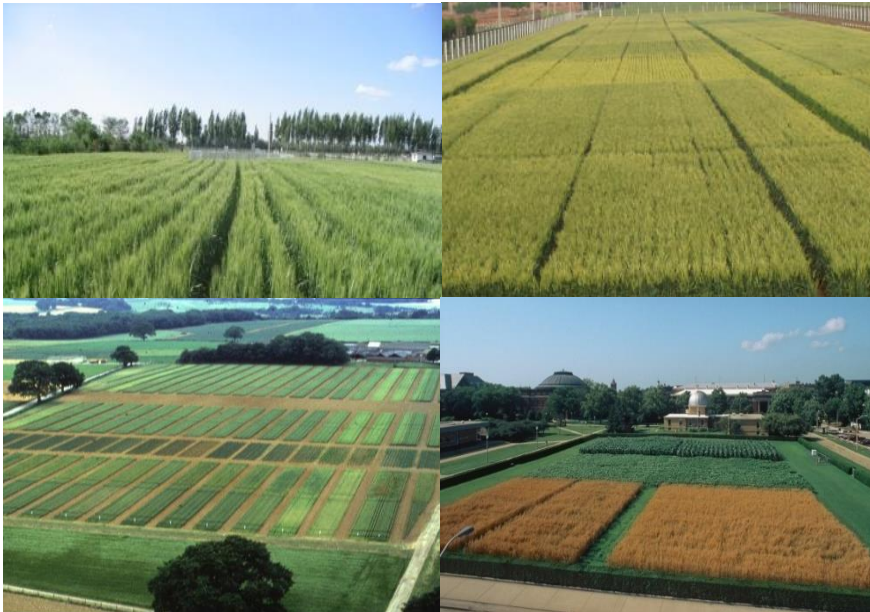


Differences and Simulation of Soil Carbon Sequestration Efficiency of Typical Croplands in China, UK and USA

Shuo Liang



**Promotors: Prof. Gilles Colinet, Prof. Bernard Longdoz
Prof. Minggang Xu (CAAS, China)**

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**Differences and Simulation of Soil Carbon
Sequestration Efficiency of Typical Croplands
in China, UK and USA**

Shuo Liang

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Abstract

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The escalating pressures of food demand and climate change have heightened global interest in sequestering carbon (C) in agricultural soils. Soil organic C (SOC) increases with C input, but the C sequestration efficiency (CSE), i.e., the conversion ratio of C input to SOC, varies with the type and amount of C input. Comparing CSE across regions and/or fertiliser treatments is challenging due to complex interactions between climate, soil properties, and fertiliser practises. Therefore, a comprehensive study of the individual effects of these factors on CSE under long-term fertilisation is urgently needed. Furthermore, limited C inputs do not fully reflect the linear-to-asymptotic behaviour of SOC dynamics, and classical long-term experiments with substantial C input accumulation are essential for understanding soil CSE characteristics. Model predictions are necessary for investigating CSE in soils that are far from C saturation, and future climate change, a pivotal factor affecting crop growth and C cycling, must be considered in these predictions. This research first studied the soil CSE and its key drivers in the plough layer under different fertilisations based on data from eight long-term experiments established in 1980s in China and four classical long-term experiments conducted over a century in the UK and USA. Then, the SPACSYS model’s performance in simulating long-term SOC dynamics in the Broadbalk continuous winter wheat system was validated. The impact of future climate change on wheat yield and SOC stock was also examined. Finally, four long-term experiments with the same soil type (i.e., Mollisol) and similar cropping systems were chosen to compare CSE at different C input accumulation levels between Northeast China and the USA. Fertiliser practises favourable for improving soil CSE and mitigating future climate change were recommended.

The eight long-term experiments in China indicated that the CSE has remained relatively constant over nearly four decades of fertilisation, with SOC linearly increasing with C input. Variance partitioning analysis (VPA) illustrated that the CSE of the main dryland region of China was mostly controlled by edaphic characteristics, especially the soil C/N ratio and clay content, followed by C input and climate. Manure amendment had the most significant fertilisation effect on SOC sequestration with an average CSE of 14.9%, followed by chemical fertilisation (9.0%) and straw return treatment (7.9%). VPA and structural equation modelling (SEM) showed that the improvement of soil nutrients and clay content controlled CSE, highlighting the main positive direct effect of soil chemical properties in the manure amendment treatment. Soil C/N ratio and pH were main explanatory factors influencing CSE in the long-term chemical NPK treatment. The negative impact of C input was the main driver of the CSE under straw return treatment due to the low humification for crop

straw and low stabilization of straw-derived C.

Based on the four classical long-term experiments with SOC showing nonlinear changes over time, the negative exponential dynamics of soil CSE was fully captured by the model ($CSE = b + a * e^{-k * C_{input}}$, $P < 0.01$). The CSE decreased quickly when C input started to pile up, until the accumulated C input reached 123–315 t C ha⁻¹ in typical Chromic Luvisols and 93–145 t C ha⁻¹ in typical Mollisols. Subsequently, it continued to decrease more slowly towards an asymptotic value (i.e., b) as a large amount of C input had been accumulated. Extreme gradient boosted tree (XGBoost) regression modelling showed that C input, MAT and TN stock were crucial factors of soil CSE at all sites with more than a century of fertilisation. Practises with manure application had more significant fertilisation effects on SOC sequestration rates and efficiency than did those with chemical fertilisation. A larger CSE asymptotic value was also observed in the manure applied treatments (2.45–10.25%) than in the chemical fertiliser treatments (0.48–3.85%).

Parametrized by data from the longest and well-documented Broadbalk continuous wheat experiment over a century, the SPACSYS model was capable of simulating the grain and straw yields of various tall and modern short-strawed varieties of winter wheat, as well as the dynamics of SOC and TN stocks in the plough layer. Without changing cultivars and managements, wheat yield could significantly enhance by 5.8–13.5% under future climate conditions compared to the baseline scenario. Meanwhile, the SOC stock was found to increase for all studied fertiliser practises under the scenarios by 0.19–3.99%, except for the NPK fertiliser practises under RCP2.6, which showed a decrease of 0.27–1.08%. Increased C input through “CO₂-fertilisation effects” can compensate C losses by soil respiration under the RCP scenarios. Manure application practises can be considered as a sustainable strategy for enhancing wheat yield and soil C sequestration under future climate change.

Verified by the Harbin and Gongzhuling Mollisols experiments in Northeast China, the SPACSYS model well simulated the yields of corn, spring wheat and soybean, as well as the SOC stock. Without changing cultivars and managements, future climate change, particularly increased temperatures, reduced corn (14.5% for Harbin and 13.3% for Gongzhuling) and soybean yields (10.6%). SOC stocks were projected to decrease by 8.2% for Harbin and 7.6% for Gongzhuling by 2100 under the RCP scenarios. Future climates could also lead to average CSE reductions of 6.3% for Harbin and 12.1% for Gongzhuling. Manure combined with chemical fertiliser is recommended as a fertiliser practise to mitigate negative future climate effects on crop yield and SOC sequestration. Mollisol CSE also showed an exponential decrease with C input in the future. Additionally, the average Mollisol CSE at two sites in the USA (13.6–27.4%) due to manure application for all fields was higher than that in Northeast China (9.2–27.1%) during the rapid decline phase of CSE. During the asymptotically stable stage, the Mollisol CSE at two sites in the USA (0.2–6.8%), with higher temperatures and large C inputs from straw return, was lower compared to that in Northeast China (3.8–10.6%).

In conclusion, soil CSE remained constant across fertiliser treatments when SOC

linearly increased with C input. Edaphic factors were more important CSE controllers than C input and MAT. However, when SOC showed non-linear changes with the accumulation of a large amount of C input, the CSE exponentially decreased and eventually approached an asymptote. The importance of C input and MAT to soil CSE promoted ahead of soil properties. Additionally, manure application was a recommended fertiliser practise not only for improving CSE under long-term fertilisation but also for mitigating adverse future climate impacts on SOC sequestration. These findings are of great significance for guiding sustainable agricultural practises, enhancing C sequestration, and addressing climate change in the study area.

Keywords: soil organic carbon sequestration, long-term experiment, carbon sequestration efficiency, climate change, the SPACSYS model, manure application

Résumé

Shuo Liang (2024). " Différences et Simulation de l'efficacité de la séquestration du carbone dans les sol de terres cultivées typiques en Chine, au Royaume-Uni et aux États-Unis " (thèse de doctorat en Français).

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253 pages, 50 figures, 53 tableaux.

Les pressions croissantes exercées par la demande alimentaire et les changements climatiques ont accru l'importance de la question de la séquestration du carbone dans les sols agricoles. Le carbone organique du sol (SOC) augmente avec l'apport de C, mais l'efficacité de la séquestration du carbone (CSE), c'est-à-dire le rapport de conversion de l'apport de C en SOC, varie selon le type et la quantité d'apport. La comparaison de la CSE entre régions et/ou traitements fertilisants est difficile en raison des interactions complexes entre le climat, les propriétés du sol et les pratiques de fertilisation. Par conséquent, une étude approfondie des effets individuels de ces facteurs sur la CSE sous fertilisation à long terme est nécessaire. En outre, des apports limités de C ne reflètent pas pleinement le comportement linéaire à asymptotique des dynamiques d'évolution du SOC, et les expériences à long-terme classiques incluant des apports importants de C sont utiles pour évaluer les caractéristiques de la CSE du sol. La modélisation prédictive est un outil indispensable d'évaluation de la CSE dans des sols peu saturés en C et spécifiquement dans la perspective des changements climatiques à venir, qui vont affecter la croissance des cultures et le cycle du C. Cette recherche a d'abord étudié la CSE du sol et ses principaux facteurs clés dans l'horizon labouré sous différents fertilisations à partir de données provenant de huit expériences à long-terme établies dans les années 1980 en Chine et de quatre expériences à long-terme menées sur un siècle au Royaume-Uni et aux États-Unis. Ensuite, la performance du modèle SPACSYS dans la simulation des dynamiques à long terme du SOC dans le système de blé hivernal continu de Broadbalk a été validée. L'impact du changement climatique futur sur la récolte de blé et le stock de SOC a également été examiné. Enfin, quatre expériences à long-terme sur sols riches en matières organiques (Mollisol) avec des systèmes de culture similaires ont été choisies pour établir une comparaison de la CSE entre le Nord-Est de la Chine et les États-Unis. Des pratiques de fertilisation favorables pour améliorer la CSE du sol et atténuer le changement climatique futur ont pu être recommandées.

Les huit expériences long-terme en Chine ont montré que la CSE est restée relativement constante au cours de presque quatre décennies de fertilisation, avec une augmentation linéaire du SOC en fonction de l'apport de carbone. L'analyse de partition de variance (VPA) a illustré que la CSE de la principale région aride de Chine était principalement contrôlée par les caractéristiques édaphiques, en particulier le rapport C/N du sol et le contenu en argile, suivies par les apports de carbone et le climat. L'apport de fumier avait l'effet de fertilisation le plus significatif sur la séquestration du SOC avec une moyenne de CSE de 14,9%, suivi par la fertilisation

chimique (9,0%) et le traitement de retour de la paille (7,9%). La VPA et le modèle structurel d'équation (SEM) ont montré que l'amélioration des nutriments du sol et du contenu en argile contrôlait la CSE, mettant en évidence l'effet direct principal et positif des propriétés chimiques du sol dans le traitement d'apport de fumier. Le rapport C/N du sol et le pH étaient les facteurs explicatifs principaux de la CSE dans le traitement de longue durée NPK. L'impact négatif de l'apport de carbone était le principal déterminant de la CSE sous le traitement de retour de la paille en raison de la faible humification de la paille et de la faible stabilisation du carbone provenant de celle-ci.

Sur la base des quatre expériences long-terme montrant des changements non linéaires du SOC au fil du temps, une dynamique négative exponentielle de la CSE du sol a été exprimée par le modèle ($CSE = b + a * e^{-k * C_{input}}$, $P < 0.01$). La CSE a décliné rapidement jusqu'à ce que l'apport de carbone cumulé atteigne 123–315 t C ha⁻¹ dans les Luvisols et 93–145 t C ha⁻¹ dans les Mollisols. Par la suite, elle a continué à décroître plus lentement vers une valeur asymptotique (c'est-à-dire b) car une grande quantité de carbone avait été accumulée. Le modèle de régression par ascension de gradient extrême (XGBoost) a montré que l'apport de carbone, la température annuelle moyenne (MAT) et le stock d'azote total (TN) étaient des facteurs cruciaux de la CSE du sol dans tous les sites où des fertilisations de plus d'un siècle avaient été réalisées. Les pratiques d'application de fumier ont eu des effets plus importants sur les taux et l'efficacité de la séquestration du carbone SOC que les pratiques avec fertilisation chimique. Une valeur asymptotique de CSE plus élevée a également été observée dans les traitements avec application de fumier (2,45–10,25%) que dans les traitements avec fertilisation chimique (0,48–3,85%).

Paramétré à partir de données provenant de l'expérience de longue durée sur du froment de Broadbalk, le modèle SPACSYS a été capable de simuler les rendements de grain et de paille de diverses variétés de blé durant l'hiver, ainsi que les dynamiques des stocks de SOC et de TN dans la couche de labour. Sans modification des variétés cultivées et des modes de gestion, le rendement du blé pourrait augmenter de manière significative de 5,8 à 13,5% sous les conditions climatiques futures par rapport au scénario de référence. Parallèlement, le stock de SOC a augmenté pour toutes les pratiques de fertilisation étudiées sous les scénarios, de 0,19 à 3,99%, à l'exception des pratiques de fertilisation NPK sous RCP2.6, qui ont montré une baisse de 1,08 à 0,27%. L'augmentation de l'apport de carbone par l'effet "fertilisation par CO₂" pourrait compenser les pertes de carbone par respiration du sol sous les scénarios RCP. Les pratiques d'application de fumier peuvent être considérées comme une stratégie durable pour augmenter le rendement du blé et la séquestration de carbone du sol sous les changements climatiques futurs.

Sur les Mollisols de Harbin et de Gongzhuling dans le Nord-Est de la Chine, le modèle SPACSYS a bien simulé les rendements du maïs, du blé de printemps et du soja, ainsi que le stock de SOC. Sans changer les variétés et les modes de gestion, le changement climatique futur, en particulier les températures accrues, aura un impact négatif sur les rendements du maïs (14,5% à Harbin et 13,3% à Gongzhuling) et du soja (10,6%). Le stock de SOC simulé diminuera de 8,2% à Harbin et de 7,6% à

Gongzhuling en 2100 selon les scénarios RCP. Les climats futurs pourraient également entraîner une diminution moyenne de CSE de 6,3% à Harbin et de 12,1% à Gongzhuling. La combinaison d'amendements organiques et d'engrais chimiques est recommandée comme pratique de fertilisation pour atténuer les effets négatifs du changement climatique futur sur les rendements des cultures et la séquestration du carbone dans le sol. La CSE des Mollisols a également montré une diminution exponentielle avec l'apport de carbone dans le futur. De plus, l'efficacité moyenne de la CSE des Mollisols sur deux sites aux États-Unis (13,6-27,4%), en raison de l'application d'engrais organiques sur tous les champs, était plus élevée que celle dans le Nord-Est de la Chine (9,2-27,1%) pendant la phase de déclin rapide de la CSE. Au stade asymptotique, l'efficacité moyenne de la CSE aux États-Unis (0,2-6,8%), avec des températures plus élevées et de grands apports du fumier, était inférieure à celle dans le Nord-Est de la Chine (3,8-10,6%).

En conclusion, la CSE du sol demeure constante lorsque la quantité de SOC augmente de manière linéaire en fonction de l'apport de carbone. Les facteurs pédologiques sont plus importants que l'apport de carbone et la MAT pour contrôler l'efficacité de séquestration du carbone. Cependant, lorsque la quantité de carbone organique présente des changements non linéaires en raison de l'accumulation d'une grande quantité de carbone, l'efficacité de séquestration du carbone décroît exponentiellement et finit par approcher une asymptote. L'importance de l'apport de carbone et de la température moyenne annuelle est alors plus grande que celle des propriétés du sol. Enfin, l'application de fumier est recommandée comme pratique de fertilisation non seulement pour améliorer l'efficacité de séquestration du carbone sous fertilisation à long terme mais aussi pour atténuer les effets néfastes du changement climatique futur sur la séquestration du carbone dans le sol. Ces résultats sont d'une grande importance pour déterminer des pratiques agricoles durables, accroître la séquestration du carbone et répondre au changement climatique dans la zone d'étude.

Mots-clés: séquestration de carbone organique du sol, expérience à long terme, efficacité de la séquestration de carbone, changement climatique, modèle SPACSYS, amendements organiques

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List of Abbreviations

C	Carbon
SOC	Soil organic carbon
OM	Organic matter
CSE	Carbon sequestration efficiency
CSE _{IP}	CSE at the inflection point
CSE _{as}	CSE asymptotic value
CO ₂	Carbon dioxide
UK	United Kingdom
USA	United States of America
SPACSYS	Soil, plant, atmosphere continuum system
VPA	Variance partitioning analysis
C/N ratio	The ratio of Carbon to Nitrogen
SEM	Structural equation modelling
XGBoost	Extreme gradient boosted tree
ANOVA	Analysis of variance
MAT	Mean annual temperature
MAP	Mean annual precipitation
TN	Total nitrogen
GHG	Greenhouse gases
NO	Nitrogen oxide
N ₂ O	Nitrous oxide
NH ₃	Ammonia
CH ₄	Methane
CK	No fertiliser
NP	Combination of chemical nitrogen and phosphorus fertilisers
NPK	Combination of chemical nitrogen, phosphorus, potassium fertilisers
NPKM	Manure combined with chemical nitrogen, phosphorus, and potassium fertilisers
NPM	Manure combined with chemical nitrogen and phosphorus fertilisers
FYM	Manure only
FYMN	Manure combined with chemical nitrogen fertiliser
NPKS	Chemical nitrogen, phosphorus and potassium fertilisers combined with straw return
NPS	Chemical nitrogen and phosphorus fertilisers combined with straw return

NS	Chemical nitrogen fertiliser combined with straw return
RCP	Representative Concentration Pathway
SOC _{ini}	Initial soil organic carbon content
AN	Available nitrogen content
TP	Total phosphorus content
AP	Available phosphorus content
TK	Total potassium content
AK	Available potassium content
BD	Bulk density
GZL	Gongzhuling
CP	Changping
ZZ	Zhengzhou
TJ	Tianjin
XZ	Xuzhou
PL	Pingliang
YL	Yangling
UM	Urumqi
HEB	Harbin
SOC _S	SOC stock
SOC _c	SOC content
SOC _F	SOC stock of fertiliser applied treatment
SOC _{initial-F}	Initial SOC stock of fertiliser applied treatment
SOC _{CK}	SOC stock of CK
SOC _{initial-CK}	Initial SOC stock of CK
H	The depth of soil sampling
C _{input}	Total C input
C _{Stubble}	Carbon input from crop above-ground residual
C _{Root}	Carbon input from crop root
C _{Rhizosphere}	Carbon input from extra-root
C _{Manure}	Carbon input from manure
Y _{grain}	Grain yield
Y _{shoot}	Shoot biomass
Y _{root}	Root biomass
F _{manure}	Manure application amount
P _{stubble}	The ratio of above-ground residual returned to soil
C _{shoot}	Carbon content of shoot
C _{root}	Carbon content of root
C _{manure}	Carbon content of manure
M _{shoot}	The moisture content of the shoot

M_{root}	The moisture content of the root
M_{manure}	The moisture content of manure fertiliser
RSR	The ratio of root to shoot
HI	Harvest index
$C_{sequestration\ rate}$	SOC sequestration rate
Cumulative Cinput _F	Total C input of fertiliser applied treatment during experimental years
Cumulative Cinput _{CK}	Total C input of CK treatment during experimental years
R^2	The coefficient of determination
RMSE	The root mean squared error
RE	The relative error
RPD	Relative percent deviation

Chapter I

General introduction

Abstract

Improved carbon sequestration in agricultural soil is increasingly pursued in recent decades to ensure soil fertility and food security under the pressure of warming climate. The amount of added organic carbon (OC) incorporated into soil C pool is affected not only by the quantity of C input but also by its conversion efficiency (i.e., carbon sequestration efficiency (CSE), $\Delta\text{SOC}/\Delta\text{C}$ input). The CSE varies depending on the amount and type of C input among different studies. Many studies showed that CSE is influenced by climate, edaphic, and fertilisation factors, as well as their complex interactions. A better understand of the main controller of soil CSE is urgently needed. In addition, the CSE, as a dimensionless indicator representing soil C sequestration capacity, has become an important parameter in scientific research on soil C sequestration and soil productivity, especially for comparative analyses. However, the comparison of soil CSE was restricted to the different treatments or cropping systems in the same field or geographical region due to the complexity of influencing factors. Furthermore, the limited range of carbon input levels constrained the observed soil carbon accumulation, and the linear to asymptotic behaviours of SOC may not be evident. Thus, it is better to study the characteristics of soil CSE in relation to C input accumulation based on long-term experiments with different SOC states. A prediction of soil CSE for experiments that are far from SOC saturation or have short fertilisation durations (corresponding to small range of C input levels) is necessary for fully capturing the characteristics of soil CSE dynamics. Future climate change should be considered during the prediction because many researches have emphasized its effects on SOC decomposition and crop yield. In this Chapter, a general review of the definition and estimation of soil CSE under individual treatments or regions, the characteristics of CSE under different long-term fertiliser treatments or systems, and the main influencing factors and associated mechanisms controlling CSE were introduced. The response of SOC sequestration to future climate change under different fertiliser practises was also presented. The problems and knowledge gaps were described, and the research objectives and strategies of the thesis were then presented.

Keywords: soil carbon sequestration, carbon sequestration efficiency, influence factors, long-term experiment, climate change

1. Background

Soil organic carbon (SOC) is one of the largest C pools within terrestrial ecosystems. As such, its content and dynamics directly influence the balance of the global C cycle (Amundson and Biardeau, 2019). The C stored in agricultural soil accounts for approximately 10% (140–170 Pg) of the global terrestrial C storage and is the most active and vulnerable component of the terrestrial C pools (Liang et al., 2016). Over the past 150 years, human activities, including tillage and disturbances, have resulted in the loss of approximately 40–90 Pg of C from the global soil C pool (Houghton, 2002; Blanco-Canqui et al., 2006). However, agricultural soil has the capacity to sequester and store atmospheric CO₂, contributing to the mitigation of global climate change (Lal, 2004; Wang et al., 2022; Zhang et al., 2016a). It has been reported that C sequestration in agricultural ecosystems could offset 5–15% of global fossil fuel C emissions (Lal, 2004). Approximately 90% of the total mitigation potential in agriculture can be achieved through the SOC sequestration (Begum et al., 2017). Moreover, high SOC soils enhance water retention and permeability, mitigating drought stress, and support microbial diversity for nutrient cycling and biological activity (Blanco-Canqui and Lal, 2007; Yadav et al., 2020). Increasing SOC sequestration in agricultural land also contributes to reduction of soil erosion, controls pollution, and improves food quality and safety (Lal, 2004; Wang et al., 2022; Zhang et al., 2016a). Therefore, maintaining and enhancing SOC levels in agricultural soils is crucial for soil quality, food security, and climate change adaptation.

Net SOC sequestration is the dynamic between the C inputs into soil from different sources (via crop residues, rhizosphere, animal manure and straw return) and the decomposition of organic C by soil microbes. Increasing C input is generally considered as the most direct and effective way to enhance SOC content. However, it is not only the quantity of C inputs that matters but also the retention coefficient of these C inputs (i.e., CSE) (Kong et al., 2005; Maillard and Angers, 2014; Six et al., 2002; Stewart et al., 2007; Yan et al., 2013). Different from the index derived from short-term experiments, which reflects the recovery efficiency of organic materials (e.g., humification coefficient), the establishment of CSE under long-term fertilisation integrates C inputs from various sources and represents the cumulative conversion efficiency of C input. It can well reflect the cumulative humification process of different sources of C inputs over a specific period (Gregorich et al., 2017). Furthermore, the CSE, a dimensionless indicator of soil C sequestration characteristics, has become an important index in scientific research on soil C sequestration and soil productivity (Batjes and Sombroek, 1997; Six et al., 2002). It also serves as a suitable measure for regional comparison of SOC sequestration capacity (Liang et al., 2016; Yan et al., 2013; Zhang et al., 2010). Because of the complexity of factors influencing CSE, such as climate, soil type, experimental duration, and field management, it is challenging to identify the predominant factor contributing to variations in CSE. The lack of systematic comparisons of CSE across different regions results in an incomplete understanding of how factors such as soil properties and climate, which vary significantly between regions, impact CSE.

Generally, SOC content increases linearly or asymptotically with C input. The relationship between SOC changes and C input, and hence the CSE, remains constant or varies as C input accumulates. This may be explained by the conceptual model proposed by Six et al. (2002), which described soils approaching C saturation as accumulating a smaller amount of SOC at a slower rate and with reduced efficiency. Therefore, a small range of C input levels (i.e., short fertilisation durations) will not necessarily capture the full range of linear-to-asymptotic behaviours for SOC content when the soil is subjected to C saturation (Stewart et al., 2007). Correspondingly, the full response of the CSE to C input may not be revealed. Therefore, the classical long-term experiments established in the 19th century in Europe and the USA, with soils approaching C equilibrium and receiving a large amount of C inputs (Li et al., 1994; Johnston et al., 2009; Miles and Brown, 2011), are crucial for studying soil CSE characteristics and essential for enhancing soil fertility and C sequestration.

Predicting CSE in soils that have not reached C equilibrium (e.g., the long-term experiments established in 1980s in China) is necessary to fully capture the characteristics of soil CSE in response to C input accumulation in the future. As projected in the IPCC's sixth assessment report by Working Group I, the temperature is expected to increase by 0.2–4.8 °C by from 1995 – 2014 to 2081 – 2100 (Lee et al., 2021). Additionally, there will be a -0.2% to 12.9% increase in precipitation over the same period (Lee et al., 2021). Even with CO₂ removal measures taken into account, the cumulative CO₂ emissions are expected to rise as global surface temperatures increase. The report, however, with high confidence, emphasizes that CO₂ removal is partially offset by CO₂ released from the ocean and land (Canadell et al., 2021; Lee et al., 2021). CSE can be affected by those changes due to their impacts on crop production and soil ecological processes (Liu et al., 2020; Senapati et al., 2019; Tao et al., 2023). By enhancing soil CSE, it is possible to mitigate climate change by reducing the amount of CO₂ released into the atmosphere, as more C is sequestered in the soil (Yan et al., 2013). Thus, it is essential to consider the impacts of future climate change on CSE to ensure the research results are more practical and sounder.

2.Literature review

2.1. Soil carbon sequestration

Soil C sequestration is a process that involves the transfer of CO₂ from the atmosphere into the soil in the form of organic C through natural or anthropogenic interventions. It is a critical component of the C cycle, contributing to the enhancement of SOC stocks, which in turn improves soil structure, fertility, and water retention capacity, benefiting both agricultural productivity and environmental ecology (Chenu et al., 2019; Stockmann et al., 2013). As soil C sequestration intensifies, the SOC content gradually increases, potentially reaching a state of soil C equilibrium. Soil C equilibrium refers to a state where the inputs of C into the soil, such as plant residues and organic matter additions, are balanced by the outputs, including decomposition and erosion losses, resulting in a relatively stable total C

pool within the soil over time, despite the ongoing complex C transformation processes. The soil C saturation concept proposes that there is a maximum equilibrium C level that will be attained when C input is maximized (Stewart et al., 2007). At this point, while the inputs and outputs of C may still be in equilibrium, the soil is unable to sequester additional C, implying that further C inputs are unlikely to lead to a significant increase in soil C content (West and Six, 2007). Consequently, additional C inputs, such as those from fertilisation or the addition of organic materials, may not significantly increase SOC content and could lead to C losses, as any excess C may not be retained in the soil and could be emitted into the atmosphere as greenhouse gases (GHG).

2.2. Soil carbon sequestration efficiency characteristics

Soil CSE is the percentage of the added OC that is converted into the soil C pool after a certain period. In previous research, the humification coefficient was usually used to characterize the conversion efficiency of added organic materials (e.g., straw and manure) (Wolf and Janssen, 1991). The short experimental period (typically 1 year) in related studies limits the representativeness of their results when comparing CSE across different sites. For example, it is difficult to reflect the response of the soil C sequestration ability for added organic materials to climate among different sites (Cai et al., 2018). In addition, the established method might introduce some errors; for example, straw that is too coarse or too fine, respectively, could reduce or increase its contact with microorganisms (Siedt et al., 2021), potentially leading to overestimation or underestimation of the C conversion efficiency.

In recent years, some researchers have attempted to estimate soil CSE through linear or nonlinear regression analysis based on long-term experimental data. Different from the short-term experiment that uses this index to represent the humification coefficient of a specific organic material, the CSE derived from long-term experiments, i.e., the cumulative C input conversion efficiency, reflects the cumulative humification process of different C sources over several years (Cai et al., 2018; Gregorich et al., 2017). Obviously, the soil CSE of long-term experiments can represent the response of the soil C sequestration capability to the long-term average climate condition at a specific study site or region. This makes the CSE an appropriate indicator for evaluating and comparing soil C sequestration capacity across different regions (sites) and/or fertiliser treatments (Yan et al., 2013; Zhang et al., 2010).

There are two methods for calculating soil CSE in long-term experiments with different fertiliser practises. One is by obtaining the percentage of SOC change between the current year and the initial year per unit of cumulative C input under a specific treatment (Liang et al., 2016; Liu et al., 2019; Purakayastha et al., 2008; Stewart et al., 2008; Zhao et al., 2016), and the other is to determine the conversion ratio of SOC change to the difference in carbon input between different fertiliser practises (e.g., different sources of exogenous C) (Hua et al., 2014; Jiang et al., 2018a; Maillard and Angers, 2014). The former method is applicable to soils with SOC showing linear increases over time. It can calculate the soil CSE for an individual treatment and emphasizes the average CSE over a specific experimental period.

However, this method could result in an underestimation of the CSE when the SOC content approaches an equilibrium level due to the positive exponential relationship between SOC content and C input (Cai, 2016; Stewart et al., 2007). In addition, studies using this method also calculate the soil CSE for a region by fitting data on C inputs and SOC stock changes derived from the combination of all fertiliser practises (Zhang et al., 2010, 2016a). The latter method is suitable for soils with SOC showing nonlinear changes over time. It focuses on the CSE caused by the differences in organic material inputs or fertiliser practises. For example, Jiang et al. (2018a) studied the soil CSE caused by manure application by calculating the incorporated ratio of cumulative manure C inputs into SOC stock changes between NPKM and NPK treatments. Furthermore, the former method is only suited for calculating CSE in soils with increasing SOC, while the latter can also be used to estimate CSE in soils experiencing SOC loss, such as Mollisols in some unsustainable agricultural regions (Nafziger and Dunker, 2011). Thus, it is crucial for researchers to select an appropriate method to estimate the CSE for specific study treatments or regions.

2.3. Research progress on soil carbon sequestration efficiency

Based on the aforementioned two methods, studies have been conducted to investigate the CSE in soils with different organic materials, fertiliser treatments, and cropping systems within the same region, as well as the average CSE across different sites or regions. Soil CSE characteristics varied across different studies. A meta-analysis based on 130 observations, considering 4–82 years of manure application, indicated an average global manure-C retention coefficient of $12\% \pm 4\%$ (Maillard and Angers, 2014). Similar values from 8.80% to 10.40% were predicted for the soil CSE of manure application treatments across 20 long-term experiments in China by 2100 (Jiang et al., 2018a). However, a higher value of approximately 23% was found in England, which was derived from 8 sites with 8–144 years of manure application (Bhagal et al., 2007). A 23-year long-term experiment in an Indian Agricultural Research Institute farm with maize–wheat–fodder–cowpea cropping system clarified that both balanced chemical fertiliser (NPK) (21%) and unbalanced chemical fertiliser (NP) (22%) treatments exhibited higher CSE values compared to treatments combining manure with chemical fertilisers (NPKM) (17%) (Dhaliwal et al., 2020). In contrary, compared with chemical fertilisers, a higher CSE was found in manure application treatments at some wheat–corn cropping sites in North China within three-decade fertilisations (Liang et al., 2016). In addition, the type of organic material could influence soil CSE (Liang et al., 2016). A study based on 29 years of long-term fertilisations found that the soil CSE in the treatment combining NPK with wheat straw (17%) was higher than that with pig manure (15%), but lower than that with cattle manure (19%) (Hua et al., 2014). Another study on soil CSE from two 25-year long-term experiments located in the same winter wheat-summer maize planting region of the China Loess Plateau revealed that manure amendments generated a higher CSE (21%) than straw returning (14%) (Wang et al., 2020b).

