

Balancing non-CO₂ GHG emissions and soil carbon sequestration in U.S. rice paddies: implications for natural climate solutions

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Abstract: The U.S. rice paddy systems play an increasingly vital role in ensuring food security, which also contribute massive anthropogenic non-CO₂ (CH₄ and N₂O) greenhouse gas (GHG) emissions with expanding cultivation area. Yet, the full assessment of GHG balance, considering trade-offs between soil organic carbon (SOC) sequestration and non-CO₂ GHG emissions, is lacking. Integrating an improved agricultural ecosystem model with a meta-analysis of multiple field studies (a total of 322 site-year data representing 43 locations for U.S. rice paddies and 3402 site-year data representing 1113 locations for global rice paddies), we found that U.S. rice paddy was a rapidly growing net GHG emission source, increased 138% from 3.73 ± 1.16 Tg CO₂eq yr⁻¹ in the 1960s to 8.88 ± 2.65 Tg CO₂eq yr⁻¹ in the 2010s. CH₄ emission made the most significant contribution (10.12 ± 2.28 Tg CO₂eq yr⁻¹) to this increase in net GHG emissions in the 2010s, but increasing N₂O emissions, accounting for ~2.4% (0.21 ± 0.03 Tg CO₂eq yr⁻¹), cannot be ignored. SOC sequestration could offset about 14.0% (1.45 ± 0.46 Tg CO₂eq yr⁻¹) of the climate-warming effects of soil non-CO₂ GHG emissions in the 2010s. The aggravation of net GHG emissions stemmed from intensified land use/cover changes, rising atmospheric CO₂, and heightened synthetic N fertilizer and manure application. Climate change also exacerbated around ~21% of soil N₂O emissions and ~10% of soil CO₂ release in the 2010s. Nonetheless, adopting no/reduced tillage resulted in a substantial decrease of ~10 % in net soil GHG emissions, and non-continuous irrigation exhibited the potential to mitigate around 39% of soil non-CO₂ GHG emissions. Overall, the cost of this net GHG emissions for achieving increased U.S. rice yield markedly declined from 1960 to 2018, resulting in 0.84 ± 0.18 kg CO₂eq ha⁻¹ of net soil GHGs on average for each kilogram of grain produced in the 2010s. There would be a great potential to reduce emissions intensity in the mid-South U.S., especially the Mississippi Delta region, through optimization of the synthetic N fertilizer and manure ratio, reduction of tillage practices, and implementation of non-continuous irrigation. Our findings underscore the importance of net CO₂ GHG mitigation in U.S. rice paddies for achieving net zero-emission and climate-friendly rice production.

Keywords: Rice paddies; Methane; Nitrous oxide; Soil Organic Carbon (SOC); Nature climate solution; DLEM

1 Introduction

Rice paddy, a flooded agricultural system that grows rice (*Oryza sativa* L.), is a significant source of vital greenhouse gases (GHG) like CH₄ and N₂O (Linguist et al., 2018; Sauniois et al., 2020; Tian et al., 2016; Hussain et al., 2015; Gupta et al., 2021). Around 30% and 11% of global agricultural CH₄ and N₂O are emitted from rice paddies (Hussain et al., 2015; Gupta et al., 2021; Sauniois et al., 2020; Tian et al., 2020; Zhao et al., 2011). It is projected that up to 2030, emissions of both non-CO₂ GHGs from global rice paddies could experience a rise of 35-60% (Smith et al., 2007). This increase is attributed to a growing demand for rice production, expected to surge by 40% due to the expanding global population. These projections raise serious environmental concerns.

The United States is one of the top grain yield producers in the world and is among the top five countries for rice exports, with an expanding cultivation area (FAO, 2017). According to data from the USDA National Agriculture Statistics Service, approximately 80% of the rice produced in the U.S. is grown in the mid-South states (Arkansas, Louisiana, Texas, Mississippi, and Missouri), with the remaining 20% mainly produced in the Sacramento Valley in California. As the United States is expanding its rice paddy area, an urgent need is to quantify N₂O and CH₄ fluxes and improve our understanding of these gases from U.S. rice paddies to develop effective mitigation strategies.

There have been considerable efforts made toward broadly quantifying GHG emissions from rice systems and quantifying the effects of agronomic management and biogeochemical variables on the emissions. These efforts have been driven by data mostly observed and measured in Asia which produces ~90% of the world's rice (Akayama et al., 2005; Yan et al., 2005, 2009), and have been used to form guidelines to estimate GHG inventories for global rice paddies

including those in the United States (Eggleston et al., 2006). However, overwhelmed by Asia data, these learnings from previous studies on how to quantify GHG emissions and what are the major drivers of the emissions are unlikely valid for U.S. rice systems, which differ inherently from those typically found in Asia in agronomic practices. Differences include but are not limited to improved water management due to laser leveling and reliable water supply, a greater degree of mechanization, direct seeded rather than transplanted, and others (Linguist et al., 2018).

Rice paddy also has a considerable potential to be harnessed at a large scale to sequester carbon (Eswaran et al., 1993; Smith et al., 2007, 2008; Josep et al., 2021; Tian et al., 2016; Wollenberg et al., 2016). Extensive reviews have yet to be conducted for the U.S. rice system. Due to possible trade-offs between SOC sequestration and non-CO₂ GHG emissions under different agricultural management practices (Guenet et al., 2021; Tian et al., 2015; Tian et al., 2011; Runkle et al., 2018), simultaneous quantification of SOC sequestration and non-CO₂ GHG emissions is crucial to accurately assess the overall climate abatement potential of mitigation measures. Furthermore, whether SOC sequestration in U.S. rice paddies can offset non-CO₂ GHG emissions and how far we are from achieving carbon-neutral agriculture still need to be determined. Net soil GHG balance, defined as the sum of SOC sequestration and emissions of N₂O and CH₄, can be used to measure the overall climate effect resulting from cumulative radiative forcing of non-CO₂ GHG emissions and CO₂ uptake (Robertson & Grace, 2004; Tian et al., 2015). Therefore, it is crucial to advance our understanding of the magnitude and spatiotemporal variations of net GHG balance in rice paddies soils, as well as the drivers of these changes. Such knowledge is essential for developing effective climate change mitigation strategies for rice cultivation without sacrificing grain production.

Given the complexity of net GHG emissions, a process-based model would be ideal for quantifying and evaluating potential mitigation options. By utilizing process-based terrestrial biosphere models that accurately depict crop growth processes and incorporate agricultural management practices along with detailed assessments of hydrological, biophysical, and biogeochemical processes, we can gain a better understanding of how management practices and environmental changes affect net soil GHG balance at regional scales (Bondeau et al., 2007; McDermid et al., 2017; You et al., 2022). However, model simulation performance is primarily limited by input data and process parameterization. Conversely, field experiments offer practical and dependable avenues for unraveling intricate correlations between shifts in net soil GHG balance and agricultural management practices amidst various environmental changes (Plaza-Bonilla et al., 2018). However, extending site-specific findings directly to extensive geographical areas amplifies result uncertainties due to the distinct environmental and management conditions at each site (Huang et al., 2022; Fer et al., 2021; Peng et al., 2011). Until recently, there has been insufficient data to quantify emissions from the U.S. rice system and evaluate the effects of significant practices over large regional scales. Therefore, combining the strengths of field observations and models while addressing their respective limitations may offer a promising approach to overcoming current bottlenecks.

Here we quantified the combined effects of multiple management practices and environmental changes on the magnitude and spatiotemporal variations of net soil GHG balance in U.S. rice paddies using a model-data integration approach. The model used here is the Dynamic Land Ecosystem Model v4.0 (DLEM v4.0), which is a highly integrated process-based terrestrial biosphere model that is capable of simultaneously depicting biosphere-atmosphere exchanges of CO₂, N₂O, and CH₄ as driven by multiple environmental forcings and management practices (You

et al., 2022). A meta-analysis was conducted over U.S. and global rice paddies to compile field observations of SOC stock/sequestration and non-CO₂ GHG (i.e., N₂O and CH₄) emissions under various management practices and environmental conditions. Global meta data was employed to enhance existing data concerning the effects of U.S. agricultural management practices and to facilitate parallel comparisons with U.S. results. We used the compiled dataset to calibrate, validate, and corroborate model simulations. Our study aimed to achieve three objectives: (1) estimate the net soil GHG balance of U.S. rice paddies from 1960 to 2018, considering multiple environmental changes (e.g., synthetic N fertilization, manure, tillage, irrigation, climate conditions, historical land use, atmospheric CO₂ concentration, and N deposition); (2) quantify the contributions of different drivers to the spatial and temporal variations in net soil GHG balance across the country; and (3) estimate the temporal-spatial changes in the net GHG emission intensity of U.S. rice paddies, a measure of GHG emissions per unit rice production.

2 Materials and methods

2.1 Data sources for meta-analysis

We conducted a comprehensive literature search to identify peer-reviewed publications reporting *in-situ* soil GHG emissions (CH_4 , N_2O , and CO_2) from the U.S. and global rice paddies under different management practices and environmental conditions. Several databases, such as Google Scholar, Web of Science, and Scopus, were used to search literature. Search keywords included “rice field or rice paddies,” “the United States or America or U.S. or USA,” “soil organic carbon or SOC,” “nitrous oxide or N_2O ,” “methane or CH_4 ,” and “greenhouse gases or GHG.” To ensure the quality of the compiled dataset, we collected papers further refined by the following criteria: (1) measurements were made in the field rather than in the laboratory; (2) ancillary information such as cropping systems, experimental year and duration, and applied management practices (e.g., N fertilizer use rate, tillage type, and irrigation) were provided; and (3) replicated field experiments were performed.

A total of 322 site-year data representing 43 locations for U.S. rice paddies and 3402 site-year data representing 1113 locations for global rice paddies were collected (Fig. 1 and Table S1 in the supplementary material). The dataset for U.S. rice paddies included 74 observations of N_2O emissions, 322 observations of CH_4 emissions, and 225 observations of rice yield (Fig. 1 and Table S1 in the supplementary material). Since there are few records of SOC stock/sequestration experiments in U.S. rice paddies, we adopted 192 observations of SOC stock that were obtained from the WoSIS (Batjes et al., 2016, <http://dx.doi.org/10.17027/isric-wdcsoils.20160003>). The global dataset included 724 records of N_2O emissions, 1238 records of CH_4 emissions, 3402 records of SOC stock (3256 records of SOC sequestration), and 1006 records of rice yield (Fig. 1),

156 obtained from Bo et al. (2022) and Liu et al. (2021). Multiple management practices were involved
157 in these observations, such as tillage type, N fertilization, irrigation, manure application, and cover
158 cropping. In addition, GetData Graph Digitizer software was used to extract exact values when
159 data was presented in graphical form.

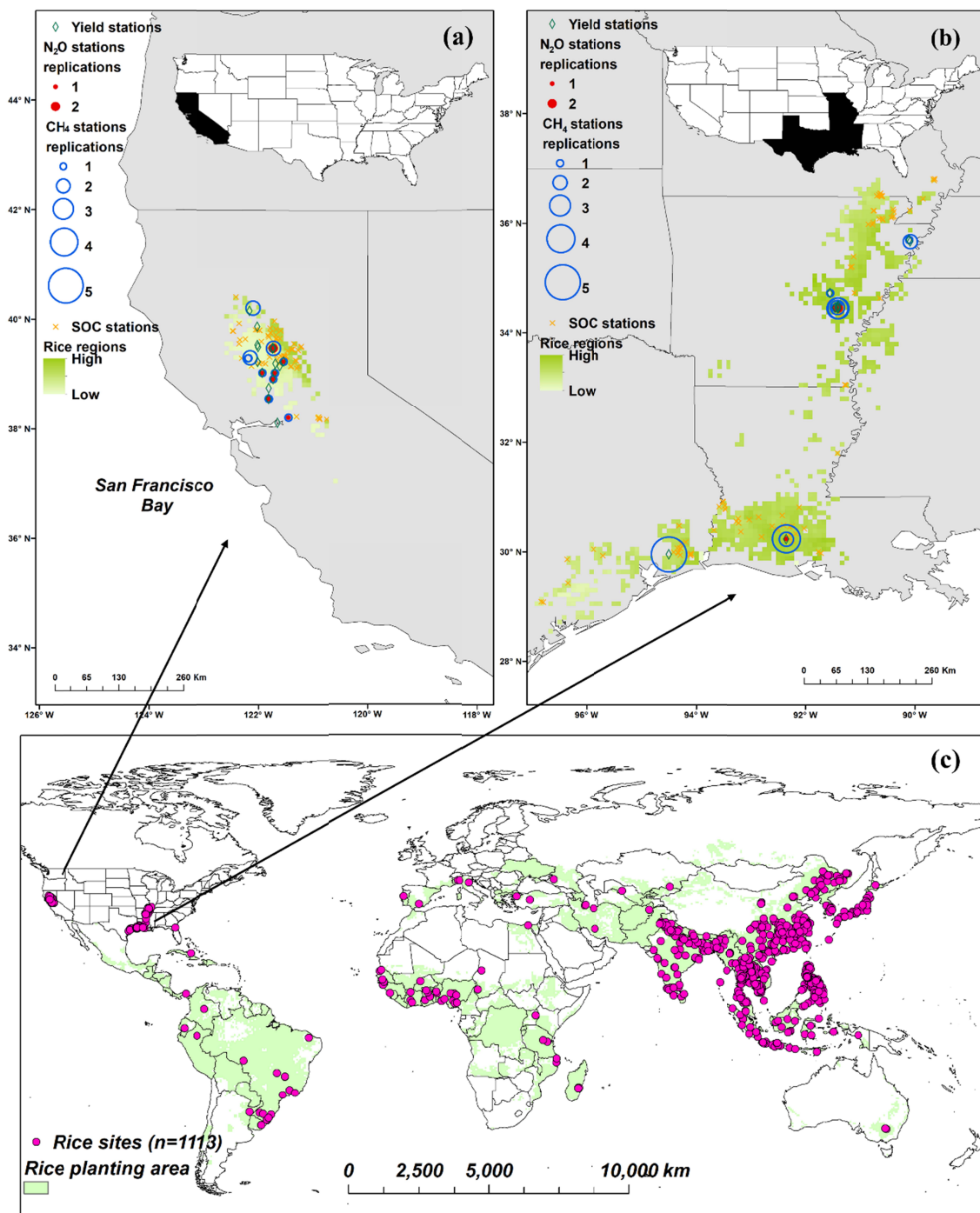


Figure 1. Spatial distribution of rice experimental sites in this study. **a** and **b** represent rice experimental/observation sites in U.S. rice paddies used for model calibration, validation, and meta-analysis. **c** represents global rice experimental/observation sites used for the meta-analysis. Rose circles include CH₄ flux, N₂O flux, SOC stock/sequestration, and yield experimental stations, obtained from Bo et al. (2022) and Liu et al. (2021); the blue circle represents the CH₄ emissions experimental station; the red circle represents the N₂O emission experimental station, and fork shape means SOC observations obtained from Batjes et al. (2016) (<http://dx.doi.org/10.17027/isric-wdcsoils.20160003>). Point size represents the number of replications/observations at each site. More detailed about sites in **a** and **b** were shown in Table S1 in supplemental information.

