Hao et al., 2012). This is because the combination of synthetic fertilizer with manure simultaneously inputs carbon and N, increasing the nitrification and denitrification rates and subsequently increasing N<sub>2</sub>O emissions (Aguilera et al., 2013). However, Charles et al. (2017) and Wang et al. (2018) found that a mixed application of synthetic fertilizer and manure did not influence N<sub>2</sub>O emissions and emission factors (EFs), compared with synthetic N fertilizer alone in plastic-shed soils.

In addition to fertilization, other measures such as the irrigation water quantity (Yao et al., 2019), nitrification inhibitors (Wu et al., 2018b; Zhao et al., 2017), and soil properties (Ding et al., 2019) have also been reported to significantly affect N<sub>2</sub>O emissions in plastic-shed vegetable production. However, because of the limited amount of published research on this topic, or because

a mixture of farming measures such as flood and drip irrigation are compared, there have been conflicting findings among meta-data studies (Gu et al., 2020; Wang et al., 2018; Wu et al., 2020; Yang et al., 2019). With the implementation of China's 13<sup>th</sup> National Five-Year Scientific Research Program, there have been many studies on plastic-shed vegetable systems in China since 2015. This paper describes a state-of-the-art meta-data study including the most recent publications on N<sub>2</sub>O emissions under plastic-shed vegetable production. The purposes of our study were to examine the utilization efficiencies of fertilizer N and the quantitative relationship with N<sub>2</sub>O emission, and to investigate the impacts of soil properties and farming measures such as nitrification inhibitors and irrigation on N<sub>2</sub>O emission under plastic-shed vegetable production. The findings will provide sound technologies with adequate scientific support to optimize plastic-shed vegetable production in China.

## 2 Materials and Methods

### 2.1 Data collection

We used ISI-Web of Science (Thomson Reuters, New York, NY, USA), the China Knowledge Resource Integrated database (CNKI), and Google Scholar (Google Inc., Mountain View, CA, USA) to conduct a survey for publications before 31 October 2020. Keywords including 'plastic-shed' or 'greenhouse,' 'vegetable,' 'N<sub>2</sub>O,' 'global warming potential (GWP),' 'greenhouse gas intensity (GHGI),' 'carbon footprint,' 'nitrogen,' 'N application rate,' 'fertilizer N rate', 'N rate,' and combinations of these were used in the publication search.

To reduce deviation, we implemented the following criteria to select studies for this systematic analysis: 1) Data must have been obtained via field monitoring in China, excluding those from pot

and soil incubation experiments; 2) Field experiments must have involved plastic-shed vegetables under conventional irrigation (flood irrigation), excluding open-field vegetables and leguminous crops; 3) Data concerning N<sub>2</sub>O emissions must have been measured by the closed chamber method; 4) The cumulative N<sub>2</sub>O emissions should be available for an entire season of vegetable crop; and 5) N sources must have been synthetic fertilizer or composted animal manure.

On the basis of these criteria, we collected 235 field measurements of N<sub>2</sub>O emissions from 40 experiments (Data S1). The information collected from each study were: experimental year, study location, vegetable species, N fertilization rate, cumulative N<sub>2</sub>O emissions over an entire vegetable season, yield, soil organic carbon (SOC) content, soil pH, soil total N content (TN), soil SOC/TN (C/N) ratio, irrigation water quantity, and growing period (day) during the vegetable season. We used GetData Graph Digitizer V2.25 (<http://getdata-graph-digitizer.com/>) to get data from graphical form.

## 2.2 Indicator calculation

In this study, four fertilizer N efficiency indicators were quantified: partial factor productivity of fertilizer N (PFP<sub>N</sub>, kg kg<sup>-1</sup>), agronomic efficiency of fertilizer N (AE<sub>N</sub>, kg kg<sup>-1</sup>), recovery efficiency of fertilizer N (RE<sub>N</sub>, %), and apparent N use efficiency (ANUE, %). These indicators were calculated as follows (Liu et al., 2019; Nie et al., 2019; Zhang et al., 2017):

$$PFP_N = Y/N \times 1000 \quad (1)$$

$$AE_N = (Y - Y_0)/N \times 1000 \quad (2)$$

$$RE_N = (U - U_0)/N \times 100\% \quad (3)$$

$$ANUE = N_E/N \times 100\% \quad (4)$$

where Y and Y<sub>0</sub> are the harvested fresh vegetable yield (t) of N fertilized and unfertilized treatments,

respectively;  $U$  and  $U_0$  are the total N uptake by aboveground vegetables ( $\text{kg N ha}^{-1}$ ) for N fertilized and unfertilized treatments, respectively;  $N_E$  is the N quantity of the economic crop part (fruit for fruit vegetables and aboveground stems and leaves for leafy vegetables) of N fertilization treatments; and  $N$  ( $\text{kg N ha}^{-1}$ ) is the fertilizer N rate of N fertilization treatments. All fertilizer N rates in this paper are per vegetable season.

The EF of  $\text{N}_2\text{O}$  was calculated as follows according to Wang et al. (2018):

$$\text{EF} = (E - E_0)/N \times 100\% \quad (5)$$

where  $E$  and  $E_0$  are cumulative  $\text{N}_2\text{O}$  emissions during the entire vegetable season ( $\text{kg N ha}^{-1} \text{ season}^{-1}$ ) for N fertilized and unfertilized treatments, respectively.

The contribution of fertilizer N to total  $\text{N}_2\text{O}$  emissions was calculated using the equation:

$$\text{N contribution} = (E - E_0)/E \times 100\% \quad (6)$$

We also calculated  $\text{N}_2\text{O}$  intensity using the following equation developed by Chen et al. (2014):

$$\text{N}_2\text{O intensity} = E/Y \times 100\% \quad (7)$$

As studies of the impacts of fertilizer formula, irrigation type, and crop residue incorporation on  $\text{N}_2\text{O}$  emissions under plastic-shed vegetable production were rare, we only analyzed the impacts of fertilizer N rate and type, nitrification inhibitors, irrigation amount, growth period, and soil properties (SOC, soil pH, soil C/N ratio) in this study. The entire dataset of vegetable crops was divided into two categories: fruit vegetables (including tomatoes, cucumbers, peppers, and eggplants) and leafy vegetables (including celery, cauliflower, coriander, spinach, baby bok choy, radish, lettuce, cabbage, and water spinach). We grouped the SOC content ( $\text{g kg}^{-1}$ ) into four levels ( $\leq 10$ , 10–20, 20–30,  $> 30$ ) and the soil C/N ratio into three levels ( $< 6$ , 6–10,  $\geq 10$ ). The soils were categorized into acidic ( $\text{pH} < 6.5$ ), neutral ( $\text{pH} = 6.5\text{--}7.5$ ), and alkaline ( $\text{pH} > 7.5$ ). Irrigation water

quantity (mm) was divided into five levels ( $\leq 200$ , 200–400, 400–600, 600–800,  $> 800$  mm), and growth period (d) was divided into three levels ( $< 100$ , 100–150,  $\geq 150$  d), respectively.

### 2.3 Relationship between N utilization efficiencies, N<sub>2</sub>O emissions, and fertilizer N rate

Regression analysis was adopted to recognize the best-fitting model between N efficiencies, N<sub>2</sub>O emissions, and fertilizer N rate, depending on the adjusted R<sup>2</sup> (adj. R<sup>2</sup>) value of the regression equation. An F-test was used to examine the regression relationship, with a  $P < 0.05$  considered to indicate significance. Two-rate inflection point model method (Zhang et al., 2014a) was adopted to recognize the inflection point of the nonlinear regression equation except quadratic equation for seeking the optimal fertilizer N rate. The common solution ( $X_0$ ,  $Y_0$ ) was obtained by simultaneously derived functions of linear and nonlinear equations between N<sub>2</sub>O emission, EF, and fertilizer N rate, where  $X_0$  was the inflection point of the nonlinear regression equation. Multiple linear regression analysis was performed to examine the contribution of fertilizer N input, irrigation water quantity, and soil properties on N<sub>2</sub>O emissions, and to identify the principal factors controlling N<sub>2</sub>O emission. We implemented one-way analyses of variance (ANOVA) to evaluate experimental effects such as total N<sub>2</sub>O emissions as affected by type and rate of fertilizer N and by the nitrification inhibitor. Means and corrected biases (i.e., 95% confidence intervals [CI]) for each category generated using boot strapping (10,000 iterations) were calculated with SPSS version 20.0 (SPSS Inc., Chicago, IL, USA). We used the least significant difference (LSD) at a 5% level of probability to compare means. All analyses were conducted using SPSS version 20.0 (SPSS Inc., Chicago, IL, USA).

### 2.4 Meta-analysis

We performed a pairwise meta-analysis of the random effects model using MetaWin 2.1

software (Han et al., 2018) to evaluate the impacts of soil properties (SOC, soil pH, soil C/N ratio) and farming practices (irrigation water quantity and growing days) on cumulative N<sub>2</sub>O emissions.

A weighted method was conducted according to Xu et al. (2017):

$$W_i = (n \times f) / \text{obs} \quad (8)$$

where  $W_i$  is the weight for observations at the  $i^{\text{th}}$  site;  $n$  and  $f$  are the number of replicates and N<sub>2</sub>O samplings per month in the field experiment, respectively; and  $\text{obs}$  is the total number of observations at the  $i^{\text{th}}$  site. To prevent extreme weights caused by high sampling frequencies, a maximum value  $f = 5$  was assigned when N<sub>2</sub>O fluxes were measured more than once per week.

The natural logarithm of the response ratio ( $\ln R$ ) was calculated as an indicator of the effect size:

$$\ln R = \ln(X_t / X_c) \quad (9)$$

where  $X_t$  and  $X_c$  represent cumulative N<sub>2</sub>O emissions for the experimental (N fertilized) and control (unfertilized) treatments, respectively.