When considering soil CSE of different sites or regions, previous research has focused on their comparison under region-specific management or cropping systems.

A summary of 17 long-term experiments established across China in 1979 or 1989/1990, with commonly used fertiliser practises in their regions, demonstrated that the CSE values for sites in North China (13.1%) were higher than those in South China (9.3%) (Zhang et al., 2016a). Yan et al. (2013) focused on the CSE in two adjacent agroecosystems with over 30 years of fertilisation and found that, for a given C input, the CSE was higher in paddy soil than in upland soil. At the same site, Liu et al. (2019) discovered that the sum of the CSE from all aggregate components in upland soil (16.02%) was greater than that in paddy soil (15.12%). Based on two 25-year long-term experiments on the China Loess Plateau under irrigated vs. rain-fed conditions, Wang et al. (2020b) confirmed that the CSE was higher under rain-fed (28%) than under irrigated (19%) conditions.

Overall, soil CSE varied greatly among different studies, even in fields with the same fertiliser treatments or organic material applications. Due to the complexity of factors affecting the CSE, the results of different studies are not directly comparable (Liang et al., 2016, 2019). Therefore, comparing soil CSE under a common condition (e.g., the same soil type, the same land use, the same climate region, a similar stage of soil C accumulation, etc.) may make the results more referable.

2.4. Factors influencing carbon sequestration efficiency

The greatly varied CSE mentioned above is a consequence of different dominant influencing factors. Generally, soil CSE is influenced by climate conditions, fertiliser managements, soil properties, and experimental duration.

2.4.1. Climate factors

Many studies have suggested that temperature and precipitation are the key climate factors contributing to soil CSE (Li et al., 1994; Liang et al., 2016; Silver and Miya, 2001; Triberti et al., 2008). These factors mainly affect soil CSE by controlling crop biomass growth, which is related to C input, and by influencing the decomposition of organic C (Aguilera et al., 2018; Liang et al., 2016). Elevated temperatures led to a shorten growing period and a reduced residual C accumulation in soil (Liu et al., 2020; Senapati et al., 2019). More frequent rainfall events decreased the N transfer rate by reducing N content in vegetative organs, which could alter plant physiological processes and lead to yield decline (Asseng et al., 2004). These conditions are not conducive to high C input. Additionally, studies have suggested that the SOC decomposition rate may serve as an external indicator of soil microbial activity (Hararuk et al., 2015; Wieder et al., 2015a). Microbial decomposition of SOC is almost linearly temperature-dependent between 5°C and 30°C, but starts to decrease at 37°C (Wu et al., 2021). Soil moisture changes, driven by temperature, humidity, and precipitation, significantly impact the utilization of OC by microorganisms, primarily by altering soil aeration conditions (Sajedi et al., 2012). Consequently, regions with higher temperatures and greater precipitation tend to experience faster C mineralization, leading to lower CSE, due to the typically enhanced activity of microbial communities under these conditions (Chen et al., 2021). For instance, a study conducted in China has confirmed that the CSE values observed in long-term

experiments (20-30 years) with mild temperatures and semi-humid climates (15.8–31.0%) were significantly higher than those with warm temperatures and semi-humid climates (6.8–7.7%) (Zhang et al., 2010).

2.4.2. Management strategy

Fertilisation stands out as a key management practise on farmlands that significantly impacts soil CSE. By enhancing nutrients availability in the soil, fertilisation can promote both above-ground crop growth and root development, thereby increasing the amount of stubble litter and root exudates (Majumder et al., 2008; West and Six, 2007). Additionally, fertilisation can affect the biomass, structure, and activity of soil microorganisms, consequently influencing the SOC decomposition (Chu et al., 2007; Tang et al., 2017). Although fertiliser management affects soil CSE through both of these pathways, as mentioned above, significant differences are found between chemical fertilisers and organic amendments. When considering chemical fertilisers, (1) long-term chemical N application can reduce the soil C/N ratio, which generally accelerated the mineralization of C sources from humus by soil microorganism and led to the decomposition of OC (Ashraf et al., 2020). (2) The increased N availability affects the C mineralization of exogenous organic materials by influencing the microbial biomass and extracellular enzyme activity, which are involved in microbial C cycling (Jesmin et al., 2021). This effect on CSE is more effective under chemical fertilisation, as manure application is likely to reduce N restriction on crop growth and mitigate subsequent effects on OC turnover (Liang et al. 2016). When considering chemical fertilisers combined with organic materials, the return of straw to the soil influences its CSE as follows: (1) the fresh OC input from straw return stimulates the mineralization of native SOM because of the increase of microbial activity associated with the availability of more easily decomposable compounds, such as phytosterols and policosanol (Kuzyakov et al., 2000). (2) Crop straw is characterised by slow biodegradation, and the C derived from its litter is in low stability (Berhane et al., 2020; Poeplau et al., 2015). These characteristics could lead to a low CSE (Zhao et al., 2016). Manure application is recommended as a sustainable fertiliser practise for improving soil structure and fertility, owing to its beneficial effects from the additional organic matter and nutrient inputs. (Guo et al., 2019; Parham et al., 2003). Manure amendments primarily result in two benefits: (1) a high C input from crop residue and a favourable environment for microbial activity associated with C sequestration (Jiao et al., 2006; Parham et al., 2003), and (2) a significantly increase of macroaggregate component-associated C and aggregation stability by increasing the number of colloids and microbial-derived agents (Jiang et al., 2018b). Compared to other physical fractions, macroaggregate component-associated C has been proved to have a greater CSE (Liu et al., 2019).

2.4.3. Soil properties

Physicochemical properties play an essential role in SOC sequestration. Soil physicochemical properties can restrict the increase of SOC caused by exogenous C addition and determine the maximum C sequestration potential (Six et al., 2002). Generally, the CSE of soil with higher clay content is significantly higher than that of

soil with higher sandy and silty contents, which is attribute the stronger organic–mineral complex adsorbed by soil clay particles (Ekschmitt et al., 2005). Clay soils usually have more SOC storage than sandy soils when the same amount of organic material is added (McConkey et al., 2003). Crop residues decompose more quickly in sandy soils than in clay soils due to the difference in the physical protection of SOC (Ladd et al., 1985; McConkey et al., 2003). In addition, the adsorption strength of the 2:1 interstratified mineral for OC is higher than that of the 1:1 type mineral because of its higher cation exchange capacity and specific surface area (Das et al., 2023). Vertisol and Mollisol soils, rich in clay and dominating the 2:1 type fine smectitic minerals, exhibit higher CSE than Inceptisol and Alfisol soils, which have the lower clay contents and are dominated by 1:1 type clay minerals (Das et al., 2023; Hua et al., 2014). Higher soil bulk density (BD), coupled with poor air permeability and restricted hydraulic conductivity, generally results in lower SOC turnover and a higher CSE (Lu et al., 2018; Thomsen et al., 1999). Moreover, the CSE values for all aggregates are higher than that of the bulk soil (Liu et al., 2019). Macroaggregates and microaggregates exhibited higher CSE due to their greater SOC stock change rates, compared to other physical fractions (Liu et al., 2019). Soil microorganisms play a pivotal role in soil C cycling. Soil pH values that are too low (pH <5.5) or too high (pH >8.5) often restrain the growth and reproduction of soil microorganisms, as well as slow down the decomposition rate of C inputs (Jones et al., 2019; Sun et al., 2019), which is favourable for achieving a higher CSE. For example, a study involving an extensive range of soils (970 samples) and a large geographical area (i.e., 200,000 km²) explicated a critical pH threshold (i.e., pH=5.5) at which the C use efficiency began to decrease (Jones et al., 2019).

2.4.4. Experimental period

The experimental period can be represented by the cumulative amount of exogenous organic C. The relationship between SOC change and C input varies in different studies (Yan et al., 2013). Some reported a linear relation (Bhattacharyya et al., 2010; Liu et al., 2019; Maillard and Angers, 2014; Majumder et al., 2008; Zhang et al., 2010), whereas others found an increasing asymptotic correlation based on long-term experiments (Cai and Qin, 2006; Fan et al., 2005; West and Six, 2007; Yan et al., 2013). Correspondingly, the linearity demonstrated a constant soil CSE (Yan et al., 2013; Zhang et al., 2010). In contrast, the asymptotic relation reflected the decline in CSE over time (Hassink and Whitmore, 1997; Six et al., 2002). The reason may be that the incorporation of added C into SOC could increase in a nearly fixed proportion when the initial SOC level is far from soil C saturation, whereas a phenomenon of ‘diminishing returns’ is observed as the soil approaches C saturation with C inputs. However, this hypothesis has not been extensively tested due to the limitations in either the C inputs or experimental periods.

In summary, despite many research efforts on CSE influencing factors and their underlying mechanisms, the complexity and interactions involved make comparing CSE across regions and/or fertiliser treatments a challenging task (Liang et al., 2016). Hence, a comprehensive study of the individual effects of different types of factors on

CSE is urgently needed, especially under different long-term experimental durations.

2.5. Response of soil carbon sequestration of cropland to fertilisation and climate change

Climate change impacts on the SOC sequestration in agricultural soils is increasingly pursued under the press of global warming (Soussana et al., 2019; Sykes et al., 2020). As projected in the IPCC, future temperatures would increase by 0.3–4.8 °C with increasing precipitation of (1%–3%)/°C by the end of the 21st century (Stocker et al., 2013), and the atmospheric CO₂ concentration would increase to more than 900 ppm if no action was taken to decrease CO₂ emissions (Meinshausen et al., 2011; Nazarenko et al., 2015). These changes may disrupt C cycling, increase water runoff, and soil erosion, and thus result in adverse effects on crop production, soil fertility, and the environment (Amelung et al., 2020; Knops et al., 2007). Notably, the soil C sequestration capacity under different fertiliser treatments would respond differently to future climate change related threats. An analysis of SOC stock changes in Bavaria indicated that no fertiliser input would decrease SOC stocks by 11–16% in the end of the 21st century (Wiesmeier et al., 2016). Similarly, in North China, SOC stocks in cropland surface soils were predicted to decrease by 6.6–17.8% by the 2080s compared with the 1980s without additional organic materials (Wan et al., 2011). However, if appropriate amounts of manure and stubble were applied, their surface soils across more than 90% of North China's cropland would gain a net C sink, and the average SOC stocks would increase by ~58% by the 2060s (Wang et al., 2014). The crop stubble conservation practise would increase the SOC storage in Ohio's cropland in the USA at a rate of 42.0 kg C ha⁻¹ yr⁻¹ by 2100, whereas the practise without crop straw input would increase it at a rate of 24.6 kg C ha⁻¹ yr⁻¹ (Evrendilek and Wali, 2004). Although manure amendment and straw return are two effective methods for improving SOC content, they can lead to increased potential N loss through leaching and runoff (Tadesse et al., 2013). In addition, terrestrial C and N losses, via CH₄ and N₂O emissions, would increase by approximately 80% and 45%, respectively, resulting in terrestrial lands becoming a net C source globally by 2100 (Stocker et al., 2013). These results highlight the importance of identifying an optimal fertiliser strategy to increase C sequestration in croplands and mitigate the risks of climate change in the future.

2.6. Importance of long-term experiments for studying soil carbon sequestration characteristics

Long-term experiments are typically initiated to evaluate the impact of fertiliser management practises on crop production, guiding the selection of the best fertiliser type and amount for a region over a set period (Johnston and Poulton, 2018; Macdonald et al., 2018). If any treatment fails to maintain or increase yields, reflective adjustments can be made to ensure sustainable crop production (Johnston and Matingly, 1976). As society and technology evolve, long-term experiments not only continue to assess the sustainability of farming systems for maintaining high productivity but also contribute to a range of other applications. They can be

summarized as follows: (1) providing field resources for studies on plant growth and soil properties, in particular for characteristics that develop slowly (e.g., soil fertility); (2) archiving soil and plant samples that are invaluable sources of materials for further research; and (3) developing models to describe the cycling and processes of nutrients among soil, plant, and atmosphere, informed by detailed management records and datasets from both current and archived samples.

Many long-term field experiments have been established for above aims worldwide, with the majority of them being situated in Europe and North America. Notable examples include a series of classical long-term experiments with more than a century of fertilisations located in Rothamsted and Woburn (UK), Uppsala (Sweden), Grignon (France), Versailles (France), Bad Lauchstädt (Germany), Askov (Denmark), Morrow (USA) and Sanborn Field (USA) (Johnston and Poulton, 2018; Lefèvre et al., 2014). In addition, sites with shorter experimental durations have been established in China since 1979 and 1989/1990, and in Australia since 1982 (Jiang et al., 2014; Latta and O’Leary, 2003), but there are fewer sites in South America and Africa (Beneduzi et al., 2019; Fynn et al., 2004).

However, many of these sites may not meet our research requirements for nonlinear SOC dynamics, which are characterized by a large amount of C inputs, generally indicating an approach towards SOC equilibrium or saturation. The Broadbalk and Hoosfield experiments at Rothamsted Research in the UK, as well as the Sanborn fields and Morrow plots in the USA, are the oldest and most representative long-term studies in Europe and the United States, respectively (Johnston and Mattingly, 1976). They may fulfil our research requirements (Li et al., 1994; Johnston et al., 2009; Miles and Brown, 2011). Furthermore, when considering the long-term experiments in China and Australia, the representativeness of their C sequestration characteristics is limited with a relatively short period (30–40 years). Model predictions can be used to study the full characteristics of soil CSE in response to C input accumulation, based on the fact that a steady-state soil C content is reached over a duration of 50 to 100 years (Mosier, 1998; Rasmussen et al., 1998). Therefore, an analysis of the four classical long-term experiments, along with processes-based model predictions for experiments with relatively short durations, is necessary to fully reflect the response of soil CSE to C input accumulation. These also correspond to the uses (1) and (2) and use (3) of long-term experiments mentioned in the preceding paragraph, respectively.

2.7. Mollisol (black soil) long-term experiments

Mollisol is the common soil type in both the classical long-term experiments in the UK and USA and those conducted in China, including Harbin, Gongzhuling, Morrow, and Sanborn long-term experiments. When considering the comparison of the soil CSE in different regions, this study focused on those Mollisol sites. These soils, characterized by their thick, dark, and well-structured surface horizon, are globally recognized as the most fertile and productive soil (Durán et al., 2011; Liu et al., 2012; Xu et al., 2020). They are primarily distributed in four regions: the majority of Russia and Ukraine (45%), the central plains of North America (29%), Northeast China

(10%), and a significant portion of South America (10%) (Liu et al., 2012; Xu et al., 2020).

The C sequestration of Mollisols is of great significance for food security and climate change mitigation. The high productivity of Mollisols, due to their fertile soil conditions and suitable climate, forms the foundation for global food supply. Those regions are major producers of wheat, corn, and other staple crops, such as contributing to 43% of China's corn, 30% of soybean, and 8% of wheat production (Xu et al., 2020), as well as nearly half of Russia's wheat and a quarter of its barley output (Romanenkov et al. 2019). Additionally, the high C sequestration capacity of Mollisols helps to reduce the concentration of CO₂ in the atmosphere, thus mitigating the effects of climate change. A fifteen-year experiment found that chemical protection by mineral association within macroaggregates and non-aggregated silt + clay-sized fractions, as well as physical protection by the occlusion of particulate organic matter within free microaggregates, contributes to SOC stabilization under a long-term warming-dominated climate change in Mollisols (Zhou et al., 2023). However, this positive effect typically occurs under suitable fertilisation or management practises, such as conservation tillage and manure application (Durán et al., 2011; Liu et al., 2012; Xu et al., 2020).

Notably, a significant depletion of C stocks in Mollisols has occurred during the last several decades because of active agricultural utilization, low organic return, and intensive tillage (Durán et al., 2011; Liu et al., 2012; Xu et al., 2020). Reports indicated Mollisols in Russia and Ukraine have lost approximately 15–40% of their original organic C pool, while those in America and Northeast China have seen a decline of around 50% (Gollany et al., 2011; Xu et al., 2020). This accelerated land degradation is often accompanied by decreases in crop yield. For example, crop yields in North America have declined by 20–40% over the past century, as a result of unsustainable cultivation practises (Posner et al., 2008). Similarly, Mollisols in Northeast China have experienced a decrease in thickness of approximately 30–50 cm, with each 1 cm reduction corresponding to a 2% reduction in crop yield (Liu et al., 2013a). The declining SOC not only negatively impacts soil productivity but also becomes an important factor for climate change due to GHG emissions (Tong et al., 2017). For instance, the average C release rate from Mollisols in Northeast China was 2 to 4 times the global average rate from the soil pedological C pool (Lal et al., 2007; Li et al., 2013). Therefore, maintaining and enhancing the C sequestration of Mollisols is of strategic importance for achieving food security and addressing climate change.

2.8. SPACSYS model introduction and application

The SPACSYS (Soil-Plant-Atmosphere-Continuum-SYStem) model is a multi-dimensional, field-scale, weather-driven, flexible time step (from minute up to daily), and process-based model that quantifies the biogeochemical processes of C, N, and P cycling, and the water and heat budgets in soil, plant and ruminant animal. The description of soil C and N processes is similar to that in a number of existing models (Wu and McGechan, 2001; Wu et al., 2007), but it offers more detail in relation to nutrient cycling from decaying root material. The water, heat, and soil N components

are conceptualized as two-dimensional submodels, with the water component incorporating a horizontal water flow that influences the movement of heat and nitrate. The soil C cycling is treated as a one-dimensional component. The state variables within a soil layer that are influenced by root systems are determined by considering the values of each root segment within that soil layer. The processes related to the C and N cycles and water movement in the SPACSYS model are presented in Figures 1-1, 1-2, and 1-3.

In the SPACSYS model, the plant growth module includes plant development, assimilation, respiration, and the allocation of photosynthates and plant N uptake. The soil organic C and N pools are composed of five sub-pools: the fresh litter pool, the microbial pool, the humus pool, the dissolved organic matter pool, the fresh OM pool (if manure was applied). The primary processes affecting the soluble N pool are mineralization, nitrification, denitrification, and plant N uptake. Additionally, the transfer of N from the organic matter pool to the microbial pool and the conversion processes between organic matter pools are directly related to C transformations, and the estimates of N transformation destinations are based on corresponding C fluxes and C/N ratios (Wu et al., 2007).

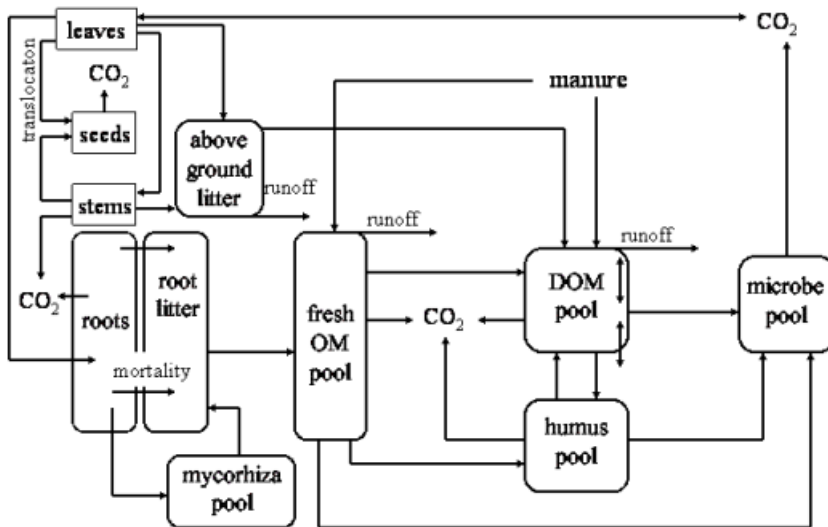


Figure 1-1 Conceptual diagram of C cycling in the model (from SPACSYS manual 6.0).

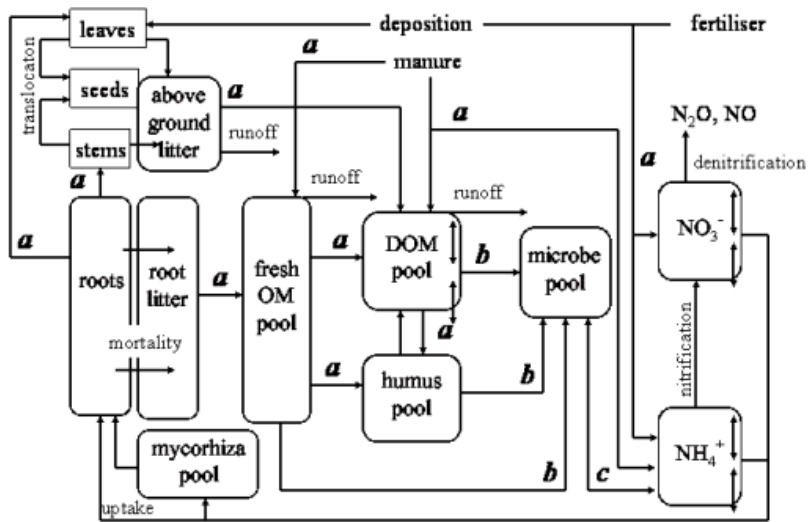


Figure 1-2 Conceptual diagram of N cycling in the model. a represents partition of decomposed organic matter; b represents the decomposition process and c represents the mineralization/immobilization process (from SPACSYS manual 6.0).

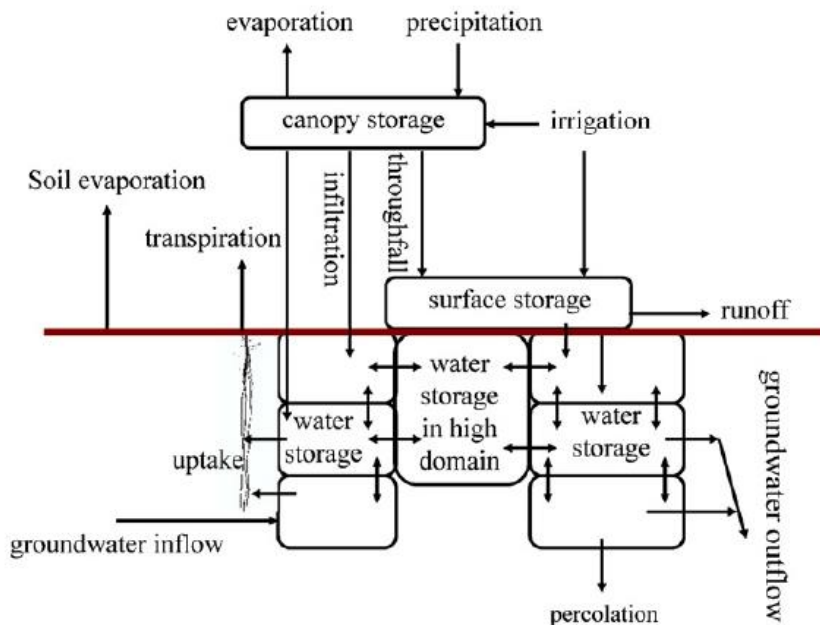


Figure 1-3 Conceptual diagram of water movement in the model (from SPACSYS manual 6.0).

Table 1-1 Comparison of involved processes between the SPACSYS model with other models (modified from Wu et al., 2007; Brilli et al., 2017; Lutz et al., 2019; Wowra et al., 2021)

		SPACSYS	DAISY	APSIM	DAYCENT	DNDC	EPIC
	SOM pools number	5	5	4	4	3	3
	Microbial biomass	√	√	√	√	√	√
	Humus	√	√	√	√	√	√
	DOC	√	√	-	√	√	√
	DON	√	√	-	-	√	√
<i>Main processes explicitly simulated</i>							
	Mineral fertiliser	√	√	√	√	√	√
	Organic fertiliser	√	√	√	√	√	√
Management	Irrigation	√	√	√	√	√	√
	Tillage	√	√	√	√	√	√
	Sowing	√	√	√	√	√	√
	Harvest/cutting	√	√	√	√	√	√
	Decomposition	√	√	√	√	√	√
	Mineralization-immobilization	√	√	√	√	√	√
	Nitrification	√	√	√	√	√	√
C, N processes	Denitrification	√	√	√	√	√	√
	Nitrate leaching	√	√	√	√	√	√
	NH ₄ ⁺ leaching	√	√	√	-	-	-
	NO ₃ ⁻ surface loss	√	-	√	-	-	√
	Volatilization	√	-	-	-	√	√
Heat transfer		√	√	-	-	√	√
	Photosynthesis	√	√	√	√	√	√
	Respiration	√	√	√	√	√	-
Plant growth	Phenological stage	√	√	√	√	√	√
	Partitioning	√	√	√	√	√	-
	Translocation	√	-	√	-	√	-

	Root architecture	√	√	√	√	√	-
	Water uptake	√	√	√	√	√	√
	Nitrogen uptake	√	√	√	√	√	√
	Nitrogen fixation	√	-	√	√	-	√
	Infiltration	√	√	√	√	√	√
Water cycling	Ground water	√	√	√	√	√	√
	Evapotranspiration	√	√	√	√	√	√
	Erosion	-	-	√	√	√	√

Numerous models that simulate plant growth and development, C and N cycles, as well as water and heat transfers, have been widely applied, such as APSIM, DNDC, DAISY, DAYCENT and EPIC. Among these, the APSIM model is capable of simulating plant growth, soil water, and C and N cycling, and it has good performance in simulating crop yields under extreme climate conditions (Holzworth et al., 2014; Ebrahimi-Mollabashi et al., 2019). However, it doesn't include the DOC and DON pools. The DNDC model can simulate soil temperature, moisture, pH, redox potential, and the emissions of NO, N₂O and NH₃ associated with nitrification and denitrification processes, but it considers a smaller number of SOM pools (Gilhespy et al., 2014). DAISY is mainly used for simulating water and C and N cycling, and it can be used to simulate crop growth and grain yield, as well as N mineralization, nitrification, denitrification, and crop N uptake, but it does not consider ammonia volatilization (Hansen et al., 2012; Gyldengren et al., 2020). Therefore, SPACSYS demonstrates significant advantages over other extensively cited simulation models in terms of the processes and number of pools considered for C and N cycling (Table 1-1). Additionally, the SPACSYS model has advantages in simulating crop growth and the movement of water and heat. For instance, the DAYCENT, EPIC and DAISY models are less detailed in their simulation of crop growth and development processes than SPACSYS and APSIM, although they have been proved to accurately simulate crop yields (Stehfest et al., 2007; Timlin et al., 2024). SPACSYS also simulates N fixation, a process that is not accounted for in models such as DAISY and DNDC. Most notably, it can simulate root architecture, providing a 3D root structure. A well-simulated root architecture can more clearly reflect the absorption of nutrients and water from the soil by the roots, as well as the transport processes to the aboveground and belowground parts of the plant, which are particularly important for plant growth (Bingham and Wu, 2011).

Since the development of the SPACSYS model in 1996, it has been proven to have a strong ability to accurately simulate plant growth, N uptake, SOC and TN stocks, and CO₂, CH₄ and N₂O emissions of cropland and grassland across Europe and China. For example, Wu et al. (2007) validated the model's accuracy in simulating spring barley grain yield and N uptake, as well as cumulative drainage flows and N leaching in an arable land at Bush Estate near Edinburgh, highlighting that SPACSYS's detailed root architecture description enhanced its performance in estimating N cycling over

SOIL-SOILN. Wu et al. (2015) validated a new microbial growth-based module for both nitrification and denitrification, and proved SPACSYS's accuracy in simulating soil moisture, aboveground biomass, N uptake, and N₂O emissions in a Scottish grassland. Li et al. (2017) confirmed the applicability of SPACSYS for simulating soil moisture, the C and N balance in a reseeded grassland in southeast England. Perego et al. (2016) studied the effects of tillage and different fertilisations on N₂O emissions from a rotation of forage and bioenergy crops in the Povalley of northern Italy. Zhang et al. (2016b; 2016c; 2018) and Liang et al. (2018) used SPACSYS to validate wheat and corn yields, SOC and TN stocks, and GHG emissions in six long-term experiments in China. Wu et al. (2020) investigated the responses to different management practises in seed yield, biological N fixation, protein yield and soil N budgets based on a four-year soybean cropping system in Northeast China. Liu et al. (2023) evaluated the utility of measured saturated soil hydraulic conductivity to enhance SPACSYS's accuracy in simulating water and soil mineral N content for a lowland UK grazed field. Clearly, the SPACSYS model has a strong foundation in nutrient cycling within the soil-plant-atmosphere system, demonstrating robust simulation results and the capability to predict different crop yields and SOC stocks under various climate change scenarios and fertiliser treatments.

3. Scientific question and objectives

This study aims to address the following question: What are the differences in soil carbon sequestration efficiency among typical croplands in China, the United Kingdom, and the United States under long-term different fertilisations? The general objectives of this study are to reveal the characteristics of soil CSE in response to different C input levels and to identify the controlling factors of these CSE characteristics based on long-term experiments with typical fertilisation and soil types in China, UK and USA. Meanwhile, fertilisation practises that are favourable to improve SOC stocks and efficiency are recommended.

The technology roadmap of our research is shown in Figure 1-1. First, through eight dryland long-term experiments with nearly four decades of fertilisation, the soil CSE characteristics of the plough layer and their main controllers were studied (red colour, Chapter II). Second, based on four classical long-term experiments with more than a century of fertilisation and a large amount of carbon input accumulation, the CSE of the plough layer, its relation with C input, and the main influencing factors were confirmed (blue colour, Chapter III). Then, the SPACSYS model was validated for its performance in simulating the long-term dynamics of SOC stock using data from the longest and well-documented continuous Broadbalk wheat system in southeastern England. Based on this validation, a sustainable strategy for enhancing carbon sequestration under future climate change was determined (orange colour, Chapter IV). Finally, SPACSYS predicted the future response of CSE in two typical Mollisols in Northeast China to climate change. The characteristics of their CSE in response to C input accumulation in the future were also studied (green colour, Chapter V). A comparison of the CSE between the two Mollisol sites in Northeast China and the two

Mollisol sites in the USA was conducted in the general discussion to highlight the regional differences in CSE.

In this study, we assumed a uniform conversion efficiency of C input to SOC within the targeted soil layer. This simplifying assumption allows for the estimation of an average CSE, although it may not account for the potential spatial and temporal heterogeneity in the actual SOC sequestration process. Additionally, the study focuses mainly on the plough layer, which accumulates the majority of carbon inputs and is directly susceptible to human activities and climate change. The CSE of the subsurface soil during long-term fertilisation is constrained by the availability of experimental data and the challenge of quantifying organic C inputs distributed across different soil layers.

To expound on the main objectives above, the specific questions were formulated:

1. What are the characteristics of soil CSE in the plough layer under different fertilisations based on long-term experiments with different durations?

Hypothesis: Soil organic carbon exhibits linear-to-asymptotic behaviours as carbon input increase. The linear relation indicates that the soil has constant C sequestration efficiency and no C equilibrium level. In contrast, the asymptotic relation emphasizes a decrease of C sequestration efficiency when the SOC approaches the equilibrium level.

2. Is the SPACSYS model appropriate for simulating the long-term dynamics of soil organic carbon stocks in the Broadbalk long-term experiment under different fertiliser treatments?

Hypothesis: Based on the model's detailed description of crop growth and C and N cycling within the soil-plant-atmosphere system, the SPACSYS model can well simulate the SOC dynamics of the Broadbalk wheat continuous cropping system for more than a century.

3. What are the differences in soil CSE between Mollisol long-term experiments in Northeast China and those in the US?

Hypothesis: Due to manure application during the first five decades of the Sanborn and Morrow experiments in the US, the CSE was higher than that in the Gongzhuling and Harbin experiments in Northeast China during the rapid decline phase of CSE. However, during the asymptotically stable stage, the Mollisol CSE in the Sanborn and Morrow experiments, which had warmer climates and received substantial C input from straw return in all plots, was lower than that in the Harbin and Gongzhuling experiments.

4. What is the recommend fertiliser practise that is favourable to improve soil CSE and mitigate future climate change?

Hypothesis: Because of the improved soil nutrient environment and the enhanced macro-aggregate components associated with higher CSE through the direct fresh organic matter input, manure application is a recommended fertiliser practise for enhancing CSE under long-term fertilisation and mitigating negative climate effects on crop yield and SOC sequestration in the future.