2.2 Model description

DLEM v4.0 is a comprehensive process-based terrestrial biosphere model that integrates major biophysical, biogeochemical, and hydrological processes to quantify daily, spatially explicit stocks and fluxes of carbon, water, and nutrients in terrestrial ecosystems and inland water systems at site, regional, and global scales (Pan et al., 2021; Tian et al., 2010, 2020; Yao et al., 2020; You et al., 2022; Zhang et al., 2018, 2020). In addition to meeting cross-scale agricultural needs such as management guidance, climate change mitigation, and adaptation, DLEM v4.0 also includes dynamic representations of crop growth and development processes, such as crop-specific phenological development, carbon allocation, yield formation, and biological N fixation, as well as agricultural management practices such as N fertilization, irrigation, tillage, manure application, dynamic crop rotation, cover cropping, and genetic improvements (You et al., 2022). The detailed representation of crop growth and management practices in DLEM v4.0 allows for the simulation of crop yield, crop state variables, biogeochemical fluxes, and pools of carbon, N, and water related to agroecosystems across various spatial and temporal scales, driven by multiple environmental forces such as climate change, atmospheric CO₂ fertilization and N deposition, tropospheric ozone pollution, and land use and land cover change. For more information on the representation of crop growth and agricultural management practices in DLEM v4.0, please refer to You et al. (2022).

2.3 Model forcing dataset

To drive DLEM v4.0, four types of long-term spatial datasets at a resolution of 5×5 arc-min were developed: Natural environmental changes (daily climate conditions, monthly atmospheric CO₂, and monthly N deposition); Yearly agricultural management practices (annual N fertilizer

use rate, crop rotation, tillage, irrigation, manure application, and crop phenology); Yearly land use and land cover change; and soil properties and other auxiliary data. Further details on this dataset were provided in supplementary Text S1.

2.4 Model calibration, validation, and uncertainty analysis

DLEM has undergone extensive validation and application to estimate daily and yearly N_2O flux, CH_4 flux, and SOC stock/sequestration as well as crop yield across various sites and regions globally (e.g., Huang et al., 2020; Lu et al., 2021; Yu et al., 2018; Friedlingstein et al., 2020; Ren et al., 2020; Saunio et al., 2020; Tian et al., 2020a; You et al., 2022; Zhang et al., 2020). For this study, we have calibrated and validated DLEM v4.0 using field observations compiled by meta-analysis to better simulate SOC stock/sequestration and emissions of N_2O and CH_4 in U.S. rice paddies. We used various metrics, such as coefficient of determination (R^2), root mean square error (RMSE), and normalized root mean square error (NRMSE), to quantitatively evaluate the model performance.

A total of 322 site-year measurements from 43 U.S. rice paddy sites were utilized to calibrate, validate, and confirm model simulations in this study (see Fig. 1). In general, DLEM v4.0 exhibited good performance in simulating yearly CH_4 and N_2O emissions as well as SOC stock/sequestration in rice paddies when compared with field observations from meta-analysis. The RMSE (NRMSE) values between model simulations and observations were 156.10 kg $\text{CH}_4\text{-C ha}^{-1} \text{ yr}^{-1}$ (16.16%), 0.28 kg $\text{N}_2\text{O-N ha}^{-1} \text{ yr}^{-1}$ (14.76%), 39.08 Mg C $\text{ha}^{-1} \text{ yr}^{-1}$ (25.72%), and 2339.59 kg $\text{ha}^{-1} \text{ yr}^{-1}$ (18.93%), respectively, while the corresponding R^2 values were 0.66, 0.84, 0.63, and 0.93, respectively (see Fig. 2).

In previous studies (e.g., Tian et al., 2011; Xu, 2010), we conducted a global sensitivity and uncertainty analysis to quantify uncertainties in the simulated regional SOC sequestration rate and

215 fluxes of N_2O and CH_4 in U.S. croplands. To do this, we used the Sobol method (Sobol, 1993) to

216 perform a variance-based global sensitivity analysis to determine the relative importance of model

217 parameters in simulating SOC sequestration rate and emissions of N_2O and CH_4 . We then

218 identified parameters that significantly affected the simulation results and generated an ensemble

219 of 100 parameter sets by randomly varying their values within 30% of their calibrated values using

220 a Monte Carlo sampling scheme (Tian et al., 2011; You et al., 2022). Finally, we used the

221 ensemble of parameter sets to drive DLEM v4.0 to simulate regional SOC sequestration rate and

222 emissions of N_2O and CH_4 from U.S. rice paddies.

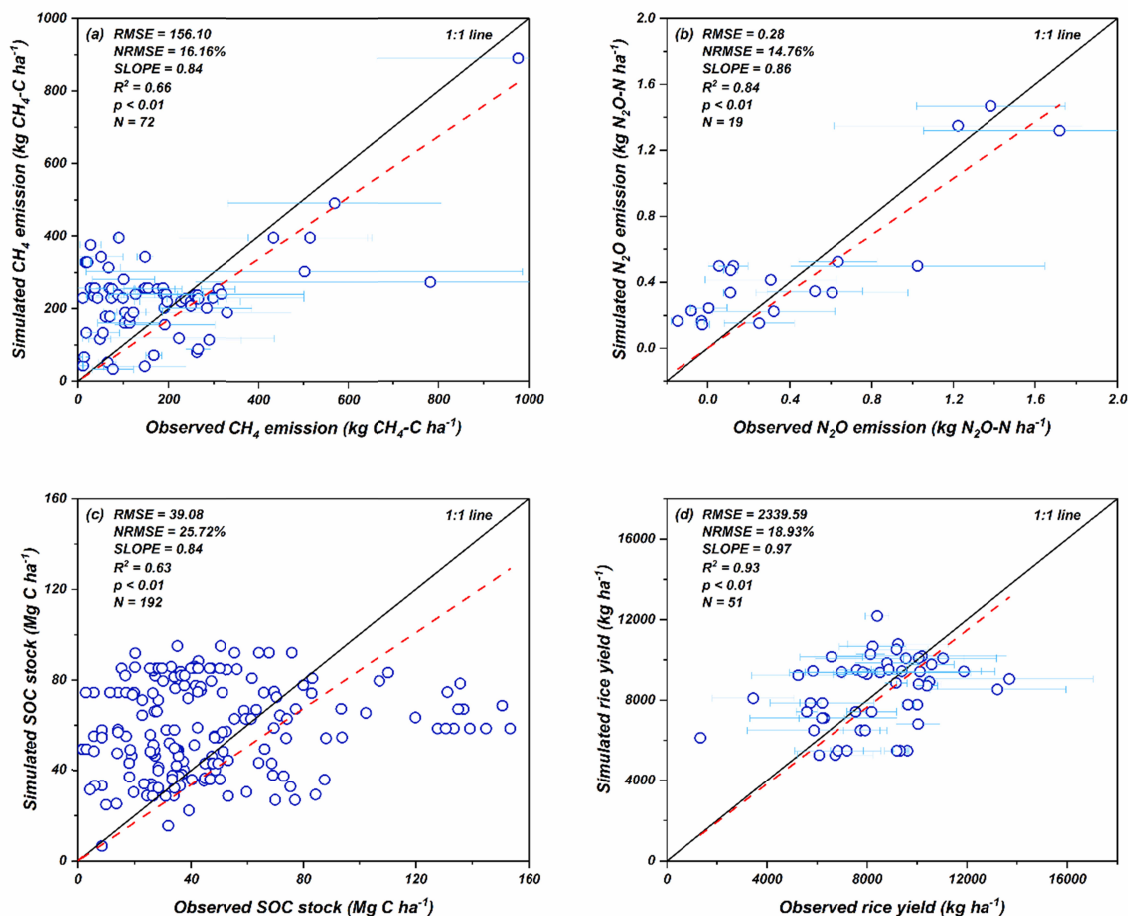


Figure 2. Site-scale comparisons of model estimates and field observations of CH_4 (a), N_2O (b), SOC stock (c), and rice yield (d) across different agricultural managements (e.g., fertilizer, irrigation, tillage, and others). The dashed line is the regression of observed data and modeled results, and the solid line is the 1:1 line. Note that we outputted the simulation results at the corresponding observed period for validation.

228 2.5 Model implementation and experimental design

229 The implementation of DLEM v4.0 consists of three main stages: an equilibrium
230 run, a spin-up run, and a transient run. During the equilibrium run, 30-year average
231 climate conditions from the 1900s to the 1920s and other environmental factors in
232 1900 were used. The equilibrium state was considered reached when changes in C, N,
233 and water pools between two consecutive 50-years were less than $0.5 \text{ g C m}^{-2} \text{ yr}^{-1}$, 0.5
234 $\text{g N m}^{-2} \text{ yr}^{-1}$, and 0.5 mm yr^{-1} , respectively. To eliminate model fluctuations caused by
235 the transition from the equilibrium run to the transient run, the spin-up run was driven
236 by detrended climate data from the 1900s to the 1920s. Finally, the transient run was
237 driven by historical data from 1900 to 2018.

238 We conducted 11 simulation experiments (Table 1) to identify the distinct roles
239 played by various drivers in influencing the net soil GHG balance of U.S. rice paddies
240 during 1960-2018. The factors considered for attribution included N fertilization,
241 tillage, irrigation, manure application, climate change, atmospheric CO_2 concentration
242 and N deposition, and LULC. To evaluate model fluctuations resulting from internal
243 system dynamics, a reference run (S0) was carried out by maintaining all factors at
244 the 1900 level (climate data in the 1900 level means the 30-year average climate
245 condition from the 1900s to the 1920s). To determine the overall impact of all the
246 factors on SOC sequestration and N_2O and CH_4 emissions, an all-combine run (S1)
247 was conducted using all historically varying input forcings during 1900-2018. The
248 difference between S1 and S0 simulations was calculated to estimate the net changes

249 in SOC sequestration rate and emissions of N₂O and CH₄ driven by all factors.
250 Furthermore, we performed 7 additional simulations (S2-S8) to examine the
251 individual contributions of each factor to annual variations in SOC sequestration rate
252 and fluxes of N₂O and CH₄. In each simulation, one specific factor was kept at the
253 1900 level, while all other factors were varied over time, and the contribution of this
254 factor was obtained by subtracting the simulation from the "All Combined"
255 simulation (S1). Additionally, since LULC is often associated with changes in the
256 overall input of management practices (e.g., manure and mineral fertilizer application),
257 we calculated the contribution of LULC by maintaining all management factors at the
258 1900 level while varying other environmental factors (Lu et al., 2021). Thus, the
259 difference between S9 and S10 was used to determine the contribution of LULC.

260

Table 1. Factorial experiments to quantify the relative contributions of different drivers to changes in SOC, N₂O, and CH₄ emissions from U.S. rice paddies.