We used  $\ln R$  values from each study to calculate the mean of the response ratios ( $\bar{R}$ ) using equation (10):

$$\bar{R} = \exp \left( \sum (\ln R_i \times W_i) / \sum W_i \right) \quad (10)$$

Mean effect sizes and bias-corrected 95% CIs were generated by a bootstrapping procedure (4999 iterations) for each categorical variable. The relative change, such as relative N<sub>2</sub>O increment, was reported as  $[\bar{R} - 1] \times 100$  to illustrate the influential effects compared with the control. We considered the mean effect sizes to be significantly different when their 95% CIs did not overlap, and to be significantly different from the control if their 95% CIs did not overlap with zero. The mean effect sizes of the categories were accepted to be significantly different between the levels of

the factors when the  $P$  values of the between-group heterogeneity ( $Q_b$ ) were less than the 0.05 level ( $P < 0.05$ ).

## 3 Results

### 3.1 Fertilizer N efficiencies

Fertilizer N efficiencies decreased with increasing fertilizer N rates for all types of vegetables. There was a significant and negative relationship between  $PFP_N$  and fertilizer N rate for the overall data (fruit and leafy vegetables), fruit vegetables, and leafy vegetables under plastic-shed production (Fig. 1a–c). Except for fruit vegetables, there was a significant and negative logarithmic relationship between  $AE_N$  and fertilizer N rate ( $P < 0.001$ ) for the overall and leafy vegetable datasets (Fig. 1d–f). The  $RE_N$  values ranged from  $-14.9\%$  to  $38.4\%$  and the ANUE values ranged from  $4.3\%$  to  $318\%$ , with high variations among studies (Fig. 1g–i).  $RE_N$  significantly decreased with increasing N rate for the overall, fruit vegetable, and leafy vegetable datasets, from  $11\%$  at fertilizer N rate of  $200 \text{ kg N ha}^{-1}$  to  $4.8\%$  at  $1000 \text{ kg N ha}^{-1}$ . The negative relationships between ANUE and fertilizer N rate were also significant ( $P < 0.001$ ) for the overall, fruit, and leafy vegetable datasets. There were inflection points in the fertilizer N rates at which N efficiencies decreased to a stable level, such as at  $\sim 500 \text{ kg N ha}^{-1}$  for  $PFP_N$  and ANUE for fruit vegetable dataset. However, for leafy vegetables, the fertilizer N efficiencies continuously decreased as the fertilizer N rates increased, although the decrease in N efficiencies was much higher at a low fertilizer N rate.

### 3.2 $N_2O$ emissions from plastic-shed vegetable fields

The mean total  $N_2O$  emission per vegetable season for the overall data set was  $5.26$  (CI:  $4.45$ –

6.17) kg N ha<sup>-1</sup>, which was similar to that of fruit vegetables (mean: 6.45; CI: 5.29–7.76 kg N ha<sup>-1</sup>) and significantly higher than that of leafy vegetables (mean: 2.83; CI: 2.37–3.34 kg N ha<sup>-1</sup>) (Fig. S1a). Correspondingly, the mean net N<sub>2</sub>O emissions were 4.48 (CI: 3.64–5.45), 5.44 (CI: 4.28–6.76), and 2.28 (CI: 1.72–2.87) kg N ha<sup>-1</sup> for the overall, fruit, and leafy vegetable datasets, respectively (Fig. S1b). The mean background (no fertilizer N input) N<sub>2</sub>O emissions in the overall, fruit, and leafy vegetable systems were 1.55 (CI: 1.11–2.07), 1.92 (CI: 1.33–2.68), and 0.75 (CI: 0.20–1.02) kg N ha<sup>-1</sup>, respectively (Fig. S1c). The total, net, and background N<sub>2</sub>O emissions from plastic-shed fruit vegetable and leafy vegetable production differed significantly (Fig. S1a–c).

The average fertilizer N rate for the overall, fruit, and leafy vegetable datasets was 583 (CI: 528–638), 667 (CI: 599–737), and 411 (CI: 348–478) kg N ha<sup>-1</sup>, respectively (Fig. S1d). Compared with the leafy vegetable, the fruit vegetable dataset emitted 1.3–1.6 fold greater total, net, and background N<sub>2</sub>O while receiving only 60% more fertilizer N (Fig. S1a–d). The mean EF values were 0.63% (CI: 0.55%–0.73%), 0.67% (CI: 0.56%–0.80%), and 0.55% (CI: 0.44%–0.67%) for the overall, fruit, and leafy vegetable datasets, respectively (Fig. S1e), and no significant differences in EF values were observed among the three datasets. Similarly, the three datasets had comparable fertilizer N contributions to total N<sub>2</sub>O emissions, with averages of 69%, 68%, and 70% being observed for the overall, fruit vegetable, and leafy vegetable datasets, respectively (Fig. S1f).

The relationships between total N<sub>2</sub>O emissions, net N<sub>2</sub>O emissions, and fertilizer N rate were significantly positive based on an exponential model of the overall and fruit vegetable datasets (Fig. 2a, b, d, and e), but the relationship was at a linear model of the leafy vegetable dataset (Fig. 2c, f). The relationship between EF and fertilizer N rate was significantly positive in an exponential model of the overall and fruit vegetable datasets, while this relationship could be described by a quadratic

equation for the leafy vegetable set (Fig. 2g–i). The inflection points of nonlinear responses between EF and fertilizer N rate in overall, fruit and leafy vegetable datasets were 574, 666 and 517 kg N ha<sup>-1</sup>, respectively (Table S1).

As the fertilizer N rate increased, the contribution of fertilizer N to total N<sub>2</sub>O emissions increased for the overall, fruit, and leafy vegetable datasets, and these relationships could be described by a positive logarithmic equation for the overall and fruit vegetable datasets (Fig. S2a, b), and a linear relationship for the leafy vegetable dataset (Fig. S2c). The fertilizer N contribution exceeded 80% at a fertilizer N rate of >1030 for the fruit dataset and 636 kg N ha<sup>-1</sup> for the leafy vegetable datasets. The N<sub>2</sub>O emission intensity also increased exponentially as the N rate increased for the overall and fruit vegetable datasets and it increased linearly for the leafy vegetable datasets (Fig. S2d–f).

### 3.3 Effects of fertilizer N type and nitrification inhibitor on N<sub>2</sub>O emissions

The effects of fertilizer N type on total and net N<sub>2</sub>O emissions varied under different fertilizer gradients (Fig. 3a, b). No significant differences were found in N<sub>2</sub>O emissions among the three fertilization types at low N rate (i.e., synthetic fertilizer, animal manure fertilizer, and synthetic + animal manure fertilizer (<800 kg N ha<sup>-1</sup>); however, at >800 kg N ha<sup>-1</sup>, the combination of synthetic N fertilizer with animal manure resulted in much larger N<sub>2</sub>O emissions compared with synthetic fertilizer or animal manure alone (Fig. 3a, b). Similar to total and net N<sub>2</sub>O emissions, the combination of synthetic N fertilizer with manure significantly increased EF at >800 kg N ha<sup>-1</sup> (Fig. S3). Compared with conventional fertilization alone, nitrification inhibitor addition significantly reduced N<sub>2</sub>O emissions. As the N rate increased, the N<sub>2</sub>O reduction rate increased linearly (Fig. 4a)

within the range of  $-1.89\%$  to  $54.5\%$  (mean:  $24.0\%$ , CI:  $19.2\%$ – $28.9\%$ ). Additionally, the reduction in emissions ranged from  $-0.01$  to  $4.16 \text{ kg N ha}^{-1}$ , with an average reduction of  $1.10 \text{ kg N ha}^{-1}$ . No significant differences were found in the  $\text{N}_2\text{O}$  emission reduction ratios between types and contents of nitrification inhibitors (Fig. 4b).

### 3.4 Impacts of soil properties, irrigation, and growth period on $\text{N}_2\text{O}$ emissions

Overall, fertilizer N significantly increased  $\text{N}_2\text{O}$  emission by  $308\%$  (CI:  $262\%$ – $362\%$ ) relative to no fertilizer N input in plastic-shed vegetable production (Fig. 5a). As SOC increased, both the net and background  $\text{N}_2\text{O}$  emissions increased; however, the background  $\text{N}_2\text{O}$  emissions increased at a higher rate (Fig. S4a, f) and the relative  $\text{N}_2\text{O}$  emission increments (%) were similar among the four SOC content levels, i.e.,  $373\%$  (CI:  $267\%$ – $508\%$ ),  $269\%$  (CI:  $212\%$ – $342\%$ ),  $384\%$  (CI:  $260\%$ – $551\%$ ), and  $269\%$  (CI:  $185\%$ – $425\%$ ) in soils with SOC content of  $\leq 10$ ,  $10\text{--}20$ ,  $20\text{--}30$ ,  $>30 \text{ g kg}^{-1}$ , respectively (Fig. 5b). In the neutral ( $6.5\text{--}7.5$ ) and alkaline ( $\geq 7.5$ ) conditions, both absolute (in  $\text{kg N N}_2\text{O ha}^{-1}$ , Fig. S4b) and relative  $\text{N}_2\text{O}$  emission increment (% , Fig. 5c) were significantly higher than those at acidic pH ( $<6.5$ ), whereas no significant differences were found in background  $\text{N}_2\text{O}$  emissions ( $\text{kg N N}_2\text{O ha}^{-1}$ , Fig. S4g).

The greatest relative  $\text{N}_2\text{O}$  emission increment (mean:  $525\%$ ; CI:  $380\%$ – $664\%$ ) was observed in soils with a low C/N ratio ( $<6$ ), compared with a soil C/N ratio of  $6\text{--}10$  (mean:  $220\%$ ; CI:  $175\%$ – $271\%$ ) and  $\geq 10$  (mean:  $380\%$ ; CI:  $267\%$ – $528\%$ ) (Fig. 5d). However, the soil C/N ratio had no significant effect on net  $\text{N}_2\text{O}$  emissions (Fig. S4c). Soils with a medium C/N ratio ( $6\text{--}10$ ) had higher background  $\text{N}_2\text{O}$  emissions compared with soils with a C/N ratio of  $<6$  and  $\geq 10$  (Fig. S4h).