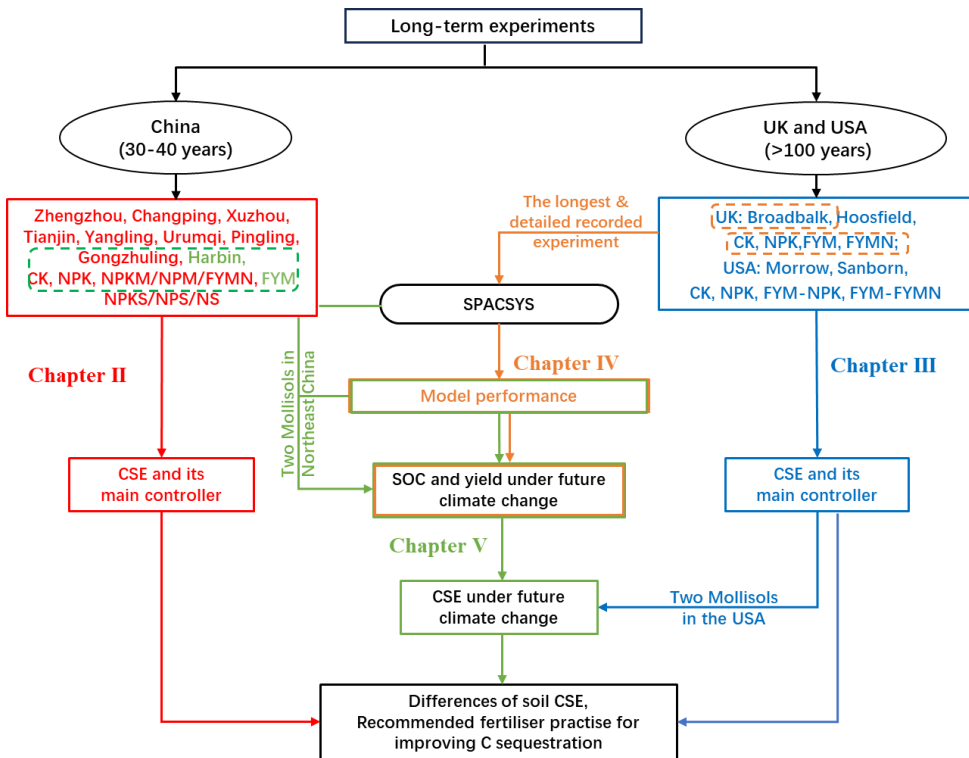


Figure 1-4 The technology route of this thesis.

4. Overview of the chapters

Chapter I General introduction

In this chapter, general information on the research progresses and objectives was described. The main content included the development of CSE calculation methods, research progresses of CSE under long-term fertilisations, and the main influencing factors and mechanisms that control CSE. The response of SOC stock to future climate change under different fertiliser treatments was also presented. The problems and knowledge gaps were described in this section. The importance and suitability of studying SOC sequestration based on long-term experiments were also presented.

Chapter II Manure amendment acts as a recommended fertilisation for improving carbon sequestration efficiency in soils of typical drylands of China

In this chapter, based on 8 long-term experiments established in 1979 or 1989/1990 across the dryland region of China, the objectives were to clarify the characteristics of soil CSE in the plough layer and its main controller with a small amount of C input (in comparison to the four classical experiments). Appropriate fertiliser practises for

improving soil CSE were recommended. In this study, we calculated CSE annually under four treatments, including no fertiliser (CK), chemical nitrogen, phosphorus and potassium combination (NPK), chemical fertilisers plus manure (NPKM/NPM/FYMN), and straw (NPKS/NPS/NS). Variance partitioning analysis (VPA) and structural equation modelling (SEM) were applied to quantify the contributions of different influencing factors to the soil CSE across the whole region and under different fertilisations.

Chapter III Soil carbon sequestration efficiency decreased exponentially with carbon input - Based on four classical long-term experiments

In this chapter, the objectives were to identify the CSE characteristics of the plough layer and its main influencing factors with a large amount of C inputs, and to confirm the relationship between the CSE and cumulative C input based on four classical long-term experiments: Broadbalk (1843–), Hoosfield (1852–), Sanborn (1888–) and Morrow (1904–). We calculated the SOC sequestration rate, C input, and CSE in the year with measured data under four treatments, including CK, NPK, manure only (FYM), and chemical N plus manure (FYMN). Extreme gradient boosted tree (XGBoost) regression modelling was applied to evaluate the importance of the examined climate variables, management factors, and edaphic properties on soil CSE.

Chapter IV Climate change impacts on crop production and soil carbon stock in a continuous wheat cropping system in southeast England

In this chapter, the SPACSYS model's performance to simulate the long-term dynamics of SOC stocks was validated using data from the longest and well-documented Broadbalk wheat system in southeast England. Whether continuous manure application over 170 years can still be considered a sustainable strategy for increasing SOC sequestration under future climate change was evaluated. Six treatments were used: CK, a combination of chemical nitrogen, phosphorus and potassium with three nitrogen application rates (N1PK, N3PK and N5PK), FYM, and FYMN. Four climate scenarios were selected to investigate the impacts of varying levels of climate change on crop production and SOC sequestration: baseline, Representative Concentration Pathway (RCP) 2.6, 4.5 and 8.5.

Chapter V Both soil productivity and carbon sequestration of Mollisols decreased under future climate change

In this chapter, the objectives were to demonstrate the response of SOC stock and CSE to future climate change in two Mollisol long-term experiments in Northeast China, and describe the full CSE characteristics when a large amount of C has been accumulated in the future. We focused on the same soil type in both the four classical long-term experiments and those in China, i.e., Mollisol, thereby establishing the foundation for a more reliable comparison of soil CSE among different regions. Four fertilisations, including CK, NPK, FYM and NPKM/FYMN, were chosen. Model predictions were performed under the baseline, RCP 2.6, RCP 4.5 and RCP 8.5 scenarios. The CSE of the two Mollisols in northeastern China were calculated based on the predicted yield and SOC stock under future climate scenarios. A beneficial strategy for simultaneously sustaining crop yield and improving CSE under future

climate change was presented.

Chapter VI General discussion and conclusions

In this chapter, the distinct characteristics of CSE in two Mollisol long-term experiments conducted in China, as well as in two others in the USA at varying stages of cumulative carbon input, have been identified and thoroughly discussed. In addition, the meaning, importance, and relevance of the general results were discussed. We stated the answers to the main research question and made recommendations for future research on our topic.

Chapter II

Manure amendment acts as a recommended fertilisation for improving carbon sequestration efficiency in soils of typical drylands of China

From: Liang, S., Sun, N., Wang, S., Colinet, G., Longdoz, B., Meersmans, J., Wu, L., Xu, M., 2023. Manure amendment acts as a recommended fertilisation for improving carbon sequestration efficiency in soils of typical drylands of China. *Frontiers in Environmental Science* 11, 1173509. doi: 10.3389/fenvs.2023.1173509

Abstract

It is generally known that soil organic carbon (SOC) stocks tend to increase with increasing C input, whereas the C sequestration efficiency (CSE), i.e. the conversion ratio of C input to SOC, differs depending on the amount and type of C input. However, there still exists the need to better understand the impact of various fertilisation practises on CSE. We studied the data from 8 long-term experiments located in the main dryland region of China in order to clarify the responses of CSE to different fertilisations and to comprehensively assess the key drivers of CSE in the plough layer considering nearly four decades of various fertiliser treatments, i.e., no fertiliser (CK), chemical nitrogen, phosphorus and potassium (NPK/NP), chemical fertilisers plus manure (NPKM/NPM/FYMN) and straw (NPKS/NPS/NS). Our results showed that manure amendment had the most significant fertilisation effect on SOC sequestration with the average CSE of manure amendment (14.9%), which was significantly higher than that of chemical fertilisations (9.0%) and straw return treatments (7.9%). And manure amendment also had the highest average SOC increase rate of 684 kg C ha⁻¹ yr⁻¹. Variance partitioning analysis (VPA) illustrated that CSE of the main dryland region of China was mostly controlled by edaphic characteristics (32.2%), especially the soil C/N ratio and clay content. The VPA and structural equation modelling (SEM) revealed that the magnitude and influencing factors driving CSE varied among different fertiliser treatments. Soil total N was the limiting factor for CSE in the CK treatment, whereas the soil C/N ratio and pH were the main explanatory factors for CSE in the long-term chemical NPK fertiliser treatment. The negative impact of C input from straw was the main driver of CSE under straw return treatments, though C input had a positive effect on soil physical properties improvement. However, when considering manure amendments, the improvement of soil nutrients and clay content controlled CSE, underlining the main positive direct effect of soil chemical properties. In a nutshell, our results recommend manure plus chemical fertilisers as a sustainable practise for improving C sequestration rate and efficiency in dryland cropping systems.

Keywords: Soil carbon sequestration efficiency, dryland, long-term fertilisation, influence factor, organic amendments.

1. Introduction

Recently, enhancing soil organic carbon (SOC) sequestration in agroecosystems has been considered as an important strategy for maintaining soil productivity and mitigating climate change globally (e.g., Amundson and Biardeau, 2019; Sun et al. 2020). It should be noted that considerable SOC losses, taking place continuously across many regions on Earth, are often characterized by either remarkable land use change or intensified agricultural practises, including the unreasonable intensive usage of fertilisers (Chen et al., 2021; Pugh et al., 2015). Generally, manure combined with mineral fertiliser has been considered as an effective strategy to improve SOC storage because of the combined effect of (i) enhanced residual C input by increasing the biomass production from the mineral fertilisers and (ii) the higher direct organic C input from the manure (Gross and Glaser, 2021; Jiang et al., 2018a; Triberti et al., 2016). However, when considering the incorporation of these sources of C to the soil C pool, it's not only the quantity that matter but also the retention coefficient of those C inputs, which has been defined as carbon sequestration efficiency (i.e. CSE) (Hua et al., 2014; Kong et al., 2005; Stewart et al., 2007). Recent studies highlighted that, regardless the applications rate, the effects of different types of fertilisers (either mineral or organic types) on SOC sequestration are remarkably different (Aguilera et al., 2013; Li et al., 2020a). Consequently, an assessment of region and/or fertiliser treatment specific CSE could be very useful in order to identify a better management strategy with the objective to enhance the soil's C sequestration ability (Hua et al., 2014; Liang et al., 2019).

Research indicates that CSE depends on climate conditions, soil types and fertiliser practises (Triberti et al., 2008; Wang et al., 2020b; Yan et al., 2013; Zhao et al., 2016). More specifically, a faster mineralization of C input, and hence lower CSE, usually appear in regions characterized by both higher temperature and precipitation amounts because of the typically more active microbial community in these environments (Chen et al., 2021; Liu et al., 2019). Soils with low initial SOC contents might have a greater potential and efficiency to sequester freshly added organic C, because these soils are still far away from their C saturation level (Stewart et al., 2008). The CSE of soils with higher clay content could be significantly higher due to the adsorption ability of soil clay particles, and hence the formation of stronger organic-mineral complexes as compared to soil with more sand and silt (Ekschmitt et al., 2005; Hua et al., 2014). A higher soil bulk density (BD) typically results in a lower SOC turnover, and therefore, higher CSE, because of the generally slower soil-atmosphere-water cycling associated with poor air permeability and restricted hydraulic conductivity (Lu et al., 2018; Thomsen et al., 1999). Soil pH could affect CSE indirectly by controlling SOC solubility and affecting plant nutrient absorption and microorganism growth (Evans et al., 2008, 2012). In addition, CSE might vary significantly relying on the different magnitudes of SOC changes in response to animal manure and crop residue amendments (Liu et al., 2014; Maillard and Angers, 2014), which mainly depends on the quality of the newly added carbon. The composted cattle organic fertiliser with high amount of stabilized component-lignin could have a higher CSE than that of uncomposted pig material (Hua et al., 2014), while they were lower than

that of dairy manure (Wang et al., 2020b). Moreover, the C input amount has been suggested negatively effecting CSE (Stewart et al., 2008). The CSE of soil with the high rate of wheat straw return was lower than that of soil with a half input rate (Hua et al., 2014), and a similar result was also found in soil with manure amendments (Wang et al., 2020b). The amounts of added substrates as related to microbial biomass C could also affect C sequestration by stimulating microbial growth and altering the composition of microbial community (Blagodatskaya and Kuzyakov, 2008). However, there is in particular a lack of precise knowledge concerning the impact of different fertilisers on CSE. In this context, it is important to note that a comparison of CSE among regions and/or fertiliser treatments is very difficult due to the existence of complex interactions between climate, edaphic and fertilisation factors (Liang et al., 2016). Hence, a comprehensive study of the individual effects of those factors on CSE under long term fertilisations is urgently needed.

The long-term experiments network of China (from 1979/1989) based on regional specific common soil and fertiliser practises, has effectively monitored the evolution of SOC stocks (Jiang et al., 2014). Hence, this network could be considered as a good opportunity to study whether a given fertiliser treatment can sustain or improve C sequestration on the long-term. The well-document and quite detailed records concerning the fertiliser managements, soil properties and climates are also supporting a comprehensive assessment of the drivers of CSE. In addition, CSE is a function of fertilisation duration (Smith, 2004; Stewart et al., 2008). When considering a period of 30 to 40 years of fertilisers application, many studies indicate that observed changes in SOC are significantly, positively and linearly correlated with C input, resulting in a constant CSE with the limited range of C input (Bhattacharyya et al., 2010; Jiang et al., 2014; Maillard and Angers, 2014; Majumder et al., 2008; Wang et al., 2020b; Zhang et al., 2010). The present study aims to (1) quantify CSE in the plough layer (0–20 cm) and to (2) identify the main drivers of CSE considering 8 long-term experiments in dryland of China.

2. Materials and methods

2.1. Study sites and experimental design

For this study eight classical long-term experiments across a wide range of pedoclimatological conditions were selected, which were based in Gongzhuling (GZL), Changping (CP), Zhengzhou (ZZ), Tianjin (TJ), Xuzhou (XZ), Pingliang (PL), Yangling (YL) and Urumqi (UM) (Figure 2-1). Detailed information about these particular study sites can be found in Jiang et al. (2018a) and Jiang et al. (2014), which are located in major wheat and corn planting areas. However, key background information, including the locations, initial soil properties, cropping systems and climates, can be found in Table 2-1 and Table 2-S1.

2.2. Fertilisation

Four different fertiliser treatments, present in all of the eight studied sites, were

considered, i.e. (1) no fertiliser (CK), (2) balanced application of chemical nitrogen (N), phosphorus (P) and potassium (K) fertilisers (NPK), (3) chemical fertilisers combined with manure (NPKM), (4) chemical fertilisers combined with straw (NPKS) (except for XZ) (Table 2-S2). Specially, CK, NP, NPM and NPS were used for PL, and No K was applied in PL due to its high soil K content. The (3) and (4) treatments in TJ were FYMN and NS because of the fully supply of P and K from manure and straw.

The total amount of N applied (chemical + organic) was equal for the NPK and NPKM, NPKS treatments at GZL, ZZ, YL and UM sites, but this amount was higher in NPKM/NPM/FYMN and NPKS/NPS/NS treatment at the other sites due to an additional manure application or straw return. The types of manure were depended on the local availability (Table 2-S2). Considering the straw return treatment, crop residues were chopped and put into the soil by rotary tillage. These crop residues were only returned during the corn season in CP, ZZ, XZ and YL.

2.3. Soil sampling and analysis

Soil samples from plough layer (0–20 cm) for all experiments were collected in approximately 15 days after harvest (the corn season in double cropping systems) in each long-term experiment (Table 2-1). The soil was air-dried and then passed through a 2-mm sieve prior. A part of the sub-sample was dried, crushed and sieved at 0.25 mm for the determination of total nutrient and 1.0 mm for nutrient analyses. Concentration of SOC was analysed with dichromate oxidation by a modified Walkley & Black method (i.e. heated for 12 minutes at 220 °C) and a correction factor of 1.1 was applied (Hu et al., 2016; Walkley and Black, 1934). Total nitrogen (TN), phosphorus (TP) and potassium (TK) were determined by the classical method of Black (1965), Murphy and Riley (1962) and Kundsén et al. (1982), respectively. Available nitrogen is measured following Lu (1999). Available phosphorus (Olsen-P) was quantified by Olsen et al. (1954), and available potassium by Shi (1976). The clay content, soil pH (considering a 1:1 water/soil mixture) and BD were also measured following Lu (1999).

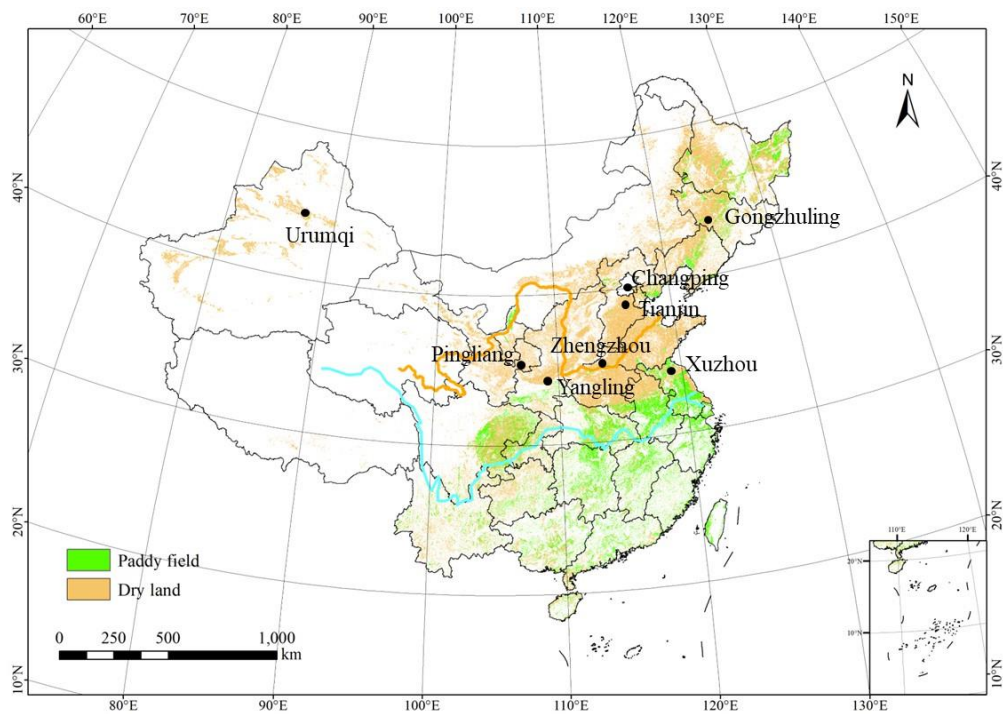


Figure 2-1 The locations of the 8 long-term dryland experiments.

Table 2-1 Initial soil characteristics of plough layer of long-term fertilisation experiments at the 8 dryland long-term experimental sites across China.

Site	Gongzhuling (GZL)	Changping (CP)	Zhengzhou (ZZ)	Tianjin (TJ)	Xuzhou (XZ)	Urumqi (UM)	Pingliang (PL)	Yangling (YL)
Strat year	1989	1990	1990	1979	1980	1990	1979	1990
Soil type (FAO)	Luvic Phaeozems	Haplic Luvisol	Calcaric Cambisol	Calcaric Cambisol	Calcaric Cambisol	Haplic Calcisol	Calcic Kastanozem	Cumulic Anthrosol
Initial SOC [#] , g kg ⁻¹	13.23	7.10	6.60	10.96	6.26	9.86	6.09	7.44
Total N ^{&} , g kg ⁻¹	1.40	0.79	0.65	1.06	0.66	0.87	0.95	0.83
Available N, mg kg ⁻¹	114	49.7	76.6	75.1	68.0	55.2	55.5	61.3
Total P [‡] , g kg ⁻¹	1.39	0.69	0.65	1.59	0.74	0.67	0.58	0.83
Olsen P, mg kg ⁻¹	27.0	4.6	6.5	16.6	12.0	3.4	7.0	9.6
Total K ^ℓ , g kg ⁻¹	22.1	14.6	16.9	16.1	22.7	19.8	20.5	21.6
Available K, mg kg ⁻¹	190	65.4	74.0	173.3	62.0	288	164.7	194.0
pH	7.6	8.2	8.3	8.1	8.2	8.1	8.2	8.6
Bulk density, g cm ⁻³	1.19	1.58	1.55	1.28	1.25	1.25	1.30	1.35
Clay, %	32	10	13	31	6	21	34	17

[#] Soil organic carbon; [&] Nitrogen; [‡] Phosphorus; ^ℓ Potassium.

2.4. Soil carbon sequestration efficiency calculation

In each site, soil C sequestration efficiency was calculated by using following equations:

SOC stocks

The SOC stocks (SOC_s , t C ha⁻¹) were calculated as:

$$SOC_s = SOC_c \times BD \times H \times 10 \quad (1)$$

where SOC_c is the SOC content (g kg⁻¹); BD is the soil bulk density (g cm⁻³); H is the depth of soil sampling (cm).

Carbon input

The C inputs included C from crop above-ground residual ($C_{Stubble}$), root (C_{Root}), extra-root ($C_{Rhizosphere}$) and manure (C_{Manure}) (Bolinder et al., 2007a). Among these C inputs, extra-root C included root exudates and other material derived from root-turnover, which was referred to as ‘rhizodeposition’ ($C_{Rhizosphere}$). The $C_{Rhizosphere}$ of small-grain cereals, corn, and soybean was assumed as 0.65 (i.e., 33/50) times the C input from the roots (Bolinder et al., 2007a), based on the finding of tracer studies (Kuzyakov and Domanski, 2000; Kuzyakov and Schneckenberger, 2004). These studies indicated that approximately 33% of the below-ground C in wheat and barley is released by living roots and remains in soil, and that 50% of the below-ground C remains in roots. Thus, the annual C input (C_{input} , t C ha⁻¹) was calculated using the following:

$$\begin{aligned} C_{input} &= C_{Stubble} + C_{Root} + C_{Rhizosphere} + C_{Manure} \\ &= Y_{shoot} \times C_{shoot} \times P_{stubble} \times 0.001 \times (1 - M_{shoot}) \\ &\quad + Y_{root} \times C_{root} \times P_{root} \times 0.001 \times (1 - M_{root}) + 0.65 \times C_{Root} \\ &\quad + F_{manure} \times C_{manure} \times (1 - M_{manure}) \quad (2) \end{aligned}$$

where Y_{shoot} is the shoot biomass (t ha⁻¹); Y_{root} is the root biomass (t ha⁻¹); $F_{manure/straw}$ is the application amount of manure or straw (t ha⁻¹); $P_{stubble}$ is the ratio of above-ground stubble returned to soil with 100% for NPKS and 15% (wheat) or 3% (corn) for other treatments (Jiang et al., 2014; Zhang et al., 2010); C_{shoot} , C_{root} and $C_{manure/straw}$ are C content of corresponding plant tissues and fertilisers, respectively (Table 2-S3); P_{root} is ratio of the root biomass in plough layer with 75.3% for wheat and 85.1% for corn, respectively (Jiang et al., 2014; Khan et al., 2007); The average moisture content of the air-dried crop samples of the shoot (M_{shoot}) and root (M_{root}) is based on the observations (%); otherwise, it is an average value of 14% (Jiang et al. 2014; Jiang et al. 2018a); M_{manure} is the moisture content of manure (%).

Considering the shoot biomass, we assumed it was approximately equal to straw dry matter production. When sites hadn’t measured straw production (XZ), it was established as follow:

$$Y_{shoot} = \frac{Y_{grain}}{HI} - Y_{grain} \quad (3)$$

where Y_{shoot} and Y_{grain} are the shoot biomass and grain yield ($t\ ha^{-1}$), respectively; HI is the harvest index (Table 2-S3).

The root biomass could be estimated by:

$$Y_{root} = Y_{shoot} \times RSR \quad (4)$$

where RSR is the root to shoot ratio (Table 2-S3).

Soil carbon sequestration rate

After calculating the SOC stocks in the plough layer across the eight long-term experiments, we found that the SOC stocks exhibited linear increases over the entire experimental period (Figure 2-2). Therefore, this study used the difference between SOC stocks in the current year and the initial year to calculate the soil C sequestration rate and efficiency.

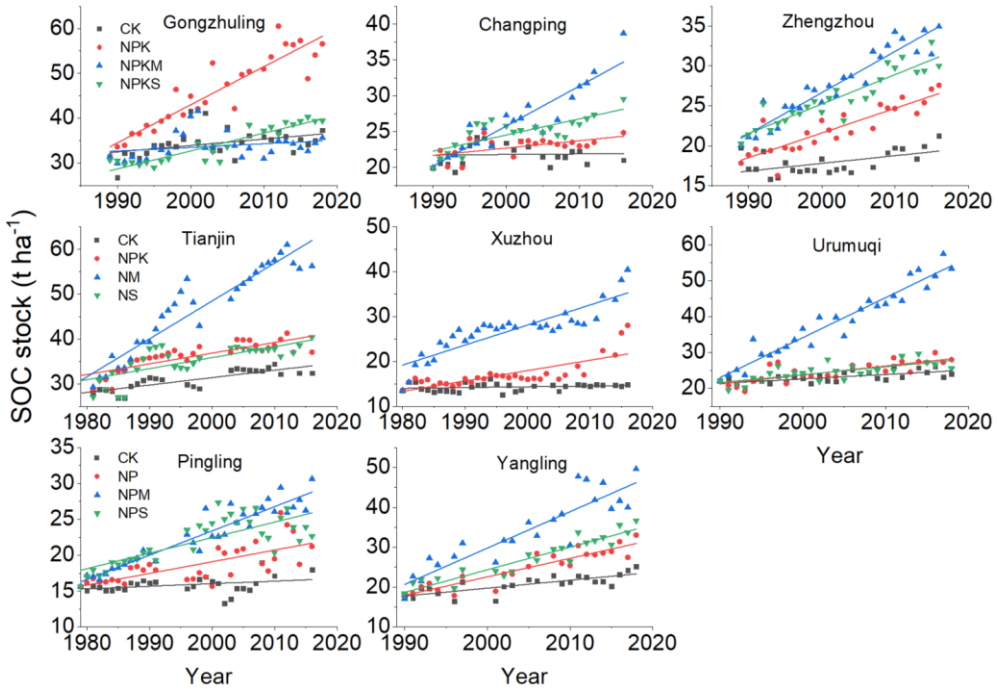


Figure 2-2 Soil organic carbon stocks ($t\ C\ ha^{-1}$) in the plough layer of the eight long-term experiments in China.

The soil C sequestration rate ($C_{sequestration\ rate}$, $t\ C\ ha^{-1}\ yr^{-1}$) in the plough layer (0–20 cm) during the experimental period was calculated as follow:

$$C_{sequestration\ rate} = \frac{SOC_t - SOC_{initial}}{t} \quad (5)$$

where SOC_t is the SOC stocks in the t-th year, and $SOC_{initial}$ is the average SOC

stocks of the initial 3 years; t is the experimental duration.

Carbon sequestration efficiency

The carbon sequestration efficiency (CSE, %) is the conversion ratio of unit carbon input to SOC stock.

$$CSE = \frac{SOC_t - SOC_{initial}}{Cinput_t} \times 100\% \quad (6)$$

where $Cinput_t$ is the total C input during t years.

2.5. Influence factors

To gain insight to factors controlling CSE among the studied dryland soils under different long-term fertiliser treatments, climate variables, edaphic properties and management factors were selected for each site. Mean annual temperature (MAT) and mean annual precipitation (MAP) as main climate factors influencing crop growth and C decompositions were collected from the National Meteorological Information Center ([http:// data.cma.cn/](http://data.cma.cn/)). Considering edaphic factors, soil chemical properties (i.e., measured total N stock (TN), available N (AN), total P (TP), Olsen P (AP), total K (TK), available K (AK), soil C/N ratio, initial SOC content and pH) as well as physical properties (i.e., clay content and BD) were selected for further analysis. As regards the management factors, C inputs from crop residue (stubble, root, rhizosphere) and organic manure/straw return were calculated for further analysis.

2.6. Statistical analysis

Figure 2-3 presents the methodological flowchart of this study. One-way analysis of variance (ANOVA) and the least significant difference (LSD; $P < 0.05$) test were applied to compare the fertilisation effects on the SOC sequestration rate, annual C input and CSE within an individual site. One-way ANOVA was also used to identify the duration when CSE of studied treatments has no significant difference with the latest record year of the experiment. Paired-samples t tests was used to compare the difference of CSE between NPK/NP and CK, NPKM/NPM/FYMN and NPKS/NPS/NS across different sites, respectively. The statistical analyses were performed with SPSS 22.0 (SPSS, IBM, 2015, Chicago, USA). Graphs were prepared using Origin profession version 2019 (OriginLab, Northampton, USA).

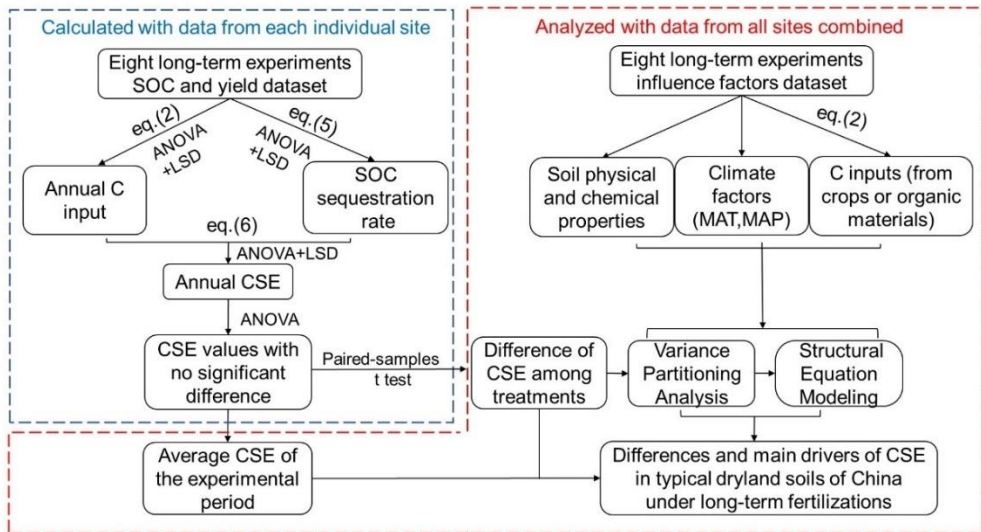


Figure 2-3 The methodological flowchart of this study.

Variance partitioning analysis (VPA) was applied to quantify the contributions (%) of (i) total selected edaphic, C input and climate factors and their interactions, as well as (ii) each individual variable within each factor when considering the CSE differences among different fertiliser treatments and sites. Only factors that passed the test of collinearity were considered when conducting the VPA by using R-software, version 4.1.2. Subsequently, selected soil variables that significantly ($P < 0.05$) influenced CSE following the results of the VPA, were used to build the structural equation modelling (SEM). This SEM was used to investigate the direct and indirect factors regulating CSE under manure amendments and straw return treatments. The chi-square statistic to the respective degrees of freedom ratio ($\chi^2/df < 3$ for good fit, < 5 for reasonable fit), probability level ($P > 0.05$), comparative fit index (CFI > 0.9), root mean square error of approximation (RMSEA < 0.08) and low Akaike information criterion (AIC) were used to assess the overall performance of the model (Browne and Cudeck, 1992).

3. Results

3.1. Annual SOC sequestration rate and C input under long-term fertilisation

Over a time span of 30 to 40 years of fertilisations, chemical fertiliser combined with manure resulted in significantly higher annual SOC sequestration rates ($684 \text{ kg C ha}^{-1} \text{ yr}^{-1}$) in the top 20cm of the soil as compared to other treatments ($P < 0.05$) (Table 2-2). Furthermore, the average annual SOC sequestration rates of chemical

fertiliser with straw return (i.e. 333 kg C ha⁻¹ yr⁻¹) were higher than those of only chemical fertilisers (244 kg C ha⁻¹ yr⁻¹).

Table 2-2 Average SOC change rate (kg C ha⁻¹ yr⁻¹) of typical croplands under different long-term fertiliser treatments.

Site	CK	NPK/NP	NPKM/NPM/FYMN	NPKS/NPS/NS
GZL	139.14 b	194.54b	988.04 a	262.57 b
CP	114.67 c	157.06 bc	498.32 a	286.71 ab
ZZ	148.78 c	311.74 b	495.11 a	345.03 ab
TJ	113.24 c	246.77 b	726.32 a	255.03 b
XZ	160.08 b	240.88 b	560.75 a	-
UM	102.41 c	213.20 b	810.35 a	259.55 b
PL	109.86 b	122.22 b	322.91 a	253.93 ab
YL	297.34 d	468.33 c	1068.12 a	667.06 bc
Average	148.19	244.34	683.74	332.84

- means no this kind of treatment. Numbers with different lowercase indicate significant difference among different treatments in an individual site ($P < 0.05$).

The annual C input from crop residues was significantly higher in the chemical fertilisers plus manure or straw treatments (i.e. 0.9–2.9 t C ha⁻¹ yr⁻¹) as compared with CK and chemical treatments (i.e. 0.6–2.3 t C ha⁻¹ yr⁻¹) (Figure 2-4). The C inputs from manure and straw return were 0.9–5.3 t C ha⁻¹ yr⁻¹ (Figure 2-4). Generally, the average annual C input under an individual treatment from the highest to the lowest value was as follows: NPKM/NPM/FYMN (2.6–7.5 t C ha⁻¹ yr⁻¹) > NPKS/NPS/NS (2.8–5.6 t C ha⁻¹ yr⁻¹) > NPK/NP (1.4–3.9 t C ha⁻¹ yr⁻¹) > CK (0.9–1.5 t C ha⁻¹ yr⁻¹) ($P < 0.05$). However, this trend was slightly different in GZL, ZZ and PL where the highest annual C input was found in the NPKS treatment.