No.	Scenario	Nfer ^a	Tillage ^b	Irrigation ^c	Manure	Climate ^d	CO ₂	Ndep	LULC
S0	Reference	1900	1900	1900	1900	1900	1900	1900	1900
S1	All Combined	1900-2018	1900-2018	1900-2018	1900-2018	1900-2018	1900-2018	1900-2018	1900-2018
S2	Without N fertilization (Nfer)	1900	1900-2018	1900-2018	1900-2018	1900-2018	1900-2018	1900-2018	1900-2018
S3	Without Tillage	1900-2018	1900	1900-2018	1900-2018	1900-2018	1900-2018	1900-2018	1900-2018
S4	Without Irrigation	1900-2018	1900-2018	1900	1900-2018	1900-2018	1900-2018	1900-2018	1900-2018
S5	Without Manure	1900-2018	1900-2018	1900-2018	1900	1900-2018	1900-2018	1900-2018	1900-2018
S6	Without Climate	1900-2018	1900-2018	1900-2018	1900-2018	1900	1900-2018	1900-2018	1900-2018
S7	Without CO ₂	1900-2018	1900-2018	1900-2018	1900-2018	1900-2018	1900	1900-2018	1900-2018
S8	Without N deposition (Ndep)	1900-2018	1900-2018	1900-2018	1900-2018	1900-2018	1900-2018	1900	1900-2018
S9	Climate+CO ₂ +Ndep	1900-2018	1900-2018	1900-2018	1900	1900	1900	1900	1900
S10	Climate+CO ₂ +Ndep+LULC	1900-2018	1900-2018	1900-2018	1900	1900	1900	1900	1900-2018

261 ^aWe assumed N fertilization rate before 1910 was kept constant at the 1910 level; ^bWe assumed tillage data before 1960 was kept constant at the 1960 level; ^cWe assumed
262 irrigation data before 1950 was kept constant at the 1950 level; ^dClimate data in 1900 was the 30-year average climate condition from the 1900s to the 1920s.

263 2.6 Global warming potential calculation

264 The global warming potential (GWP) is a metric used to quantify the cumulative
265 radiative forcing resulting from the emission of 1 kg of a trace gas compared to the
266 emission of 1 kg of CO₂ (Myhre et al., 2013). In croplands, the GWP value of the net
267 soil GHG balance is determined by calculating the sum of CO₂ equivalents from SOC
268 sequestration and emissions of N₂O and CH₄:

$$GWP = F_{CO_2-C} \times \frac{44}{12} + F_{N_2O-N} \times \frac{44}{28} \times GWP_{N_2O} + F_{CH_4-C} \times \frac{16}{12} \times GWP_{CH_4} \quad (1)$$

$$F_{CO_2-C} = -SOC_{SR} \quad (2)$$

269 where F_{CO_2-C} , F_{N_2O-N} , and F_{CH_4-C} were annual fluxes of CO₂, N₂O, and CH₄,
270 respectively; SOC_{SR} was SOC sequestration rate; molecular weight conversion
271 fractions 44/12, 44/28, and 16/12 were used to convert the mass of CO₂-C, N₂O-N,
272 and CH₄-C into CO₂, N₂O, and CH₄, respectively; GWP_{N_2O} and GWP_{CH_4} were
273 GWP constants indicating radiative forcing of N₂O and CH₄ in terms of their CO₂
274 equivalents, and this study used the GWP values integrated over a time horizon of 100
275 years for N₂O and CH₄, which were 273 and 27, respectively (IPCC AR6).

3 Results

3.1 Temporal-spatial changes of net GHG balance in U.S. rice paddies

According to our estimates, the annual soil non-CO₂ greenhouse gas (GHG) emissions from U.S. rice paddies experienced a notable increase over the years. In the 1960s, these emissions were approximately 3.89 ± 1.13 Tg CO₂eq yr⁻¹. By the 1990s, they had surged to 9.79 ± 1.08 Tg CO₂eq yr⁻¹, showing a significant growth rate of 181.13 Mg CO₂eq yr⁻¹. Subsequently, in the 2010s, the emissions reached a level of 10.33 ± 2.30 Tg CO₂eq yr⁻¹, demonstrating a slightly weaker upward trend with an increase of 57.37 Mg CO₂eq yr⁻¹. The annual soil CH₄ emissions originating from rice paddies represented a substantial portion, accounting for 97.93% (equivalent to 10.12 ± 2.28 Tg CO₂eq yr⁻¹ or 7511.47 ± 1611.45 kg CO₂eq ha⁻¹) in the 2010s. This was accompanied by a growth rate of 134.44 Mg CO₂eq yr⁻¹ (61.72 kg CO₂eq ha⁻¹ yr⁻¹) (Fig. 3a). While soil N₂O emission in paddies soil was initially modest, it displayed a substantial and noteworthy increase, reaching 0.21 ± 0.03 Tg CO₂eq yr⁻¹ (167.68 ± 24.84 kg CO₂eq ha⁻¹) in the 2010s. This growth trend is particularly significant at a rate of 2.69 Mg CO₂eq yr⁻¹ (1.33 kg CO₂eq ha⁻¹ yr⁻¹) (Fig. 3b). The simulations aligned closely with the findings from our meta-analysis, showing similar results for CH₄ emission (8792.17 ± 1577.95 kg CO₂eq ha⁻¹ vs. 8809.26 ± 1507.44 kg CO₂eq ha⁻¹) and N₂O emission (198.74 ± 13.24 kg CO₂eq ha⁻¹ vs. 208.02 ± 264.13 kg CO₂eq ha⁻¹) over the same period. Notably, the annual soil CH₄ emission from U.S. rice paddies surpassed the global average (7787.57 ± 10771.4 kg CO₂eq ha⁻¹). Conversely, the soil N₂O emission from U.S. rice paddies was lower than the global average (521.63 ± 729.49 kg CO₂eq ha⁻¹). The distribution of CH₄ flux resulting from rice cultivation exhibited remarkable polarization, with a high CH₄ emission of approximately 7200 kg CO₂eq ha⁻¹ observed in the

majority of the mid-South States, particularly in the Mississippi Delta Arkansas region, as well as in the northern part of the Sacramento Valley region in California (Fig. 3a). In contrast, other areas, such as Texas, displayed lower rates, measuring less than 5400 kg CO₂eq ha⁻¹. Similarly, the spatial pattern of N₂O emissions showed limited variation across the mid-South States, with emissions higher than 128.7 kg CO₂eq ha⁻¹ (Fig. 3b). In contrast, the simulations indicated low N₂O emissions for the Sacramento Valley region.

The soil stock in U.S. rice paddies measured 200.80±3.58 Mg CO₂eq ha⁻¹ in the 2010s, surpassing the global average of 161.98±84.85 Mg CO₂eq ha⁻¹ based on meta-analysis. This outcome stems from U.S. rice paddies exhibiting a significant capacity to sequester about 1.45±0.46 Tg CO₂eq yr⁻¹ (equivalent to 1305.76±460.19 kg CO₂eq ha⁻¹ yr⁻¹) during this period, accounting for approximately 14.03% of soil non-CO₂ GHG emissions. This soil CO₂ uptake demonstrated an average growth rate of 30.67 Mg CO₂eq yr⁻¹ (equivalent to 24.16 kg CO₂eq ha⁻¹ yr⁻¹) from 1960 to 2018 (Fig. 3c). The majority of U.S. rice paddies acted as carbon sinks, with relatively higher rates of SOC sequestration (>290 kg CO₂eq ha⁻¹ yr⁻¹) observed in the Sacramento Valley region and lower rates (<150 kg CO₂eq ha⁻¹ yr⁻¹) across the mid-South States (Fig. 3c). Conversely, the global rice paddies exhibited a carbon source, with an average emission of 6828.77±37238.78 kg CO₂eq ha⁻¹ yr⁻¹ of soil CO₂ throughout the study period.

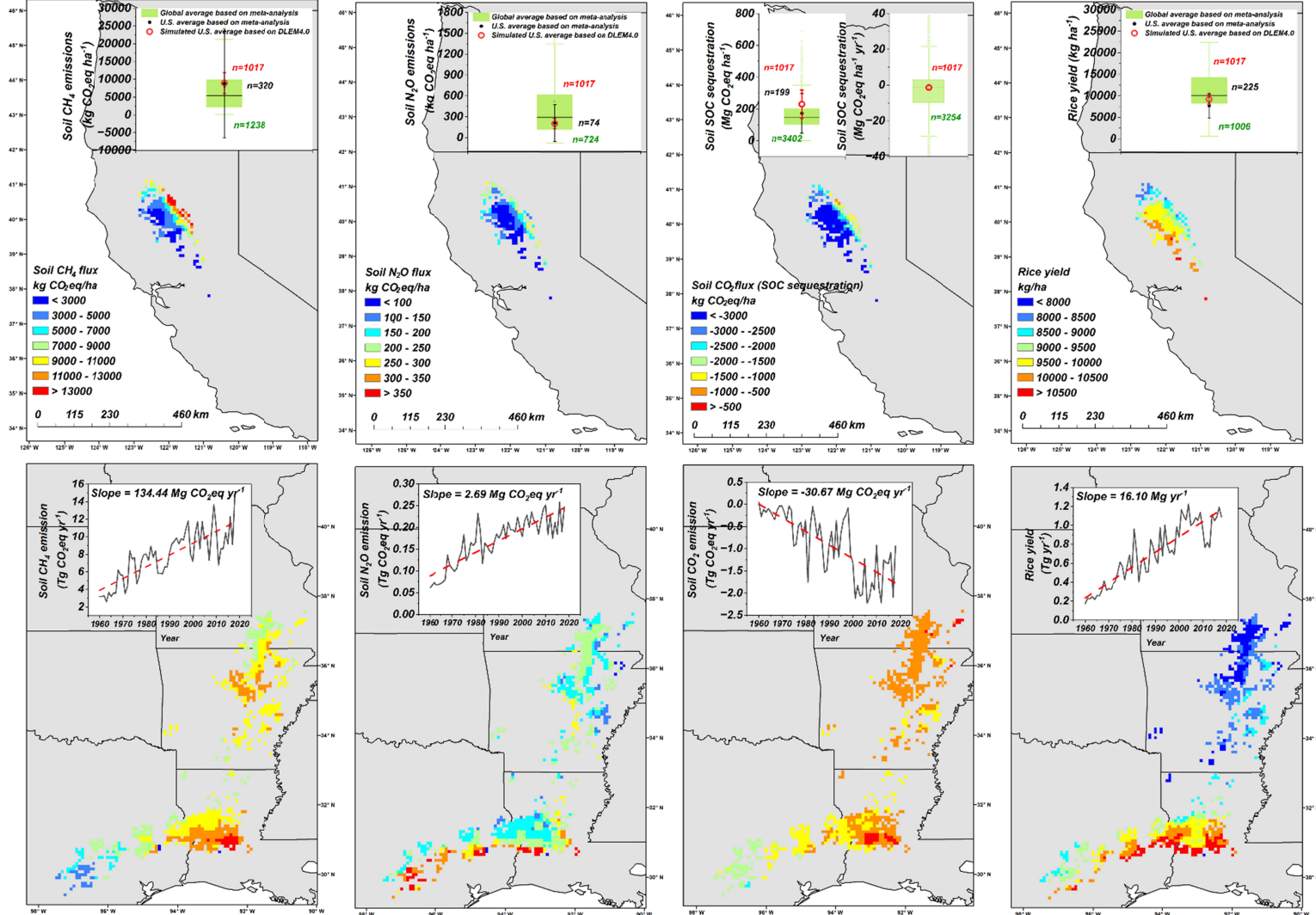


Figure 3. Spatial patterns of average annual soil CH₄ flux, N₂O flux, and CO₂ flux (SOC sequestration) in rice paddies from 2010 to 2018. The insets in the top row depicted a comparison between observations and simulations of soil CH₄ flux, N₂O flux, and CO₂ flux (SOC sequestration) in global rice paddies (illustrated by the box) and U.S. rice paddies (represented by the circle). The insets in the bottom row showed annual changes in soil CH₄ flux, N₂O flux, and CO₂ flux (SOC sequestration) in rice paddies from 1960 to 2018. Note that negative values in soil fluxes represent uptake, positive values represent release, and negative SOC sequestration rate indicates soil CO₂ flux.

The U.S. rice paddy thus showed a rapidly increasing in net soil GHG emissions at a growth trend of $161.37 \text{ Mg CO}_2\text{eq yr}^{-1}$ ($57.71 \text{ kg CO}_2\text{eq ha}^{-1} \text{ yr}^{-1}$) from the 1960s to the 1990s, and then leveled off to $8.89 \pm 2.65 \text{ Tg CO}_2\text{eq yr}^{-1}$ ($7405.11 \pm 1665.07 \text{ kg CO}_2\text{eq ha}^{-1}$) by the 2010s (Fig. 4). The distribution of net soil GHG balance showed considerable spatial heterogeneity, with hotspots in the north of Sacramento Valley region and the Mississippi Delta region where peak net soil GHG emissions were estimated to be higher than $8000 \text{ kg CO}_2\text{eq ha}^{-1}$ (Fig. 4). In contrast, some U.S. rice paddies (primarily located in the southeast of the Sacramento Valley region) acted as a net sink of GHGs during the study period (representing $<5\%$ of U.S. rice paddies area), suggesting that sequestered SOC in these regions completely offset non- CO_2 GHG emissions (Fig. 4).

We further analyzed the spatial distribution of the relative contribution of SOC sequestration and N_2O and CH_4 emissions to the net GHG balance of U.S. rice paddies (Fig. 5). Over the study period, soil CH_4 emissions played a dominant role in controlling the net GHG balance of most rice paddies (e.g., most of the Mississippi Delta and the north of the Sacramento Valley region), while SOC sequestration and CH_4 emissions synergistically controlled the net GHG balance in the southwest of mid-South States (mainly in Texas). Meanwhile, the proportion of regions dominated by SOC sequestration that increased over time in most of the Sacramento Valley region is noteworthy and indicates an increasing role of SOC sequestration in controlling the net GHG balance across U.S. rice paddies (Fig. 5).