The relative  $\text{N}_2\text{O}$  emission increment increased from  $92\%$  (CI:  $47\%$ – $163\%$ ) under the lowest

irrigation quantity level (<200 mm) to 587% (CI: 317%–1112%) under the highest irrigation quantity level (>800 mm, Fig. 5e). Meanwhile, net N<sub>2</sub>O emissions significantly increased when irrigation water quantity exceeded 800 mm (Fig. S4d). Irrigation water quantity had no significant effect on background N<sub>2</sub>O emissions (Fig. S4i). A longer growth period (100–150 and ≥150 d) was correlated with higher net and background N<sub>2</sub>O emissions compared with <100 d (Fig. S4e, j).

A multiple linear regression model was established between the total N<sub>2</sub>O emission and N rate, irrigation water quantity (I), soil TN content, SOC content, soil pH, and soil C/N ratio:  $Y \text{ (kg N}_2\text{O-N ha}^{-1}\text{)} = -12.207 + 0.010 N \text{ (kg N ha}^{-1}\text{)} + 0.004 I \text{ (mm)} - 1.067 TN \text{ (g kg}^{-1}\text{)} + 0.461 SOC \text{ (g kg}^{-1}\text{)} - 0.446 pH + 0.393 \text{ C/N ratio}$  (adj.  $R^2 = 0.60$ ,  $P < 0.001$ ,  $n = 76$ ).

## 4. Discussion

### 4.1 Comparison of N efficiencies for plastic-shed vegetable production and cereal crops

In this study, we used four indicators, i.e., PFP<sub>N</sub>, AE<sub>N</sub>, RE<sub>N</sub>, and ANUE, to evaluate the N efficiencies of Chinese plastic-shed vegetable production. PFP<sub>N</sub> and AE<sub>N</sub> reflect the crop productivity per unit kg of fertilizer N applied, while RE<sub>N</sub> and ANUE are the recovered fertilizer N per unit kg of fertilizer N applied (Liu et al., 2019; Nie et al., 2019; Zhang et al., 2017).

The average N rate for Chinese plastic-shed fruit vegetable production per season was approximately three-fold the global rate; however, the average PFP<sub>N</sub>, AE<sub>N</sub>, RE<sub>N</sub>, and ANUE values of the fruit vegetable dataset in China were 180 kg kg<sup>-1</sup>, 22 kg kg<sup>-1</sup>, 7.5%, and 29%, respectively (Table 1). The PFP<sub>N</sub> for the fruit vegetable system in China was 47%–70% of tomato production in the United States (256–383 kg kg<sup>-1</sup>) (Ayankojó et al., 2018; Ozores-Hampton et al., 2015; Zotarelli et al., 2009a), and only 15%–22% of those in European countries (814–1198 kg kg<sup>-1</sup>) (Elia and

Conversa, 2012; Farneselli et al., 2015; Gallardo et al., 2020). The average ANUE of fruit vegetable production in China was 54%–94% of that of tomato production in the United States (31%–54%) (Ayankojo et al., 2018; Ozores-Hampton et al., 2015; Zotarelli et al., 2009b). The average  $AE_N$  and  $RE_N$  values for fruit vegetable production in China were only 9%–14% and approximately 10% of those of tomato production in Italy (Elia and Conversa, 2012; Farneselli et al., 2015). Similarly, the mean  $RE_N$  and ANUE values in Chinese plastic-shed vegetable production were 6.8% and 33%, respectively, which were only 14%–31% (Duan et al., 2019c; Jiang et al., 2018; Nie et al., 2019; Wang et al., 2019; Zhou et al., 2019b) and 50% (Chen et al., 2014; Zhang et al., 2017) of those for Chinese cereal crop production, respectively (Table 1).

Unlike  $PFP_N$  and ANUE, which quantify the overall fertilizer N efficiencies (including the fertilizer N retained in soil stock),  $AE_N$  and  $RE_N$  both define the real fertilizer N efficiencies by subtracting the soil N contribution from the CK treatment. The extremely low  $AE_N$  and  $RE_N$  values also indicate that soil N stock increased to a high level after long periods of intensive plastic-shed vegetable production. While this is beneficial for land productivity, high irrigation may intensify the risk of N leaching (Lv et al., 2019) and water pollution (Li et al., 2018b).

Fertilizer N efficiencies for the Chinese plastic-shed vegetable system were lower than those of cereal crops in China and vegetable production abroad. The primary reason was that there was an extremely high N rate for Chinese plastic-shed vegetable production that greatly exceeded the N demand of vegetables (Liang et al., 2015; Min et al., 2012). During the past four decades, farmers in China have been continuously pursuing high crop yield via high N input and irrigation as well in addition to other farming practices. The vegetable supply has been ensured in China, which has one of the highest vegetable consumption levels in the world (FAO, 2020); however, resources and

energy consumption were also high. Therefore, Chinese government has been implementing a series of actions (Su et al., 2020; Zhang et al., 2019a) to eliminate some negative impacts such as soil acidification (Sun et al., 2020), soil salinization (Bai et al., 2020), eutrophication (Bai et al., 2020), and N<sub>2</sub>O emissions (Yao et al., 2019) in vegetable production.

#### 4.2 N<sub>2</sub>O emission and the relationship with fertilizer N input

The adj. R<sup>2</sup> values of the regression equation between the total and net N<sub>2</sub>O emission and fertilizer N rate for the overall dataset were 0.55 and 0.52, respectively (Fig. 2a, d), indicating that fertilizer N input may explain 55% and 52% of the total and net N<sub>2</sub>O emission changes, respectively. During crop production, such as plastic-shed vegetable production, the fertilizer N rate is always a dominant factor affecting N<sub>2</sub>O emissions (Mosier and Kroeze, 2000). However, almost half of the N<sub>2</sub>O emission changes were not accountable, demonstrating that other environmental factors such as temperature, soil water-filled pore spaces (WFPS), and soil inorganic N content may have significant impacts (Yao et al., 2019). Our analysis also indicated that the average contribution of fertilizer N to total N<sub>2</sub>O emissions in plastic-shed vegetable production was 69%, confirming that fertilizer N was the dominant factor influencing N<sub>2</sub>O emissions (Wang et al., 2018; Yao et al., 2019).

The total and net N<sub>2</sub>O emissions both had exponential responses to fertilizer N rate in the overall and fruit vegetable models, unlike the linear response observed in the leafy vegetable dataset. The main reason for this discrepancy was that the mean fertilizer N rate in fruit vegetable production was 667 kg N ha<sup>-1</sup>, which was much higher than that for leafy vegetables (411 kg N ha<sup>-1</sup>), indicating that there was a higher N surplus for fruit vegetable production (Fig. S1d and Table S2). Additionally, this was because the fertilizer N rate for most leafy vegetable production in our study was < 800 kg N ha<sup>-1</sup> season<sup>-1</sup>, and if the fertilizer N increased further, there might be an exponential relationship

as the N surplus (N input-crop harvested N) was much higher. Excessive N input may have stimulated soil mineralization and accelerated the soil nitrification-denitrification rate, increasing N<sub>2</sub>O emissions greatly at the higher N rate (Wang et al., 2014; Yao et al., 2019). Greater increases in N<sub>2</sub>O emissions with small increases in N input will occur when N is improperly applied above the optimal rate (Kim et al., 2013; Wang et al., 2014). This highlighted that the exponential relationship was more appropriate to predict the responses of N<sub>2</sub>O emission to fertilizer N under plastic-shed vegetable production in China.

Responses of EF to fertilizer N rate differed between fruit and leafy vegetable production (Fig. 2h, i) because there was lower N inputs and surpluses in leafy vegetable production compared with fruit vegetable production (Table S2). At a fertilizer N rate of 500 kg N ha<sup>-1</sup>, leafy vegetables achieved peak crop yield and N uptake and N<sub>2</sub>O emissions declined (Li et al., 2015b; Zhang et al., 2014b), which was different from the results observed in response to continuously increasing the fertilizer N rate and EF during fruit vegetable production (Fig. 2b, e, h).

Rapid increase of total N<sub>2</sub>O emissions and net N<sub>2</sub>O emissions were observed at the fertilizer N rate of ≥ 800 kg N ha<sup>-1</sup> for fruit vegetables under plastic-shed production (Fig. 2b, e, h, Table S1). The inflection points of the fertilizer N were far higher than the N uptake by harvested fruit vegetables (318 kg N ha<sup>-1</sup>) and the estimated N surplus was 412 kg N ha<sup>-1</sup> (Table S2). These findings were quite different from the results in previous studies of cereal crop production (Chen et al., 2014), in which N<sub>2</sub>O emissions increased rapidly as the N surplus exceeded zero. Decreased mineral N in the topsoil because of extremely high irrigation and a low manure N release rate (Ding et al., 2013; Wang et al., 2018; Yao et al., 2019) reduced N<sub>2</sub>O emissions, which explains why the inflection points of the fertilizer N rate increased further. Notably, both the overall and fruit vegetable datasets

showed similar relationships between the total N<sub>2</sub>O emissions, net N<sub>2</sub>O emissions, EF, and fertilizer N rate. This was because the ratio of the sample number of fruit vegetables to the overall vegetable dataset was 67% (Table S2).