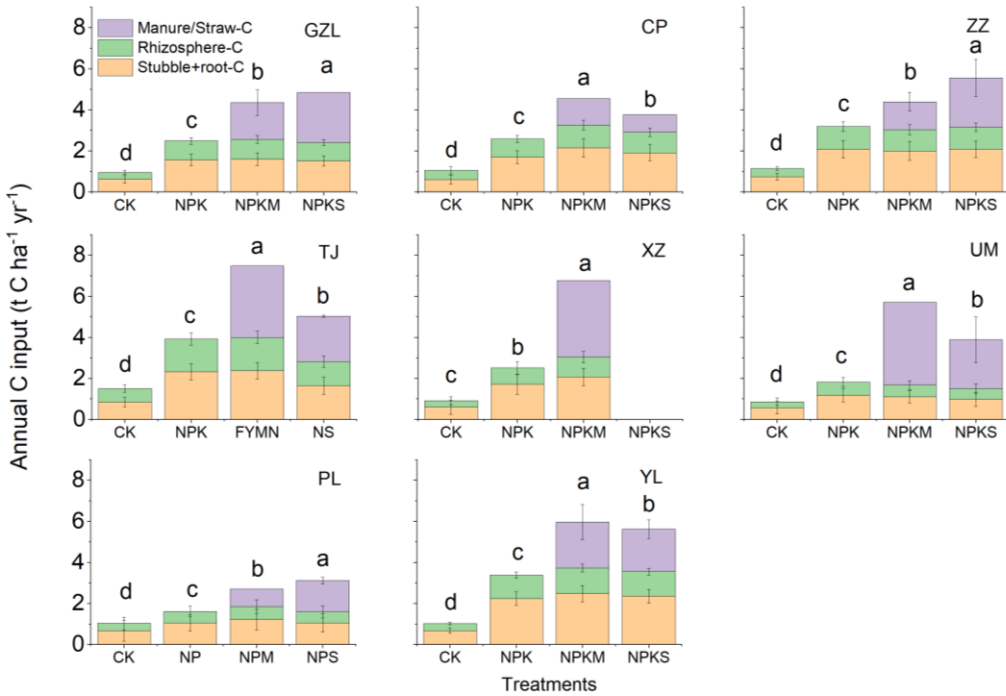


Figure 2-4 The average annual C input from different sources under different fertiliser treatments at all long-term experimental sites. Numbers with different lowercase letters indicate significant difference ($P < 0.05$) for total annual C input under different treatments within an individual site.

3.2. Carbon sequestration efficiency under long-term fertilisation

After calculating the annual CSE of each fertiliser treatment in each long-term experiment (Figure 2-S1), we took an average value of the last 9 years in which CSE had no significant difference with the latest record year for all sites (Tables 2-3, 2-4, Table 2-S4). Generally, the CSE in GZL, UM, PL and YL, located across Northeast and Northwest China and typically characterized by high clay contents (i.e. 17–34%), was higher (i.e. CSE of 4.6–27.7%) than those in other sites located across the North China plain with typically lower clay contents (except TJ) (i.e. CSE of 3.5–15.2%) (Table 2-3, Table 2-S1). The CSE of chemical NPK fertilisers, with an average value of 9.0%, was significantly lower than that of treatments with manure amendments with the average of 14.9% ($P < 0.01$) (Tables 2-3, 2-4). However, NPK/NP had a significantly higher CSE as compared to NPKS/NPS/NS (7.9%) ($P < 0.01$) (Tables 2-3, 2-4, Table 2-S5).

Table 2-3 Average carbon sequestration efficiency (%) of typical drylands under different long-term fertiliser treatments.

Site	CK	NPK/NP	NPKM/NPM/FYMN	NPKS/NPS/NS
GZL	13.78 b	8.78 c	27.56 a	6.14 d
CP	6.79 c	6.73 c	11.08 a	9.21 b
ZZ	3.96 c	9.40 b	15.16 a	9.37 b
TJ	10.12 b	9.09b	11.54 a	5.93 c
XZ	3.53 c	4.23 b	8.31 a	-
UM	10.51 b	10.46 b	17.41 a	4.52 c
PL	4.64 c	10.05 b	13.05 a	9.65 b
YL	13.97 b	12.90 b	15.44 a	10.25 c
Average	8.41	8.96	14.94	7.87

- means no this kind of treatment. Numbers with different lowercase indicate significant difference among different treatments in an individual site ($P < 0.05$)

Table 2-4 Paired-samples t tests of CSE under different fertiliser treatments.

Treatments		Correlation	t	df	Sig. (two-tailed)
NPK/NP	CK	0.57**	1.003	71	0.319
NPK/NP	NPKM/NPM/FYMN	0.40**	-9.49	71	0.000**
NPK/NP	NPKS/NPS/NS	0.16 ^Φ	-	63	-

^Φ there was no significant relation between CSE of NPK and NPKS, and independent samples t-tests was used for the analysis (Table 2-S5); ** $P < 0.01$.

3.3. Factors influencing carbon sequestration efficiency

The VPA results indicated that 37.9% of the variations could be explained by edaphic, C input and climate factors as well as their interactions across all sites and treatments (Figure 2-5). However, the edaphic factors contributed the most (32.2%) to the CSE of studied region, with in particular the soil C/N ratio (4.9%) and clay content (3.8%), followed by C input (3.2%) and climate (2.8%). Similarly, the contribution of edaphic factor (21.6–63.6%) was much higher than that of C input (1.2–20.2%) and climate factors (0.1–18.3%) under each fertiliser treatment or site (Figure 2-5). For the contribution of each individual variable within each factor, soil C/N ratio and clay content were important controllers to CSE as being indicated by significant relations for most sites (except for GZL and UM for C/N and CP and XZ for clay) (Table 2-5). Furthermore, soil C/N ratio only had significant influence on the CSE of CK and chemical fertilisers (Table 2-5).

For different treatments, the VPA results showed that TN was the main controller for CSE of CK with a much higher contribution of 9.6% as compared to other variables that had a significant effect on CSE (1.4–5.3%, $P < 0.05$) (Table 2-5). When considering chemical NPK treatment, soil C/N ratio (5.1%) and pH (1.3%) were the main explanatory factors (Table 2-5). Furthermore, many edaphic factors had a significant effect on CSE with contributions ranging between 0.8 and 3.9% and between 1.4% and 5.2% for manure and straw amendments, respectively (Table 2-5) ($P < 0.05$). The SEM illustrated that the selected variables were able to predict 66% and 29% of variances in CSE under NPKM/NPM/FYMN and NPKS/NPS/NS of the studied region, respectively (Figure 2-6). Soil chemical properties had the largest direct effect on CSE for manure amendments with a positive standardized coefficient of 0.76, whereas C input had the largest indirect effect with a positive standardized coefficient of 0.44 through its association with soil chemical factors and clay content (Figure 2-6 a&c, Figure 2-S2 a&c). C input had the largest direct effect on CSE with a negative standardized coefficient of -0.69 for straw return treatments, whereas climate and C input had similar indirect effects with a positive standardized coefficient of 0.16 and 0.14, respectively, through their interactions with soil chemical and physical properties (Figure 2-6 b&d, Figure 2-S2 b&d).

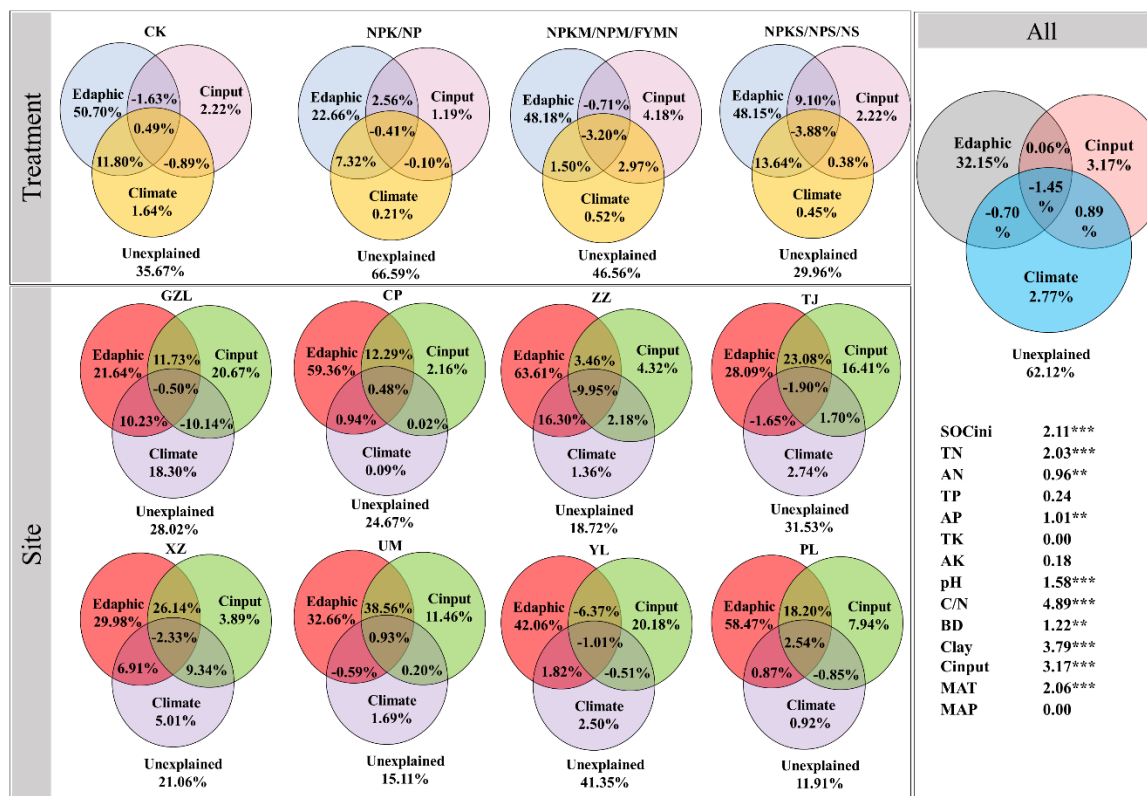


Figure 2-5 The proportional contributions (%) of edaphic, climate, C input factor and their interactions on CSE under each treatment, each dryland long-term experimental site and across all fertiliser treatments and sites in China based on VPA method.

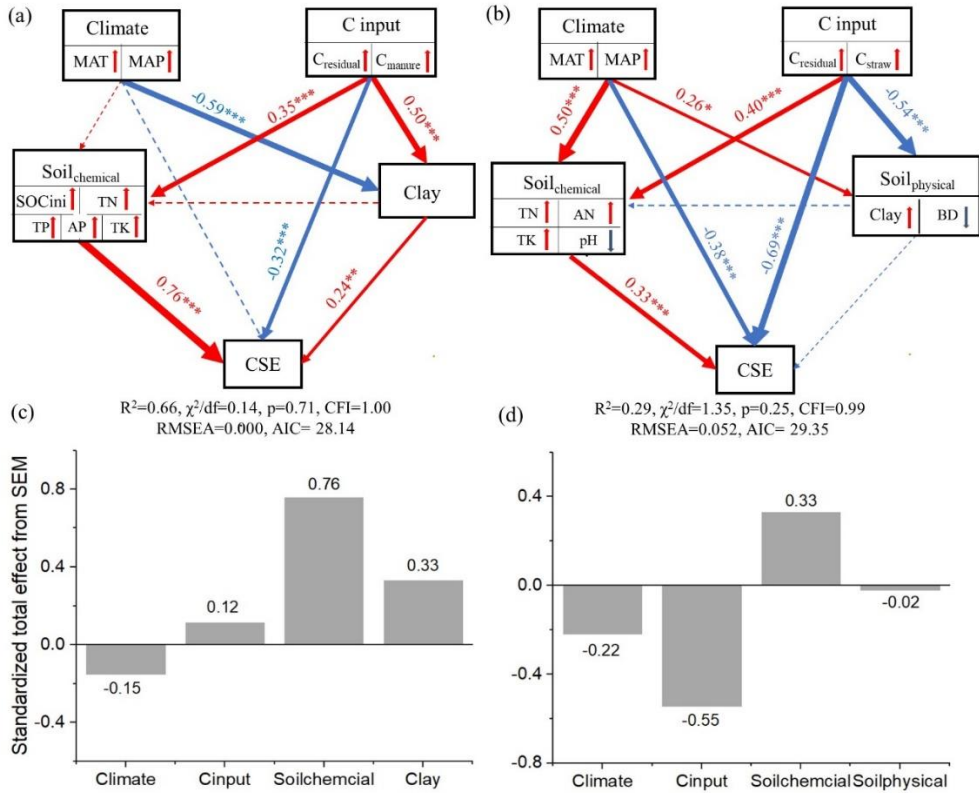


Figure 2-6 Structural equation model showing the effects of climate, carbon (C) input, soil chemical properties and soil physical properties on soil carbon sequestration efficiency (CSE) under NPKM/NPM/FYMN (a&c) and NPKS/NPS/NS (b&d). The red and blue lines in (a) and (b) represent positive and negative effects, respectively. The line width and the numbers above the lines (standardized path coefficient) corresponding to the strength of the path. Solid and dashed lines represent significant and non-significant paths, respectively ($*P < 0.05$, $**P < 0.01$, $***P < 0.001$). Multi-layer rectangles represent the first component from the principal component analysis of climate factors, soil chemical and physical properties, C inputs, and the vertical red (/blue) arrows within it represent the positive (/negative) relationships between adjacent variables and the corresponding PC1. MAT: mean annual temperature, MAP: mean annual precipitation, SOC_{ini}: initial SOC content, TN: soil total nitrogen content, AN: available N content, TP: total phosphorus content, AP: Olsen P content, TK: total potassium content, AK: available K content, Clay: soil clay content, BD: soil bulk density, pH: soil pH, C_{residual}: C input from stubble, root and exudates, C_{manure}: C input from manure, C_{straw}: C input from straw. χ^2 : chi-square values, d.f.: degree of freedom, GFI: goodness of fit index, RMSEA: the root mean square error of approximation.

Table 2-5 The proportional contribution (%) of individual edaphic, carbon input, and climate factors on variance of carbon sequestration efficiency based on variance partitioning analysis.

Category	Factors	Treatments				Sites							
		CK	NPK/NP	NPKM/ NPM/FYMN	NPKS/ NPS/NS	GZL	CP	ZZ	TJ	XZ	UM	YL	PL
Edaphic	SOCini	5.31 ***	1.03	3.87 ***	0.47	3.28 *	0.00	13.32 ***	11.26***	0.00	0.00	3.66 *	1.46 **
	TN	9.56 ***	0.32	1.49 *	1.78 *	0.34	8.57 ***	2.38 **	2.85*	1.32	0.00	3.89 *	3.74 ***
	AN	0.56	0.89	0.14	1.40 *	5.16 **	0.16	0.00	0.44	1.46	0.44	0.00	0.74
	TP	0.00	1.08	4.34 ***	0.33	0.00	3.85 ***	2.88 **	1.88	0.00	0.28	3.12*	0.17
	AP	0.00	1.14	0.96 *	1.02	4.78 *	0.00	2.42 **	0.00	0.00	0.00	0.94	0.73
	TK	1.92 **	0.34	0.80 *	0.01	0.00	0.22	0.00	0.80	0.32	0.00	0.00	3.07 ***
	AK	0.38	0.28	0.13	5.23 ***	0.00	2.76 **	0.00	0.00	0.00	0.41	0.12	0.00
	pH	0.99	1.29 *	0.20	2.45 **	0.00	0.00	0.00	16.42 ***	0.00	0.43	3.01 *	0.52

Difference and Simulation of Soil Carbon Sequestration Efficiency of Typical Cropland in China, UK and USA

Cinput	C/N	1.35 *	5.13 **	0.75	0.01	0.02	13.59 ***	2.45 **	3.94 *	3.65**	0.83	3.18 *	4.45 ***
	BD	0.13	1.17	0.37	2.31 *	1.49	0.00	0.00	1.24	0.14	0.36	9.37 ***	2.01 **
	Clay	4.91 ***	1.10	3.31 ***	2.47 ***	3.32 *	0.34	18.26 ***	2.80 *	0.03	2.03**	8.36 ***	5.59 ***
	C _{residual}	2.22 **	1.19	0.96 *	1.71 *	18.28 ***	0.93	1.61 *	1.98	1.18	2.50 **	1.24	0.89 *
	C _{manure/straw}	-	-	0.65	2.18 **	0.37	0.09	4.06 ***	13.75 ***	0.89	9.09 ***	1.93	0.66 *
Climate	MAT	1.45 *	0.21	0.37	0.22	13.21 ***	0.07	1.33 *	2.02 *	4.80 **	0.38	2.47 *	0.40
	MAP	0.00	0.00	0.00	0.00	6.44 **	0.00	0.00	1.84	0.39	1.69	0.00	0.51

SOC_{ini} means the initial SOC content; C_{residual} means the C input from stubble, root and rhizosphere; C_{manure/straw} means the C input from manure or straw return; * $P < 0.05$, ** $P < 0.01$, *** $P < 0.001$. The proportional contribution of total edaphic, carbon input, and climate factors on variance of carbon sequestration efficiency is shown in Figure 2-5.

4. Discussion

4.1. Response of SOC sequestration rate and C input to long-term fertilisation

Over nearly 40 years of combined application of chemical fertilisers and manure significantly ($P < 0.05$) increased SOC stocks in the top 20cm of the dryland soils of China under both single cropping and double cropping systems (Table 2-2), which is in agreement with previous studies (Jiang et al., 2018a; Liang et al., 2016; Ren et al., 2021). The average SOC sequestration rates (i.e., 148–684 kg C ha⁻¹ yr⁻¹) across our studied sites for all fertilisers treatments were very close to the results of 30–590 kg C ha⁻¹ yr⁻¹ derived by 45 field trials (20–37 years) across Chinese drylands as being reported by Ren et al. (2021). The highest SOC sequestration rate under the NPKM treatment can be explained by the fact that this treatment is characterized by the highest average plant derived C-input (stubble + root + rhizosphere) of 1.7–4.0 t C ha⁻¹ yr⁻¹ in combination with the additional organic C input from manure (0.85–4.05 t C ha⁻¹ yr⁻¹) (Figure 2-4). Moreover, an accurate C input estimation is important for the analysis of CSE differences among different treatments. In our study, the annual wheat and corn derived C-inputs for all treatments are ranging between 0.51 and 2.46 t C ha⁻¹ yr⁻¹ and between 0.72 and 2.63 t C ha⁻¹ yr⁻¹, respectively (Table 2-S6). Those are comparable to C input values of 0.58–2.86 t C ha⁻¹ yr⁻¹ for wheat and of 0.90–2.39 t C ha⁻¹ yr⁻¹ for corn reported for China, Belgium, France and Canada, despite the different methods for C input calculation (Bolinder et al., 2007b; Jiang et al., 2018a; Meersmans et al., 2013).

4.2. Main drivers for carbon sequestration efficiency in typical dryland of China

Edaphic characteristics, especially soil C/N ratio and clay content, were the main factors driving CSE after 30–40 years of fertilisations across the main dryland regions of China (Figure 2-5, Table 2-5). Soil C/N ratio is an indication of soil quality as it is typically inversely proportional to the decomposition rate of SOC (Zinn et al., 2007). Agricultural management practises, especially fertilisation, tend to affect this ratio in agroecosystem (Deng et al., 2020). In this study, compared to CK, the long-term intensified N fertiliser application slightly decreased the soil C/N ratio (Figure 2-S3). The reduction of soil C/N ratio usually suggested a potential SOC decrease and GHG emissions with high mineralization rates (Majumder et al., 2008; Xu et al., 2016). However, the soil C/N ratio of manure amendments were significantly higher than that of NPK/NP ($P < 0.05$) and slightly higher than that of chemical fertilisers plus straw return (Figure 2-S3). A meta-analysis of globally published studies indicated that the quality of the organic amendment was the main driver determining CSE (Maillard and Angers, 2014), which might be the major reason explaining the

difference of soil C/N ratios and their effects on CSE. It seems that the high C/N ratio of straw (i.e. on average 67 for wheat and 50 for corn across the entire study area) contributed to the slow decomposition of organic matter and the deficiency of available mineral nitrogen due to the competition of microbes with crops (Triberti et al., 2016), and which, therefore, generally results in increasing SOC stocks (Mahmoodabadi and Heydarpour, 2014). However, organic substances originating from aboveground residues are more resistant to decomposition than those of crop roots (Rasse et al., 2005), which could impede the high CSE. The low C/N ratio of manure (i.e. on average 22 across the entire study area) might result in an increased organic matter decomposition, whereas on the other hand the input of direct organic C, and the associated formation of stable soil organic complexes from the resistant constituents in manure (e.g. lignin and polyphenol), might lead to an accumulation of SOC (Majumder et al., 2008). Furthermore, the application of manure, compared to straw return and chemical NPK applications, was beneficial to the enhancement of the size and functional group diversity of soil microorganisms that contributed to OC sequestration, like *Proteobacteria* (Wang et al., 2020a), and inherent C substances decomposition, like *Dothideomycetes* (Freedman et al., 2015). Therefore, the mechanisms and efficiencies for the sequestration of C into SOC pool among the studied treatments were significantly different.

In our study, the high clay content generally contributed to high CSEs. The CSE in GZL, UM, PL and YL with high clay contents (17–34%) (except TJ with high contributions of C input and pH to CSE) was higher (4.5–27.6%) than that of CP, ZZ and XZ sites (3.5–15.2%) with relatively low clay contents (6–13%) (Table 2-3, Table 2-S1). The SEM of NPKM also showed that clay content had a positive effect on CSE (Figure 2-6 a & c). A study focused on CSE in Vertisols with a clay content of around 40%, highlighted that a soil with a high clay content has typically high CSE irrespective of the type of C input (wheat straw, pig manure or cattle manure) (Hua et al., 2014). Soil clay influences the sequestration of organic C due to the strong adsorption capacity of the clay particles by organo-mineral bonding reactions (Chen et al., 2010; Ekschmitt et al., 2005). Hence, soils with high clay contents are generally characterized by low SOC decomposition rates as SOC could be chemical stabilized after being absorbed onto clay minerals (Franzluebbers et al., 1996; Mikha and Rice, 2004). This chemical protection of SOC by association with organo-mineral complexes was reinforced with the increase in clay content (Hassink, 1997).

4.3. The variation of key influence factor of carbon sequestration efficiency among different fertiliser treatments

Notably, the main controlling factor of CSE varied considerably among different treatments (Table 2-5, Figure 2-5). Based on the results of VPA, soil TN was the most important factor for CSE of CK. When considering the CK treatment, N supply depended only on the basic soil fertility, whereas N as a limiting factor on crop growth and belowground C transformations has an important influence on the amount of C

input and thus the CSE (Liang et al., 2016). Soil C/N ratio (5.1%) and pH (1.3%) were main explanatory variables under long-term chemical NPK fertilisations (Table 2-5). The long-term chemical N application caused a decrease of soil C/N ratio (Figure 2-S3), which most probably accelerated soil microorganisms' mineralization of carbon sources from humus and led to the decomposition of organic carbon (Brown et al., 2014). Liang et al. (2016) showed that pH was an important factor explaining CSE variations in drylands at sites located in Northwest China, because pH was significantly inversely related to SOC content. Moreover, the decrease of pH due to the intensified mineral N fertiliser application impeded the abundance and activity of microorganisms, resulting in a reduced SOC quality (e.g., polyphenols content) (Kemmitt et al., 2006) and influencing organic C turnover and accumulation due to a reducing of microbial mineralization (Wong et al., 2010).

For treatments combining manure with chemical fertilisers, our results presented that soil chemical properties had the largest direct positive effect on CSE, whereas C input had the largest indirect positive effect, through soil chemical properties and clay content, on CSE (Figure 2-6, Figure 2-S2 a&c). Generally, manure application was recognized as a recommended practise for improving soil fertility and soil structure because of the beneficial impacts of the additional organic matter and nutrient inputs (Guo et al., 2019). Previous studies have indicated that manure amendments led to greater retention of total C, N, P and K in the soil under wheat and corn cropping systems across China (Jiao et al., 2006; Su et al., 2006), which on its turn provided favourable conditions for crop growth and microbial activity. Additionally, recent research found that manure combined with NPK chemical fertilisers significantly increased the SOC stock and the macroaggregate component associated C (Liu et al., 2019; Yan et al., 2013). Moreover, manure amendments seem to promote aggregate stability significantly as compared with long-term chemical fertilisers or no fertiliser treatments (Haynes and Naidu, 1998; Liu et al., 2019; Yan et al., 2013). In this respect it is important to note that Liu et al. (2019) showed that the CSE of macroaggregates was greater than that of other physical fractions, and as such this can be seen as a reason to why CSE in treatments with manure amendments are higher. The latter can be explained more specifically as follows: Firstly, manure directly increased mycorrhizal hyphae that, as colloids, could be associated with mineral fraction of the soil in order to bind microaggregates into macroaggregates (Bronick and Lal, 2005). Secondly, increased microbial biomass and associated activity due to manure amendments might lead to a greater production of microbial-derived agents (Pan et al., 2009; Six et al., 1999). Hence, manure applications could result in more stable SOC in both newly formed macroaggregates and microaggregates (Fonte et al., 2009).

For chemical fertilisers plus straw return treatment, the negative effect of C input was the main driver of CSE (Figure 2-6, Figure 2-S2 b&d). The negative effect can be explained by the following reasons: (1) The increased mineralization of native soil origin matter (SOM) was stimulated by fresh OC input following straw return. More

specifically, the added residuals could have increased the concentration of easily available compounds (e.g. phytosterols, policosanol), which are essential sources for microorganisms, and thus stimulates microbial activity via, causing a positive priming effect (Kuzyakov et al., 2000; Wu et al., 2019b). (2) Most of the added straw derived C would be mineralized eventually. Studies with ^{13}C -labeled corn straw has estimated that about 42–79% of the straw derived C got mineralized, and as such only c. 10% of this C input was transformed into particulate organic carbon and more stable SOC fractions (An et al., 2015; De Troyer et al., 2011; Yang et al., 2012). (3) Crop straw characterised by slow biodegradation and low stability of its litter-derived C both adverse to a high CSE (Berhane et al., 2020; Poeplau et al., 2015; Zhao et al., 2016). The root derived C could have more opportunity for physico-chemical interactions with soil particles, and hence, could be much more efficiently incorporated into the stable soil C pools (Rasse et al., 2005), which might explain the significant difference between NPK/NP and NPKS/NPS/NS in the present study. Moreover, our results indicated lower CSE in straw plus chemical fertilisers as compared to manure amendments, highlighting that the type of OC input influences CSE. A 34 years corn-wheat rotation in Italy gave a similar result with more than doubled CSE under manure combined N fertiliser than that of straw returning (Triberti et al., 2008). In addition, another study considering a long-term dryland rotation in Northeast China has shown that CSE was 30% higher under NPKM treatment than under NPKS treatment (Zhao et al., 2016). Hence, the choice of the type of organic amendment is crucial to increase soil carbon sequestration.

4.4. Uncertainties of carbon sequestration efficiency estimation

In our study, the CSE values of CK and chemical fertilisers ranged from 3.5 to 14.0% in drylands of China, which is close to the results of 2.1–17.0% derived by Zhao et al. (2016) but lower than those of Wang et al. (2020b), i.e. 14–32%. The CSE of NPKM/NPM/FYMN in our study generally (except GZL, Table 2-3) matched the range of $12\% \pm 4\%$, which was a global manure-C retention coefficient established by a meta-analysis based on 130 observations considering an average of 18 years of application (Maillard and Angers, 2014). Besides the impact of edaphic properties, climate and C input on CES variations, the uncertainties in our study may also partly intensify the difference. We obtained the CSE of the different treatments by making two assumptions. More precisely, we assumed that (i) the short-term effect of the priming effect after exogenous organic matter addition would be negligible for long-term carbon sequestration, and (ii) the soil respiration rates before and after C addition were similar during long-term fertilisations. Moreover, we estimated C input via root and stubble by using a fixed ratio of the aboveground biomass (Table 2-S3), and the C contents of manure as well as crop's shoot and root were not measured annually. The effects of different sources of manure on CSE was not considered in this study even if a three decades long-term experiment observed that CSE of cattle manure was

higher than that of pig manure (Hua et al., 2014). However, the main reason was that the cattle manure was composted and had a high lignin content of 23.7%, which was considered as stabilized component of SOC, influencing its pool-size and its turnover (Ghosh et al., 2004). When considering the straw return in different regions, the plant residue chemistry converged after the straw decomposed over nearly a decade despite of different climate zones (Wang et al., 2023), and the initial quality does not control the long-term decomposition of SOC (Schmidt et al., 2011). Thus, we should pay more attention to whether the property of manure is one of the determining factors of CSE under long-term fertilisation. Although these elements are quite common uncertainties in long-term experiment studies, it should be noted that the estimate of rhizosphere C input may help us to reduce the uncertainty.

Additionally, there was a significant linear relation between cumulative C input and SOC stock change for each treatment when combining the data from all sites (Figure 2-S4). Based on the fact that there was a curvilinear relation between SOC change and C input rates obtained by data from 14 long-term experiments with durations of 12 to 96 years (Stewart et al., 2007), a small range of C input levels (fertilisation durations) will not necessarily reflect the full range of linear to asymptotic behaviours when a soil is subject to C saturation. Future studies should focus on CSE characteristics and its main controlling factors when a large amount of C has been accumulated.

5. Conclusion

Our study revealed that nearly four-decades of combined chemical NPK fertilisers and manure application significantly promoted SOC sequestration rate and efficiency in drylands of China. The improvement of soil chemical properties, which was a consequence of the manure application, was the main driver of soil carbon sequestration efficiency in manure amendment. Additionally, the soil C/N ratio and clay content most contributed to CSE across the main dryland region of China. The controlling factors of CSE varied considerably among the different treatments. Soil total N was the limiting factor for CSE of CK by controlling C input, whereas the soil C/N ratio and pH were the main explanatory factors of the long-term chemical NPK fertilisation treatment. The negative impact of C input was the main driver of CSE with straw return due to low humification for crop straw and low stabilization of straw-derived carbon. In conclusion, our findings highlight that different factors are driving SOC sequestration efficiency when considering different fertilisation treatments across main drylands of China. Hence, our study can be considered as a reference to promote soil carbon sequestration under long-term fertilisations in wheat and corn production regions.

6. Supplementary Figures and Tables

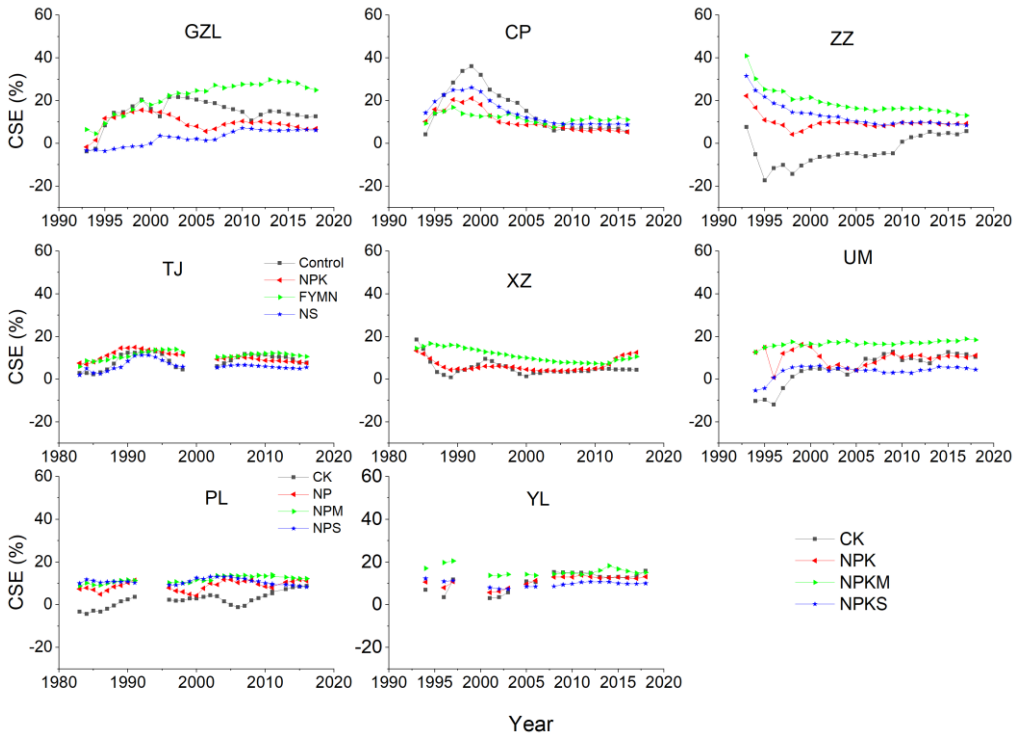


Figure 2-S1 Soil carbon sequestration efficiencies (CSE) (0–20 cm) of different treatments at the 8 long-term experiments. GZL means Gongzhuling; CP means Changping; XZ means Xuzhou; TJ means Tianjing; XZ means Xuzhou; UM means Urumqi; YL means Yangling; PL means Pingliang.