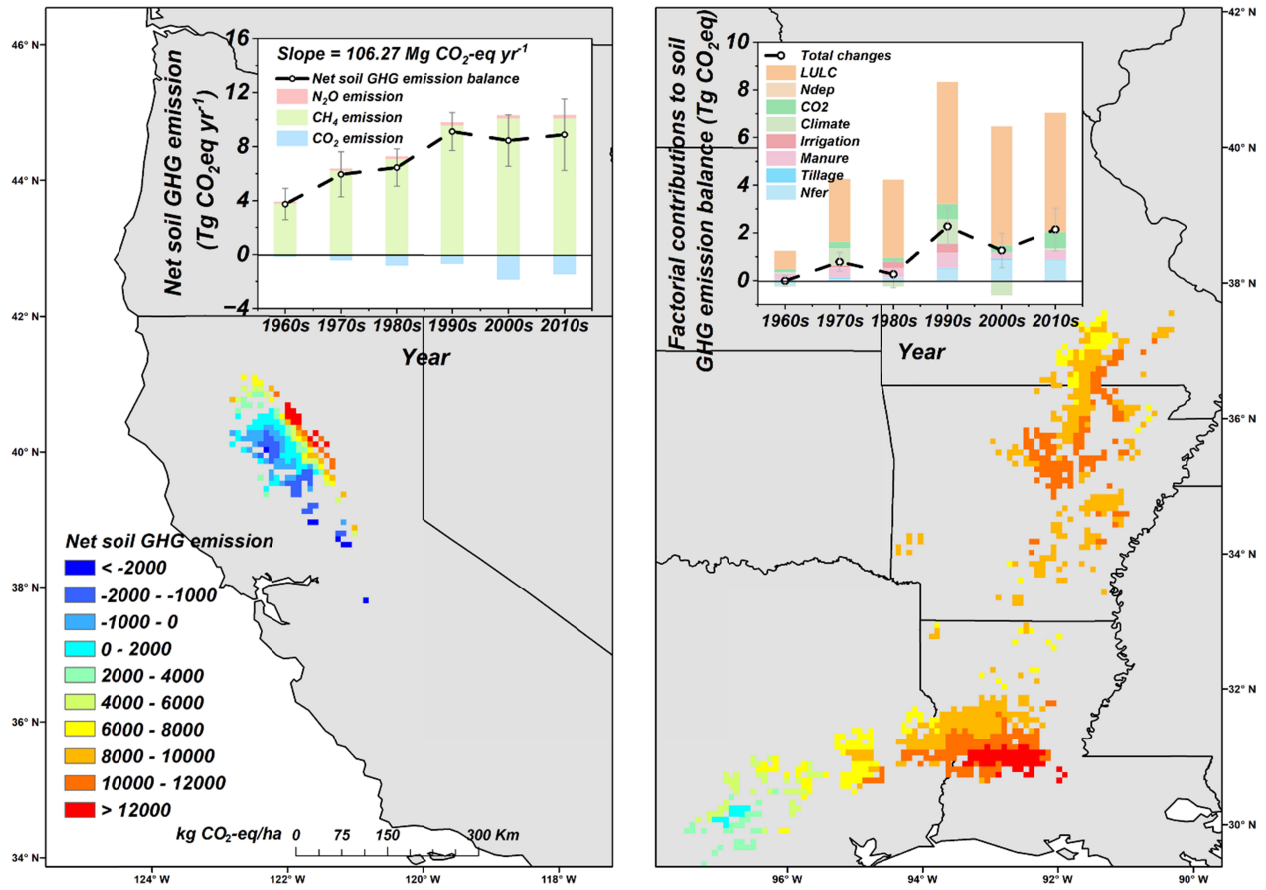


Figure 4. Spatial pattern of average annual net soil GHG balance of U.S. rice paddies in the 2010s. Inset in a represents the decadal average GWP of three gases; inset in b showed factorial contributions of multiple agricultural management practices and environmental forcing to changes in the net GHG balance of U.S. rice paddies from the 1960s to the 2010s. *Nfer* represents nitrogen fertilizer use; *Ndep* represents atmospheric nitrogen deposition; LULC represents land use and land cover change (reflecting both cropland abandonment and expansion, as well as interannual crop rotation changes); and CO₂ represents atmospheric carbon dioxide concentration. Note that the sum of factorial contributions of individual drivers (i.e., stacked bars) does not equal net changes in the net GHG balance (i.e., black line) due to interaction effects. Note that error bars in insets represent ± 1 standard deviation of the net GHG balance in each decade.

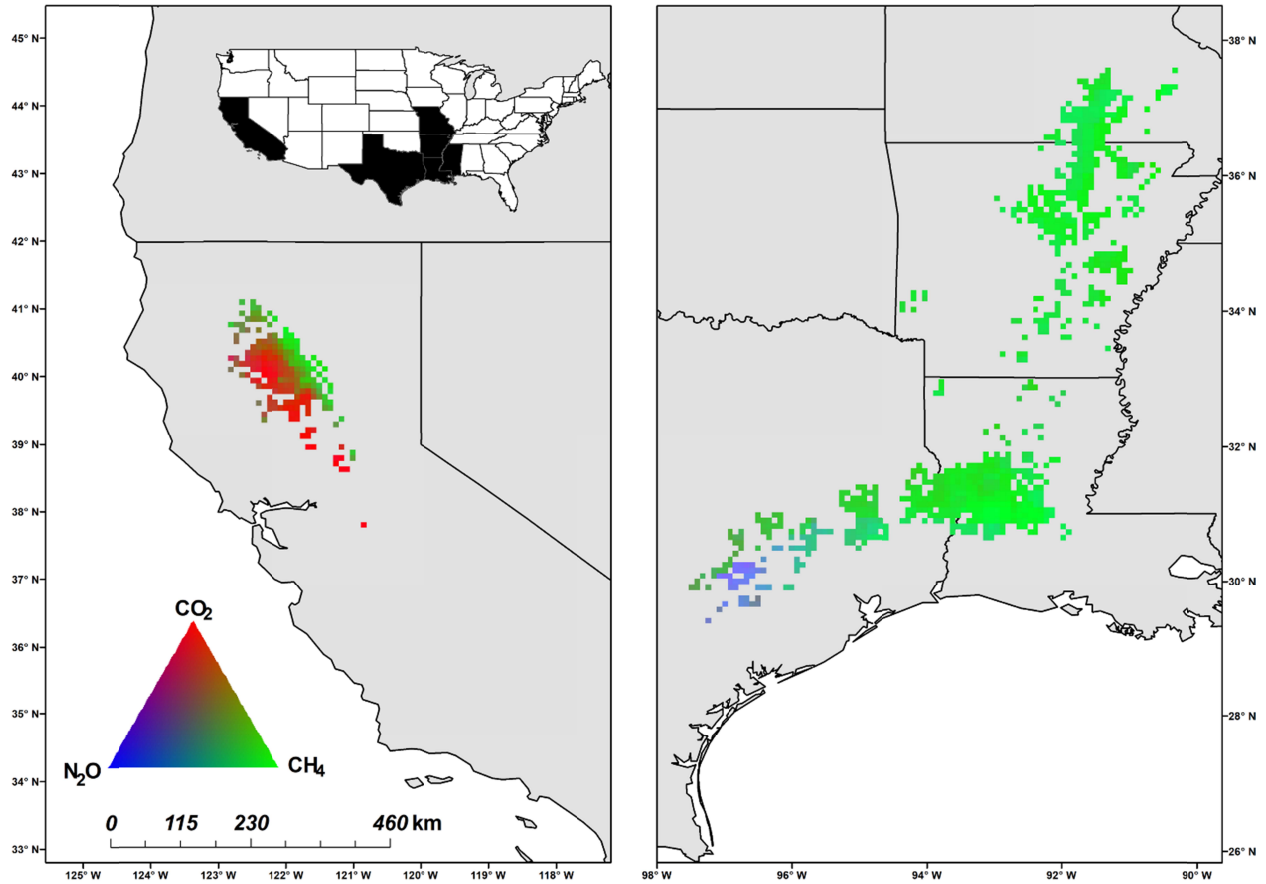


Figure 5. Spatial distributions of the relative contribution of SOC sequestration and emissions of N_2O and CH_4 to net soil GHG balance of U.S. rice paddies in the 2010s.

3.2 Factorial contributions of multi-driver changes to net GHG balance in U.S. rice paddies

Leveraging agricultural management practices to curb net GHG emissions from croplands has recently come under sharp focus (Fargione et al., 2018). Over the study period, the application of agricultural management practices in the U.S. croplands was constantly being strengthened (Fig. S1). For example, synthetic N fertilizer use in U.S. croplands increased substantially over our study period (Fig. S1a), from 2.48 Tg N year⁻¹ in 1960 to 11.8 Tg N year⁻¹ in 2015. However, it is worth noting that the upward trend has subsequently shown signs of deceleration. The proportion of U.S. croplands adopting enhanced tillage (e.g., no tillage or reduced tillage) practices increased significantly over the past three decades according to CRM tillage survey data. However, for U.S. rice paddies, conventional tillage was still dominant, and

the proportion was increasing over the study period (Figure S1c). Manure application has continuously increased from 1900, resulting in over 1.2 Tg N yr⁻¹ in manure usage after the 2000s (Figure S1d). Irrigated cropland acreage has also increased significantly, reaching 51.5 million acres by 2018 (Figure S1e).

We thus further quantified the factorial contributions of key drivers, including multiple agricultural management practices and environmental forcings, to changes in the net soil GHG balance of U.S. rice paddies from 1960 to 2018 by setting up a series of simulation experiments (Table 1). Simulation results during the study period showed that the rapid increase of synthetic N fertilizer application was the dominant factor for driving the net GHG emission exacerbation of U.S. rice paddies, contributing to 0.85 Tg CO₂eq yr⁻¹ on average in the 2010s with a rising trend of 22.89 Mg CO₂-eq yr⁻¹ in net GHG emission. These changes accounted for roughly 39.78% of the alterations in the net GHG emission (Fig. 4). Within this context, N fertilizer played a substantial role in influencing the changes in soil CH₄ emission about 46.41% (equivalent to 1.64 Tg CO₂eq yr⁻¹), consistent with findings from similar studies in U.S. rice paddies (50.60% in observations), but higher than the global average (33.86% in observations) based on the meta-analysis in this study (Fig. 6a). This contribution exhibited a growth trend of 37.18 Mg CO₂-eq yr⁻¹ over the study period (Fig. 10a). Moreover, N fertilizer also notably influenced soil N₂O emission, contributing about 0.21 Tg CO₂eq yr⁻¹ (representing 97.45% of the emissions) during the 2010s. This contribution exhibited a considerable increasing trend of 2.72 Mg CO₂-eq yr⁻¹ throughout the study period (Fig. 10b), surpassing the meta-analysis results for both U.S. rice paddies (23.12% in observations) and the globe average (48.13% in observations) (Fig. 6b). Despite these intensified GHG emissions, N fertilizer positively affected SOC sequestration simultaneously, with a growth rate of 17.01 Mg CO₂eq yr⁻¹. This contribution accounted for

63.17% (equivalent to 1.0 Tg CO₂eq yr⁻¹) of changes in SOC sequestration in U.S. rice paddies during the 2010s (Fig. 10c), surpassing the global average (29.78% in observations) as indicated in Fig. 6c.

Increasing manure application was another impact factor for driving changes in the net GHG balance of U.S. rice paddies. On average, manure application contributed approximately 0.42 Tg CO₂eq yr⁻¹ during the 2010s, showing a weak growth trend and accounting for 19.53% of changes in net GHG emissions (see Fig. 4). As the most important organic soil amendment, manure played a crucial role in enhancing SOC sequestration. Its contribution increased significantly from 0.05 Tg CO₂-eq yr⁻¹ (24.19%) in the 1960s to 0.51 Tg CO₂-eq yr⁻¹ (32.24%) in the 2010s, with a gradually increasing trend of 10.77 Mg CO₂eq yr⁻¹ (Fig. 10c). However, it's worth noting that manure application also contributed to a 26.22% increase (0.93 Tg CO₂-eq yr⁻¹) in soil CH₄ emission (Fig. 10a). On a global scale, the contribution of manure application to SOC sequestration and CH₄ emission was even more remarkable, accounting for 94.56% and 44.90%, respectively, based on meta-analysis (Fig. 7a, c). Interestingly, while manure induced only a slight increase in soil N₂O emission in U.S. rice paddies (Fig. 10b), it reduced soil N₂O emission by 14.10% in global rice paddies.