The exponential response to the fertilizer N rate in the overall and fruit vegetable production observed in our study agreed with results reported by Wang et al. (2014) for maize fields, Chen et al. (2014) for grain crop production (rice, wheat, maize) (Table S3), and Zhang et al. (2016) for vegetable production, but they were different from the linear responses reported by Wang et al. (2018) for open-field and plastic-shed vegetable production and Yang et al. (2019) and Wu et al. (2020) for vegetable production (Table S3). These differences may have occurred for several reasons, namely: 1) the average fertilizer N rate used during open-field production was 265 kg N ha<sup>-1</sup>, which was far lower than the 583 kg N ha<sup>-1</sup> used in our study (Table S2); 2) the number of datasets used by Wang et al. (2018) (n = 53) and Yang et al. (2019) (n = 223, including open-field and plastic-shed vegetable production) and Wu et al. (2020) (n = 211, including open-field and plastic-shed vegetable production) were much lower than in our study (n = 235) for plastic-shed production; 3) Wang et al. (2018) underestimated the impacts of fertilizer N rate on N<sub>2</sub>O emissions for plastic-shed production because their analyses inappropriately included mixed multi-season with single-season vegetable production. Compared with single-season production, multi-season vegetable production had less N surplus and emitted less N<sub>2</sub>O under the same fertilizer N rate; and 4) Yang et al. (2019) and Wu et al. (2020) mixed open-field with plastic-shed vegetable production (Table. S2). Overall, the above analyses indicated that the exponential model is more appropriate for N<sub>2</sub>O predictions for fruit vegetable production than the linear model developed by Wang et al. (2018), Yang et al. (2019), and Wu et al. (2020) for plastic-shed vegetable production in China.

#### 4.3 N<sub>2</sub>O emission and mitigation potential for plastic-shed vegetable production in China

The conventional seasonal N rates in plastic-shed fruit and leafy vegetable production were 816 (Huang et al, 2017) and 501 (Data S1) kg N ha<sup>-1</sup>, respectively. The seasonal N<sub>2</sub>O emission rate in plastic-shed fruit and leafy vegetable production were 4.78 and 2.71 kg N ha<sup>-1</sup> (Table 2), respectively, based on the exponential modelling (Fig. 2a). The annual N<sub>2</sub>O emission rates under typical plastic-shed fruit vegetable production (two crop seasons per year) and leafy vegetable production (three crop seasons per year) in China were 9.56 and 8.13 kg N ha<sup>-1</sup>, respectively (Table 2). Extrapolation of these N<sub>2</sub>O emission rates to the planting acreage of 2.44 million ha (fruit vegetable) and 1.48 million ha (leafy vegetable) in China (Li et al., 2018a; MOA, 2015; SOHU, 2018) gave a total estimated N<sub>2</sub>O emission of 35.3 Gg N yr<sup>-1</sup>. Similarly, depending on annual mean N<sub>2</sub>O emissions (3.00 and 3.90 kg N ha<sup>-1</sup>) (Cui et al., 2014) and planting acreage (4.11 and 5.03 million ha) for rice-rice and wheat-maize production, respectively (Jiang et al., 2019; Zhang et al., 2019b), the total N<sub>2</sub>O emissions were estimated to be 12.3 Gg N yr<sup>-1</sup> for rice-rice in Southern China and 19.6 Gg N yr<sup>-1</sup> for wheat-maize rotation in the Northern China Plain. With the 3.92 million ha of planting acreage in China, the plastic-shed vegetable production emitted 10.7% more of N<sub>2</sub>O than the 9.14 million ha of cereal crop (rice, wheat and maize) production (Table 2).

N<sub>2</sub>O emissions had a dynamic relationship with increases in fertilizer N rate, indicating that the fixed simulation of the IPCC model (1% linear emission) may not accurately predict N<sub>2</sub>O emissions and mitigation potentials for farmland soil. Therefore, we compared the reduction in N<sub>2</sub>O emissions between three grain crops (rice, wheat, maize) and two types of vegetable production (open-field, plastic-shed) per reduction of 100 kg N ha<sup>-1</sup> in China (Fig. 6) using the results determined in this and previous studies (Cui et al., 2014; IPCC, 2007; Wang et al., 2018). We found

that N<sub>2</sub>O emission mitigation gradually increased with increasing fertilizer N rate for plastic-shed vegetable production, which was different from the results reported for IPCC tier 1 and by Wang et al. (2018). The mitigation of N<sub>2</sub>O emissions was lower than that estimated by the IPCC model (i.e., 1 kg N ha<sup>-1</sup>, Fig. 6) when the fertilizer N rate was <876 kg N ha<sup>-1</sup>, while at >876 kg N ha<sup>-1</sup> the N<sub>2</sub>O emission mitigation was much greater than 1 kg N ha<sup>-1</sup>. As the N rate reached 1200 and 1500 kg N ha<sup>-1</sup> (excessive fertilization levels), N<sub>2</sub>O emissions may have been mitigated by 1.84 and 3.26 kg N ha<sup>-1</sup>, respectively, which were approximately 1.8 and 3.2 fold greater than the reductions reported for the IPCC model, 2.7 and 4.9 fold greater than the values reported by Wang et al. (2018) for plastic-shed vegetable production, and 1.5–2.6 fold and 2.7–4.7 fold greater than the values reported by Cui et al. (2014) for grain fields fertilized at 300 kg N ha<sup>-1</sup>, respectively (Fig. 6). The findings above highlight that both the IPCC model and the model developed by Wang et al. (2018) significantly underestimated N<sub>2</sub>O emissions and mitigation potential for Chinese plastic-shed vegetable production. Optimizing the excessive N input is essential as the planting acreage of plastic-shed vegetable production increases in China.

#### 4.4 Influences of fertilizer N type on N<sub>2</sub>O emissions

At high fertilizer N rates (>800 kg N ha<sup>-1</sup>), a combination of synthetic fertilizer and composted manure led to larger N<sub>2</sub>O emissions than synthetic fertilizer or manure alone under the same N rate for plastic-shed vegetable production, which was inconsistent with the results of many previous studies (Chang et al., 2020; Tang et al., 2019; Wu et al., 2018a). The most important factor driving N<sub>2</sub>O emissions under plastic-shed vegetable production was probably the availability of N and C to soil microorganisms where soil moisture was generally high (Hao et al., 2012; Lei et al., 2010). At a low N rate (<800 kg N ha<sup>-1</sup>), synthetic fertilizer alone did not provide a sufficient C supply (Ding

et al., 2013). Additionally, applying manure or a combination of synthetic fertilizer with manure did not supply sufficient N to soil microorganisms because of the slow release of nutrients from manure, resulting in competition for N between microorganisms and vegetable plants (Chen et al., 2014; Cui et al., 2014; Tang et al., 2019). Therefore, no differences were detected in N<sub>2</sub>O emissions among the three N fertilizer types (Fig. 3b). At high fertilizer N rates (>800 kg N ha<sup>-1</sup>), the combination of synthetic fertilizer with manure provided sufficient substrate N and labile organic carbon to increase microbial activity (Cai et al., 2013; Chang et al., 2020; Hao et al., 2012). This simultaneously overcame the C limitation with synthetic N fertilizer and the slow nutrient release with manure, resulting in a greater increase in N<sub>2</sub>O emissions and EFs. An interesting finding indicated that the rate of 800 kg N ha<sup>-1</sup> season<sup>-1</sup> both regulated N<sub>2</sub>O emissions (Fig. 2b, e and h) and allowed higher vegetable yield in fruit vegetable production (<0.8%) (Fig. S5b). The optimal fertilizer N rate in leafy vegetable production was approximately 500 kg N ha<sup>-1</sup> (Table S1) determined by the inflection point of the quadratic relationship between EF and N rate (Fig. 2i), as the relationships between total N<sub>2</sub>O emissions, net N<sub>2</sub>O emissions, yield, and N rate didn't exhibit inflection points (Fig. 2c, f and S5c).

#### 4.5 Effects of nitrification inhibitor, irrigation, growth period, and soil properties on N<sub>2</sub>O emissions

Nitrification inhibitors have been confirmed to inhibit the transformation of ammonium N into nitrate N, reducing N<sub>2</sub>O generation (Hao et al., 2012; Zhao et al., 2017). In our study, the average mitigation of N<sub>2</sub>O by a nitrification inhibitor was 24.0%, and there were no significant differences between the types and doses of inhibitors as a result of the limited experimental data (Fig. 4b).

Soil moisture, as measured by WFPS, in plastic-shed vegetable production is significantly higher than that in open-field vegetable production (Table S4). The mean soil temperature ranged

from 17.2 °C to 19.8 °C in plastic-shed vegetable production (Table S4); this was beneficial to N<sub>2</sub>O generation, which was mostly from denitrification and oxidizer-denitrification (Castro Silva et al., 2008; Yao et al., 2019; Ye et al., 2018). Again, these findings agree with the results of our study (Fig. 5e and S4d) and highlight that reduced irrigation can mitigate N<sub>2</sub>O emissions from plastic-shed vegetable production. Fruit vegetable production usually has a longer growth period and higher inputs of fertilizer N and irrigation water compared with leafy vegetable production (Yao et al., 2019, Min et al., 2012), therefore significantly increasing N<sub>2</sub>O emissions (Fig. S4e).

Our analysis found that high SOC content (>30 g kg<sup>-1</sup>) significantly increased N<sub>2</sub>O emission for plastic-shed vegetable production (Fig. S4a). Low SOC content decreases microbial activity and also N<sub>2</sub>O emissions (Xia et al., 2020; Schnürer et al., 1985). SOC content is usually below a threshold (20 g kg<sup>-1</sup>, Fig. S4a, f) in plastic-shed vegetable production, where the farming measures tend to accelerate the mineralization of SOM (Fan et al., 2014). Low SOC (<20 g kg<sup>-1</sup>) limits N<sub>2</sub>O emissions, but also negatively affects vegetable yield and overall soil fertility (Fan et al., 2014; Lv et al., 2019). Compared with a high C/N ratio, low soil C/N limited N<sub>2</sub>O emissions (Fig. S4 h). With the application of large amounts of manure and synthetic N fertilizers in plastic-shed vegetable production (Fan et al., 2014), soil microbial activity is enhanced and the net N<sub>2</sub>O emission is also much higher (Fig. 3b). As the length of the plastic-shed farming period increased, soil pH gradually decreased (Lv et al., 2020; Zhang et al., 2020). Acidic soils tended to have decreased N<sub>2</sub>O emissions compared with neutral soil; however, crop yields and land productivity may also be negatively affected. Besides the improvement in soil fertility, manure fertilization may also balance the soil pH and should be a priority farming practice in plastic-shed vegetable production.