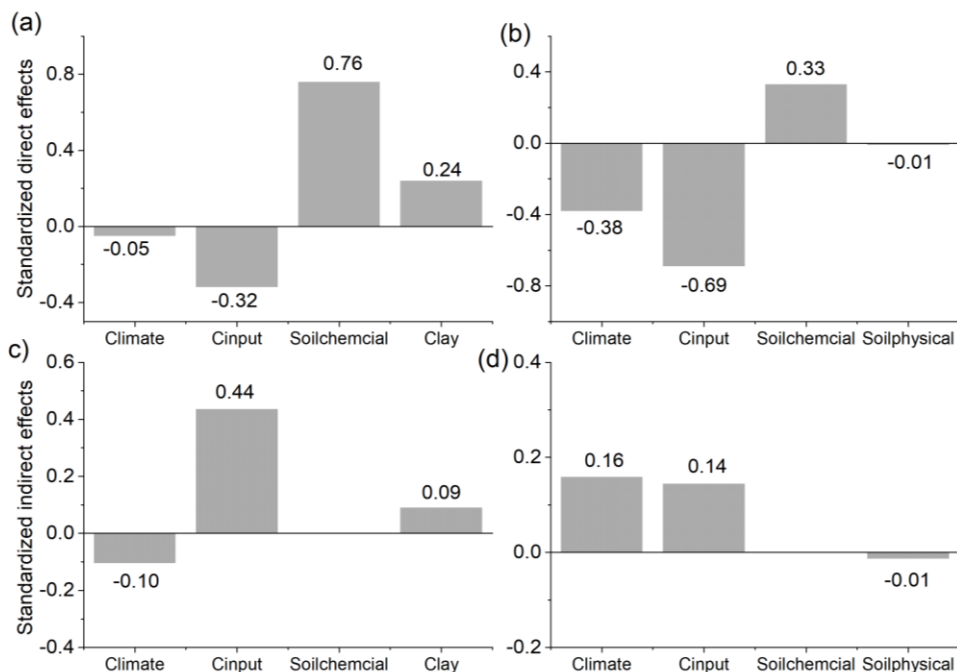


Figure 2-S2 Standardized direct and indirect effects of each variable from the structural equation model (SEM) under chemical fertilisers plus manure (a, c) and straw (b, d). The values adjacent to the column represent the standardized coefficients in SEM. Soil_{chemical} means soil chemical properties; Soil_{physical} means soil physical properties.

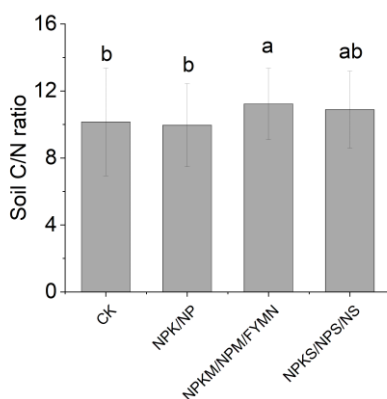


Figure 2-S3 Soil C/N ratio of different fertiliser treatments with all data from the 8 sites combined. Numbers with different lowercase indicate significant differences among different treatments by using one-way analysis of variance (ANOVA) test ($P < 0.05$).

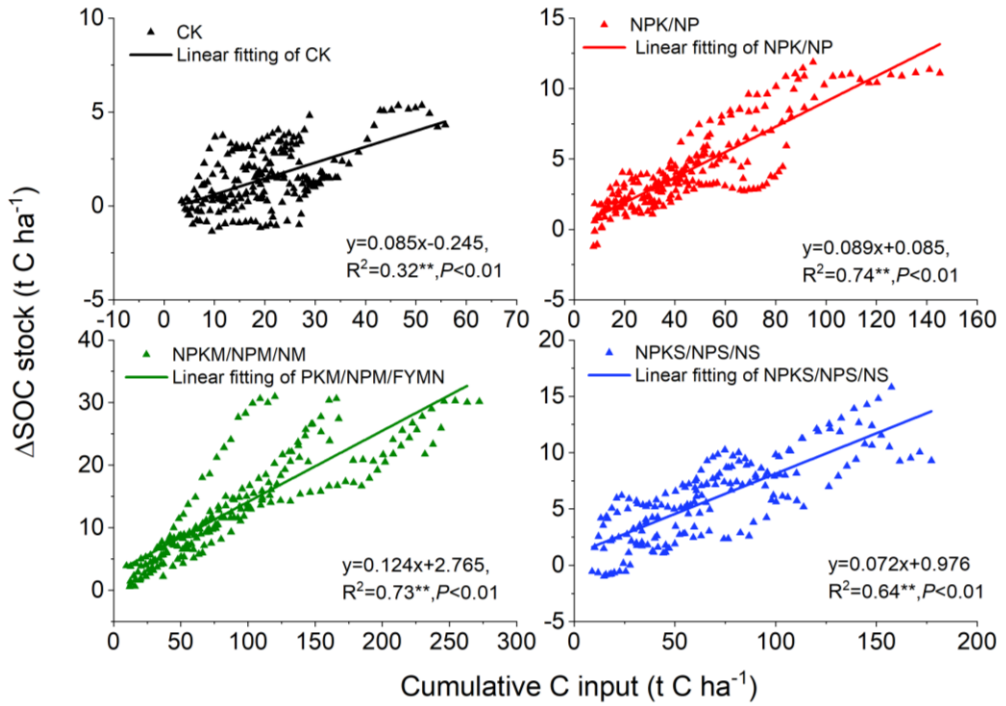


Figure 2-S4 Relation of cumulative C input and SOC stock change (Δ SOC stock: the difference as compared to initial status) for each treatment with data from all sites combined.

Table 2-S1 The locations, cropping systems and climatic characteristics of the studied sites

Site	Location	Climate type	Temperature	precipitation	Cropping system	
Gongzhuling (GZL)	124°48' E, 43°30' N	Mild temperate; Semi-humid	4.5	595	Single cropping	corn
Changping (CP)	116°15' E, 40°13' N	Warm temperate; Semi-humid	11.0	600	Double cropping;	Wheat-corn
Tianjin (TJ)	116°57' E, 39°25' N	Warm temperate; Semi-humid	11.6	607	Double cropping;	Wheat- corn
Zhengzhou (ZZ)	113°41' E, 35°00' N	Warm temperate; Semi-humid	14.1	645	Double cropping;	Wheat- corn
Xuzhou (XZ)	113°40' E, 34°47' N	Warm temperate; Semi-humid	14.0	860	Double cropping;	Wheat-Corn
Urumqi (UM)	87°46' E, 43°57' N	Mild temperate; Semi-arid	7.7	310	Single cropping;	Corn-wheat-wheat
Pingliang (PL)	107°30' E, 35°16' N	Mild temperate; Semi-arid	8.0	540	Single cropping;	Corn-wheat
Yangling (YL)	108°00' E, 34°17' N	Warm temperate; Semi-arid	13.0	634	Double cropping;	Wheat-Corn

Table 2-S2 Nitrogen, phosphorus and potassium application rates in different fertiliser treatments at the 8 long-term experimental sites across China (kg ha^{-1}).

Treatment	GZL		CP		TJ [#]			ZZ		XZ		YL		PL ^{&}	UM
	C	W	C	W	C	W	C	W	C	W	C	W/C	W/C		
	<u>Nitrogen (kg N ha^{-1})</u>														
NP/NPK	165	150	150	285	210	165	188	150	150	165	188	90	242		
NPKM	50+	150+	150	285+	210	50+	188	150+	150	50+	188	90+	85+		
	115*	146*		54/129*		115*		237*		115*		40*	240*		
NPKS	112+	150+	150	285+	210	123+	188	-	-	123+	188	90+	217+		
	53*	18*		14*		42*				42*		40*	29*		
	<u>Phosphorus (kg P ha^{-1})</u>														
NP/NPK	36	33	33	142.5	-	36	41	33	33	58	25	33	60		
NPKM	36+	33+	33	16*	-	36+	41	33+	33+	58+	25	33+	22+		
	39*	63*				66*		45*	70/51*	106*		200*	65*		
NPKS	36+	33+	33	15*	13*	36+	41	-	-	58+	25	33+	51+		
	6*	3*				8*				4*		22*	5*		
	<u>Potassium (kg K ha^{-1})</u>														
NPK	68	37	37	71.3	-	68	78	93.4	93.4	69	78	-	47		
NPKM	68+	37+	37	66*	-	68+	78	93.4+	93.4+	69+	78	-	10+		
	77*	78*				92*		200/110*	226/115*	139*		-	160*		
NPKS	68+	37+	37	239*	107*	68+	78	-	-	69+	78	-	39+		
	58*	23*				86*				86*		-	54*		

		<u>Types of manure or straw</u>						
straw	Corn	Corn	Wheat/ Corn	Corn	Corn	Corn	Wheat/ Corn	Wheat/ Corn
manure	Cow, pig	Pig	Chicken with furnace ash, Chicken	Horse, cow	Cow	Cow	Cow, pig	Sheep

C means corn; W means wheat; # No P and K application at TJ for chemical fertiliser plus manure/straw treatments; & No K application at PL;
 *The amount of nitrogen, phosphorus and potassium from manure or straw; - means no application.

Table 2-S3 Carbon (C) contents in shoot and root of cereals, root: shoot ratio (RSR) and harvest index (HI), and manure C content.

Sites	Crop	Shoot C (g/kg)	Root C (g/kg)	RSR	HI
All long-term experiments	Wheat ¹⁻⁴	434	425	0.29*	0.41-0.45*
	Corn ¹⁻⁵	433	412	0.12*	0.46-0.52*
	Manure	C: 17.45-41.38% (fresh)*			

* means measured data; 1-5 are references, and the value in this table is the average value from the references (Cai et al., 2015; E et al., 2018; Li et al., 2015; Wang et al., 1989; Wang et al., 1991).

Table 2-S4 The duration (n) when CSE had no significant difference with the latest year, and the ANOVA test results.

Sites	Latest year	Significant difference year	n	df	Mean square	F	P
Gongzhuling	2018	2007	12	1	159.90	12.25	0.03*
Changping	2016	2003	14	1	244.60	20.42	0.00***
Zhengzhou	2018	2009	10	1	1401.43	8.36	0.01**
Tianjin	2016	2003	14	1	41.10	6.60	0.02*
Xuzhou	2016	2000	16	1	237.37	79.47	0.00***
Urumqi	2018	2007	12	1	277.93	5.72	0.03*
Pingliang	2016	2008	9	1	94.20	6.45	0.03*
Yangling	2018	2008	11	1	168.12	9.10	0.01**

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

Table 2-S5 Independent samples t-tests for the CSE difference between NPK/NP and NPKS/NPS/NS.

Control	Treatment	Sig. (L)	T	Sig. (t)	n
NPK/NP	NPKS	0.784	2.663	0.009**	63

** $P < 0.01$.

Table 2-S6 Average annual C input ($\text{t C ha}^{-1} \text{ yr}^{-1}$) from crop residue (stubble + root + rhizosphere) at all sites.

Treatments	Double cropping system		Single cropping system	
	Wheat	Corn	Wheat	Corn
	Zhengzhou		Pingliang	
CK	0.63	0.72	0.59	2.00
NPK	1.90	1.46	1.36	2.19
NPKM	1.96	1.60	1.51	2.63
NPKS	1.96	1.52	1.25	2.35
	Xuzhou		Urumqi	
CK	0.60	0.73	0.51	1.43
NPK	1.93	1.46	1.60	2.29
NPKM	2.46	1.59	1.52	2.17
NPKS	-	-	1.24	2.12
	Tianjin		Gongzhuling	
CK	0.64	0.87	-	0.96
NPK	1.67	2.24	-	2.50
NPKM	1.71	2.28	-	2.57
NPKS	1.16	1.71	-	2.42
	Changping			
CK	0.59	0.76		
NPK	1.23	1.39		
NPKM	1.61	1.68		
NPKS	1.65	1.54		
	Yangling			
CK	0.61	0.76		
NPK	1.93	1.46		
NPKM	2.14	1.97		
NPKS	2.05	1.80		

Chapter III

Soil carbon sequestration efficiency decreased exponentially with carbon input - Based on four classical long-term experiments

Abstract

Soil organic carbon (SOC) is affected not only by the amount of C input but also by its conversion efficiency (i.e., CSE, $\Delta\text{SOC}/\Delta\text{C input}$), which has been observed to decrease as soils approach C saturation. A better understanding of the CSE characteristics changed with a large amount of C input accumulation (corresponding to long fertilisation durations) is urgently needed to effectively sequester C in agroecosystem, thereby improving soil fertility and mitigating climate change. We chose four classical long-term experiments, namely Broadbalk and Hoosfield in the UK (>170 years), as well as Morrow and Sanborn in the USA (>130 years). They are all dryland with similar fertiliser treatments, including no fertiliser (CK), combined mineral nitrogen, phosphorus and potassium (NPK), manure (FYM), and chemical N plus FYM (FYMN). Based on the data and records of crop yield, soil properties, and climate conditions during the whole fertilisation period, the study also assessed the key drivers of soil CSE in the plough layer. The results showed that practises with manure application had more significant fertilisation effects on SOC sequestration rate and efficiency compared with chemical fertilisation. The XGBoost regression modelling indicated that C input, MAT, and TN stock were crucial factors influencing CSE across four sites with more than a century of fertilisations. Moreover, clay content played the most important role in the Broadbalk and Hoosfield experiments. Initial SOC was another controller of soil CSE in the Morrow and Sanborn experiments. In addition, the CSE decreased rapidly in the Chromic Luvisol of the Broadbalk and Hoosfield experiments until C input reached 123–315 t C ha⁻¹, and a similar trend was observed in the Mollisols of the Morrow and Sanborn experiments before C input levels of 93–145 t C ha⁻¹. Then, the CSE decreased more slowly until it approached a more stable, equilibrium level. The relationship between CSE and cumulative C input fitted well with the negative exponential function (i.e., $\text{CSE} = b + a * e^{-k * \text{Cinput}}$, $P < 0.01$). The highest CSE asymptotic value (i.e., b) was found in practises with manure application. This study demonstrated the negative exponential change behaviour of soil CSE in response to C input accumulation and helped target the optimal fertiliser strategies to enhance both rate and efficiency of soil carbon sequestration under long-term fertilisations.

Keywords: soil organic carbon sequestration, classical long-term experiment, carbon sequestration efficiency, manure application, long-term fertilisation

1. Introduction

Soil organic carbon (SOC) is one of the most important components of soil, recognized for its significant role in soil productivity and fertility (Bauer and Black, 1994). In recent decades, the enhancement of SOC sequestration in agricultural soils has received worldwide attention as an effective strategy for mitigating climate change (Amundson and Biardeau, 2019). Generally, the SOC stock increased directly by incorporating added C. However, the relation between SOC stock and C input varies in different studies, and the slope of the relationship function is commonly considered as the soil carbon sequestration efficiency (i.e., CSE, $\Delta\text{SOC}/\Delta\text{C}$ input). Some suggested a linear relation (Bhattacharyya et al., 2010; Liang et al., 2016; Liu et al., 2019; Maillard and Angers, 2014; Majumder et al., 2008; Zhang et al., 2010), whereas others found a no-linear (e.g., decreased logarithmic or exponential) correlation based on long-term experiments (Cai and Qin, 2006; West and Six, 2007; Yan et al., 2013). The latter emphasized a decreased CSE as the soil approached C saturation (Hassink and Whitmore, 1997; Six et al., 2002). In addition, a small range of C input levels (corresponding to shorten fertilisation durations) will not necessarily reflect the full range of linear-to-asymptotic behaviours of SOC, and thus the CSE characteristics (Stewart et al., 2007). Thus, classical long-term experiments that exhibited full SOC behaviours under more than a century of fertilisation could be an essential tool to study the full characteristics of soil CSE.

Long-term experiments are typically initiated to assess the impacts of fertiliser management practises on crop production, aiming to identify the optimal fertiliser practises for a specific region (Johnston and Poulton, 2018; Macdonald et al., 2018). As society and technology evolve, long-term experiments offer a vital resource for studying plant growth and soil properties, particularly those attributes that develop incrementally over time, such as soil fertility. These experiments provide a unique opportunity to observe and understand the dynamics of ecological systems and to inform more sustainable agricultural practises. There are many long-term experiments around the world, but many of them may not meet our specific research requirements regarding the status of approaching SOC saturation or equilibrium with a broad C input accumulation. However, the classical long-term experiments established earlier in Europe and North America, such as the Broadbalk and Hoosfield at Rothamsted Research in the UK, as well as the Morrow plots and Sanborn fields in the USA, which are the oldest and most representative experiments of the two regions (Johnston and Poulton, 2018), may fulfil our research requirements (Li et al., 1994; Johnston et al., 2009; Miles and Brown, 2011). The test results from those long-term experiments on cereal production have demonstrated that both chemical fertilisation and organic manure application is capable of sustaining high yield production over a period of more than a century (Johnston and Mattingly, 1976). However, these experiments exhibited varying capacities for soil C sequestration, achieving different levels of SOC, and thus exhibited differing soil CSE. Therefore, we chose these four long-term experiments to examine how soil CSE responds to the accumulation of C inputs under different long-term fertiliser practises.

Studies have demonstrated that soil CSE mainly depends on climatic conditions, soil types, and fertilisation practises. Our results in Chapter II derived from experiments with soils far from C saturation suggested that edaphic factors, especially soil C/N ratio and clay content, were more important CSE drivers than C input and climate factors. However, a summary of 25 long-term experiment, encompassing soils in both C equilibrium and disequilibrium states, revealed that C input was the most important controller for CSE, followed by soil properties, management and climate (Cai, 2016). This may be due to the large range of C input levels observed across the 25 long-term experiments. Thus, as analysed in Chapter II, a comprehensive study of the contribution of each individual influencing factor to soil CSE with a large amount of C input accumulation is necessary to fully understand the characteristics of CSE under long-term fertilisations.

Based on the data of the Broadbalk and Hoosfield long-term experiments in the UK and the Morrow and Sanborn long-term experiments in the USA, our research focused on (1) the response of soil CSE in plough depth to different fertiliser treatments, (2) the relation between CSE and C input, and (3) the main drivers of soil CSE with a large amount of C input accumulation.

2. Materials and methods

2.1. Study sites and experimental design

Four classical long-term experiments are chosen, including 1) the Broadbalk and Hoosfield long-term experiments in the UK, and 2) the Morrow and Sanborn long-term experiments in the USA. Detailed information about the four experiments can be found elsewhere (Hutchinson, 1963; Khan et al., 2007; Liang et al., 2008; Miles and Brown, 2011; Wilhelm and Wortmann, 2004). The basic background information, including the locations, soil properties, cropping systems and climates, are shown in Table 3-1.

Table 3-1 Locations, cropping systems, climatic conditions and initial soil properties of the studied sites

Sites	Broadbalk	Hoosfield	Morrow	Sanborn
Region	Southeast England		Central plain of the USA	
Starting year	1843	1852	1904	1888
Location	51°48' N, 0°20' W	51°48' N, 0°22' W	40°06' N, 88°13' W	38°57' N, 93°20' W
Climate type	Temperate maritime climate	Temperate maritime climate	Temperate continental climate	Temperate continental climate
Temperature	10.2	10.2	11.4	12.9

Precipitation	718	718	1043	1058
Cropping system	Single cropping; Continuous wheat; Oat-corn-wheat-wheat-wheat	Single cropping; Continuous barley	Single cropping; Continuous corn; corn-oat/corn-soybean (after 1967)	Single cropping; Continuous corn/wheat; corn-wheat-red clover
Initial soil properties				
Soil type	Chromic Luvisol	Chromic Luvisol	Mollisol	Mollisol
Initial SOC, g kg ⁻¹	10.00 ^a	11.32 ^b	55.0 ^f	40.0 ^h
Total N, g kg ⁻¹	1.10 ^a	1.26 ^b	4.21 ^f	4.00 ^h
pH	7.82 ^c	7.3 ^d	5.89 ^g	5.45 ⁱ
Bulk density, g cm ⁻³	1.25 ^a	1.18 ^b	1.00 ^f	1.12 ^h
Clay, %	25 ^a	25 ^c	27 ^f	20 ^h

a: estimated value of 1843; b: measured data of 1852; c: average measured values of 1865; d: average measured values of 1965; e: estimated value of 1852; f: estimated value of 1876; g: average measured values of 1967; h: estimated value of 1888; i: average measured values of 1938.

2.2. Fertilisation

Four fertiliser treatments common to all experiments were chosen: (1) no fertiliser (CK), (2) combinations of chemical N, phosphorus (P) and potassium (K) fertilisers (NPK), (3) chemical fertilisers combined with manure (FYMN and NPKM), and (4) manure only (FYM). The application rates of chemical N, P and K fertilisers, and manure are presented in Table 3-2. The types of manure were dependent on the local availability (Tables 3-2, 3-3).

Table 3-2 Application rates of nitrogen, phosphorus, potassium (kg ha⁻¹ yr⁻¹), and manure (t ha⁻¹ yr⁻¹) in different fertiliser treatments

Sites	Treatments	N	P	K	Manure	Straw
Broadbalk	CK, NPK, FYM, FYMN	96, 144	35	90	35	/
Hoosfield	CK, NPK, FYM	0, 48, 96, 144	35	90	35	/
Sanborn	CK, NPK, FYM, FYM-NPK	0-112	0-38	0-16.6	13.4/20.2	All of residual
Morrow	CK, NPK, FYM, FYM-MNPK	224	56	336	4.5	All of residual

Note: FYM-NPK means the fertilisation changed from FYM to NPK during the experimental period; FYM-FYMN means the fertilisation changed from FYM to FYMN during the experimental period.

In the Broadbalk experiment, we chose the continuous wheat system and the 5-yr oat–corn–wheat–wheat–wheat rotation. The annual N application amount was 144 kg ha⁻¹ yr⁻¹ for the NPK treatment. The average N application rate for FYM was 224 kg ha⁻¹ yr⁻¹. Notably, the chemical N application of FYMN started in 1968, and the application rate was 96 kg ha⁻¹ yr⁻¹ before 2004 and later as 144 kg ha⁻¹ yr⁻¹. The fields were divided into subplots in 1926 and 1968. The fertiliser management in the rotation system was the same as that in the continuous wheat cropping system, while there was no N or FYM application in the oat planting years. In the Hoosfield experiment, the N application rate was 48 kg ha⁻¹ yr⁻¹ before 1968 and later as a rotation of 0-48-96-144 kg ha⁻¹ yr⁻¹ for the NPK treatment. The manure application rate in the Hoosfield experiment is the same as that in the Broadbalk experiment.

In the Sanborn experiment, we chose the continuous wheat system (CK, NPK and FYM), the continuous corn system (CK and FYM), and the 3-yr corn–wheat–red clover rotation (CK, FYM-NPK and FYM-FYMN). The average N application rate for the FYM/FYMN treatment was 41 kg ha⁻¹ yr⁻¹. For the NPK treatment in the continuous wheat system, the application rate of N, P, and K was 56, 9.9, and 18.6 kg ha⁻¹ yr⁻¹, respectively. For the FYM-NPK treatment in the rotation, the average annual application rate of N from manure was 41 kg ha⁻¹ yr⁻¹ before 1913 and increased to 62 kg ha⁻¹ yr⁻¹ by 1928. Additionally, a rate of 16 kg ha⁻¹ yr⁻¹ of phosphorus from lime was added to the plots receiving the FYM-NPK before 1950. Subsequently, an average application rate of 50, 8.7 and 16.6 kg ha⁻¹ yr⁻¹ of N, P, and K was applied, respectively, in accordance with the soil test. All crop residues were retained and incorporated into the plot of origin after 1950. In the rotation system, additional 112 and 37 kg ha⁻¹ yr⁻¹ of chemical N was applied to the FYM-FYMN treatment after 1950 for corn and wheat, respectively, in accordance with the soil test.

In the Morrow experiment, we chose the continuous corn system and the 2-yr corn–oat (before 1967)/soybean (after 1967) rotation (CK, FYM-NPK and FYM). The chosen plots except CK were all treated with manure before 1955, and the average N application rate from manure was 75 kg N ha⁻¹ yr⁻¹. For the NPK treatment, a total rate of 224-56-336 kg N-P-K ha⁻¹ yr⁻¹ was applied to NPK plots after 1954. Grain and straw were removed, while crop stubble and root residues were returned to the original plot from 1905 to 1955. All crop residues were returned to NPK plots from 1955 and to all plots from 1967.

2.3. Soil sampling and analysis

In the Broadbalk experiment, soil samples from the plough layer (0–23 cm) were collected approximately every 20 years up until 1987, after which the collection frequency was revised to every five years. Selected treatments from 1881, 1893, 1914, 1936 and 1944 were reanalysed in 2001–2004 for soil total C and total N by combustion (LECO) and for CaCO₃-C by manometry (Kalembasa and Jenkinson,

1973). The data from 1992, 1997, 2000, and 2005 were also analysed by combustion and manometry. Notably, the organic C values used in this study are the total C minus $\text{CaCO}_3\text{-C}$. The data from 1865 were derived from the original Soda lime analyses (for TN) and soil C/N ratios from 1893 (for SOC). The SOC and TN from 1966 were analysed by the Walkley-Black method (Walkley and Black, 1934) and the Kjeldahl method (Tinsley, 1950), respectively. The data from 1987/88 were analysed by the Tinsley method for SOC and by the Kjeldahl method for TN (Bremner and Mulvaney, 1982; Tinsley, 1950). The soil samples (0–23 cm) in the Hoosfield experiment were collected in 1882, 1913, 1946, 1965, 1975, 1982, 1998, 2008, 2013 and 2017. Soil TN was measured by the Kjeldahl method (Bremner and Mulvaney, 1982). Soil total C was determined gravimetrically by dry combustion, carbonate C was determined gravimetrically, and organic C was determined by difference (Kalembasa and Jenkinson, 1973). Detailed information regarding the sampling and analysis methods employed for the Broadbalk and Hoosfield long-term experiments can be found in Hutchinson (1963), Experimental Station Rothamsted (1970), Jenkinson and Johnston (1977), and the electronic Rothamsted Archive (e-RA) (<https://www.era.rothamsted.ac.uk/dataset/rbk1/02-BKSOC1843> and <http://www.era.rothamsted.ac.uk/Hoos/hfsoils#SEC5>).

The soil samples from 0–20 cm of the Sanborn experiment's selected plots in 1915, 1928, 1938, 1948, 1962, 1988, 1999, 2008 and 2014 were used for the study. The SOC content was measured with a LECO CR-12 carbon analyser following method 6A2 carbon by dry combustion (Miles and Brown, 2011). Soil samples from the surface layer (0–15 cm) were collected and analysed approximately every 10 years between 1904 and 1953, almost every five years from 1955 to 1974, and annually from 1980 to 1993 in the Morrow long-term experiment. Samples from 2005, 2007, and 2008 were also analysed. The analyses for total organic C were performed by the dichromate oxidation technique of Mebius (1960). For the aim of comparing soil CSE of long-term experiments with the same soil type (i.e., Mollisol), the SOC measurements from Morrow's 0–15 cm layer were extrapolated to a 20 cm depth based on the assumption of an equal distribution of SOC within the top layer. To obtain the SOC content for the 0–20 cm layer of the Morrow plots, the SOC content of the 0–15 cm layer was multiplied by 1.33. Details on the sampling and analysis methods employed in the Morrow and Sanborn experiments can be found in Odell et al. (1982, 1984), Smith (1942), and Liang et al. (2008).

2.4. Soil carbon sequestration efficiency calculation

This section gives the different equations used to compute soil C fluxes, stocks and sequestration efficiency.

The SOC stock and C input calculations used the same equations that were presented in Chapter II. Notably, $P_{stubble}$ is 20% for all treatments in the Broadbalk and Hoosfield experiments (pers. Comm., experience value), and 10% for all treatments in the Morrow and Sanborn experiments (Khan et al., 2007; Liang et al., 2008); $P_{stubble}$ is 100% when all crop residues are returned; P_{root} is 75.3% for wheat and oats, 85.1% for corn and 70% for soybean, bean, red clover, and cowpea, respectively (Bolinder

et al., 2007a; Jiang et al., 2018a; Jiang et al., 2014; Khan et al., 2007); The average percentage of moisture content in the shoot (M_{shoot}) and root (M_{root}) are based on the observations of the air-dried crop samples when they are available; otherwise, they are set to 14% (Jiang et al., 2018a); The percentage of moisture content of manure (M_{manure}) is set to 70% (the average value of all observations). Straw yields for the Morrow experiment in unrecorded years, as well as root biomass for all years of the four experiments, have been established. Other indexes about C input calculation are shown in Table 3-3.

Table 3-3 Carbon (C) contents in shoot and root of cereals and manure, and root: shoot ratio (RSR) and harvest index (HI) for calculating C input in the four classical long-term experiments

Sites	Cereal	Shoot C g/kg	Root C g/kg	RSR	HI	
UK	Wheat ¹⁻⁶	456	375	0.39 ¹²⁻¹³	-	
	Bean ⁷	429	363	0.10 ¹⁴⁻¹⁵	-	
	Broadbalk	Oats ⁸	428	428	0.24 ^{11, 16}	-
		Silage corn ⁹⁻¹¹	438	400	0.19 ¹⁷⁻¹⁸	-
		Manure (average)	dairy manure C: 35.80% (dry) *			
	Hoosfield	Barley ^{13,19,20}	446	397	0.40 ^{12,21}	-
		Manure (average)	dairy manure C: 35.44% (dry) *			
	Morrow ²²⁻²³	Corn	437	343	0.5	0.5
		Soybean	454	467	0.6	0.4
		Oats	440	440	0.8	0.5
Manure (average)		dairy manure C: 47.0% (dry) *				
USA	Wheat ²⁴	370	296	0.88	0.33 (average)*	
	Sanborn	Corn ²⁴	409	261	1.0	0.44 (average)*
		Red clover ²⁵⁻²⁹	431	460	0.4	/
		Cowpea ³⁰	442	430	0.2 ³¹⁻³²	/
		Manure (average)	horse manure C: 49.5% (dry)*			

*Measured data; ¹⁻³² are references, and the value in this table is the average value from the references (Angers, 1992; Berg et al., 1987; Biscoe et al., 1975; Buyanovsky and Wagner, 1986; Chapman, 1997; Chirinda et al., 2012; Christie et al., 1992; De Manzi and Cartwright, 1984; Franzluebbers et al., 1994; Greenhalf et al., 2012; Hall et al., 1914; Henriksen and Breland, 2002; Hibberd et al., 1996; Khan et al., 2007; Komainda et al., 2018; Laubach et al., 2019; Lentz and Ippolito, 2012; McKendry, 2002; Nafziger and Dunker, 2011; Rengasamy and Reid, 1993; Robinson et al., 1993; Ross et al., 2018; Seremesic et al., 2017; STÜTZEL and AUFHAMMER, 1991; Sutherland et al., 1985; Triberti et al., 2008; Van Vuuren et al., 2000; Welbank et al., 1974; Whitehead, 1970; Yang et al., 2002; Zhang et al., 2004).

After calculating the SOC stocks in the plough layer for the four classical long-term experiments, we found that the SOC stocks exhibited nonlinear increases or decreases over the entire experimental period (Figure 3-1). Therefore, this study used the relative change in SOC stocks under fertiliser treatments compared to those under the no-fertiliser to calculate the C sequestration rate and efficiency.