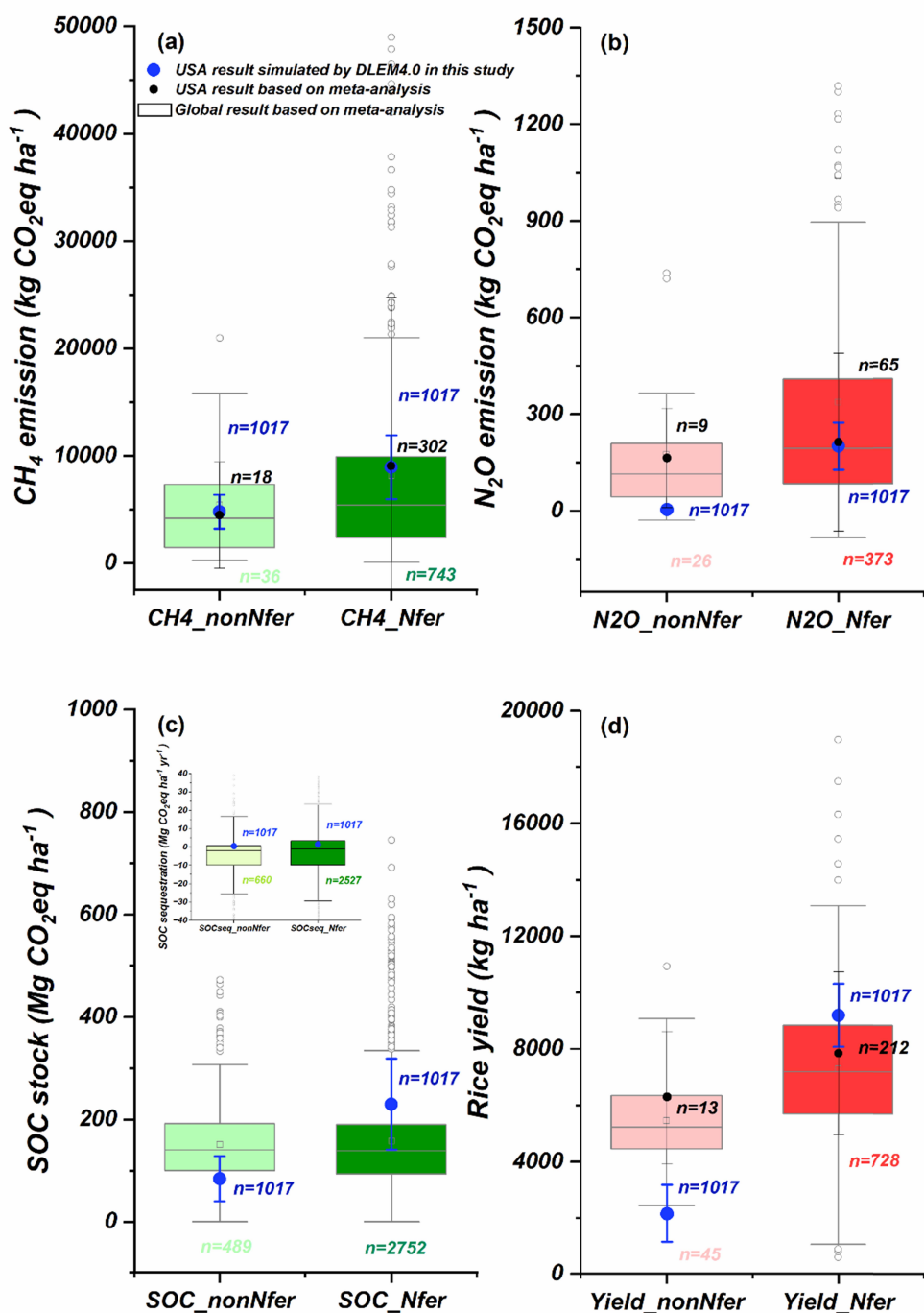


Figure 6. Effects of synthetic nitrogen fertilizer (*Nfer*) application to CH₄ emission, N₂O emission, SOC stock/sequestration, and rice yield in U.S. rice paddies over the study period based on simulation and meta-analysis. Inset in (c) represents the effect of *Nfer* to SOC sequestration. *nonNfer* and *Nfer* represent without and with nitrogen fertilizer use, respectively; *n* is the number of data records.

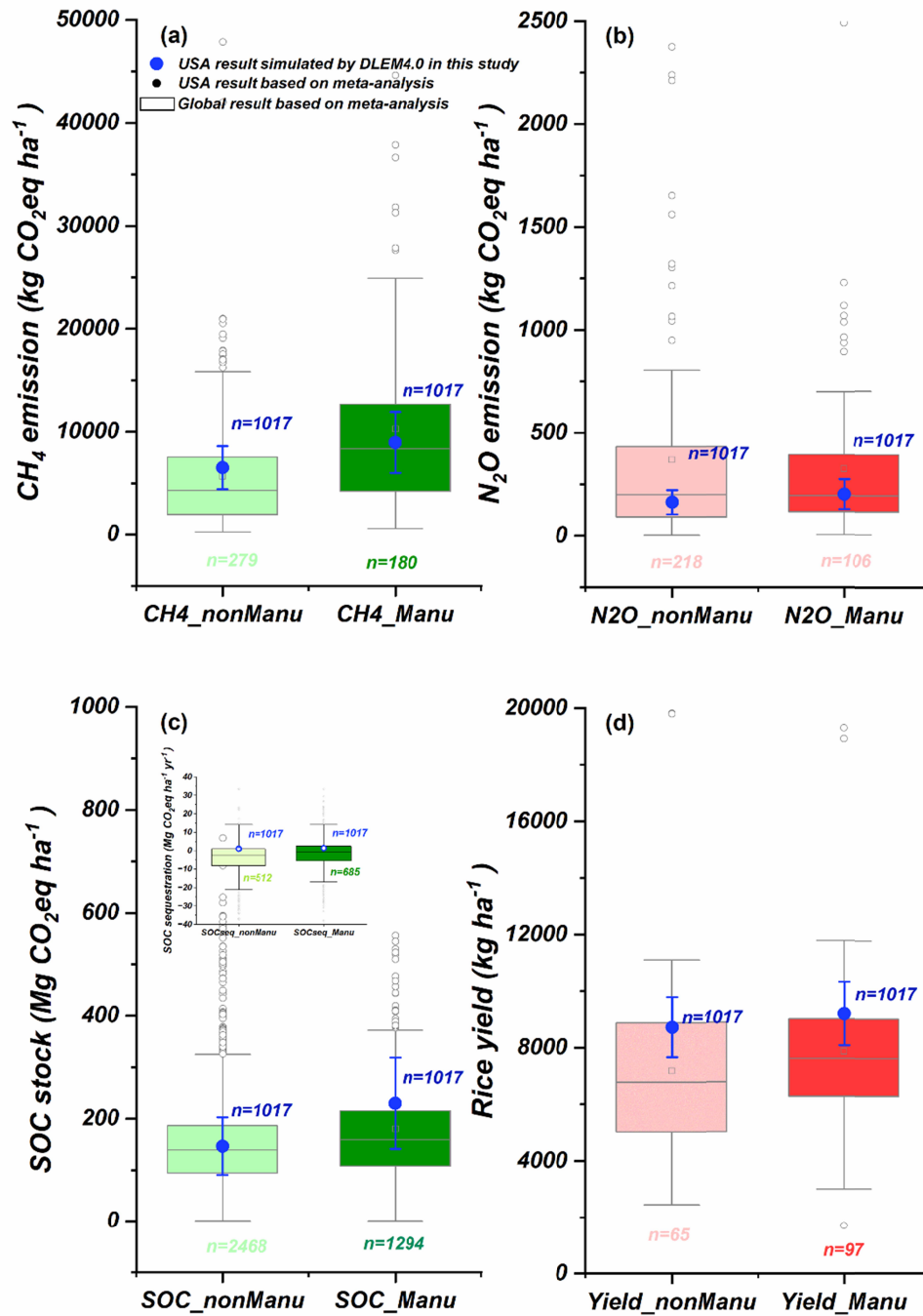


Figure 7. Effects of manure application to CH₄ emission, N₂O emission, SOC stock/sequestration, and rice yield in U.S. rice paddies over the study period based on simulation and meta-analysis. Inset in (c) represents the effect of manure on SOC sequestration. *nonManu* and *Manu* represent without or with manure input, respectively; *n* is the number of data records.

The tillage practices for U.S. rice paddies decreased net soil GHG emissions from 0.08 Tg CO₂eq yr⁻¹ in the 2000s to 0.03 Tg CO₂eq yr⁻¹ in the 2010s (Fig. 4). As a result, the relative contribution of tillage practices to net GHG emission changes steadily decreased from 6.04% in the 2000s to 1.22% in the 2010s. Within this context, tillage practices were associated with a significant decrease of 18.38% (0.04 Tg CO₂eq yr⁻¹) in soil CH₄ emission changes during the 1960s (Fig. 10a). However, this effect diminished notably in subsequent years, leading to a remarkable reduction in emissions. By the 2010s, tillage practices even facilitated a positive transformation, causing a sequestration of 1.83% (0.06 Tg CO₂eq yr⁻¹) in soil CH₄ emission changes. Regarding soil N₂O emission, tillage-induced changes increased slightly to 0.004 Tg CO₂eq yr⁻¹ in the 2010s, contributing to 2.25% of soil N₂O emission changes (Fig. 10b). However, one concerning consequence of increasing the proportion of conventional tillage practices was the continuous aggravation of SOC loss, resulting in 0.08 Tg CO₂-eq yr⁻¹ (6.07%) in soil CO₂ emission in the 2010s (Fig. 10c). On the other hand, compared to conventional tillage, adopting no/reduced tillage methods induced a significant increase of 12.77% in soil CH₄ emission changes in the U.S. rice paddies. This value fell within the range of 6.06% to 45.21% obtained from control trials in Humphreys et al. (2018) and Pittelkow et al. (2014) (Fig. 8a). Nevertheless, in U.S. rice paddies, adopting no/reduced tillage resulted in a substantial decrease of 9.21% in soil N₂O emissions while simultaneously boosting SOC sequestration by an impressive 26.96% when compared to the traditional conventional tillage practices (Fig. 8b, c). Furthermore, it is crucial to highlight that based on the meta-analysis, adopting conventional tillage practices in global rice paddies could potentially result in a considerable annual SOC loss of 7.66±38.03 Mg CO₂eq ha⁻¹ yr⁻¹. In contrast, opting for no/reduced tillage methods could lead to a substantial SOC sequestration of 33.94±26.26 Mg CO₂eq ha⁻¹ yr⁻¹.

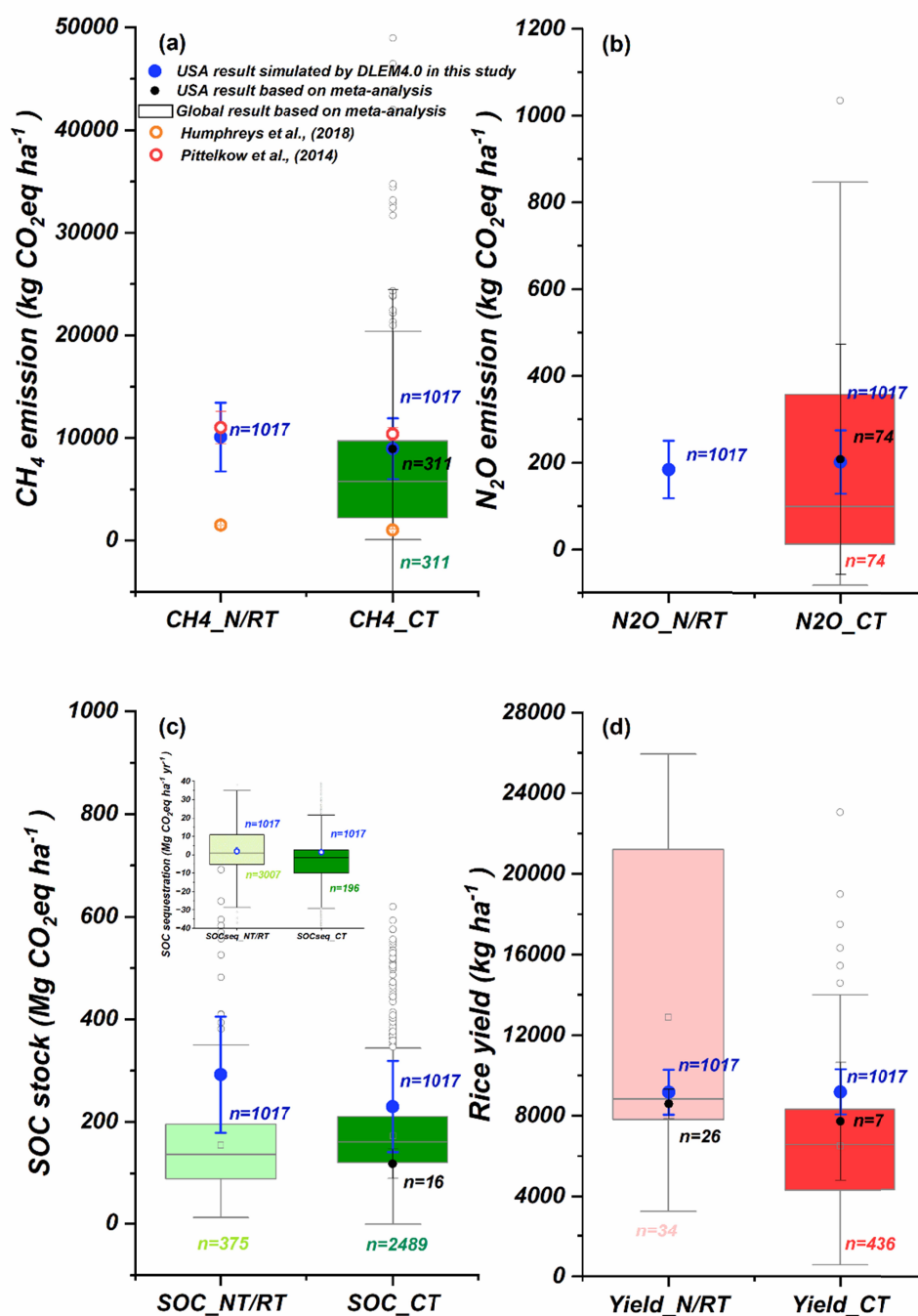


Figure 8. Effects of tillage practices to CH₄ emission, N₂O emission, SOC stock/sequestration, and rice yield in U.S. rice paddies over the study period based on simulation and meta-analysis. Inset in (c) represents the effect of tillage practices to SOC sequestration. NT/RT and CT represent no tillage or reduced tillage and conventional tillage, respectively; *n* is the number of data records.