Multiple regression analysis indicates that N<sub>2</sub>O emissions are more sensitive to changes in soil

properties (TN, SOC, C/N ratio) because of their higher coefficients, although their variation ranges are much lower than fertilizer N ( $>1500 \text{ kg N ha}^{-1}$ ) and irrigation water ( $>1000 \text{ mm}$ ) inputs (Data S1). The SOC and C/N ratio are less important factors than fertilizer N input, as they have wider range of variation ( $5.4\text{--}31.1 \text{ g kg}^{-1}$  and  $3.6\text{--}25.4$ , respectively) than TN ( $0.71\text{--}3.20 \text{ g kg}^{-1}$ ) and pH ( $4.8\text{--}8.3$ ) (Data S1). In addition to directly affecting soil  $\text{N}_2\text{O}$  emissions, increased fertilizer N and irrigation water input are able to accelerate labile SOM (or SOC) decomposition (Lv et al., 2019; Wu et al., 2017) and reduce the soil C/N ratio (Fan et al., 2014), whereas decreased SOC and soil C/N ratio increased fertilizer N and the irrigation water input (Fan et al., 2014; Lv et al., 2019), reinforcing a vicious cycle that further decreases soil pH (Lv et al., 2020) and enhances  $\text{N}_2\text{O}$  emission in plastic-shed vegetable production.

## 5. Conclusions

Fertilizer N efficiencies in intensively managed plastic-shed vegetable production facilities were much lower than those observed for open-field vegetable and grain production, although the vegetable yields in the plastic-shed system were much higher. The average contribution of applied fertilizer N to  $\text{N}_2\text{O}$  emissions was 69% in plastic-shed vegetable production, and exceeded 80% at fertilizer N rates of  $>1030$  (fruit vegetable) and  $636 \text{ kg N ha}^{-1} \text{ season}^{-1}$  (leafy vegetable). The exponential model was more appropriate than the linear relationship reported by previous studies to predict the  $\text{N}_2\text{O}$  emissions with fertilizer N rate, especially for fruit vegetables. For leafy vegetable, as the fertilizer N for most collected studies was  $<800 \text{ kg N ha}^{-1} \text{ season}^{-1}$ , a linear relationship between  $\text{N}_2\text{O}$  emissions and fertilizer N was observed and it might change to the exponential relationship, if the fertilizer N increased to a higher rate. The fertilizer N rate of  $800 \text{ kg N ha}^{-1}$

season<sup>-1</sup> for fruit vegetable and 500 kg N ha<sup>-1</sup> season<sup>-1</sup> for leafy vegetable production may effectively mitigate N<sub>2</sub>O emissions and also ensure the high vegetable yield. Nitrification inhibitor and water-saving irrigation are also effective measures to mitigate N<sub>2</sub>O emissions. N<sub>2</sub>O emissions from soils with neutral pH, high organic carbon content (> 30 g kg<sup>-1</sup>), growth period ≥100 d, and higher irrigation responded more sensitively to fertilizer N application, while maintaining high vegetable yields. Farming practices need much to be improved to balance land productivity, N<sub>2</sub>O mitigation, and other ecological services in intensive plastic-shed vegetable production.

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804

**Figure captions:**

Fig. 1 Relationship between partial factor productivity of fertilizer N ( $PFP_N$ ), agronomic efficiency of fertilizer N ( $AE_N$ ), recovery efficiency of fertilizer N ( $RE_N$ ), apparent N use efficiency (ANUE), and fertilizer N rate in the overall (a, d, g, j), fruit (b, e, h, k), and leafy (c, f, i, l) plastic-shed vegetable datasets. The significance of each equation was determined using an F-test (\*\*\*, \*\*, and \* indicate  $P < 0.001$ ,  $< 0.01$ , and  $< 0.05$ , respectively). The number of data points is given in each equation.

Fig. 2 Relationship between total and net  $N_2O$  emissions, emission factor (EF), and fertilizer N rate in the overall (a, d, g), fruit (b, e, h), and leafy (c, f, i) plastic-shed vegetable datasets. The significance of each equation was determined using an F-test (\*\*\*, \*\*, and \* indicate  $P < 0.001$ ,  $< 0.01$ , and  $< 0.05$ , respectively). The number of data points is given in each equation.

Fig. 3 Total  $N_2O$  emissions as affected by rate and type of N fertilizer (a), and the relationship between net  $N_2O$  emissions and fertilizer N rate (b) in plastic-shed vegetable production system in China. Black solid and dotted lines indicate medians and means, respectively. Box boundaries indicate upper and lower quartiles, whisker caps indicate 95th and 5th percentiles, and circles indicate outliers. Means in each plot with different lowercase letters indicate statistical significance at  $P < 0.05$ . The number of data points is given below each box. The significance of each equation was determined using an F-test (\*\*\*, \*\*, and \* indicate  $P < 0.001$ ,  $< 0.01$ , and  $< 0.05$ , respectively). The number of data points is given in each equation.

Fig. 4 Relationship between  $N_2O$  emission reduction and fertilizer N rate (a) and  $N_2O$  emission reduction ratio as

affected by rate and type (DCD: dicyandiamide and CP: chlorinated pyridine) of nitrification inhibitor (b) in Chinese plastic-shed vegetable production. Black solid and dotted lines indicate medians and means, respectively. Box boundaries indicate upper and lower quartiles, whisker caps indicate 95th and 5th percentiles, and circles indicate outliers. Means in each plot with different lowercase letters indicate statistical significance at  $P < 0.05$ . The number of data points is given below each box. The significance of each equation was determined using an F-test (\*\*\*, \*\*, and \* indicate  $P < 0.001$ ,  $< 0.01$ , and  $< 0.05$ , respectively). The number of data points is given in each equation. “Unknown DCD” means the DCD content was not indicated in article.

Fig. 5 Responses of N<sub>2</sub>O emissions to fertilizer N compared with no fertilizer N input (a), categorized into soil organic carbon (SOC) (b), soil pH (c), soil C/N ratio (d), irrigation water quantity (e), and growing days (f). N<sub>2</sub>O emission responses are expressed as the relative increase (%) compared with no fertilizer N input with 95% CIs represented by the error bars. Figures in parentheses indicate number of observations. Between-group heterogeneity ( $Q_b$ ) and the probability ( $P$ ) were used to describe statistical differences in N<sub>2</sub>O emission responses between different levels of the categorized factors.

Fig. 6 Comparison of N<sub>2</sub>O emission reductions between three grain crops (rice, wheat, maize) and vegetable planting pattern (open-field, plastic-shed) in China per 100 kg N ha<sup>-1</sup> decrease.

Table 1 Comparison of N efficiencies between plastic-shed vegetable production and cereal crops in China and abroad

Crop	Region	Fertilizer N rate kg N ha <sup>-1</sup>	PFP <sub>N</sub> <sup>a</sup> kg kg <sup>-1</sup>	AE <sub>N</sub> <sup>b</sup> kg kg <sup>-1</sup>	RE <sub>N</sub> <sup>c</sup> %	ANUE <sup>d</sup> %	References
Vegetable							
Whole vegetable dataset	Plastic-shed, China	583 CI: 528-638	196 CI: 165-233	21 CI: 16-27	6.8 CI: 5.1-8.7	33 CI: 28-40	This study
	Fruit vegetable	667 CI: 599-737	180 CI: 156-209	22 CI: 16-27	7.5 CI: 5.6-9.5	29 CI: 25-34	
Leafy vegetable	Plastic-shed, China	411 CI: 348-478	235 CI: 145-346	20 CI: 8.4-36	5.0 CI: 1.7-9.0	45 CI: 28-68	This study
	Tomato	224	383			31	(Ayankajo et al., 2018)
Tomato	USA	291	340			41	(Ozores-Hampton et al., 2015)
Tomato	USA	242	256				(Zotarelli et al., 2009a)
Tomato	USA	242				52	(Zotarelli et al., 2009b)
Tomato	Italy	200	814	161	70		(Elia and Conversa, 2012)
Tomato	Italy	200	828	249	65		(Farneselli et al., 2015)
Cucumber	Spain	255	1198				(Gallardo et al., 2020)
Cereal crops							
Rice	China	209	41				(Chen et al., 2014)
Rice	China	150	44	10	22		(Nie et al., 2019)
Rice	China	155	59	18	49		(Wang et al., 2019)
Wheat	China	210	33			57	(Chen et al., 2014)
Wheat	China	240	33	17	40		(Duan et al., 2019c)
Wheat	China	223	37			74	(Zhang et al., 2017)
Maize	China	270	43			51	(Chen et al., 2014)
Maize	China	300	32	12	36		(Zhou et al., 2019b)
Maize	China	180	52	19	50	55	(Jiang et al., 2018)
Maize	China	223	41			59	(Zhang et al., 2017)
Wheat-Maize	China	446	39			67	(Zhang et al., 2017)

<sup>a</sup>: partial factor productivity of fertilizer N (PFP<sub>N</sub>)

<sup>b</sup>: agronomic efficiency of fertilizer N (AE<sub>N</sub>)

<sup>c</sup>: recovery efficiency of fertilizer N (RE<sub>N</sub>)

<sup>d</sup>: apparent N use efficiency (ANUE)

Table 2 Comparison of N<sub>2</sub>O emissions, fertilization and acreage between vegetable and cereal production in China

Crop system	Region	Fertilizer N	N <sub>2</sub> O	Planting	Total N <sub>2</sub> O	References
		rate	emission	acreage	emission	
		kg N ha <sup>-1</sup>	kg N ha <sup>-1</sup>	million ha	Gg N	
<b>Seasonal</b>						
PFV <sup>a</sup>	China	816	4.78			(Huang et al., 2017; this study)
PLV <sup>b</sup>	China	501	2.71			(Data S1; This study)
Rice	Yangtze River watershed	214	1.50			(Cui et al., 2014)
Wheat	North China Plain	284	1.90			(Cui et al., 2014)
Maize	North China Plain	229	2.00			(Cui et al., 2014)
<b>Annual</b>						
PFV-PFV	China		9.56	2.44	23.3	(SOHU, 2018; MOA, 2015)
PLV-PLV-PLV	China		8.13	1.48	12.0	(SOHU, 2018; Li et al., 2018a)
Total PV <sup>c</sup>	China			3.92	35.3	(Li et al., 2018a; MOA, 2015)
Rice-Rice	Southern China		3.00	4.11	12.3	(Jiang et al., 2019)
Wheat-Maize	North China Plain		3.90	5.03	19.6	(Zhang et al., 2019b)

<sup>a</sup>: PFV indicates plastic-shed fruit vegetable production. PFV-PFV indicates two seasons of plastic-shed fruit vegetable production per year.