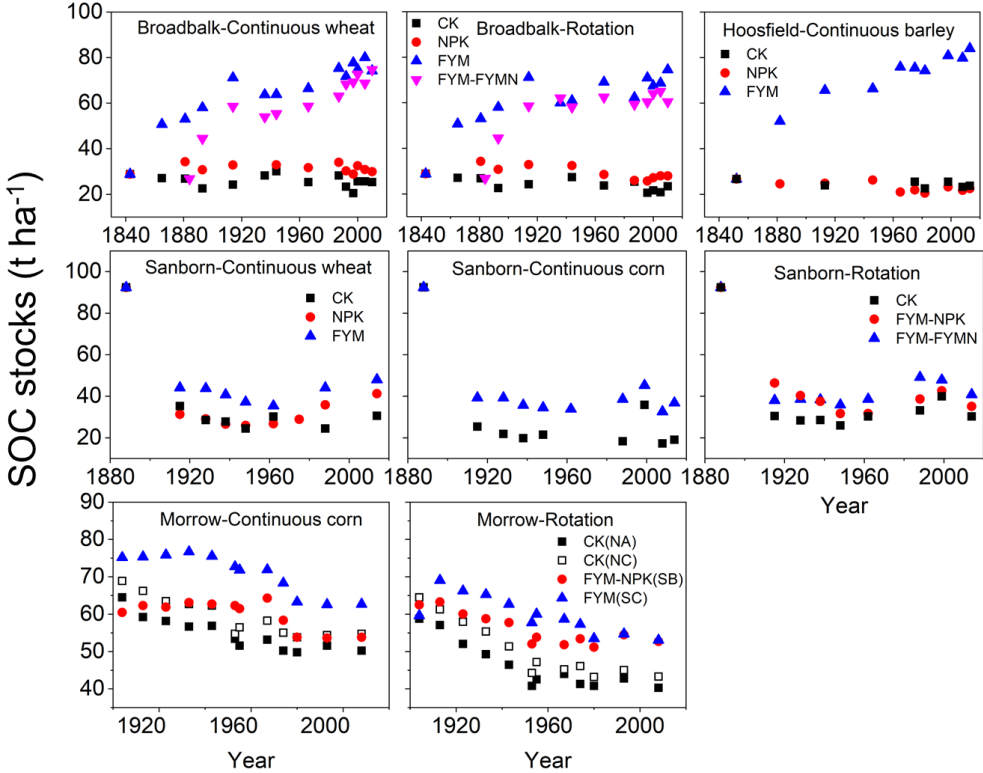


Figure 3-1 Soil organic carbon stocks ($t C ha^{-1}$) in the plough layer of the four classical long-term experiments. FYM-NPK means the fertilisation changed from FYM to NPK during the experimental period for the rotation; FYM-FYMN means the fertilisation changed from FYM to FYMN during the experimental period for the rotation.

SOC sequestration rate

The SOC sequestration rate with fertilisation ($C_{sequestration\ rate}$, $t C ha^{-1} yr^{-1}$) in the plough layer during the experimental period was calculated using the following:

$$C_{sequestration\ rate} = \frac{(SOC_F - SOC_{initial-F}) - (SOC_{CK} - SOC_{initial-CK})}{t} \quad (1)$$

where SOC_F are the SOC stocks of NPK, FYMN and FYM treatments in the t-th year, and SOC_{CK} is the SOC stock of CK treatment in the t-th year; t is the experimental duration. $SOC_{initial-F}$ is the initial SOC stock of plot with fertilisations and $SOC_{initial-CK}$ is the initial SOC stock of plot without fertilisation. If the calculation

showed a negative value, the SOC content of each treatment used the average value of three consecutive measurements around the t-th year to avoid this measurement errors.

Carbon sequestration efficiency

The carbon sequestration efficiency (CSE, %) of fertilisation treatments is the ratio between the SOC stock change and C input both relative to the CK equivalent value (Jiang et al., 2018a).

$$CSE = \frac{SOC_F - SOC_{CK}}{cumulative\ Cinput_F - cumulative\ Cinput_{CK}} \times 100\% \quad (2)$$

where *cumulative Cinput_F* and *cumulative Cinput_{CK}* are the total C input during t years for treatments with fertilisers (NPK, FYMN and FYM) and CK, respectively.

2.5. Influence factors

To identify the factors deriving CSE under long-term fertilisations when a large amount of C has been accumulated in studied regions, climate variables, management factors, and edaphic properties were collected. MAT and MAP as key climate factors controlling crop growth and OC mineralization were selected for analysis. Data for these variables were collected from the weather station at Rothamsted Research for the Broadbalk and Hoosfield experiments, and from the National Centers for environmental information (<https://www.ncdc.noaa.gov/cdo-web/>) for the Morrow and Sanborn experiments. For management factors, total C input from stubble, root, rhizosphere, and organic manure was calculated for further analysis. In consideration of edaphic properties, several key soil characteristics were chosen for analysis based on the limited available data from each site. These included initial SOC content, TN, soil C/N ratio, pH, BD and clay percentage. For the clay content, the average value was used for analysis when there was no record. For the years without measured BD in the Morrow and Sanborn experiments, the following function was used for the calculation:

$$BD = 100 / \left(\frac{1.727 \times SOC}{BD_{OM}} + \frac{100 - 1.727 \times SOC}{BD_{MIN}} \right) \quad (3)$$

where SOC is soil organic carbon content (%), BD_{OM} and BD_{MIN} are the bulk densities ($g\ cm^{-3}$) for organic matter and mineral matter, respectively (Adams, 1973). In this study, the specific BD_{OM} and BD_{MIN} values for the Morrow (0.185 and 1.85, respectively) are from Kwon and Hudson. (2019). The specific BD_{OM} and BD_{MIN} values for the Sanborn experiment (0.249 and 1.52, respectively) were derived using the same method described in Khan et al. (2007). The measurements from soil samples (n=72) collected on 8th May 2014 and 1st April 2016 were used for model calibration, and the measurements from soil samples (n=72) collected on 4th September 2014 and 18th August 2016 were used for model validation. This study used three statistical criteria to evaluate the model performance: (1) the coefficient of determination (R^2) that measures the degree of agreement between simulation and observation; (2) the

root mean squared error (RMSE), indicating the average deviation of estimated values from observed ones; and (3) the relative error (RE) that reflects the overall difference between simulated and observed data. The results are presented in Figure 3-2.

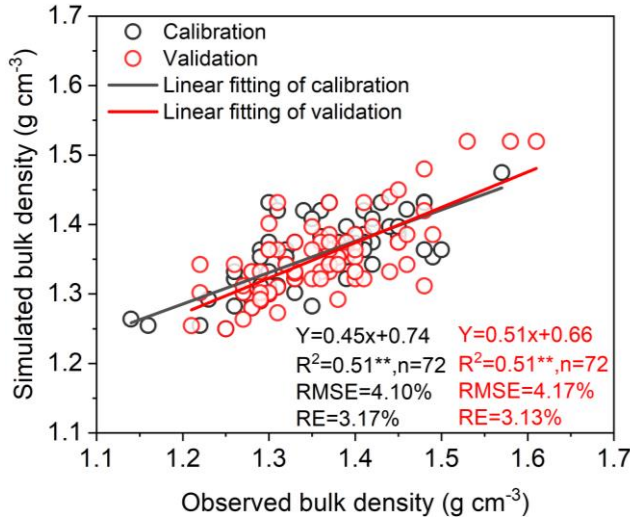


Figure 3-2 Relationship between simulated and observed bulk densities (g cm⁻³) in the plough layer of the Sanborn long-term experiment.

2.6. Statistical analysis

The study presented the results of SOC sequestration rates, C inputs and CSE by dividing the whole experimental phase into different periods. This division was based on the particular modifications in fertiliser application and/or field management that occurred during the long-term experimental period. By doing this, we were able to assess the effects of these changes on C sequestration in the soil over time. One-way ANOVA and the LSD ($P < 0.05$) test were applied to compare the effects of fertilisation on the annual C input from residues in an individual period. One-way ANOVA and the LSD ($P < 0.05$) test were also applied to compare the effects of cropping system on annual total C input in an individual treatment.

Generally, the threshold of a model that describes the relation between soil biodiversity/functional attribute (y) and influence factors (x) can be identified by two steps (Zhang et al., 2023). First, choosing suitable model from linear and nonlinear model (i.e., quadratic and generalized additive model), with the nonlinear model indicating the presence of a threshold point. Secondly, choosing suitable piecewise linear regression from four types of threshold effects that included step, hinge, segmented and stegmented regression based on the characteristics of the nonlinear model identified in step one. In our study, we utilized the Generalized Additive Model (GAM) to capture the complex trends in the data through the use of smoothing parameters. This model allows for a more nuanced description of the CSE dynamic curve by accounting for non-linear relationships (Fong et al., 2017; Zhang et al., 2023).

Following the GAM analysis, we applied segmented piecewise linear regression, a technique that divides the data into distinct segments and fits linear models to each segment (Fong et al., 2017). This approach was informed by the continuous threshold characteristics identified by the GAM regression, enabling us to pinpoint the inflection points of the CSE dynamic curve (CSE_{IP}) for each treatment. These inflection points represent significant changes in the CSE and are crucial for understanding the treatment effects over time (Fong et al., 2017; Zhang et al., 2023). We used the “gam” package and “segmented” package of R, version 4.3.0, for above analyses.

Extreme gradient boosted tree (XGBoost) regression modelling was applied to evaluate the feature importance of the studied climate variables, management factors, and edaphic properties to soil CSE, particularly considering the nonlinear changes of key factors, such as SOC and TN stocks. We used the “xgboost” package of R, version 4.3.0. Before conducting XGBoost analysis, the test of collinearity was considered by computing the variance inflation factor ($VIF < 10$) with SPSS 24.0 (SPSS, Inc., 2017, Chicago, USA). The XGBoost regression modelling refers to a tree ensemble model that can be used for classification and regression (Zhao et al., 2022). We chose the XGBoost regression because of the following two main advantages: (1) investigating complex and nonlinear relationships while maintaining high prediction accuracy and (2) estimating and ranking the feature’s relative importance and visualizing their marginal effects (Chen et al., 2023). The grid search method was used for the hyperparameter optimization of XGBoost, which estimated the model performance for each combination of the specified hyperparameters. The hyperparameters used for the XGBoost in this study are shown in Table 3-4. Due to limited measurements from each individual experiment, we combined data of experiments from the same country with similar soil types and weather conditions to explore the controllers of soil CSE in these two regions under long-term fertilisations.

Table 3-4 Hyperparameters in the XGBoost regression model

Index	Meaning	Value	
		Broadbalk & Hoosfield	Morrow & Sanborn
n rounds	The number of iterations in the training process	150	100
max_depth	Maximum depth of a tree	2	2
eta	The learning rate	0.4	0.4
gamma	Minimum loss reduction required to make a further partition on a leaf node of a tree	0	0

	colsample_	Column sampling rate of	0.8	0.8
	bytree	features when building each tree		
	min_child_	Minimum sum of the instance		
	weight	weights contained in child nodes	1	1
	subsample	Sampling rate of the training samples	0.75	1
Model performance	R ²	The coefficient of determination	0.86	0.60
	RMSE	The root mean squared error	2.74	4.41
	RPD	Relative percent deviation	2.70	1.62

3. Results

3.1. Changes in SOC stocks and C inputs of Broadbalk and Hoosfield

During the whole experimental period of the Broadbalk and Hoosfield long-term experiments, the SOC sequestration rates decreased with time for all treatments (Tables 3-5, 3-6). When considering the change of SOC stocks with different fertiliser treatments relative to CK, the SOC sequestration rate of FYM and FYMN was greater than that of NPK. Correspondingly, the annual C inputs of FYM and FYMN, with averages of 3.87–4.34 t C ha⁻¹ yr⁻¹ in Broadbalk and 3.65–3.72 t C ha⁻¹ yr⁻¹ in Hoosfield, were significantly higher than those of NPK and CK (Figure 3-3).

In the Broadbalk experiment, the fertiliser treatments did not indicate consistent characteristics in their carbon sequestration rates across different cropping systems (Table 3-5). The NPK and FYM treatments demonstrated very close values between the continuous wheat system and rotation. However, when considering the FYMN, the C sequestration rate of the rotation was lower than that of the continuous wheat system. In the Hoosfield experiment, the C sequestration rate for the NPK treatment in the continuous barley system was lower than that of both the continuous wheat and rotation systems in the Broadbalk. The C input from the crop in the continuous barley system (0.16–0.94 t C ha⁻¹ yr⁻¹) for all treatments was lower than that for both the continuous wheat (0.18–1.32 t C ha⁻¹ yr⁻¹) and rotation (0.19–1.45 t C ha⁻¹ yr⁻¹) systems in the Broadbalk experiment. The average C input from manure over the whole period was 2.89 t C ha⁻¹ yr⁻¹ in the Broadbalk and Hoosfield experiment.

3.2. Changes in SOC stocks and C inputs of Morrow and Sanborn

Similarly, compared with the SOC sequestration rate of the NPK (Morrow: 0.07–0.08 t C ha⁻¹ yr⁻¹, Sanborn: 0.04–0.27 t C ha⁻¹ yr⁻¹), FYM and FYMN had higher SOC change rates of 0.07–0.29 t C ha⁻¹ yr⁻¹ in the Morrow experiment and 0.08–0.38 t C ha⁻¹ yr⁻¹ in the Sanborn experiment (Tables 3-7, 3-8) associated with higher annual C inputs during the whole experimental periods (Figures 3-4 and 3-5). The SOC

sequestration rates decreased with time for most studied cropping systems in the Morrow (except continuous corn) and Sanborn (except continuous wheat with NPK) experiments.

The annual C input from crops for all treatments in the continuous corn of the Morrow experiment (without straw return: 0.45–1.49 t C ha⁻¹ yr⁻¹; with straw return: 2.01–6.26 t C ha⁻¹ yr⁻¹) was lower than that of the rotation (without straw return: 0.51–1.67 t C ha⁻¹ yr⁻¹; with straw return: 3.05–6.82 t C ha⁻¹ yr⁻¹) ($P < 0.05$) (Figure 3-4). However, the difference in the annual C input from crops between/among cropping systems in the Sanborn experiment varied among treatments (Figure 3-5). When considering the FYM/FYMN, rotation (without straw return: 1.20–1.37 t C ha⁻¹ yr⁻¹; with straw return: 3.11 t C ha⁻¹ yr⁻¹) > continuous wheat (without straw return: 1.02–1.17 t C ha⁻¹ yr⁻¹; with straw return: 2.96 t C ha⁻¹ yr⁻¹), continuous corn (without straw return: 1.05–1.08 t C ha⁻¹ yr⁻¹; with straw return: 2.80 t C ha⁻¹ yr⁻¹) ($P < 0.05$). In the NPK treatment where straw was not returned, the annual carbon input from crops was very close between the rotation (1.20–1.23 t C ha⁻¹ yr⁻¹) and continuous wheat (1.15–1.18 t C ha⁻¹ yr⁻¹) systems. However, when all straw was returned, the annual carbon input in the continuous wheat system (3.08 t C ha⁻¹ yr⁻¹) exceeded that of the rotation system (2.38 t C ha⁻¹ yr⁻¹). The average C input from manure over the whole period was 2.12 and 1.36 t C ha⁻¹ yr⁻¹ in the Morrow and Sanborn experiment, respectively. Similar to the Broadbalk and Hoosfield experiments, the Morrow and Sanborn experiments also did not yield consistent conclusions regarding SOC sequestration rates across the different cropping systems for all treatments (Tables 3-7, 3-8).

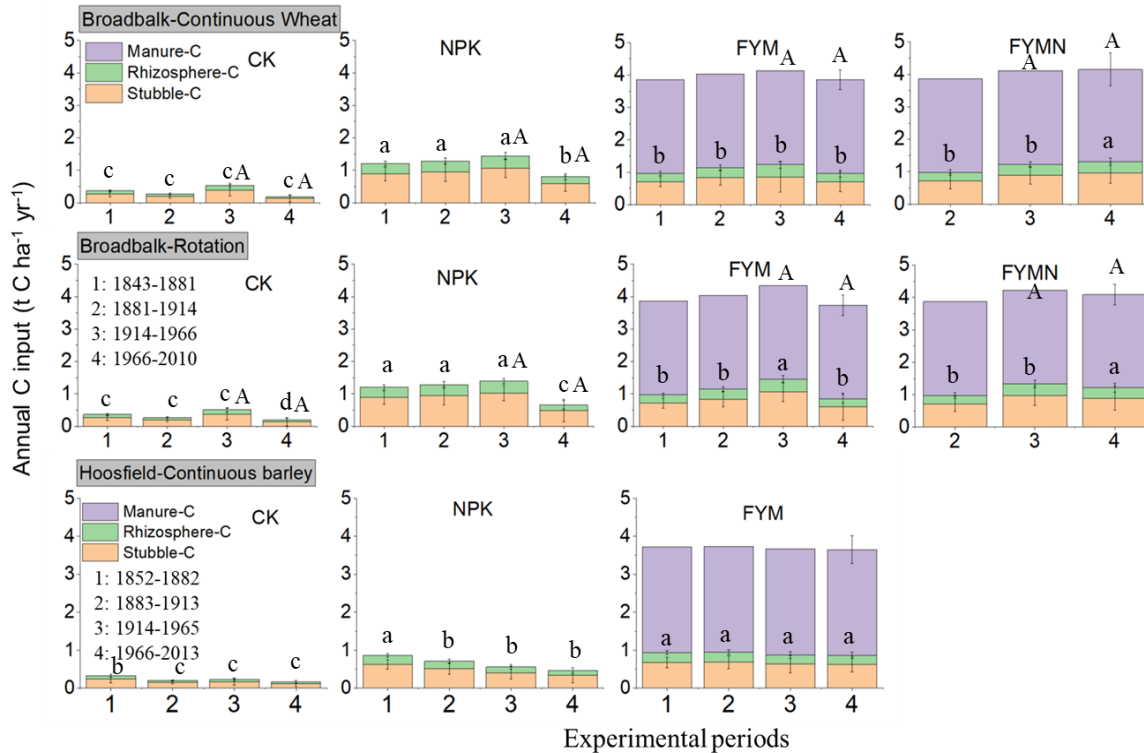


Figure 3-3 Average annual C input from different sources in Broadbalk continuous wheat system and rotation and Hoosfield continuous barley system. Numbers with different lowercase letters indicate significant difference ($P < 0.05$) for annual C input from residues (stubble + root + rhizosphere) under different treatments within an individual experimental period. Numbers with different capital letters indicate significant difference ($P < 0.05$) for annual C input between different cropping systems with the same fertilisation within an individual experimental period. The same as below.

Table 3-5 Soil organic carbon stocks (t C ha⁻¹) in some specific years and average sequestration rates (t C ha⁻¹ yr⁻¹) in the plough layer (0–23 cm) of the Broadbalk long-term experiment within an individual experimental period

Treatment	Initial SOC stock	<u>1881</u>		<u>1914</u>		<u>1966</u>				<u>2010</u>			
		Continuous wheat		Continuous wheat		Continuous wheat (Section I)		Continuous wheat (Section II)		Continuous wheat		Rotation ‡	
		stocks	rate	stocks	rate	stocks	rate	stocks	rate	stocks	rate	stocks	rate
CK	28.80	26.78	-	24.20	-	25.35	-	23.62	-	25.35	-	23.33	-
N3PK		34.27	0.20	32.83	0.12	31.68	0.05	28.51	0.04	29.96	0.04	27.93	0.04
FYM		53.04	0.69	71.17	0.66	66.41	0.33	69.08	0.37	74.22	0.40	74.49	0.42
FYMN	26.78			58.59	1.19	58.64	0.41	62.45	0.48	74.67	0.61	60.42	0.46

‡ The plots are located in the section II, which is divided from the continuous wheat system in 1926, and the section II was divided into subplots for rotations in 1968.

Table 3-6 Soil organic carbon stocks (t C ha⁻¹) in some specific years and average sequestration rates (t C ha⁻¹ yr⁻¹) in the plough layer (0–23 cm) of the Hoosfield long-term experiment within an individual experimental period

Treatment	Initial SOC stock	<u>1882</u>		<u>1913</u>		<u>1965</u>		<u>2013</u>	
		stocks	rate	stocks	rate	stocks	rate	stocks	rate
CK		26.48	-	26.80	-	24.15	-	26.90	-
NPK	30.71	28.30	0.06	27.80	0.03	24.11	0.004	25.83	0.003
FYM		59.90	1.11	75.50	0.80	87.20	0.56	96.60	0.43

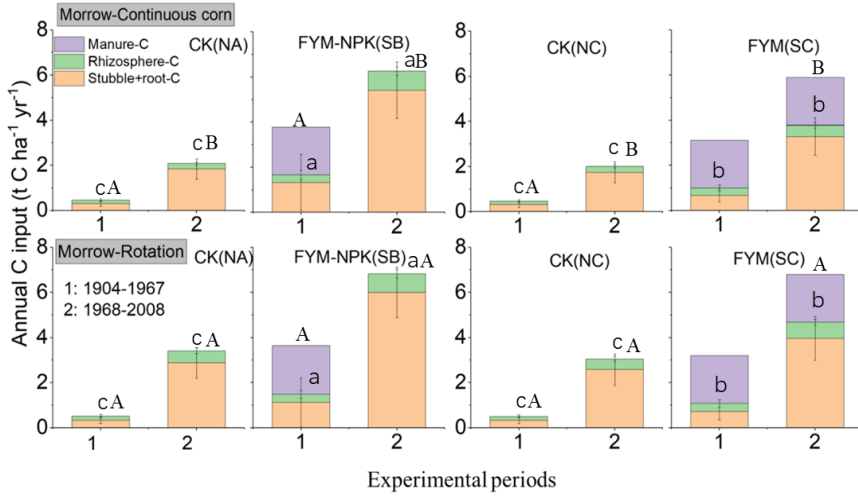


Figure 3-4 Average annual C input ($t C ha^{-1} yr^{-1}$) from different sources in continuous corn system and rotation system of the Morrow experiment. NA, SB, NC and SC mean different plots where the treatments are located.

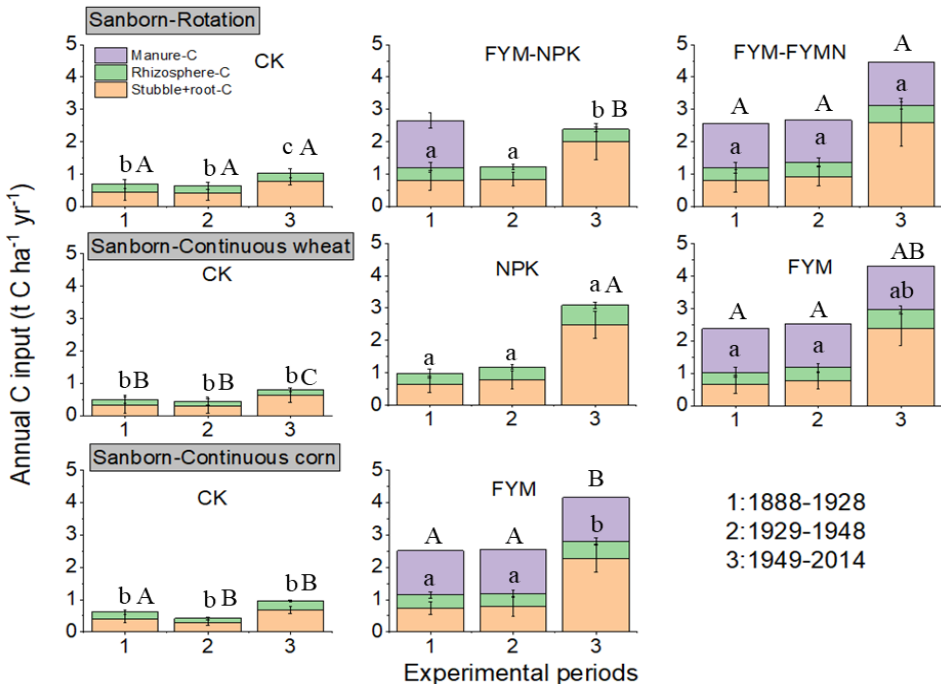


Figure 3-5 Average annual C input ($t C ha^{-1} yr^{-1}$) from different sources in continuous corn system, continuous wheat system and rotation system of the Sanborn experiment.

Table 3-7 Soil organic carbon stocks (t C ha^{-1}) in some specific years and average sequestration rates ($\text{t C ha}^{-1} \text{ yr}^{-1}$) in the plough layer of the Morrow (0–20 cm) long-term experiment within an individual experimental period

Treatment	Initial SOC stock (1904)		<u>1968</u>				<u>2008</u>			
	Continuous corn	Rotation	Continuous corn		Rotation		Continuous corn		Rotation	
			stock	rate	stock	rate	stock	rate	stock	rate
CK (NA)	58.79	64.48	44.28	-	50.91	-	40.27	-	50.21	-
FYM-NPK (SB)	62.48	60.45	52.93	0.08	61.07	0.22	52.72	0.08	53.83	0.07
CK (NC)	63.48	70.89	44.64	-	55.15	-	43.29	-	54.71	-
FYM (SC)	59.62	73.18	59.21	0.29	70.37	0.20	53.05	0.14	64.69	0.07

Note: NA, SB, NC and SC are the plots where the treatments are located; FYM-NPK means the fertilisation changed from FYM to NPK during the experimental period.

Table 3-8 Soil organic carbon stocks (t C ha⁻¹) in some specific years and average sequestration rates (t C ha⁻¹ yr⁻¹) in the plough layer (0–20 cm) of the Sanborn long-term experiment within an individual experimental period

Treatment	Initial SOC stock (1888)	Year	Continuous wheat		Continuous corn		Rotation	
			stock	rate	stock	rate	stock	rate
CK		1928	28.5	-	24.8	-	28.3	-
NPK/FYM-NPK			29.1	0.01	-	-	31.7	0.30
FYM/FYM-FYMN			43.8	0.38	38.3	0.34	38.6	0.26
CK		1948	24.5	-	21.4	-	25.9	-
NPK/FYM-NPK	92.40		25.9	0.04	-	-	31.7	0.10
FYM/FYM-FYMN			37.3	0.32	34.5	0.22	35.9	0.17
CK		2014	30.6	-	21.0	-	30.3	-
NPK/FYM-NPK			41.2	0.27	-	-	35.1	0.04
FYM/FYM-FYMN			48.0	0.44	36.8	0.13	40.8	0.08

Note: FYM-NPK means the fertilisation changed from FYM to NPK during the experimental period for rotation; FYM-FYMN means the fertilisation changed from FYM to FYMN during the experimental period for rotation.

3.3. Carbon sequestration efficiency and its relation with C input

After calculating the sequestration rate and C input for each long-term experiment, we obtained the soil CSE and its correlation with the C input under each treatment (Tables 3-9 to 3-12; Figures 3-6, 3-7). We found that the soil CSE with manure application (FYM and FYMN) was higher compared to that with chemical fertilisers. In addition, the relationship between soil CSE and cumulative C input could be well described by the exponential function ($CSE = b + a * e^{-k * C_{input}}$, $P < 0.01$) across most treatments. Soil CSE decreased exponentially as the C input accumulated, eventually approaching a near-equilibrium level (i.e., the asymptotic value). In each long-term experiment, the asymptotic value of the soil CSE was found to be higher for the treatments with manure application (2.45–10.25%) compared to the NPK treatment (0.48–3.85%). Additionally, our analysis revealed a rapid decrease stage in soil CSE (Figures 3-8 and 3-9). The inflection points for each treatment are presented in Table 3-13, indicating the onset of the slower phase of CSE decline. For the Chromic Luvisol in the Broadbalk and Hoosfield long-term experiments, the soil CSE decreased rapidly before the cumulative C input reached 123–315 t C ha⁻¹. For Mollisols in the Morrow and Sanborn experiments, the soil CSE decreased rapidly before the cumulative C input reached 93–145 t C ha⁻¹.

Table 3-9 Carbon sequestration efficiency (%) in the plough layer (0–23 cm) of the Broadbalk long-term experiment within an individual experimental period

	1865	1881	1914	1966		2010	
Treatments	Continuous wheat	Continuous wheat	Continuous wheat	Continuous wheat (Section I)	Continuous wheat (Section II)	Continuous wheat	Rotation
N3PK	18.71	19.87	9.00	4.55	3.35	3.99	4.16
FYM	25.82	15.95	13.13	8.86	8.57	8.01	7.49
FYMN	-	-	25.82	12.41	12.53	9.78	9.87

Note: Section I and section II mean the plot where the cropping systems were located.

Table 3-10 Carbon sequestration efficiency (%) in the plough layer (0–23 cm) of the Hoosfield long-term experiment within an individual experimental period

Treatment	1882	1913	1965	2013
NPK	9.52	4.79	4.13	0.12
FYM	24.24	16.65	12.32	10.91

Table 3-11 Carbon sequestration efficiency (%) in the plough layer (0–20 cm) of the Morrow long-term experiment within an individual experimental period

Treatments	1913		1968		2008	
	Continuous wheat	Rotation	Continuous wheat	Rotation	Continuous wheat	Rotation
FYM-NPK (SB)	30.27	16.19	5.10	6.15	3.31	1.37
FYM (SC)	36.45	37.70	8.05	8.56	3.13	2.55

Note: SB and SC are the plots where the treatments are located; FYM-NPK means the fertilisation changed from FYM to NPK during the experimental period.

Table 3-12 Carbon sequestration efficiency (%) in the plough layer (0–20 cm) of the Sanborn long-term experiment within an individual experimental period

Treatments	1915			1828			1948			2014		
	CW	CC	Ro	CW	CC	Ro	CW	CC	Ro	CW	CC	Ro
NPK/FYM-NPK	7.40	-	31.75	3.32	24.48	17.81	2.71	-	7.62	5.86	5.94	3.60
FYM/FYM-FYMN	25.22	34.55	19.61	19.02	-	13.74	8.98	13.56	8.98	5.02	-	3.47

Note: CW means continuous wheat system; CC means continuous corn system; Ro means rotation; FYM-NPK means the fertilisation changed from FYM to NPK during the experimental period for the rotation; FYM-FYMN means the fertilisation changed from FYM to FYMN during the experimental period for the rotation.

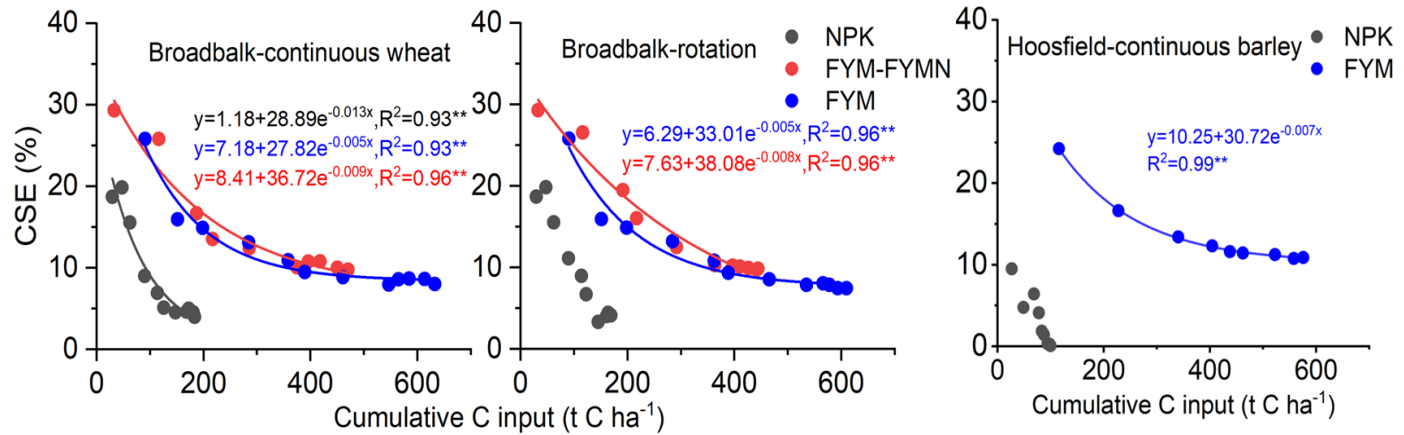


Figure 3-6 Relation between cumulative carbon (C) input (t C ha⁻¹) and carbon sequestration efficiency (CSE, %) for all treatments in studied cropping systems of Broadbalk and Hoosfield. ** means the relation fitted the negative exponential function well ($P < 0.01$).

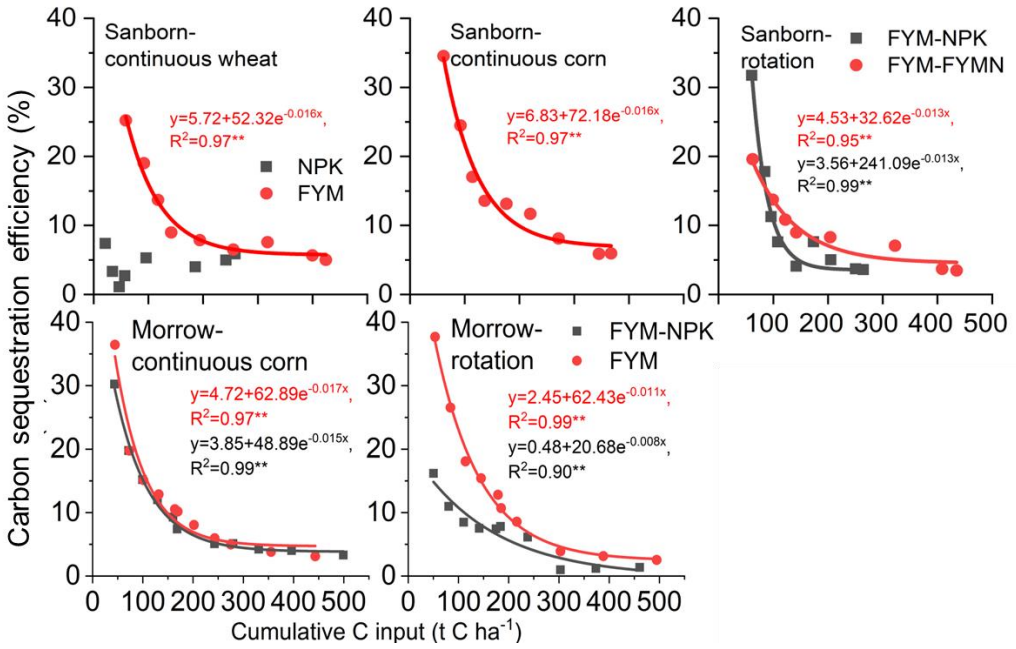


Figure 3-7 Relation between cumulative carbon (C) input (t C ha⁻¹) and carbon sequestration efficiency (CSE, %) for all treatments in studied cropping systems of Morrow and Sanborn.