The importance of irrigation in determining the net soil GHG balance of U.S. rice paddies cannot be overstated. During the 1990s, irrigation contributed approximately 0.37 Tg CO₂eq yr⁻¹ on average, with a slight growth rate of 11.07 Mg CO₂eq yr⁻¹ (Fig. 4). However, due to the decreasing adoption of continuous irrigation practices, the irrigation-induced net GHG emissions of U.S. rice paddies significantly reduced, resulting in irrigation sequestering approximately 0.07 Tg CO₂eq yr⁻¹ in the 2010s (Fig. 4). Continuous irrigation had a notable impact on soil CH₄ emission changes in U.S. rice paddies, steadily increasing to 0.68 Tg CO₂eq yr⁻¹ in the 2010s, roughly explained 19.07% of soil CH₄ emission changes (Fig. 10a). A meta-analysis conducted in this study revealed that non-continuous irrigation in U.S. rice paddies led to a substantial reduction of 57.27% (3668.70 kg CO₂eq ha⁻¹) in soil CH₄ emissions compared to continuous irrigation (Fig. 9a). Furthermore, on a global scale, the adoption of non-continuous irrigation practices resulted in an even more significant decrease of 42.06% (3688.21 kg CO₂eq ha⁻¹) in soil CH₄ emissions (Fig. 9a). In contrast, irrigation played a crucial role in curbing soil N₂O emission changes in U.S. rice paddies, leading to a notable reduction of 7.84% (equivalent to 0.014 Tg CO₂eq yr⁻¹) during the 2010s (Fig. 10b). Globally, non-continuous irrigation practices mitigated soil N₂O emission by an average of 53.40% (equivalent to 144.60 kg CO₂eq ha⁻¹) relative to continuous irrigation, as illustrated in Fig. 9b. On the whole, non-continuous irrigation exhibited the potential to mitigate around 39.2% of soil non-CO₂ GHG emissions on global average. Over the study period, irrigation had a notably enhanced effect on SOC sequestration in U.S. rice paddies, contributing to 0.73 Tg CO₂eq yr⁻¹ in the 2010s, accounting for 46.39% of the total changes in SOC sequestration (Fig. 10c). Other driving factors, such as LULC, CO₂ concentration, and climate change increased net GHG emission at an average rate of 5.01 Tg CO₂eq yr⁻¹ (233.61%), 0.67 Tg CO₂eq yr⁻¹ (31.37%), and 0.06 Tg CO₂eq yr⁻¹ (3.01%) in the

2010s, respectively (Fig. 4). It is noteworthy that climate change led to a 3.8% decrease
 (equivalent to 0.13 Tg CO₂eq yr⁻¹) in soil CH₄ emissions, while simultaneously causing a 20.69%
 increase (equivalent to 0.38 Tg CO₂eq yr⁻¹) in soil N₂O emissions and a 10.17% increase
 (equivalent to 1.16 Tg CO₂eq yr⁻¹) in soil CO₂ emissions during the 2010s.

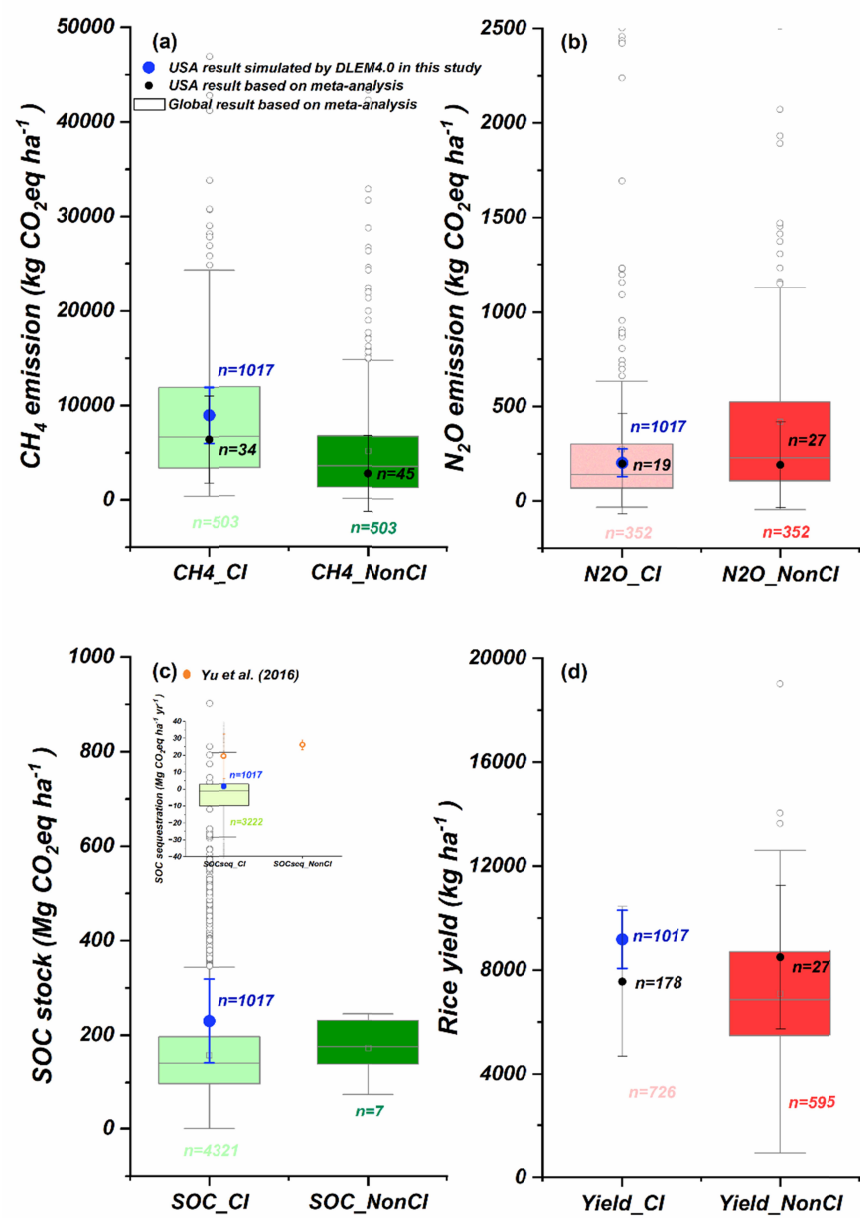


Figure 9. Effects of irrigation on CH₄ emission, N₂O emission, SOC stock, and rice yield in U.S. rice paddies over the study period based on simulation and meta-analysis. Inset in (c) represents the effect of irrigation on SOC sequestration. *nonCI* and *CI* represent no continuous irrigation and continuous irrigation, respectively.

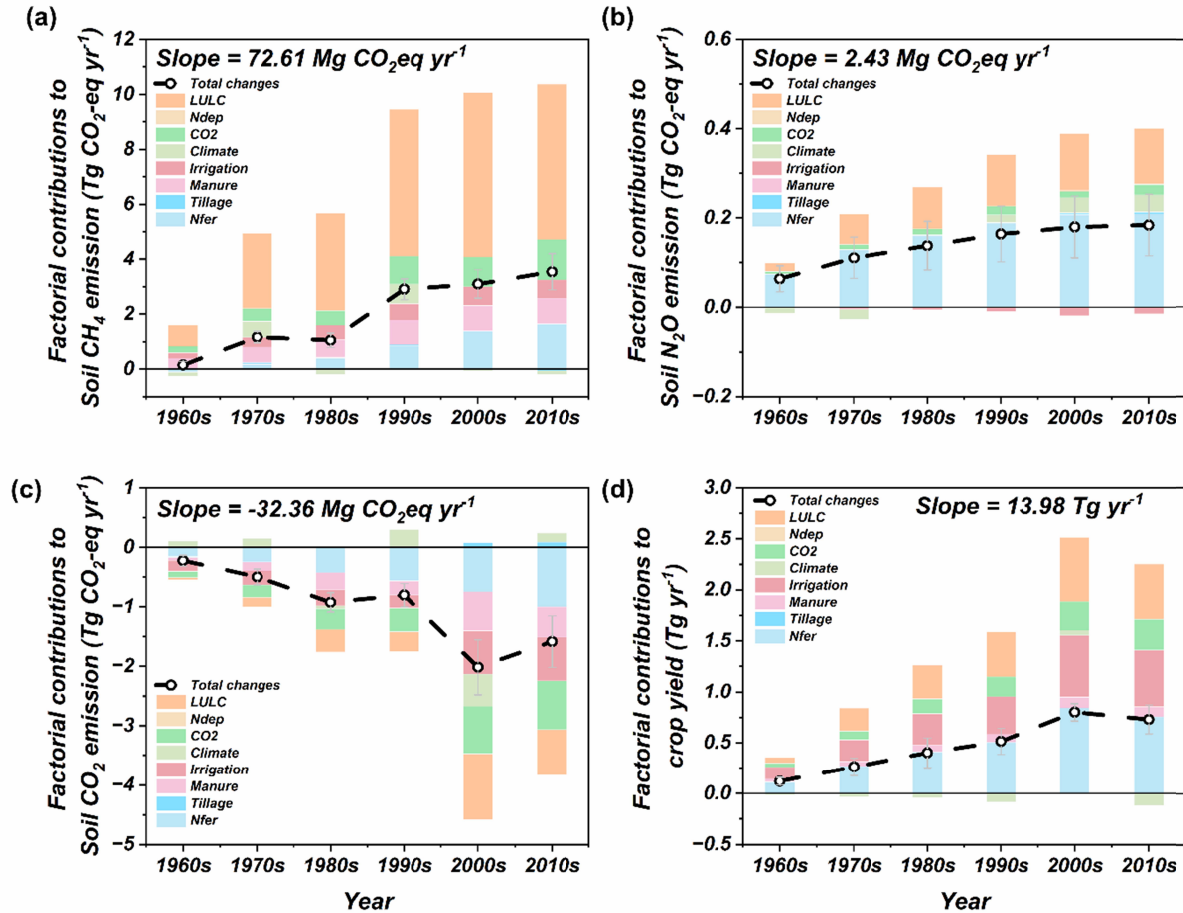


Figure 10. Factorial contributions of multiple agricultural management practices and environmental factors to changes in U.S. rice paddies' net GHG emission and crop yield from the 1960s to the 2010s. *Nfer* represents nitrogen fertilizer use; *Ndep* represents atmospheric nitrogen deposition; *LULC* represents land use and land cover change (reflecting both cropland abandonment and expansion, as well as interannual crop rotation changes); and *CO₂* represents atmospheric carbon dioxide concentration. Note that the sum of factorial contributions of individual drivers (i.e., stacked bars) does not equal net changes in the net GHG balance (i.e., black line) due to interaction effects. Note that error bars in insets represent ± 1 standard deviation of the net GHG balance in each decade.

3.3 Temporal-spatial changes of net GHG emissions intensity in U.S. rice paddies

The enhancement of agricultural management practices across U.S. croplands resulted in a significant increase in rice production to 0.98 ± 0.31 Tg per year (8823.43 ± 1569.82 kg ha⁻¹ yr⁻¹) in the 2010s. This growth equated to a rise of 16.10 Mg per year (107.52 kg ha⁻¹ yr⁻¹) over the study period (Fig. 3d). While slightly lower than the global average (11339.8 ± 4314.65 kg CO₂eq ha⁻¹), these changes underscored the positive impact of various factors on rice production. In this context, the augmentation of N fertilizer application, the expansion of irrigation area, the increase in atmospheric CO₂ concentration, and the incorporation of manure contributed to these rice production changes by 0.75 (103.26%), 0.56 (76.31%), 0.30 (41.57%), and 0.10 (13.52%) Tg ha⁻¹ yr⁻¹, respectively (Fig. 10d). However, the influence of climate change somewhat hindered these production changes by 0.12 (16.48%) Tg ha⁻¹ yr⁻¹. These trends were consistent with both the U.S. and global averages based on the meta-analysis. For instance, N fertilizer led to a 20.03% increase in rice yield in U.S. rice paddies and a 25.23% increase on the global average (Fig. 6d). Furthermore, manure input enhanced rice yield by 8.5% on the global average (Fig. 7d). Comparatively, non-continuous irrigation, as opposed to continuous irrigation, improved U.S. rice yield by 12.08% according to the meta-analysis findings (Fig. 9d).