<sup>b</sup>: PLV indicates plastic-shed leafy vegetable production. PLV-PLV-PLV indicates three seasons of plastic-shed leafy vegetable production per year.

<sup>c</sup>: Total PV indicates the whole plastic-shed vegetable production, including FPV-FPV and LPV-LPV-LPV.

Fig. 1

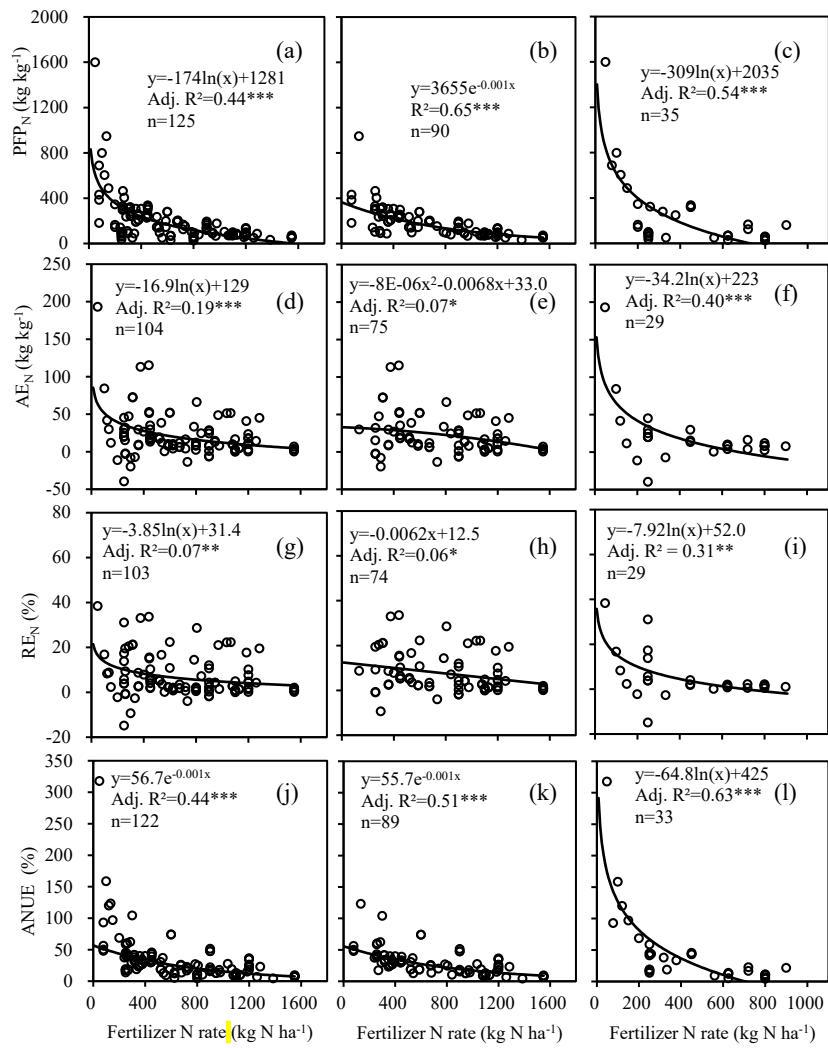


Fig. 2

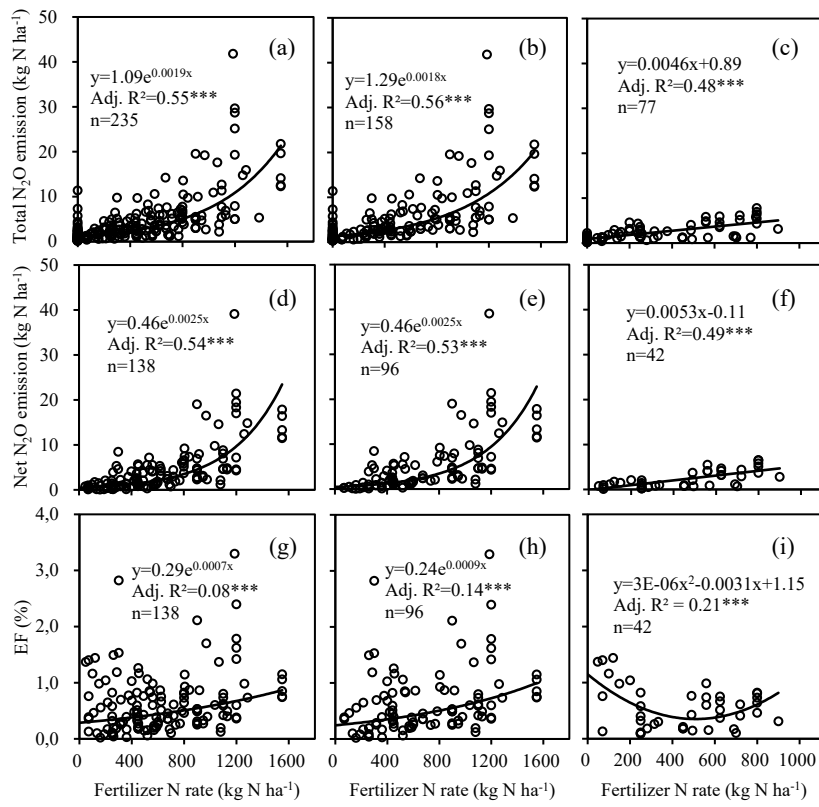


Fig. 3

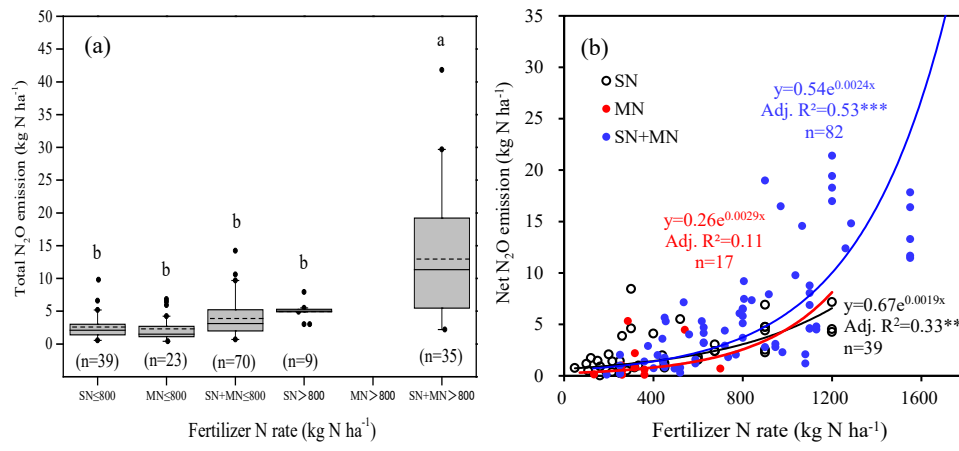


Fig. 4

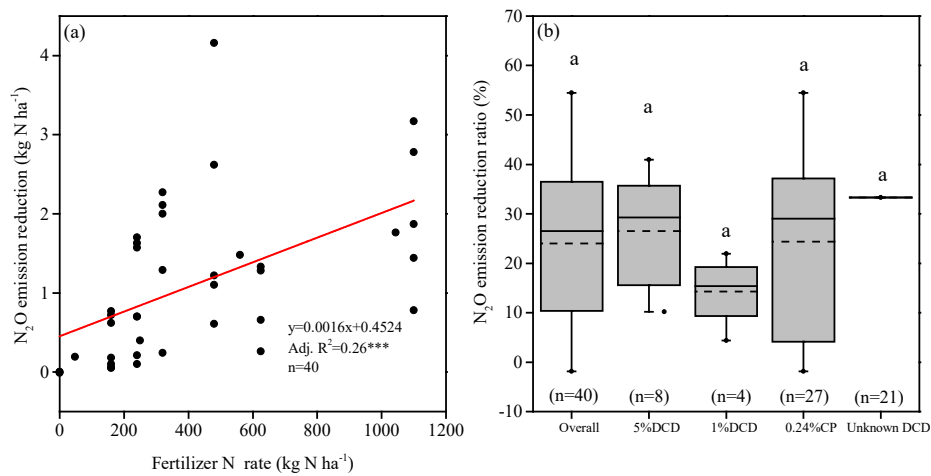


Fig. 5

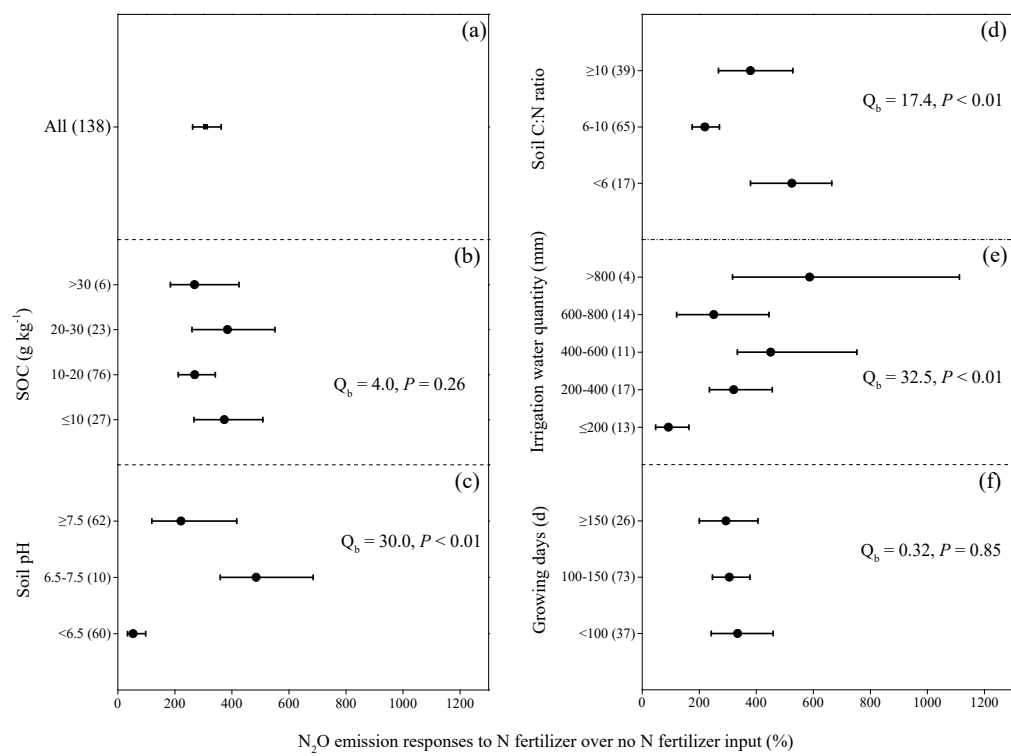
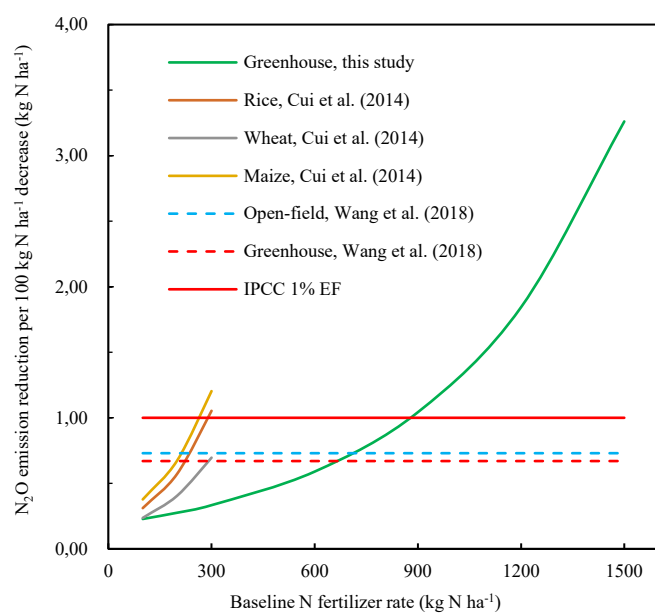


Fig. 6



## Captions of supplementary figures

Fig. S1. Total (a), Net (b), Background (c) N<sub>2</sub>O emissions, fertilizer N rate (d), emission factor (EF) (e), N contribution to N<sub>2</sub>O emissions (f) for the overall, fruit, and leafy vegetable datasets. Black solid and dotted lines indicate medians and means, respectively. Box boundaries indicate upper and lower quartiles, whisker caps indicate 95th and 5th percentiles, and circles indicate outliers. Means in each plot with different lowercase letters indicate statistical significance at  $P < 0.05$ . The number of data points is given below each box.

Fig. S2. Relationship between N contribution to total N<sub>2</sub>O emissions, N<sub>2</sub>O intensity, and fertilizer N rate in the overall (a, d), fruit (b, e), and leafy (c, f) plastic-shed vegetable datasets. The significance of each equation was determined using an F-test (\*\*\*, \*\*, and \* indicate  $P < 0.001$ ,  $< 0.01$ , and  $< 0.05$ , respectively). The number of data points is given in each equation.

Fig. S3. Emission factors (EFs) as affected by rate and type of N fertilizer. Black solid and dotted lines indicate medians and means, respectively. Box boundaries indicate upper and lower quartiles, whisker caps indicate 95th and 5th percentiles, and circles indicate outliers. Means in each plot with different lowercase letters indicate statistical significance at  $P < 0.05$ . The number of data points is given below each box.

Fig. S4. Net N<sub>2</sub>O emissions (a–e) and background N<sub>2</sub>O emissions (f–j) as affected by soil organic carbon (SOC), soil pH, soil C/N ratio, irrigation water quantity, and growing days in plastic-shed vegetable production system in China. Black solid and dotted lines indicate medians and means, respectively. Box boundaries indicate upper and lower quartiles, whisker caps indicate 95th and 5th percentiles, and circles indicate outliers. Means in each plot with different lowercase letters indicate statistical significance at  $P < 0.05$ . The number of data points is given below each box.

Fig. S5. Relationship between fresh vegetable yield and fertilizer N rate in the overall (a), fruit (b), and leafy (c)

plastic-shed vegetable datasets. The significance of each equation was determined using an F-test (\*\*\*, \*\*, and \*

indicate  $P < 0.001$ ,  $< 0.01$ , and  $< 0.05$ , respectively). The number of data points is given in each equation.

Supplementary Tables

Table S1 Optimal fertilizer N rate and inflection points of non-linear response between total N<sub>2</sub>O emissions, net

N<sub>2</sub>O emissions, emission factor (EF), yield, and fertilizer N rate in the overall, fruit, and leafy plastic-shed

vegetable datasets.

Indicator	Inflection point of fertilizer N rate (kg N ha <sup>-1</sup> )		
	Overall	Fruit vegetable	Leafy vegetable
Total N <sub>2</sub> O emission	763	767	ND <sup>a</sup>
Net N <sub>2</sub> O emission	859	883	ND
EF	574	666	517