** means the relation fitted the negative exponential function well ($P < 0.01$).

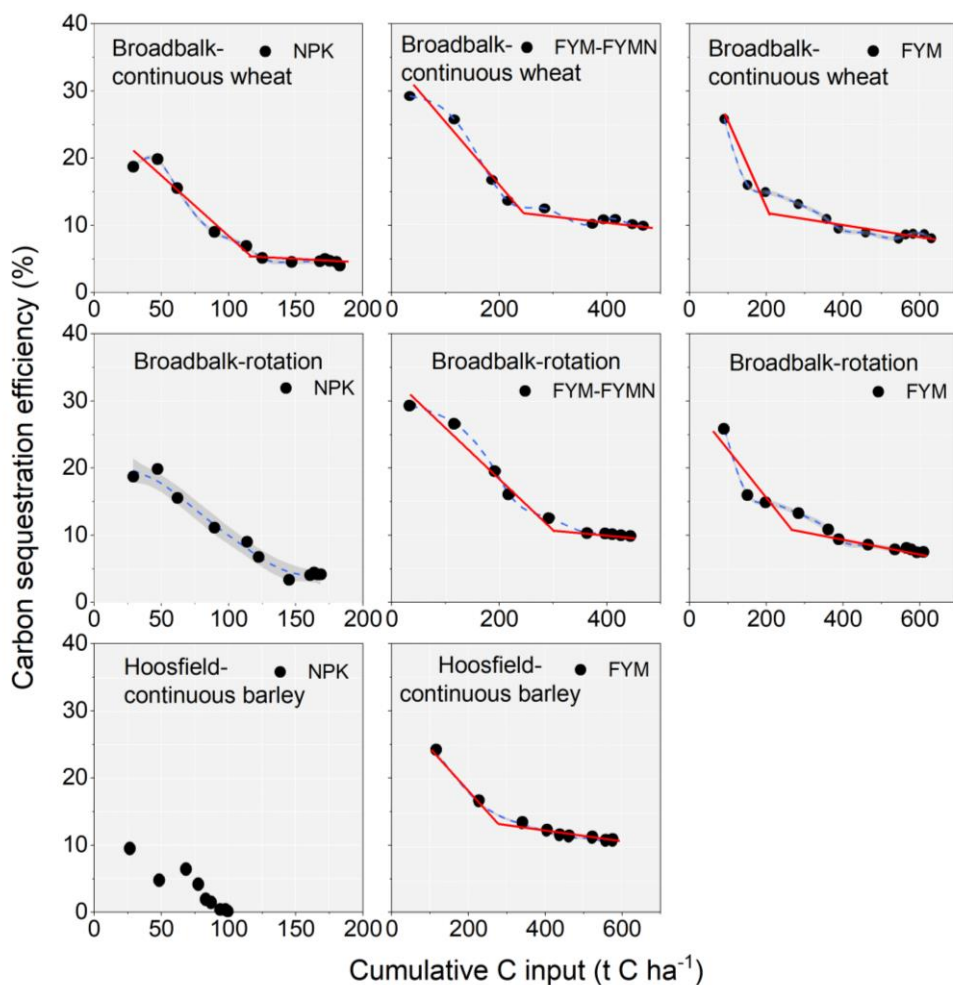


Figure 3-8 The generalized additive model and segmented piecewise linear regression for the relation between carbon sequestration efficiency (%) and cumulative C input (t C ha⁻¹) in the Broadbalk and Hoosfield experiments. AIC means Akaike information criterion. *** $P < 0.001$.

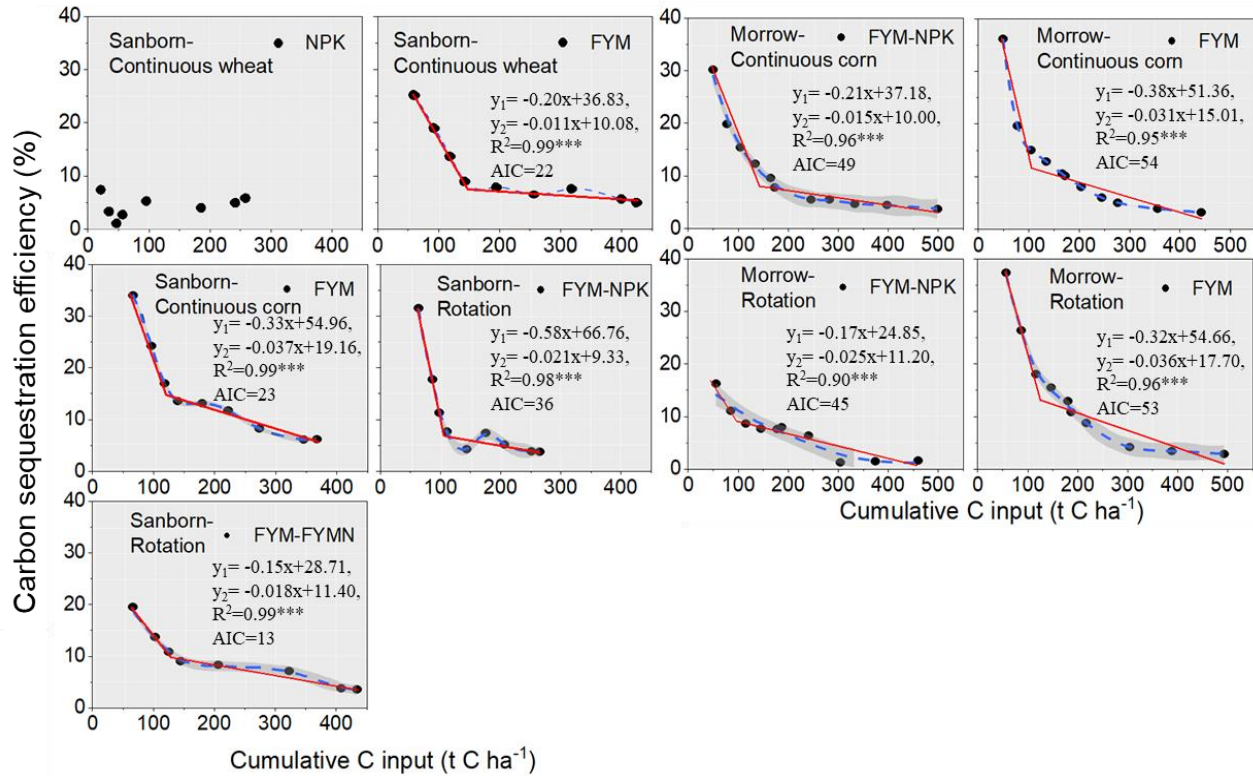


Figure 3-9 The generalized additive model and piecewise linear regression for the relation between carbon sequestration efficiency (%) and cumulative C input (t C ha⁻¹) in the Morrow and Sanborn experiments. AIC means Akaike information criterion. *** $P < 0.001$.

Table 3-13 The cumulative carbon (C) input and carbon sequestration efficiency (CSE, %) at the inflection point (CSE_{IP}), and the CSE asymptotic value (CSE_{as})

Site	Cropping systems	Treatments	Inflection point		CSE _{as}
			C input	CSE _{IP}	
Broadbalk	Wheat	NPK	123	4.4	1.2
		FYM-FYMN	235	12.4	8.4
		FYM	206	13.5	7.2
	Rotation	NPK	-	-	-
		FYM-FYMN	315	10.6	7.6
		FYM	260	8.1	6.3
Hoosfield	Barley	NPK	-	-	-
		FYM	271	13.1	10.3
Sanborn	Wheat	FYM	145	7.8	5.7
	Corn	FYM	122	14.9	6.8
	Rotation	FYM-NPK	103	7.1	3.6
		FYM-FYMN	133	9.9	4.5
	Morrow	Corn	FYM-NPK	141	7.5
FYM			105	12.1	4.7
Rotation		FYM-NPK	93	8.7	0.5
		FYM	128	13.0	2.5

3.4. Main factors influencing carbon sequestration efficiency

The impact of C input, soil properties and climatic variables on the CSE of the two typical regions are shown in Figure 3-10. The XGBoost regression indicated that the most important controller for soil CSE of the Broadbalk and Hoosfield experiments in southeastern England was clay percentage, with the highest feature importance of 0.36, followed by C input (0.23), TN (0.15) and MAT (0.10). Nevertheless, the C input (0.33) was the most important factor for soil CSE of the Morrow and Sanborn experiments in the Central Great Plain of America. MAT (0.29), TN (0.16) and initial SOC content (0.11) were the main explanatory factors that ranked after C input (Figure 3-10).

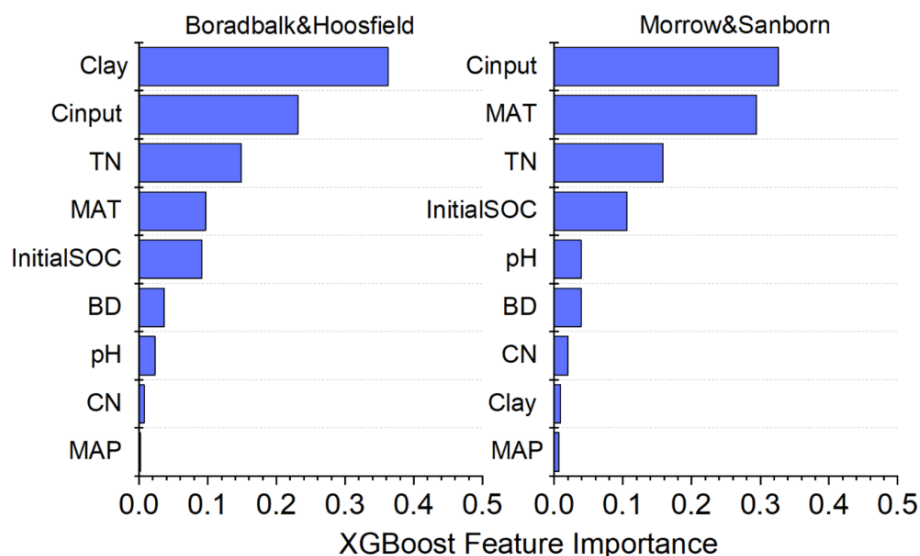


Figure 3-10 The importance value of independent variables for controlling carbon sequestration efficiency (CSE) of the plough layer in each studied region by using the extreme gradient boosted tree (XGBoost) regression modelling. Model performance shows in Table 3-4.

4. Discussion

4.1. Fertiliser effect on soil carbon sequestration

Over a century of fertilisation, manure application has a higher SOC sequestration ability compared to treatments using only chemical fertilisers, yielding higher C sequestration rates and soil CSE levels (Tables 3-5 to 3-12, Figures 3-6, 3-7). Consequently, the SOC stocks of the FYM and FYMN treatments in studied systems increased by an average of 156% and 215%, respectively, in the Broadbalk and Hoosfield experiments, as compared to the initial SOC stocks. Furthermore, they were on average 145% and 274% higher, respectively, than those of the NPK treatment. In the Morrow and Sanborn fields, despite the decline in SOC stocks over time (Figure 3-1), the SOC stocks in the manure-applied treatments were, on average across all cropping systems, 11% higher in the Morrow field and 20% higher in the Sanborn field than in the treatments using only chemical fertilisers. Therefore, regardless of whether SOC increases or decreases with C inputs, manure application consistently provides a greater advantage for SOC sequestration over chemical fertilisers when compared to the CK.

The positive effects of manure only and manure amendment treatments were mainly due to the following reasons: (1) The manure, being a mix of dung and compost from horses or cattle, likely contained more humified and resistant materials like lignin and

polyphenols, which could remain unaffected and persist for long periods (Liao et al., 2006). In the Hoosfield experiment, manured plots contained approximately 50% higher organic C than unfertilised plots after manure addition had been discontinued for 104 years (Johnston, 1986), thus indicating the long-term stability and persistence of organic material derived from manure. (2) The enhancement of activities and functional group diversity within the microbial community associated with C sequestration in soil treated with manure contributed beneficially to SOC sequestration (Blagodatskaya and Kuzyakov, 2008). As summarized by Lal (2003) based on many classical long-term experiments in North America, including Sanborn and Morrow, the addition of manure increased microbial biomass and mineralizable C. This reported indicated higher SOC concentrations and greater biological activity in organic fertiliser treatments compared to chemical fertiliser applications. (3) The macroaggregate component and aggregate stability, which are linked to a high CSE, significantly increase after manure application. Liu et al. (2019) has proved that the CSE of macroaggregates was greater than that of other physical fractions. For instance, in the Broabadlk, Morrow, and Sanborn long-term experiments, the increased SOC content due to FYM application led to enhanced stability of soil macroaggregates (Acikgoz et al., 2017; Chakraborty et al., 2014; Huggins et al., 1998; Powlson et al., 2014). Accordingly, this can be seen as a reason contributing to the higher CSE observed in treatments with manure applications.

4.2. Carbon sequestration efficiency changes with C input accumulation

Our results demonstrated that soil CSE decreased exponentially ($CSE = b + a * e^{-k * C_{input}}$, $P < 0.01$) with the accumulation of C input until it approached a near-equilibrium level (i.e., b , the asymptotic value), with the exception of NPK treatments in certain cropping systems where data limitations may have constrained the observed trends. This decreased dynamic in soil CSE with C input has been directly or indirectly supported by findings from previous studies (Jiang et al., 2018a; Stewart et al., 2007, 2008; Yan et al., 2013). Hassink and Whitmore (1997) clarified that as soil approached C saturation, it accumulated less SOC at a lower efficiency. As our results show, the SOC sequestration rate and soil CSE generally decreased over time for almost all treatments in the four classical long-term experiments (Tables 3-5 to 3-8, Figures 3-6, 3-7). Furthermore, a study discussed the soil C saturation model and proposed the concept of the C saturation deficit based on 14 long-term experiments ranging 12–96 years (Stewart et al., 2007). The conceptual model implied that the further the soil from saturation, the higher the SOC sequestration potential and efficiency (Stewart et al., 2008). This could explain the exponential decreasing trend observed between soil CSE and C input under long-term fertilisation, as derived from the four classical long-term experiments. Moreover, Jiang et al. (2018a) predicted the CSE dynamic with long-term manure application and derived the same function for the relationship between soil CSE and accumulative manure C input as our study did, using both measured and predicted data by 2100 from 20 long-term experiments across China.

4.3. Main drivers of soil carbon sequestration efficiency with a large amount of C input accumulation

The XGBoost regression modelling indicated that C input, MAT and TN were the predominant factors influencing soil CSE in the four classical long-term experiments. Specifically, carbon input is the most important factor influencing soil CSE in the Morrow and Sanborn experiments in the USA, and ranks as the second most important factor for the CSE in the Broadbalk and Hoosfield experiments in England. Stewart et al. (2007) has emphasized the necessity of a wide range of C inputs to fully capture the linear to asymptotic behaviours of SOC changes. Otherwise, a smaller section of the asymptotic curve between SOC and C input could appear linear in the early stages of SOC stock accumulation, leading to the perception of a constant CSE (Yan et al., 2013). As indicated in Chapter II, a linearly increasing SOC and a nearly constant CSE were found with 30–40 years of fertilisation. In such case, carbon input was less important than soil properties, especially clay content and soil C/N ratio, for controlling CSE. Thus, a large range of C input levels is crucial for clarifying the full CSE dynamics. Maillard and Angers (2014) summarized that a dominant impact of cumulative C input on SOC sequestration could explain at least 53% of the variances in SOC stock, as determined by a meta-analysis based on 130 observations considering 4–82 years of fertilisation. Similarly, Cai (2016) demonstrated that cumulative C input could account for 42% of the variations in soil CSE by analysing 25 long-term experiments that included both C equilibrium and disequilibrium soils in China. These results support the high importance of C input for soil CSE under long-term fertilisation for soils with SOC showing nonlinear changes.

MAT was also found as one of the key factors influencing soil CSE in the four classical long-term experiments. Temperature mainly affects soil CSE by regulating crop biomass growth (related to C input) and by influencing the decomposition of organic C (Aguilera et al., 2018; Liang et al., 2016). The latter might be the main reason for this study. According to the weather records from the Broadbalk and Hoosfield experiments, the MAT increased by approximately 1.2°C from 1878–1914 (8.97°C) to 1990–2018 (10.20°C). Similarly, the MAT increased by approximately 0.5°C from 1893–1928 (12.50°C) to 1990–2018 (12.97°C) in Sanborn and from 1900–1930 (10.77°C) to 1990–2018 (11.35°C) in the Morrow experiment. The significant increase in MAT might accelerate the mineralization of added OC and thus reduce soil CSE (Chen et al., 2021; Liu et al., 2019). Poeplau et al. (2011) developed a C response function for cultivation ages of 100–200 years by summarizing data from 205 fields converted from forest or grassland to cropland, which was consistent with the four classical long-term experiments in the present study. And their analysis revealed that the decrease rate of SOC significantly increased with temperature. In this case, the increase in temperature led to higher OC decomposition rates that were not offset by plant residues (Schlesinger, 1986). Similarly, Evrendilek and Wali (2001) found that a 0.5°C increase in MAT between 1866 and 1996 resulted in a decrease of 1% in SOC stock for continuous wheat systems in Ohio croplands in the USA. Bellamy et al. (2005) suggested that the 0.5°C increase in temperature should have been responsible for the observed 0.6% loss of SOC in England and Wales after 1978. Fantappiè et al. (2011)

clarified that temperature (with an increase of 0.28–1.01°C) had the most relevant impact on SOC sequestration in arable lands among climate factors in Italy after 1961 ($r = -0.329^{**}$).

Soil TN stock was identified as another key factor influencing soil CSE under long-term fertilisation with a large amount of C input. Soil TN content represents the balance between plant N demand and soil N supply in croplands, and it limits crop growth and the transformation of belowground C. Researchers have suggested a significant positive relation between SOC changes and TN content because N addition generally increases plant production by enhancing soil TN content and promoting N availability (Henriksen and Breland, 1999; Ren et al., 2021), which in turn supports the accumulation of SOC from crop residues (Finn et al., 2016; Jesmin et al., 2021). In addition, the increased N availability affected the mineralization of exogenous organic C by affecting the microbial biomass and extracellular enzyme activity involved in microbial C cycling (Jesmin et al., 2021). Based on six dryland experiments under long-term fertilisations, soil available N was found to be the most important limiting factor among soil properties and could explain 7.9% of the total variation in CSE (Liang et al., 2016). As demonstrated by an incubation experiment conducted in the Morrow field, high N availability, compared to soils with low N addition, increased the activity of cellulase and protease—two key enzymes involved in OC mineralization (Jesmin et al., 2021). Furthermore, Liang et al. (2016) recognized that the impact of soil available N on soil CSE was more effective under the NPK treatment. This was attributed to the fact that manure input in the FYMN/FYM treatments was likely to alleviate N limitations on crop growth and subsequent effects on OC turnover, in comparison to the NPK treatment.

In addition, clay content was the most important factor controlling soil CSE of the Broadbalk and Hoosfield experiments. Generally, soil physiochemical properties determine the maximum potential for soil C sequestration, which can constrain the increase of SOC resulting from added C inputs (Six et al., 2002). This impact is more significant for soil that is far from C saturation (Hassink and Whitmore, 1997), as there is more available space for the adsorption of organic molecules (Boyle et al., 1989). A study conducted in the Broadbalk experiment, which focused on soils with both chemical and organic fertilisers, similar to our study, suggested that the minimum SOC concentration increment was found to be higher in soils with higher clay content (Watts et al., 2006). The soil in Broadbalk and Hoosfield experiments was initially far from C saturation when the experiments began (Johnston et al., 2009; Rasmussen et al., 1998). During the long-term fertilisation period, the availability of free space on the clay surface, along with the proportion of organic polymers bound to clay, determined the adsorption and desorption of SOC (Boyle et al., 1989). Further, the clay-protected SOC was difficult to decompose due to the strong adsorption capacity of clay particles by organo-mineral bonding reactions, and hence, soils with higher clay contents can exhibit significantly higher CSE compared to soils with higher sand and silt contents (Ekschmitt et al., 2005; Hua et al., 2014). Thus, clay content may mainly determine the space available for stabilizing OC once exogenous C is added,

particularly when the soil approaches C equilibrium in the Broadbalk and Hoosfield experiments (Johnston et al., 2009). Meanwhile, our results indicated that soil physical properties (e.g., clay content) and their protection seemed to be limited by their characteristics, which was consistent with the phenomenon of C saturation observed during long-term soil C accumulation (Hassink, 1997). In the Morrow and Sanborn experiments, the initial SOC content was another important influential factor influencing soil CSE. Research in the Central Great Plain of America, where the Morrow and Sanborn experiments were located, revealed that the high initial SOC level together with the high clay content contributed to the formation of extremely stable and durable soil aggregates. As a result, the addition of SOM may not be as effectively protected within these already highly stable aggregates (Puget and Lal, 2005). Thus, the high initial SOC content in the Morrow and Sanborn experiments might have led to a low potential and efficiency for sequestering freshly added organic C.

4.4. Recommended fertilisation and its uncertainties

Based on the results of Chapter II and Chapter III, manure amendment is an effective fertiliser practise for enhancing SOC storage and soil CSE compared to chemical fertilisers. However, our findings are limited to the plough layer. It is important to note that SOC was changed by fertiliser practises in both surface and subsurface soils. Accounting only for soil C change in the surface layer may misrepresent the C sequestration capacity across the soil profile. For example, long-term NPKM application in Gongzhuling resulted in a significantly greater increase in SOC stock in the 20–40 cm and 40–60 cm layers compared to the 0–20 cm layer, when compared to the initial values. Conversely, in Zhengzhou, NPKM had a negative effect on SOC stock in the subsoil, decreasing it by 5% in the 40–60 cm layer, while SOC stock increased by 18% in the 0–20 cm layer and by 10% in the 0–40 cm layer compared to the initial values (Liang et al., 2019). Similarly, an increase in SOC stock in the 0–20 cm layer and a decrease in the 20–60 cm layer were observed with FYM application from 1915 to 1988 in the corn cropping system of the Sanborn long-term experiment (Buyanovsky and Wagner, 1998; Miles and Brown, 2011). Although research on CSE in subsurface soils is limited due to the scarcity of experimental data and the challenge of quantifying C input allocation across different soil layers, the influence of fertilisation on CSE in both surface and subsoil remains controversial. A study reported that, over three decades of fertilization, sites with high initial SOC contents (e.g., Gongzhuling) exhibited lower CSE in the 0–20 cm layer than in the 0–60 cm under NPK and NPKM treatments. In contrast, at sites with lower initial SOC contents (e.g., Zhengzhou), a higher CSE in the 0–20 cm layer compared to the 0–60 cm layer was observed (Liang et al., 2019). Those uncertainties necessitate that future research should focus on the characteristics of CSE in subsoil in response to different long-term fertilisations.

5. Conclusion

Based on four classical long-term experiments with more than a century of fertilisation, the full behaviour of the negative exponential dynamics of soil CSE was captured in this study. The results demonstrated that soil CSE decreased rapidly once C input began to accumulate and then continued to decrease more slowly up to an asymptotic value when a large amount of C input was accumulated. This suggests that managers should take appropriate strategies before the C input reaches the inflection point, ensuring a timely improvement in CSE. When considering the factors controlling soil CSE, carbon input, MAT, and TN stock were crucial factors across the four experiments with more than a century of fertilisation. Clay content played an important role for soil CSE only in the Broadbalk and Hoosfield experiments. The initial SOC significantly controlled soil CSE only in the Morrow and Sanborn experiments. Furthermore, the classical long-term experiments clarified that manure application had a higher SOC sequestration rate and efficiency compared with chemical fertilisation. The classical long-term experiments revealed that manure application yielded a higher asymptotic value for soil CSE relative to chemical fertilisation practises. These results help identify optimal fertiliser strategies to promote CSE under long-term fertilisation.

Chapter IV

Climate change impacts on crop production and soil carbon stock in a continuous wheat cropping system in southeast England

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Abstract

Understanding dynamics of soil organic carbon (SOC) stock in agroecosystems under climate change is imperative for maintaining soil productivity and offsetting greenhouse gas emissions. Simulations with the SPACSYS model were conducted to assess the effects of future climate scenarios (RCP2.6, RCP4.5 and RCP8.5) and fertilisation practises on crop yield and SOC stock by 2100 for a continuous winter wheat cropping system in southeast England. Weather data between 1921 and 2000 was considered as the baseline. SPACSYS was first calibrated and validated with the data of the Broadbalk continuous winter wheat experiment for over a century. Six treatments were used: no fertiliser, a combination of chemical nitrogen, phosphorus and potassium with three nitrogen application rates (N1PK, N3PK and N5PK), manure only (FYM, close N application rate to N5PK) and a combination of manure and chemical nitrogen application (FYMN, the same chemical N application rate as N3PK). Compared with the observations, SPACSYS was able to simulate grain yields and dynamics of SOC and TN stocks. Our predications showed that wheat yield would increase by 5.8–13.5% for all the fertiliser application treatments under future climate scenarios compared to that under the baseline because of a gradual increase in atmospheric CO₂ concentration. Meanwhile, the SOC stock can increase for the practises under the scenarios except the NPK fertiliser practises under RCP2.6. Increased C input through “CO₂-fertilisation effects” can compensate C losses by soil respiration under the RCP scenarios. We concluded that manure application practises can be considered as a sustainable strategy for enhancing wheat yield and soil C sequestration under the future climate scenarios.

Key words: Yield; Soil organic carbon stock; Broadbalk wheat experiment; SPACSYS; Climate change

1. Introduction

Sequestering more organic C in agricultural soils plays a critical role in mitigating climate change (Sykes et al., 2020). It has been reported that about 90% of the total mitigation potential in agriculture could be achieved by SOC sequestration (Begum et al., 2017). SOC changes can be manipulated by agronomic management practises, especially fertilisation. It has been shown that types and rates of applied fertiliser have various effects on SOC stock under different climatic conditions (Ma et al., 2022; Wan et al., 2011; Wiesmeier et al., 2016), and hence, a more detailed quantification of these effects is required. Furthermore, changes in SOC stock are very slow, and a steady-state soil C content would be realised over 50–100 years, depending on fertiliser application practises as well as soil type specific settings (Johnston et al., 2009; Rasmussen et al., 1998). Therefore, long-term experiments based on regional specific common soil and crop types as well as fertiliser practise conditions (Smith et al., 1997), may be the best choice to estimate whether a given fertiliser practise can sustain crop productivity and improve soil health under climate change on the long-term.

Winter wheat is currently the most extensively grown arable crop in the UK with approximately 40% of the arable area (Cho et al., 2012; Harkness et al., 2020). Future climate projections suggested a hotter and drier summer and warmer and wetter winter across the UK (Harkness et al., 2020). CO₂ concentration would even increase to more than 900 ppm by 2100 under the worst scenario (Meinshausen et al., 2011). Because of uncertainty in climate change in the future and a slow response of SOC to the change, it would be difficult to observe the consequences with field or controlled experiments timely.

Modelling is a powerful option for predicting SOC changes under different climate and fertilisation scenarios (Begum et al., 2017). Previous predictions showed that climate change would increase winter wheat yield by ca. 5–33% in most regions of the UK (Cho et al., 2012; Ghaffari et al., 2002; Semenov and Shewry, 2011; William et al., 2018) and also increase SOC stock by 2.5–10 t C ha⁻¹ in the following decades (Lugato et al., 2014; Smith et al., 2005; Yigini & Panagos, 2016). However, those positive responses were mainly derived from uncoupled crop growth models and soil C models that consider an altered C-input value using estimates of net primary productivity (NPP). They didn't take into consideration the interaction of C and N among soil, plant and atmosphere. Thus, models that are able to simulate the interactions between crop, soil nutrients cycling, management practises and other environmental variables are needed to conduct a more realistic and comprehensive assessment about crop yield, and the C sequestration capacity considering various fertiliser management practises and/or different climate scenarios. Process-based models could be used as decision support tools in order to assess the impacts of climate change and agronomic management practises on agroecosystems, as they allow an integration of various sources of data and knowledge on crop and/or environmental variables to evaluate hypotheses (Arulnathan et al., 2020).

The SPACSYS model has been proven with a strong ability to accurately simulate

plant growth, N uptake, SOC and TN stocks, and CO₂, CH₄ and N₂O emissions of cropland and grassland across Europe and China (Liu et al., 2018, 2020; Perego et al., 2016; Wu et al., 2015; Zhang et al., 2016b, 2016c). Compared with other popular process-based models, it considers more processes in C, N and P cycling (Table 1-1). The Broadbalk continuous wheat winter experiment at Rothamsted Research in England is the longest continuous experiment (>170 years) in the world and has well-documented records about crop and field managements (Johnston and Poulton, 2018). If simulations by SPACSYS can be verified by the data from the experiment, then the prediction on crop yield and the dynamics of SOC under future climate scenarios would be much more credible. Although the same dataset has been used for model validation with CENTURY/DailyDayCent (Falloon and Smith, 2000; Begum et al., 2017), Roth-CNP (Muhammed et al., 2018), C-TOOL (Taghizadeh-Toosi et al., 2014) and Roth-C (Falloon and Smith, 2000), the impacts of future climate change on the yield of winter wheat and SOC stock have not been explored simultaneously. This is important because a question needs to be answered whether continuous FYM amendment over 170 years would impede the goal of C neutrality under future climate scenarios as a low SOC sequestration rate is expected when approaching SOC saturation (Stewart et al., 2007).

In this study, we tried to identify the response of crop yield and SOC stock to future climate change in the winter wheat continuous system in southeast England and recommend a sustainable strategy for both soil productivity and carbon sequestration. To achieve this, firstly we calibrated and validated the SPACSYS model using the data collected from the Broadbalk continuous winter wheat experiment over a century on the wheat yield, and SOC and TN stocks, then quantified the yield response of winter wheat to future climate change scenarios with different fertiliser practises, and finally assessed the dynamics of SOC stock and C balance with the continuous winter wheat cropping system and various fertiliser application practises under future climate scenarios.

2. Materials and methods

2.1. Site description and experiment treatments

The data from the Broadbalk continuous winter wheat experiment (<http://www.era.rothamsted.ac.uk/experiment/rbk1>) which started from 1843 at Rothamsted Research, Harpenden, UK (0°22'30" W, 51°48'36" N, 128 m a.s.l.) are used for model calibration and validation in this study. The soil type corresponds to a Chromic Luvisol when considering FAO soil classification. The site has a cool temperate climate with an average temperature of 10.2°C and an average total annual precipitation amount of 793 mm (according to 1991–2017 period). Its original purpose was to test the response of crop yields on various combinations of fertilisers. The experiment remains its long-term integrity but has been modified to address current agriculture challenges (Johnston and Poulton, 2018). The original plots were subdivided twice, occurred in 1926 and 1968, respectively. There are 10 sections since

the last subdivision. The grain and straw yields and soil data, as used in this study, were from Section 1 of Broadbalk. Because of availability of historic daily weather data, only the data from 1914 onwards were used in this study.

The treatments that were basically established in 1852 were kept largely unaltered until 1968. Some of the fertiliser treatments were updated to better reflect modern agriculture after 1968. There are 20 treatments in total. In our study, six treatments were chosen: no fertiliser application (CK), a combination of chemical N, phosphorus (P) and potassium (K) fertilisers with three N application rates (thereafter, N1PK, N3PK and N5PK), and farmyard manure (FYM) application only or with N chemical fertiliser (thereafter, FYM and FYMN). All details about fertiliser applications for the selected treatments are summarized in Table 4-1.

Table 4-1 Nitrogen fertiliser and manure application rates for different treatments of the Broadbalk continuous winter wheat experiment

Treatment	N [†] (kg N ha ⁻¹ yr ⁻¹)	Manure [‡] (t ha ⁻¹ yr ⁻¹)	Form
CK	-	-	before 1967: ammonium sulphate and ammonium chloride (1:1);
N1PK	48	-	
N3PK	144	-	1968 -1985: calcium ammonium nitrate;
N5PK	96 (1852-1967)/ 144 (1968-1984)/ 240 (from 1985)	-	After 1985: ammonium nitrate;
FYM	-	35 (from 1843)	
FYMN [#]	96 (from 1968)/ 144 (from 2005)	35 (from 1885)	Farmyard manure (cow)

[†] Before 1968, a fixed rate of 24 kg N ha⁻¹ chemical N fertiliser was applied in the autumn and the remainder in spring each growing season, and then all applied in spring.