Achieving increased U.S. rice yields over the past six decades has come with a trade-off of boosting soil GHG emissions. On average, producing a kilogram of grain in the 1960s emitted 1.27 ± 0.38 kg CO₂eq of net soil GHGs. Nonetheless, this intensity exhibited a substantial reduction to 0.84 ± 0.18 kg CO₂eq kg⁻¹ in the 2010s, highlighting a remarkable trend of decline at 0.013 kg CO₂eq kg⁻¹ yr⁻¹. This trend signifies an increasingly efficient rice production process in emitting fewer GHGs. It is crucial to emphasize the mounting concern regarding the intensity of N fertilizer, which escalated from 0.29 kg CO₂eq kg⁻¹ in the 1970s to 1.13 kg CO₂eq kg⁻¹ in the

2010s (Fig. S2). Conversely, the net GHG intensity of manure exhibited a significant reduction, indicating a notably improved emission generation efficiency. The majority of U.S. rice paddies functioned as net sources of GHGs, as depicted in Fig. 11. Regions exhibiting net soil GHG emissions intensities higher than $0.9 \text{ kg CO}_2\text{eq kg}^{-1}$ were predominantly situated in the northeast of the mid-South States, encompassing Arkansas, Louisiana, Mississippi, and Missouri. Conversely, Texas displayed comparatively lower net soil GHG emissions intensities, spanning from $0.30 \text{ kg CO}_2\text{eq kg}^{-1}$ to $0.90 \text{ kg CO}_2\text{eq kg}^{-1}$, as indicated in Fig. 11. However, certain U.S. rice paddies, primarily located in the southeast of the Sacramento Valley region, acted as minor net sinks of GHGs during the production of a kilogram of grain in the 2010s (Fig. 11).

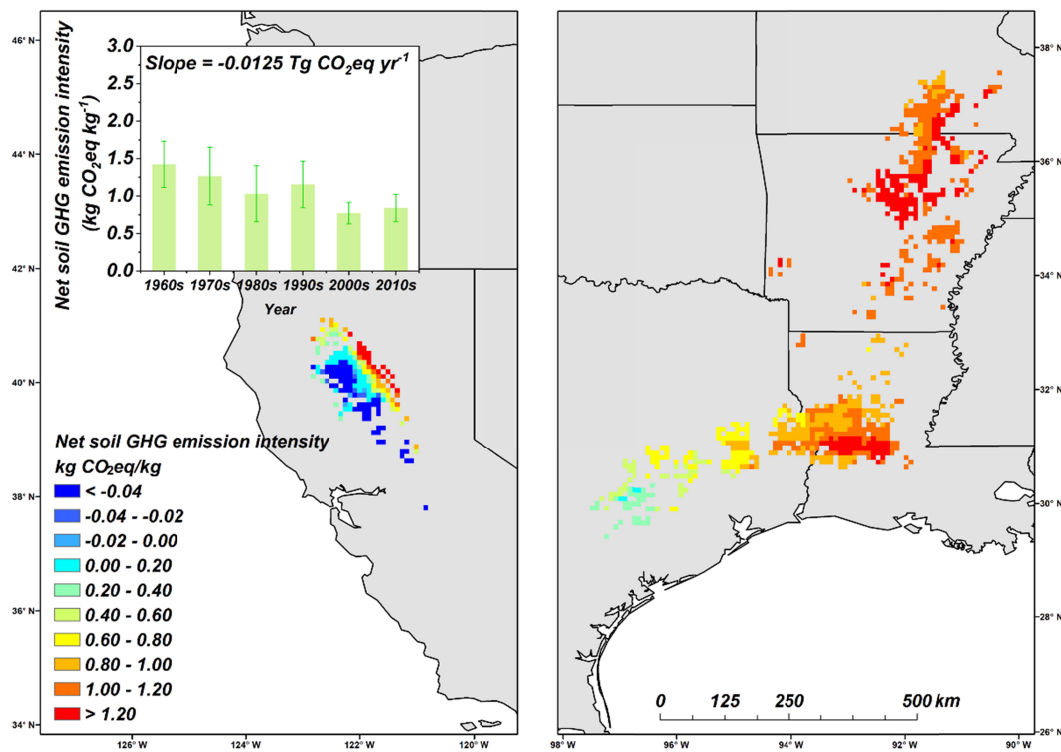


Figure 11. Spatial pattern of average annual net soil GHG emission intensity of U.S. rice paddies in the 2010s. Inset represents U.S. rice paddies' decadal average net soil GHG emission intensity. Note that error bars in insets represent the ± 1 standard deviation of the net GHG emission intensity each decade.

4 Discussion

4.1 Comparison with previous studies

We compared our estimates of SOC stock/sequestration and emissions of N₂O and CH₄ in U.S. rice paddies and similar estimates from various regions (Table 2). Our estimate of CH₄ emissions, 0.28 ± 0.06 Tg C yr⁻¹ or 224.61 ± 40.17 kg C ha⁻¹ yr⁻¹ in the 2010s, was closely aligned with earlier assessments centered around an annual emissions rate of approximately ~ 0.25 Tg C yr⁻¹ or ~ 226.2 kg C ha⁻¹ yr⁻¹ (EPA, 2015; Sass et al., 1999; Tian et al., 2015; Huang et al., 1998; Sass & Fisher Jr., 1999; Linquist et al., 2018). Our estimated N₂O emissions in U.S. rice paddies amounted to 0.42 ± 0.04 kg N ha⁻¹ yr⁻¹ in the 2010s, which was lower than values reported in prior studies (ranging from 1.19 to 8.4 kg N ha⁻¹ yr⁻¹, Mummey et al., 1998; Linquist et al., 2018), but comparable to the outcomes of the meta-analysis in this study (0.48 ± 0.62 kg N ha⁻¹ yr⁻¹). Over the last six decades, our estimated average SOC stock in U.S. rice paddies during the 2010s stood at 54.76 ± 0.98 Mg C ha⁻¹ yr⁻¹, aligning with the reported ranges from WoSIS and previous studies (Rogers et al., 2014; Vasques et al., 2014; Zhong & Xu, 2014; Ruark et al., 2010). On the whole, our estimates of SOC stock and emissions of N₂O and CH₄ in U.S. rice paddies showed similar magnitudes and trends to those of other regional estimates. However, disparities persist, likely attributable to uncertainties in input data and variations in estimation methodologies. For example, the records of N₂O emissions from rice fields in the meta-analysis were generally slightly higher than those indicated by the model. This difference could stem from the inclusion of N₂O emissions induced by non-continuous irrigation in some experiments, whereas our model considers only conventional continuous irrigation. Furthermore, certain experiments captured emissions solely during the growing season and disregarded emissions during the fallow period, a significant peak of N₂O emissions (Linquist et al., 2018), whereas the

539 model results provide a comprehensive annual emission total. By integrating the model with
540 empirical data, we can gain a more accurate comprehension of how agricultural management
541 practices and environmental alterations influence the net soil GHG balance on a continual
542 regional scale (Bondeau et al., 2007; McDermid et al., 2017; You et al., 2022). Thus, this study's
543 approach of integrating the model and data offers a reasonable method for estimating the net soil
544 GHG balance in U.S. rice paddies.

Table 2. Comparisons of SOC stock and N₂O and CH₄ emissions from other studies.

Fluxes	Reported value	Reported region	Time period	Approaches	References
SOC stock (Mg C ha ⁻¹ yr ⁻¹)	20.82	Arkansas	2011-2012	Experiment	Rogers et al., (2014)
	44.21	Florida	2003-2005	Observation	Vasques et al., (2014)
	31.36	Louisiana	2001-2010	STATSGO database	Zhong and Xu, (2014)
	33.55	California	2006-2008	Experiment	Ruark et al., (2010)
	51.57 ± 46.86	Entire U.S. rice paddies	1960-2012	WoSIS	WoSIS
	54.76 ± 0.98	Entire U.S. rice paddies	1981-2018	Process-based model	This study
N ₂ O (kg N ha ⁻¹ yr ⁻¹)	7.6~8.4	Entire U.S. rice paddies	/	the NGAS model	Mummey et al., (1998)
	1.29~2.57	Entire U.S. rice paddies	1980-2013	Meta-analysis	Linguist et al., (2018)
	0.48±0.62	Entire U.S. rice paddies	Until to 2019	Meta-analysis	This study
	0.42±0.04	Entire U.S. rice paddies	1980-2016	Process-based model	This study
CH ₄ (kg C ha ⁻¹ yr ⁻¹)	261.0~394.0	Entire U.S. rice paddies	1980-2013	Meta-analysis	Linguist et al., (2018)
	249.9±121.5	Texas	1991-1995	Meta-analysis	Huang et al., (1998)
	263.3±134.1	Texas	1991-1995	Semi-empirical model	Huang et al., (1998)
	226.2±101.0	Texas	1989-1993	Meta-analysis	Sass et al. 1999
	244.71±425.2	Entire U.S. rice paddies	Until to 2019	Meta-analysis	This study
	224.61±40.17	Entire U.S. rice paddies	1980-2018	Process-based model	This study
CH ₄ (Tg C yr ⁻¹)	0.04 ~ 0.47	U.S. rice paddies	/	IPCC Guidelines	Sass et al. 1999
	0.3	North America croplands	1979-2018	Process-based model	Tian et al. 2015
	0.276	U.S. rice paddies	1990	IPCC Guidelines	EPA 2015
	0.267	U.S. rice paddies	2005	IPCC Guidelines	EPA 2015
	0.255	U.S. rice paddies	2011	IPCC Guidelines	EPA 2015
	0.279	U.S. rice paddies	2012	IPCC Guidelines	EPA 2015
	0.249	U.S. rice paddies	2013	IPCC Guidelines	EPA 2015
	0.28 ± 0.06	Entire U.S. rice paddies	2010s	Process-based model	This study

* Some stations only recorded N₂O emissions during the growth period of rice. In this paper, the annual emissions of these stations were estimated according to the ratio of emissions during the growth period and the fallow period in Linguist et al., (2018).

4.2 Impacts of natural environmental changes on net GHG balance

Changes in natural environmental factors, encompassing climatic conditions, the rise in atmospheric CO₂ concentration, and heightened atmospheric N deposition, exerted significant contributions to the increase of net GHG emissions within U.S. rice paddies across the study period (Fig. 4). Despite notable interannual variability, there exists a positive correlation between soil GHG emission and climate warming at a specific temperature threshold, as evidenced by various studies (e.g., Aben et al., 2017; Griffis et al., 2017). This threshold generally corresponds to increased activity among soil microorganisms, leading to a heightened pace of soil organic matter degradation and release of inorganic nitrogen (Avrahami & Conrad, 2003; Boonjung & Fukai, 1996; Laborte et al., 2012; H. Zhang et al., 2016; Carey et al., 2016; Pärn et al., 2018; Weier et al., 1993; Yvon-Durocher et al., 2014), ultimately exacerbating the flux of soil CH₄, N₂O, and CO₂. Precipitation can directly change soil moisture levels, thereby influencing anaerobic conditions and (de)nitrification by affecting soil oxygen content, which in turn contributes to the production and emission of CH₄ and N₂O (Butterbach-Bahl et al., 2013; Turner et al., 2015; L. Zhang et al., 2010). The oxidation rate of CH₄ in the soil has a critical water content value, which determines its maximum oxidation rate. If the soil moisture content goes above this critical value, the oxidation capacity of CH₄ significantly reduces, leading to a considerable increase in CH₄ emissions (Oh et al., 2020; Gupta et al., 2021; Saunio et al., 2020; Tian et al., 2016). Concerning N₂O emission, the highest levels occur during alternating soil wetting and drying when soil moisture content (water-filled porosity, WFPS) falls within the range of 45% to 75% (Ciarlo et al., 2008; Kuang et al., 2019; H. Liu et al., 2022). Soil water content levels above or below these thresholds can reduce soil oxygen status, which indirectly affects (de)nitrification and soil microorganism activity (Butterbach-Bahl et al., 2013; Turner et

al., 2015), ultimately leading to decreased N₂O emission rates (Dalal et al., 2003; Khalil & Baggs, 2005). Moreover, it's noteworthy that the population status, quantity, and activity of CH₄-producing, CH₄-oxidizing, and (de)nitrification bacteria are significantly impacted by not only the status but also fluctuations in soil water content, thereby profoundly influencing CH₄ and N₂O emissions. For instance, during the initial stages of rice drying, CH₄ emissions don't decrease as soil water content drops; instead, a peak emission occurs. Similarly, during the early flooding stage, considerable N₂O emissions still occur in the soil (G. Tian et al., 2002; Majumdar et al., 2000; Bo et al., 2022). In this study, the ongoing rise in climatic warming and variable precipitation patterns in U.S. rice paddies since 1960 (see Fig. S1) indicate a positive response of net GHG emissions to climate change, contributing to a 20.7% increase in soil N₂O emissions and a 10.2% rise in soil CO₂ release, alongside a 3.8% increase in soil CH₄ emissions during the 2010s. Similar positive responses have been documented in other studies (Liu et al., 2020; Guo et al., 2023). For example, a global meta-analysis by Liu et al. (2020) found that experimental warming of approximately 1.5°C in rice paddies accelerated SOC decomposition by 12.9% and stimulated N₂O emissions by 35.2%.