Yield	1125	967	ND
Optimal N rate	≈ 800	≈ 800	≈ 500

<sup>a</sup>: ND = no data.

Table S2 Comparison of fertilizer N rate, N<sub>2</sub>O emissions, net N<sub>2</sub>O emissions, emission factor (EF), vegetable N

uptake, N surplus, and number of observations for vegetable production in China.

Crop	Region	Fertilizer N rate kg N ha <sup>-1</sup>	N <sub>2</sub> O emission kg N ha <sup>-1</sup>	Net N <sub>2</sub> O emission kg N ha <sup>-1</sup>	EF %	Vegetable N uptake kg N ha <sup>-1</sup>	N surplus <sup>a</sup> kg N ha <sup>-1</sup>	Number of observations (n)
<b>This study</b>								
Vegetable (Fruit + Leafy vegetables)	Plastic-shed, China	583	5.26	4.48	0.63	261	391	235
Fruit vegetable	Plastic-shed, China	667	6.45	5.44	0.67	318	412	158
Leafy vegetable	Plastic-shed, China	411	2.83	2.28	0.55	106	333	77
<b>Reference: Wang et al., 2018</b>								
Vegetable (Open-field + Plastic-shed)	China	428	3.91	2.61	0.69	ND <sup>b</sup>	ND	153
Vegetable	Open-field, China	265	2.62	1.83	0.71	ND	ND	100
Vegetable	Plastic-shed, China	719	6.22	4.55	0.65	ND	ND	53

<sup>a</sup>: Nitrogen surplus was defined as fertilizer N rate minus aboveground nitrogen uptake.

<sup>b</sup>: ND = no data

Table S3 Comparison of total N<sub>2</sub>O emission equations between this study and other studies.

Crop	Region	Y kg N ha <sup>-1</sup>	X kg N ha <sup>-1</sup>	Equation	Equation type	References
Vegetable	Plastic-shed, China	Total N <sub>2</sub> O emission	Fertilizer N rate	$Y = 1.09e^{0.0019X}$	Exponential	This study
Fruit vegetable	Plastic-shed, China	Total N <sub>2</sub> O emission	Fertilizer N rate	$Y = 1.29e^{0.0018X}$	Exponential	This study
Leafy vegetable	Plastic-shed, China	Total N <sub>2</sub> O emission	Fertilizer N	$Y = 0.0046 X + 0.89$	Linear	This study
Vegetable	China	Total N <sub>2</sub> O emission	Fertilizer N rate	$Y = 0.0073 X + 0.93$	Linear	(Wang et al., 2018)
Vegetable	Open-field, China	Total N <sub>2</sub> O emission	Fertilizer N rate	$Y = 0.0073 X + 0.75$	Linear	(Wang et al., 2018)
Vegetable	Plastic-shed, China	Total N <sub>2</sub> O emission	Fertilizer N rate	$Y = 0.0067 X + 1.56$	Linear	(Wang et al., 2018)
Vegetable	China+Other 6 countries	Total N <sub>2</sub> O emission	Fertilizer N rate	$Y = 0.015 X - 0.270$	Linear	(Yang et al., 2019)
Vegetable	China	Total N <sub>2</sub> O emission	Fertilizer N rate	$Y = 0.08 X + 1.56$	Linear	(Wu et al., 2020)
Vegetable	China	Total N <sub>2</sub> O emission	Fertilizer N rate	$Y = 18.1e^{0.0007X}$	Exponential	(Zhang et al., 2016a)
Rice	China	Total N <sub>2</sub> O emission	N surplus	$Y = 0.74e^{0.011X}$	Exponential	(Chen et al., 2014)
Wheat	China	Total N <sub>2</sub> O emission	N surplus	$Y = 0.54e^{0.0063X}$	Exponential	(Chen et al., 2014)
Maize	China	Total N <sub>2</sub> O emission	N surplus	$Y = 1.13e^{0.0071X}$	Exponential	(Chen et al., 2014)
Maize	China	Total N <sub>2</sub> O emission	Fertilizer N rate	$Y = 0.51e^{0.0051X}$	Exponential	(Wang et al., 2014)
Maize	China	Total N <sub>2</sub> O emission	N surplus	$Y = 1.18e^{0.0048X}$	Exponential	(Wang et al., 2014)
Rice	China	Total N <sub>2</sub> O emission	Fertilizer N rate	$Y = 0.37e^{0.0061X}$	Exponential	(Cui et al., 2014)
Wheat	China	Total N <sub>2</sub> O emission	Fertilizer N rate	$Y = 0.33e^{0.0054X}$	Exponential	(Cui et al., 2014)
Maize	China	Total N <sub>2</sub> O emission	Fertilizer N rate	$Y = 0.48e^{0.0058X}$	Exponential	(Cui et al., 2014)

Table S4 Comparison of soil temperature and water-filled pore space (WFPS) in topsoil between plastic-shed and open-field vegetable production in China.

Crop	Mean soil temperature °C	Mean WFPS %	References
<b>Plastic-shed</b>			
Cucumber-Cucumber	19.8	67	(Yao et al., 2019)
Green soybean-Pepper-Broccoli	17.2	77	(Yao et al., 2015)
Red pepper-Chrysanthemum	19.7	82	(Liu et al., 2013)
Tomato-Chinese cabbage-Green soybean	18.7		(Zhang et al., 2016b)
<b>Open-field</b>			
Amaranth-Turnip-Garlic	15.1	61	(Yao et al., 2015)
Chinese cabbage-celery-celery cabbage	17.9	71	(Liu et al., 2013)
Green vegetables-Lettuce-Chinese cabbage-Chinese cabbage-Green vegetables- Green vegetables	16.9	56	(Deng et al., 2012)

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Fig. S1

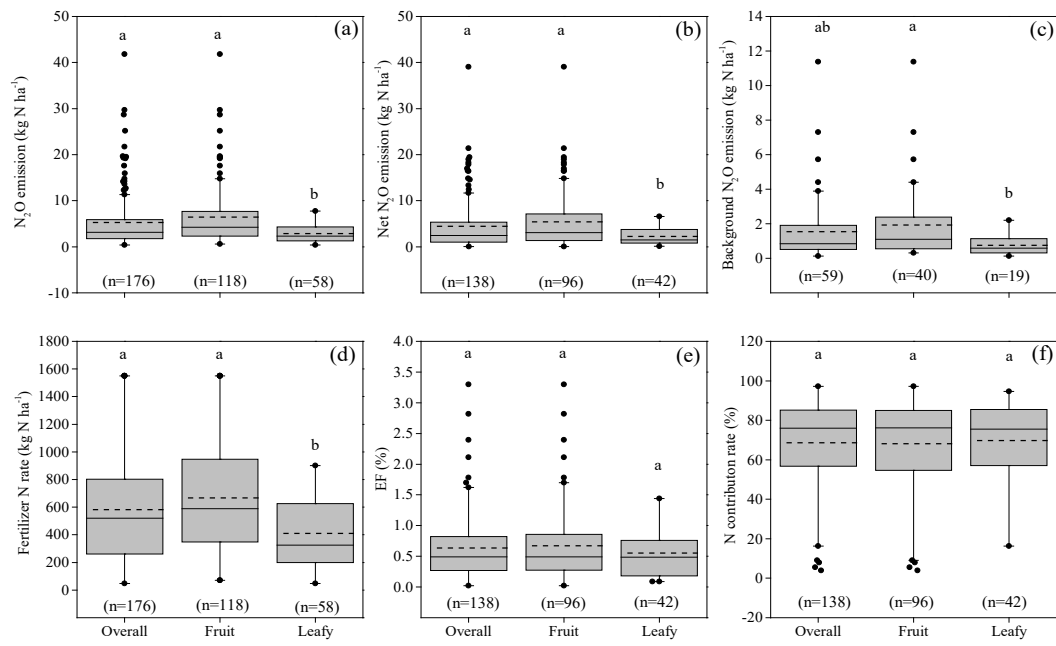


Fig. S2

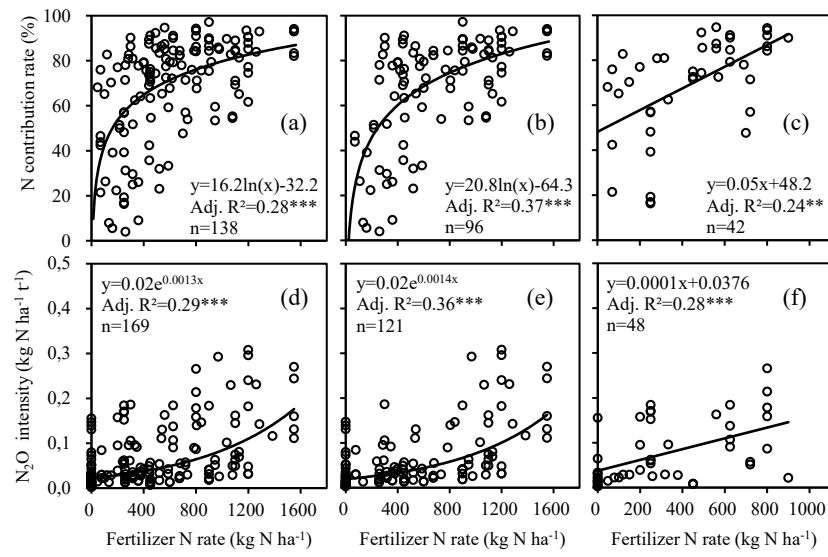


Fig. S3

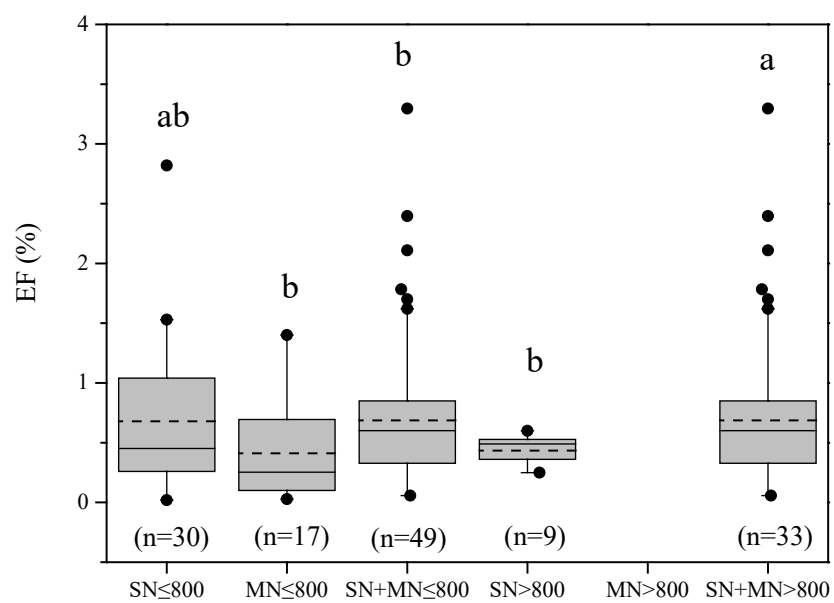


Fig. S4

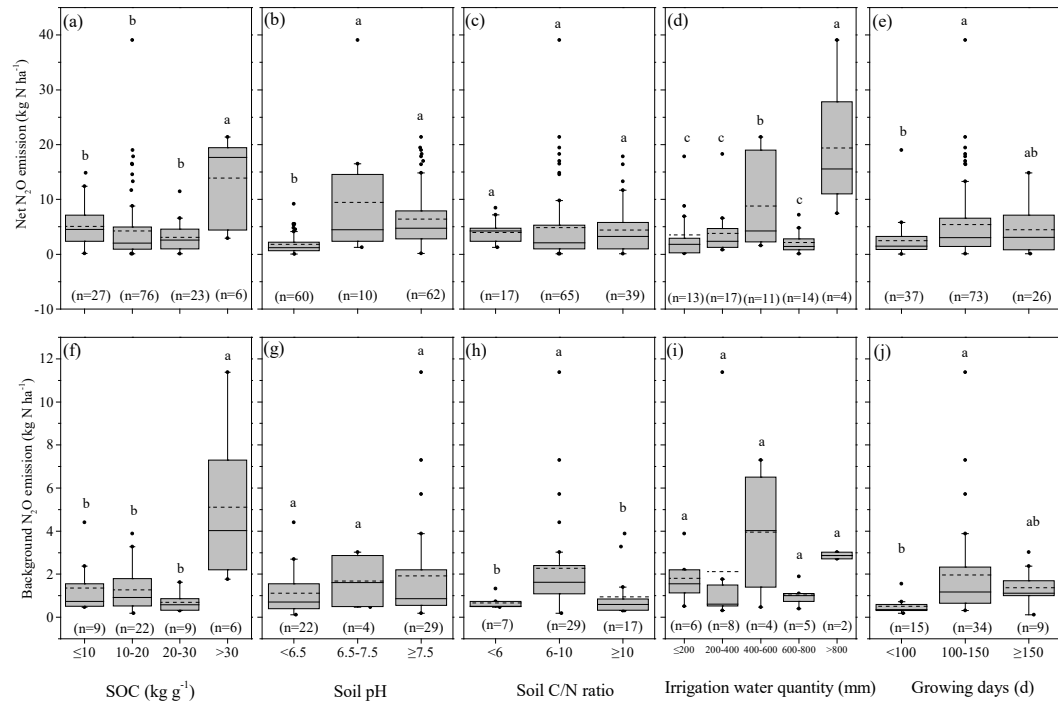


Fig. S5