In our investigation, the enrichment of atmospheric CO₂ concentration has led to a 31.4% increase in net soil GHG emissions annually across global rice fields from the 1960s to the 2010s (Fig. 4). Elevated CO₂ levels are known to promote belowground carbon production, which both improves organic carbon sequestration and provides a heightened substrate for (de)nitrification and methanogens' activity (Allen et al., 2003; Jackson et al., 2009; Pregitzer et al., 2008; Zak et al., 2000). Similar to the findings of others (Shen et al., 2023; Bai et al., 2023), our study found that the enrichment of atmospheric CO₂ concentration improved SOC sequestration by 51.8% but exacerbated 41.5% of soil CH₄ emission. Field observations have corroborated these findings,

demonstrating that rice fields subjected to free-air CO₂ enrichment experiments exhibited significantly higher CH₄ production and N₂O emissions compared to those under ambient conditions (Dijkstra et al., 2012; Inubushi et al., 2003). Chen et al. (2013) identified an increasing trend in CH₄ emissions from rice fields in China attributed to elevated atmospheric CO₂ concentrations. A meta-analysis of data on the effect of elevated CO₂ on CH₄ emissions highlighted that CO₂ enrichment could enhance CH₄ emissions in rice fields by 43.4% (van Groenigen et al., 2011).

During the study period, U.S. nitrogen deposition exhibited an upward trend, increasing at a rate of 0.04 kg N ha⁻¹ yr⁻¹ (Fig. S1). The stimulative impact of N deposition on CH₄ emission in this study is notably constrained within environments characterized by high nitrogen fertilizer levels (Fig. 10a). Similar findings of the positive impact of heightened N deposition on net GHG emissions have been documented in other studies (Xu et al., 2020; Yang et al., 2021). This effect arises from the increased availability of nitrogen, which can foster processes like nitrification and denitrification, consequently leading to heightened N₂O emissions. Additionally, nitrogen addition can bolster crop growth, providing more carbon substrate for microbial activity, thereby stimulating CH₄ emissions and SOC sequestration (Zhang et al., 2016).

4.3 Impacts of agricultural management practices on net GHG balance

Numerous field investigations and meta-analyses have provided evidence that intensified agricultural management practices significantly exacerbate soil GHG emissions in croplands (Cui et al., 2013; Reay et al., 2012; Lu et al., 2021; Davidson et al., 2009; Bai et al., 2019; Dutta et al., 2023; Bo et al., 2022; Gupta et al., 2021). However, these practices also hold the potential to confer advantages for SOC sequestration in croplands due to their substantial mitigation benefits, cost-effectiveness, and additional positive outcomes such as improved soil and water

quality and preservation of biodiversity (Fargione et al., 2018; Li et al., 2022). For example, the increased application of synthetic N fertilizer not only directly supplements nitrogen, thereby contributing to N₂O emissions, but also stimulates higher litter input, increased root biomass, and greater root exudation, providing carbon substrates for methanogens and stimulating CH₄ production (Zhang et al., 2016). In this study, it has been identified as the primary driver promoting non-CO₂ GHG emissions (with a 46.4% increase in CH₄ emissions and a 113.6% increase in N₂O emissions) and enhancing SOC sequestration by 63.2% (Fig. 10a, b). Similar findings have been reported in other studies (Crutzen et al., 2016; Cui et al., 2013; Galloway et al., 2008; Gao et al., 2018; Grassini & Cassman, 2012; Reay et al., 2012; Van Groenigen et al., 2010; Gerber et al., 2016; Lu et al., 2021; Li et al., 2022). Furthermore, excessive application of N fertilizer can lead to detrimental effects on soil structure, resulting in increased bulk density, reduced porosity, altered soil pH, and decreased or imbalanced nutrient content. This can also lead to a reduction in the number of beneficial microorganisms, ultimately resulting in a surge of N₂O and CH₄ emissions and a slowdown or even reversal of SOC sequestration (Liu & Greaver, 2009; Tian, Lu, et al., 2016; Zaehle, Ciais, Friend, & Prieur, 2011; Zhang et al., 2020; Cui et al., 2021). For instance, the application of more than 200 kg ha⁻¹ of N fertilizer induced a 90.4% increase in N₂O emissions in U.S. rice paddies and a 1.97-fold increase globally, while SOC sequestration showed only a marginal increase compared to the 100-200 kg ha⁻¹ N fertilizer application (see Fig. S3). Optimizing N fertilizer use rates is an imminent need for achieving maximum benefits, enhancing SOC sequestration, improving crop yields, and curbing non-CO₂ GHG emissions (Gerber et al., 2016; Xia et al., 2017). In addition to decreasing the amount of N fertilizer applied, changing the timing of N fertilizer application and deep fertilization can also improve N use efficiency and decrease GHG emissions (X. Chen et al., 2014; Cui et al., 2013; Ju

et al., 2009; Xia et al., 2017).

The influence of manure was particularly pronounced in this study, especially concerning CH_4 production (which increased by 26.2%) and SOC sequestration (rising by 32.2%). This effect can be attributed to the introduction of carbon-rich substrates through humus input, which in turn stimulates microbial growth, metabolic processes, and methane-producing microbial activity. Consequently, this leads to a substantial rise in SOC content and CH_4 production (Amon et al., 2001). The carbon-nitrogen ratio present in manure plays a role in shaping N_2O emissions by impacting microbial nitrogen processes, leading to an increase in (de)nitrification (Davidson et al., 2009). However, this contribution is considerably less significant compared to synthetic N fertilizer. For instance, in our study, manure only contributed to a 0.2% increase in soil N_2O emissions in U.S. rice paddies in the 2010s (see Fig. 10b). In comparison to synthetic N fertilizer, manure stimulates microorganisms to assimilate more ammonium nitrogen into the active organic nitrogen pool in the soil. Our study revealed that in soils treated with both synthetic N fertilizer and manure, SOC stock was 9.2% higher than in soils treated with synthetic N fertilizer alone (see Fig. S4). Moreover, it is important to note that regardless of whether synthetic N fertilizer or manure is applied, exceeding the carbon and nitrogen demands of crops and soil microorganisms can lead to a significant decline in the cumulative effect of SOC. For example, in the case of manure application, SOC stock in soils with 100-200 kg N ha⁻¹ increased by 14.2% compared to soils with less than 100 kg N ha⁻¹ of manure application. However, the increase was only 1.4% when manure application levels exceeded 200 kg N ha⁻¹ (Fig. S4).

Our factorial analysis revealed that enhanced tillage practices significantly contributed to an increase in soil CO_2 release by 6.1%, a finding consistent with other studies conducted in the U.S. (Bai et al., 2019; Dutta et al., 2023). This effect can likely be attributed to the fact that tillage

disrupts the soil, accelerating the rate of decomposition of soil organic matter (Mishra et al., 2010; Olson et al., 2014; Salinas-Garcia, Hons, & Matocha, 1997; Bai et al., 2019) and diminishing the biomass of fungi and earthworms (Lavelle, Brussaard, & Hendrix, 1999; Briones & Schmidt, 2017). Consequently, this disruption leads to a reduction in the stabilization of SOC. Furthermore, the disturbance caused by tillage introduces more oxygen into the soil, temporarily altering the anaerobic environment. As a result, CH₄ emissions reduced by 1.8%, while N₂O emissions increased by 2.2% in the 2010s, as observed in this study (see Fig. 10a, b). This insight is also reflected in one of our study's findings, illustrated in Fig. 8. Comparing it to conventional tillage, the adoption of no-tillage or reduced tillage practices yielded an approximately 27% increase in SOC sequestration. However, it also led to a 12.7% increase in CH₄ emissions and a 9.2% reduction in N₂O emissions.

Apart from fertilization, CH₄ emissions in rice paddies are primarily influenced by water management and organic amendments (Nayak et al., 2015; Shang et al., 2011; Wassmann, Neue, Buendia, Corton, & Lu, 2000; Zhang et al. 2016). Conventional continuous irrigation practices intensified soil CH₄ emissions by around 19% but mitigated soil N₂O emissions by approximately 7.8% in U.S. rice paddies during the 2010s (Fig. 10a, b), aligning with similar findings from other studies (Bo et al., 2022; Gupta et al., 2021). A strategy like non-continuous irrigation (e. g. midseason drainage and intermittent irrigation) has been proposed to decrease CH₄ emissions (Cheng, Ogle, Parton, & Pan, 2014; Ma et al., 2013; Nayak et al., 2015; Wassmann et al., 2000; Zhang et al., 2016; Zou et al., 2005) by promoting aerobic soil conditions and reducing CH₄ production from paddy fields by 36%–65% (Ma et al., 2013; Zou et al., 2005; Runkle et al., 2018). However, it's important to note that these measures often involve a trade-off between CH₄ and N₂O emissions (Ma et al., 2013; Wassmann et al., 2000; Zou et al., 2005). For

instance, the reduction in CH₄ emissions through midseason drainage is partly offset by increased N₂O emissions, offsetting 49.2%–67.6% of CH₄ reduction (Zou et al., 2005). Similarly, the impact of non-continuous irrigation can vary widely based on environmental and management factors, as previously documented (Carrijo et al., 2017; Jiang et al., 2019; Liu et al., 2019b). Our comprehensive global meta-analysis further demonstrates that the disparity in CH₄ and N₂O emissions between continuous and non-continuous irrigation practices in rice fields becomes more pronounced with higher synthetic N fertilizer application rates. When the application of synthetic N fertilizer surpasses 200 kg ha⁻¹, the adoption of non-continuous irrigation concurrently leads to a reduction in CH₄ emissions by roughly 48% relative to continuous irrigation, while N₂O emissions experience a corresponding increase of approximately 80% (Fig. S5). This effect of non-continuous irrigation translates to a 90% enhancement in CH₄ emissions mitigation and a substantial 4.2-fold escalation in N₂O emissions compared to scenarios with synthetic N fertilizer application below 100 kg ha⁻¹.

4.4 Uncertainty and future work

We have assessed the uncertainty in the modeled net GHG balance caused by model parameters. However, other sources of uncertainty require attention and improvement to enhance estimates. First, there could be uncertainties introduced by the model forcing datasets. For instance, the crop-specific N fertilization data used to drive DLEM v4.0 was reconstructed from state-level surveys, which may need to accurately reflect the actual spatial variations in fertilizer use in magnitude and timing. Additionally, tillage intensity data are only available for recent decades, which could also introduce uncertainties. Thus, collaborative efforts within the scientific community are vital to improve the quality of model-forcing datasets. Second, under-representing some processes in DLEM v4.0 could also result in simulation biases. For

instance, our model's representation of irrigation practices (without alternate wetting and drying) is relatively simple conventional continuous irrigation, leading to little simulated soil moisture that could impact GHG emission predictions, especially for soil N₂O emission. Last, the lack of spatialized data on cover crop practices could have biased simulation results. Addressing these limitations will lead to more accurate estimates in the estimates in future work.

5 Conclusion

Using a comprehensive model-data integration approach, we conducted a state-of-the-art estimate of the spatiotemporal variations of the net soil GHG emission in U.S. rice paddies from 1960 to 2018. Results indicated that U.S. rice paddy was a growing net GHG emissions source (from 3.73 ± 1.16 Tg CO₂eq yr⁻¹ in the 1960s to 8.88 ± 2.65 Tg CO₂eq yr⁻¹ in the 2010s). Soil CH₄ and N₂O emissions strongly contributed to this growth by about 114% and 2% of total annual net soil GHG emissions in the 2010s, respectively, whereas soil CO₂ uptake limited GHG emission growth by about 16%. The intensification of synthetic N fertilizer usage and the application of manure, coupled with the elevation in atmospheric CO₂ concentration and land use/cover area, emerged as the primary drivers behind the escalation in net soil GHG emissions. These factors significantly outweighed the compensating effect of soil carbon sequestration generated by conventional continuous irrigation practices. Notably, the net soil GHG emissions per unit of grain exhibited a substantial decline, reaching 0.84 ± 0.18 kg CO₂eq kg⁻¹ in the 2010s. This emphasizes an increasingly efficient rice production process marked by reduced GHG emissions. However, it's essential to highlight that the intensity of N fertilizer raised concerns. Our study underscores the potential for optimizing fertilizer efficiency to effectively curtail net GHG emissions per yield, especially when combined with conservation tillage. Nevertheless, addressing the intricate balance between soil CH₄ and N₂O emissions necessitates strategic interventions, such as optimal

intermittent irrigation practices. Striving for a harmonious equilibrium between food security and ecological sustainability, mitigation strategies could concentrate on refining fertilizer applications alongside improved management techniques like conservation tillage and the selection of climate-resilient crop varieties. Such measures have the potential to create synergistic benefits by simultaneously reducing GHG emissions and enhancing overall productivity.

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Data Availability Statement

All data used in this study are publicly available. Daily climate data during the period 1901-2018 derived from the Climate Research Unit-National Center for Environmental Prediction 6-hourly climate datasets is available at <https://rda.ucar.edu/datasets/ds314.3/>. Monthly atmospheric CO₂ concentration data from 1900 to 2018 were obtained from the NOAA GLOBALVIEW-CO₂ dataset derived from atmospheric and ice core measurements at https://gml.noaa.gov/webdata/ccgg/trends/co2/co2_mm_gl.txt. The N fertilizer use maps and the crop-specific N fertilizer use maps reconstructed are publicly available at <https://doi.org/10.1594/PANGAEA.883585> (Cao et al., 2017). The gridded datasets of manure N production and application in the contiguous US are available at <https://doi.org/10.1594/PANGAEA.919937> (Bian et al., 2020). The annual tillage intensity map from 1960 to 2018 was reconstructed from the county-scale tillage practices survey data (1989–2011) obtained from the National Crop Residue Management Survey (CRM) of the Conservation

Technology Information Center at <https://www.ctic.org/CRM>. All result data in this study are publicly available via <https://doi.org/10.6084/m9.figshare.24152160.v1> (Zhang et al., 2023)

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