[‡] It was estimated 224 kg N ha⁻¹ yr⁻¹.

[#] The treatment started from 1885.

Winter wheat was generally sown between September and October and harvested between late July and early September the following year. Cultivars varied over time, and short-strawed cultivars were introduced after 1967 (Table 4-S1). Herbicides and insecticides have been used as routine applications since 1964 and 1979, respectively. The plots were cultivated before sowing each year with a ploughing depth of 23 cm. Grain yield and straw yields at 85% dry matter were recorded each year. Soil samples taken within the 0–23 cm depth increment were taken and analysed periodically.

2.2. Data

Historic daily weather data for the site, agronomic management records, grain yield, straw yield, SOC and TN contents were downloaded from e-RA

(<http://www.era.rothamsted.ac.uk>). Management includes cultivation (ploughing depth and date), crop management (date and rate of seeding and harvesting date) and fertiliser application (date, amount and fertiliser type) for each treatment.

2.3. Model description

The SPACSYS model is a multi-dimensional, field-scale, weather-driven, flexible time step (from minute up to daily), and process-based model that quantifies the biogeochemical processes of C, N, and P cycling, and the water and heat budgets in soil, plant and ruminant animal. As the details of the model have been described elsewhere (Wu et al., 2007, 2015, 2019a, 2022), here we briefly described it in terms of organic pools and simulated processes. Carbon and N are held in a number of above-ground and below-ground pools, including the fresh litter pool, the microbial pool, the humus pool, the dissolved organic matter pool, the fresh OM pool (if manure was applied), the ammonium pool and the nitrate pool. Main plant growth processes are plant development, assimilation, respiration, and partitioning of photosynthate and nutrients from uptake estimated with various mechanisms implemented in the model, plus N fixation for legume plants, and root growth and development that is described either in 3D or 1D root system. Nitrogen cycling coupled with C cycling covers the transformation processes for OM and inorganic N. The main processes and transformations causing size changes to soluble N pools are mineralization, nitrification, denitrification and plant N uptake. Most of these are dependent on soil water content and temperature. Nitrate is transported through the soil profile and into field drains or deep groundwater with water movement. A biological-based component for the denitrification process has been implemented that can estimate nitrogen gaseous emissions. Fluxes between the pools occur in a particular way according to physical and biological conditions in the source and destination pools. The fresh litter, dissolved OM and humus pools receive contributions from above ground litter fall and below ground root litter. Decomposed OM from litter, dissolved OM and humus are partitioned to different pools. Meanwhile, CO₂ is released from soils with the decomposition process and microbial respiration. The Richards equation for water potential and Fourier's equation for temperature are used to simulate water and heat fluxes, which are inherited from the SOIL model.

2.4. Model input and parameterisation

The SPACSYS model was operated with a daily time-step in our study. Daily meteorological data (max temperature, min temperature, precipitation, wind speed, humidity and solar radiation) were provided as model input. Because photosynthesis was calculated on an hourly basis, hourly temperatures and solar radiation were interpolated based on the date and site location in a simulation. Soil physical properties shown in Table 4-2 were estimated by pedotransfer functions implemented the model based on soil texture and soil organic matter content prior to simulations. Initial SOC content of each treatment used the average value of three consecutive measurements around 1914 to avoid measurement errors. The atmospheric CO₂ concentration was set to 296 ppm in 1914 and 380 ppm in 2017 (IPCC, 2021) with a

linear increase over the period. Parameters about C and N cycling, soil water redistribution and heat transformation were adopted from previous studies (Bingham and Wu, 2011; Wu et al., 2015). Those for plant photosynthesis and development were based on the previous study (Liu et al., 2020). The built-in Multi-Objective Shuffled Complex Evolution Metropolis algorithm (MOSCEM-UA) (Vrugt et al., 2003) was applied for parameter optimization. The calibrated parameters with optimisation on wheat winter, and soil C and N cycling are listed in Tables 4-S3 and 4-S4, respectively.

Because wheat cultivars varied over time and some of them only planted for a short period (Table 4-S1), especially the short-strawed wheat cultivars, which are not enough to parameterise them. Thus, we assumed that the performance of cultivars grown during the 1914 to 1967 (tall-strawed varieties), the 1968 to 1995 (short-strawed varieties), and the 1996 to 2017 (short-strawed varieties) periods is similar and chose cultivars Red Standard, Cappelle Desprez, and Hereward as the dominant cultivar for each period, respectively. During each period, the grain and straw yields for the chosen dominant cultivars were for calibration, whereas the yields data of rest cultivars were used for validation. The parameters related to soil C and N cycling, water redistribution and heat transformation were kept constant through the entire period. SOC and TN stocks in the plough layer (i.e. 0–23 cm) were calculated after using the C contents and soil bulk density measurements. The SOC and TN stocks data from the three NPK treatments were used to calibrate and those from the rest of the treatments for model validation. Figure 4-1 presents the methodological flowchart of the model calibration and validation.

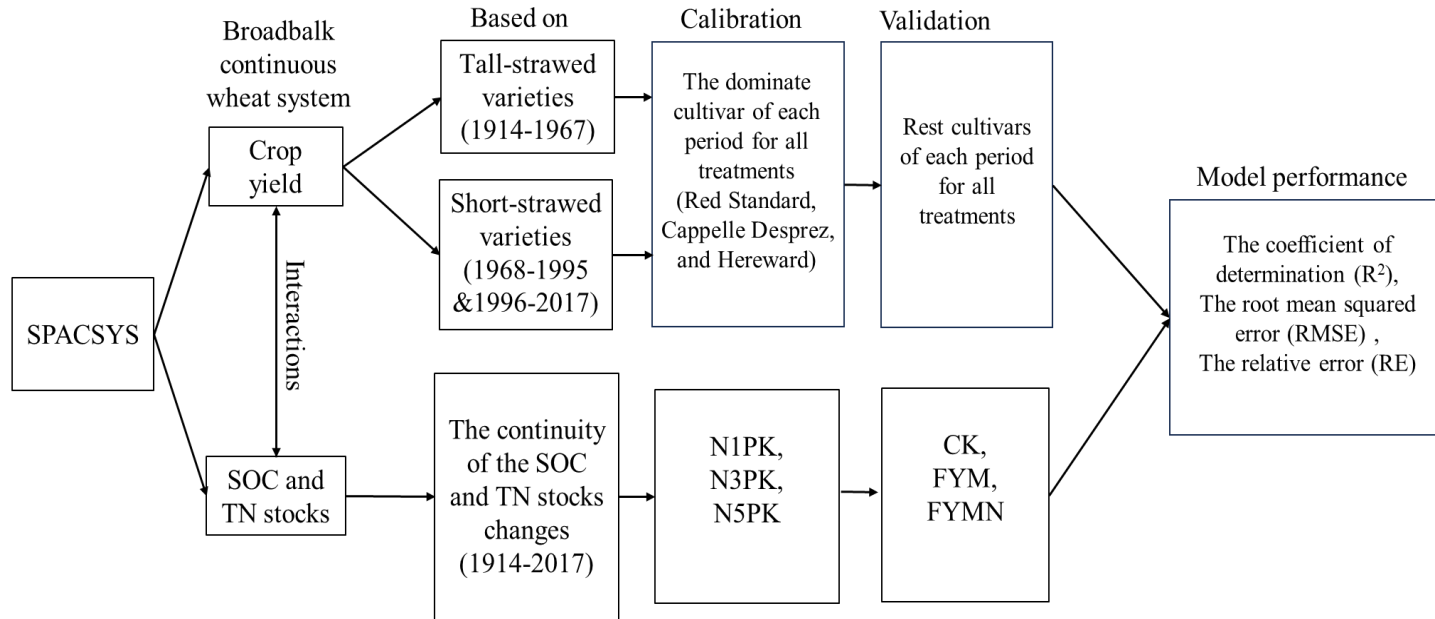


Figure 4-1 A methodological flowchart of model calibration and validation.

Table 4-2 Soil texture and pH, and estimated soil properties in different soil layers for different treatments of the Broadbalk continuous winter wheat experiment

Treatment	Upper depth (m)	Lower depth (m)	SOC [†] (t/ha)	Clay [‡] (%)	Sand [‡] (%)	Silt [‡] (%)	Saturated water content (%)	Field capacity (%)	Saturated total conductivity (mm/day)	pH [‡]
CK	0	0.20	21.8	24	8	68	50.23	41.75	105.55	7.6
	0.20	0.51	25.2	57	3	40	55.39	55.03	88.50	7.2
	0.51	0.96	40.1	71	9	20	56.17	59.87	109.33	7.0
	0.96	1.42	25.8	70	8	22	56.16	59.66	105.55	6.8
N1PK	0	0.23	29.1	19	16	65	48.36	39.83	139.91	7.6
	0.23	0.41	16.1	25	10	65	50.31	41.70	113.25	7.5
	0.41	0.66	22.4	39	3	58	53.28	47.21	88.50	7.3
	0.66	0.94	25.0	40	5	55	53.28	47.50	94.96	7.3
	0.94	1.17	13.7	36	10	54	52.33	45.26	113.25	7.4
N3PK	0	0.23	32.8	19	16	65	48.36	39.83	139.91	7.6
	0.23	0.41	17.1	25	10	65	50.31	41.70	113.25	7.5
	0.41	0.66	24.2	39	3	58	53.28	47.21	88.50	7.3
	0.66	0.94	27.2	40	5	55	53.28	47.50	94.96	7.3
	0.94	1.17	15.5	36	10	54	52.33	45.26	113.25	7.4

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N5PK	0	0.23	30.5	19	16	65	48.36	39.83	139.91	7.6
	0.23	0.41	16.9	25	10	65	50.31	41.70	113.25	7.5
	0.41	0.66	23.5	39	3	58	53.28	47.21	88.50	7.3
	0.66	0.94	26.3	40	5	55	53.28	47.50	94.96	7.3
	0.94	1.17	14.4	36	10	54	52.33	45.26	113.25	7.4
FYM	0	0.18	49.3	25	18	57	49.73	40.18	150.12	7.3
	0.18	0.48	81.0	37	12	51	52.34	45.32	121.52	7.0
	0.48	0.71	34.5	50	29	21	52.78	46.03	221.18	7.0
	0.71	1.02	46.5	43	27	30	52.09	43.88	206.13	7.0
	1.02	1.52	34.7	35	30	35	50.73	39.94	229.11	6.9
FYMN	0	0.18	38.7	25	18	57	49.73	40.18	150.12	7.3
	0.18	0.48	40.9	37	12	51	52.34	45.32	121.52	7.0
	0.48	0.71	15.4	50	29	21	52.78	46.03	221.18	7.0
	0.71	1.02	20.7	43	27	30	52.09	43.88	206.13	7.0
	1.02	1.52	15.5	35	30	35	50.73	39.94	229.11	6.9

† Soil organic carbon (SOC) content was only measured in the 0–23 cm and 23–46 cm depths in 1914 and estimated in different soil layers based on the ratio of each layer fraction to total SOC stock of the measured soil profile in 1969.

‡ measured data.

Table 4-3 Average annual maximum and minimum temperatures, precipitation and CO₂ concentration under the future climate scenarios (2021–2100) and the baseline (1921–2000) at the experimental site

Scenarios	Maximum (°C)	Minimum (°C)	Precipitation (mm)	Precipitation events (times)	Frequency daily precipitation >10 mm (times)	CO ₂ concentration [†] (ppm)
Baseline	13.3 (±0.7)	7.7 (±0.6)	829 (±149)	17171	1642	380
RCP2.6	16.0 (±0.8)	8.0 (±0.6)	730 (±135)	16127	1438	424
RCP4.5	16.7 (±0.9)	8.6 (±0.8)	688 (±112)	15249	1300	536
RCP8.5	17.8 (±1.6)	9.5 (±1.3)	656 (±112)	14669	1306	934

[†] CO₂ concentration in 2100 for different future climate scenarios.

2.5. Prediction

In order to investigate the impacts of future climate change on wheat yield, SOC stock, and C balance with different fertiliser practises, the daily bias-corrected weather data for three future climate scenarios (2021–2100) and the baseline climate (1921–2000) based on the well-cited HadGEM2-ES model with a spatial resolution of $0.5^\circ \times 0.5^\circ$ (Collins et al., 2011; Jones et al., 2011) were downloaded from the Inter-Sectoral Impact Model Intercomparison Project (www.isimip.org, Arneth et al., 2017). The future scenarios were Representative Concentration Pathway (RCP) 2.6, 4.5 and 8.5 (van Vuuren et al., 2011). The average annual precipitation, maximum and minimum temperatures ($^\circ\text{C}$) for the baseline and RCP climate scenarios are shown in Table 4-3. Their dynamic changes are shown in Figure 4-S1. Climate projections showed warmer and dryer future conditions in the study region.

Current cultivar “Crusoe” was used for the future crop variety. To simplify the impact of climate change on crop phenology and yield, the sowing date was fixed on 15th October each year in this study, and the harvest date is determined by simulated physiological maturity. To study the response of existing fertilisation practises to future climate change, field management and fertiliser practises are the same as the experimental treatments.

2.6. Statistical analysis

We used three statistical criteria to evaluate model performance: (1) the coefficient of determination (R^2) that describes the degree of fitness between simulation and observation; (2) the root mean squared error (RMSE), a measure of the average deviation of the estimates from the observed values; and (3) the relative error (RE) that reflects the overall difference between simulated and observed data.

For the predications, two-way ANOVA and Tukey ($P < 0.05$) were used in order to compare the effects of fertiliser treatments and climate scenarios on grain yield, SOC stock, NPP (gross primary productivity minus plant respiration), soil respiration and C balance. Coefficient variation (CV, %) was used to represent stability of grain yield. A low CV value means high yield stability (Berzsenyi et al., 2000). Statistical analysis was performed with SPSS 24.0 (SPSS, Inc., 2017, Chicago, USA).

3. Results

3.1. Model calibration and validation

Overall, the SPACSYS model was able to simulate wheat yields for different treatments in Broadbalk continuous wheat cropping system under more than a century of fertilisations (Table 4-4; Figures 4-2 and S4-2). However, model performance for the modern short-strawed varieties was better than the tall varieties. For the tall varieties, the main discrepancy between simulation and observation occurred for cv. Red Standard with all the treatments during the period 1918 to 1925. Discrepancies also existed for short-strawed cultivars between 1978 and 1995, however, the model

underestimated grain and straw yields only for FYMN, with approximately 11% and 35%, respectively. In addition, the SPACSYS model satisfactorily simulated the dynamics of SOC and TN stocks in the ploughing layer under different fertiliser application practises for over a century (Figure 4-3) as being sown by supportive statistical indicator values (Table 4-4). About 16% underestimation in TN with FYMN after 1992 was found (Figure 4-S3).

Table 4-4 The statistical criteria about model performance for wheat grain yield, straw dry matter and SOC and TN stocks at Broadbalk

Index	Calibration	Validation	Calibration	Validation
	<u>Grain yield</u>		<u>Straw dry matter</u>	
Tall-strawed varieties (1915-1967)				
R ²	0.24**	0.30**	0.53**	0.41**
RMSE (%)	52	34	37	32
RE (%)	-31	-10	-41	-16
n	119	131	119	131
Short-strawed varieties (1968-1995)				
R ²	0.71**	0.67**	0.55**	0.41**
RMSE (%)	26	26	37	47
RE (%)	-16	-11	4	-15
n	60	108	60	106 [†]
Short-strawed varieties (1996-2017)				
R ²	0.70**	0.65**	0.63**	0.58**
RMSE (%)	26	32	38	36
RE (%)	-16	-20	-26	-26
n	101	30	101	30
		<u>SOC stock</u>		<u>TN stock</u>
R ²	0.40**	0.98**	0.69**	0.98**
RMSE (%)	5.8	9.8	3.4	9.4
RE (%)	-4.6	-3.5	-0.5	3.5
n	9	48	9	48

[†] data missing for CK and N3PK in 1987.

** means $P < 0.01$.

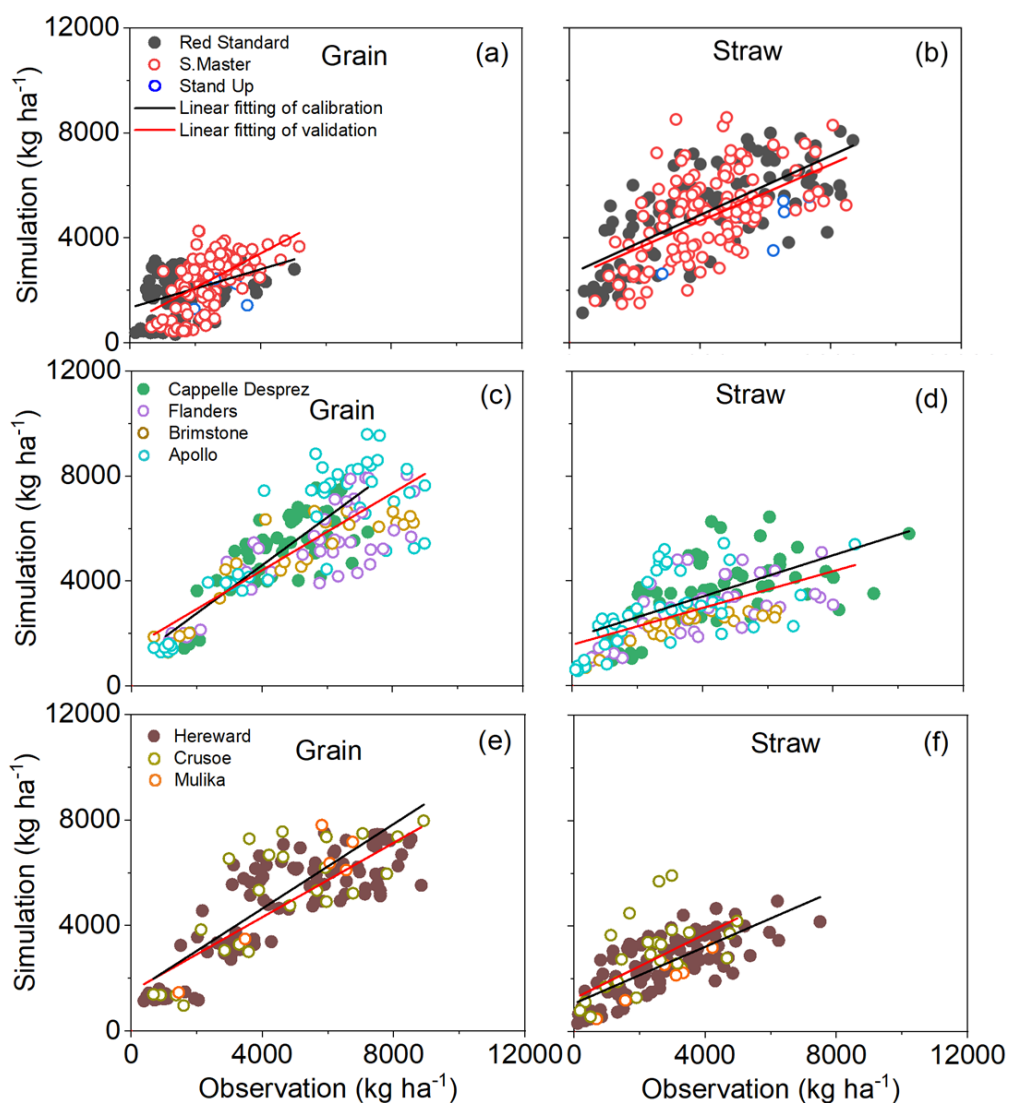


Figure 4-2 Relationship between simulated and observed grain and straw yields of winter wheat from different treatments over the 1914–1967 (a and b), 1968–1995 (c and d) and 1996–2017 (e and f) periods for calibration and validation. The cultivars with solid circle were used for model calibration, and others with open circle were used for model validation.

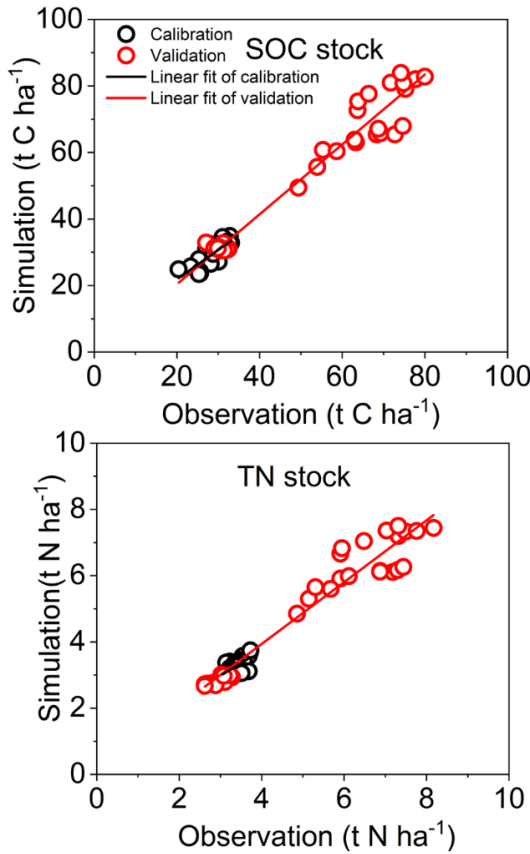


Figure 4-3 Relationship between simulated and observed soil organic carbon and total nitrogen stocks in the plough layer (0–23 cm) for calibration and validation.

3.2. Climate change effect on wheat yield under long-term fertilisations

Compared with the baseline, future climates had a positive effect on grain yield (5.81–13.51%) with all N input treatments and negative without N input (-9.12–4.26%) with a rank of: RCP8.5 \geq RCP4.5 \geq RCP2.6 ($P < 0.05$) (Figures 4-4 and 4-S4; Table 4-5). Grain yields were higher (5366–10976 kg ha⁻¹) with a higher N application rate (N5PK and FYMN) than those (2207–9663 kg ha⁻¹) with other N application rates ($P < 0.05$). Grain yields with N5PK and FYMN did not show significant differences among the three RCP scenarios ($P > 0.05$). In an individual treatment, grain yields under the RCP scenarios were in general more stable with lower CV values (8.80–11.80%) than that under the baseline (10.19–14.13%) except CK and N1PK (Table 4-5).

Table 4-5 The coefficient variation (CV) of grain yield, average SOC stock and C budget for different fertiliser application treatments under all the climate scenarios over the simulation period. Numbers with different lowercase letters indicate significant differences among climate scenarios for a treatment ($P < 0.05$) and those with different capital letters indicate significant differences among different treatments under a climate scenario ($P < 0.05$).

Treatments	Climate scenarios	Grain		SOC		C budget
		Relative changes	CV	Stocks	Relative changes	
CK	Baseline	-	9.53	24.85 bF	-	62 cD
	RCP2.6	-4.76	11.88	24.66 cF	-0.74	112 bcC
	RCP4.5	-4.26	10.68	24.89 bF	0.19	132 abC
	RCP8.5	-9.12	12.86	25.07 aF	0.92	166 aB
N1PK	Baseline	-	8.62	33.92 cE	-	242 cC
	RCP2.6	5.92	9.66	33.77 dE	-0.43	374 bcB
	RCP4.5	7.88	8.86	34.49 bE	1.70	467 abB
	RCP8.5	11.41	12.15	35.26 aE	3.99	650 aA
N3PK	Baseline	-	11.06	36.11 cD	-	443 cB
	RCP2.6	8.53	8.80	36.00 dD	-0.27	610 bcA
	RCP4.5	12.14	8.88	36.80 bD	1.93	683 abA
	RCP8.5	13.51	9.00	37.36 aD	3.49	867 aA
N5PK	Baseline	-	10.19	36.53 cC	-	504 bA
	RCP2.6	5.81	9.88	36.13 dC	-1.08	642 abA
	RCP4.5	8.10	8.85	36.72 bC	0.54	687 abA
	RCP8.5	8.88	9.89	37.36 aC	2.28	881 aA
FYM	Baseline	-	14.13	92.82 dA	-	543 bA
	RCP2.6	9.71	10.16	93.60 cA	1.56	673 abA
	RCP4.5	12.10	10.24	93.91 bA	1.63	780 abA
	RCP8.5	13.39	11.63	94.42 aA	2.34	855 aA
FYMN	Baseline	-	13.39	84.66 dB	-	593 bA
	RCP2.6	8.64	11.02	85.38 cB	0.84	694 abA
	RCP4.5	10.61	9.42	85.44 bB	0.92	701 abA
	RCP8.5	9.56	11.80	85.81 aB	1.36	774 aA

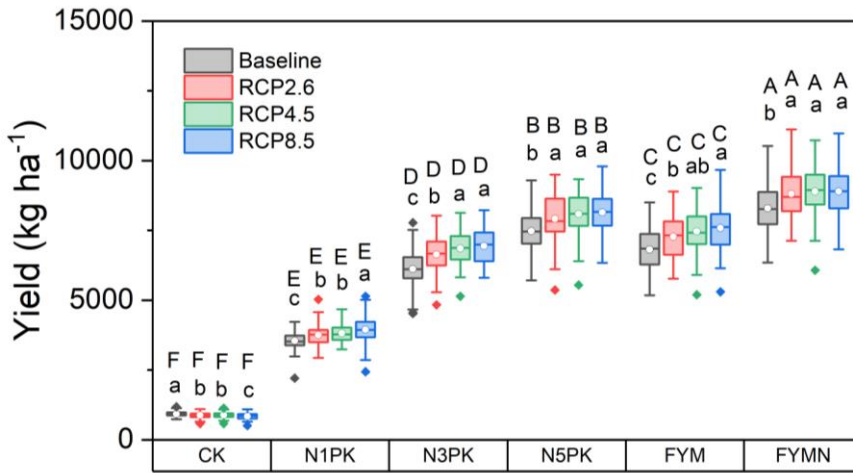


Figure 4-4 Wheat grain yield with different fertiliser practises under future climate scenarios. The lines and circles within the boxes are the median and mean values. The spots are outliers. Columns with different lowercase letters indicate significant differences among climate scenario with an individual treatment ($P < 0.05$). Columns with different capital letters indicate significant differences among different treatments under the same climate scenario ($P < 0.05$). The effects of fertilisation treatments and climate scenarios on wheat yields between 2021 and 2100 were shown in Table 4-S4.

3.3. Climate change effect on SOC stock under long-term fertilisations

The SOC stock with FYM or FYMN had positive responses to all three RCP scenarios with an average relative increase (0.84–2.34%) to that under the baseline (Figure 4-5; Table 4-5). Meanwhile, the field with FYM had the highest C sequestration rate over the simulation period (i.e. 107–142 kg C ha⁻¹ yr⁻¹) among all the fertiliser practises for both baseline and RCP scenarios (Table 4-6). In addition, when considering SOC stock under future climate scenarios with CK and NPK fertiliser practises, these SOC stocks had positive responses to the RCP4.5 and RCP8.5 scenarios during 2021–2100 with average relative increases of 0.19–3.99% (Figure 4-5; Table 4-5). However, the SOC stocks with CK and NPK fertiliser practises showed a negative response with average decreases of 0.27–1.08% to the RCP2.6 (Figure 4-5; Table 4-5).

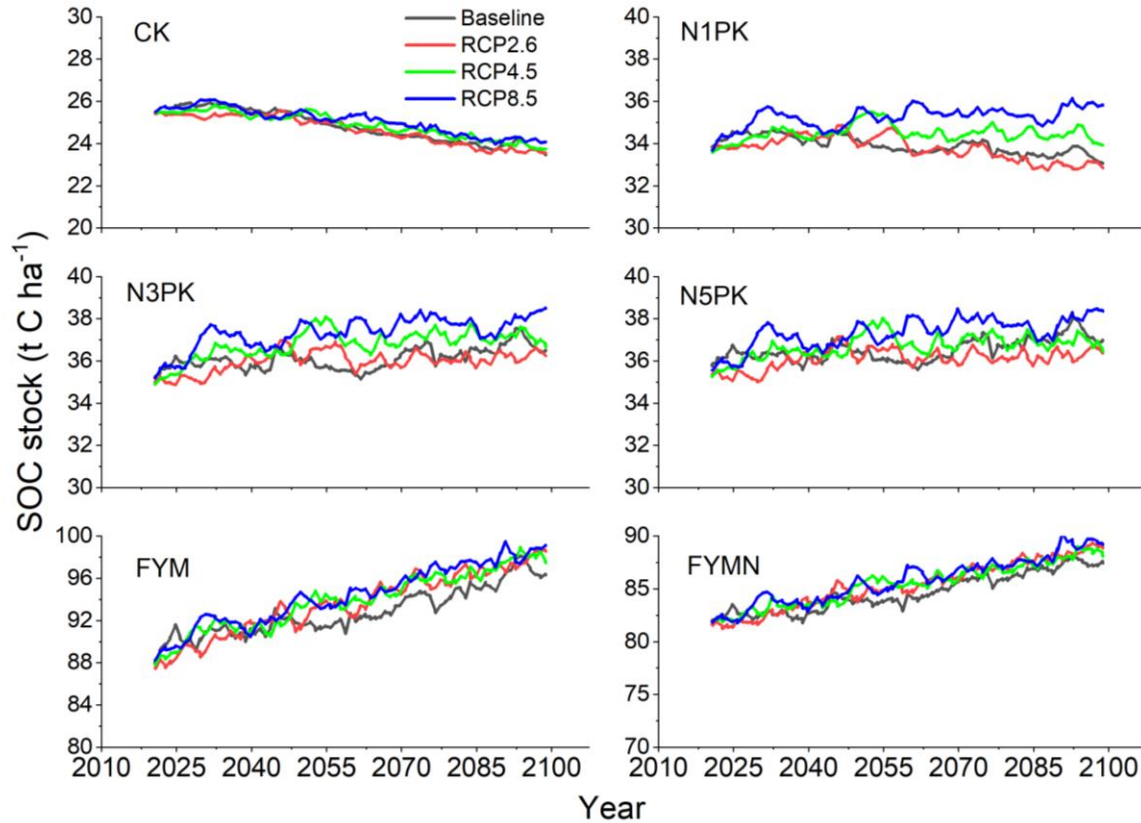


Figure 4-5 Soil organic carbon stock dynamics in the plough layer (0–23 cm) with different fertiliser practises under future climate scenarios from 2021 to 2100.

Table 4-6 The average annual change rate of soil organic carbon stock ($\text{kg C ha}^{-1} \text{ yr}^{-1}$) during 2021–2100 with various fertiliser management practises under different climate scenarios

Treatments	Baseline	RCP2.6	RCP4.5	RCP8.5
CK	-26.64	-23.62	-22.56	-18.28
N1PK	-10.37	-11.08	4.26	22.27
N3PK	15.26	15.68	22.08	41.94
N5PK	13.81	12.74	14.51	35.88
FYM	106.72	142.16	124.56	139.25
FYMN	70.97	93.80	81.98	95.22

3.4. Climate change effect on C balance under long-term fertilisations

The analysis of average annual inputs and outputs of C with different fertiliser practises under the climate scenarios indicated that all the fertiliser treatments resulted in a C sink under each climate scenario within a range of $62\text{--}166 \text{ kg C ha}^{-1} \text{ yr}^{-1}$ for CK, $242\text{--}881 \text{ kg C ha}^{-1} \text{ yr}^{-1}$ for NPK fertiliser practises and $543\text{--}855 \text{ kg C ha}^{-1} \text{ yr}^{-1}$ for FYM and FYMN (Figure 4-6; Table 4-5). Hence, future climate changes seem to result in a net C sink (i.e. $112\text{--}881 \text{ kg C ha}^{-1} \text{ yr}^{-1}$) and a relative increase of $50\text{--}424 \text{ kg C ha}^{-1} \text{ yr}^{-1}$ as compared with the baseline (i.e. $62\text{--}593 \text{ kg C ha}^{-1} \text{ yr}^{-1}$). This was especially the case under the RCP8.5 climate scenario with an average increase of $104\text{--}424 \text{ kg C ha}^{-1} \text{ yr}^{-1}$. There was no significant difference for C sink levels among N5PK, FYM and FYMN under the RCP scenarios.

4. Discussion

4.1. Model performance

The SPACSYS model is able to simulate winter wheat grain yields (R^2 of $0.24\text{--}0.71$ and RMSE of $26\text{--}52\%$) and SOC and TN stocks (R^2 of $0.40\text{--}0.98$ and RMSE of $3\text{--}10\%$), respectively, for both calibration and validation ($P < 0.05$) (Table 4-4). The values of R^2 and RMSE for the yield were close or slightly better than of those from DayCent (R^2 : $0.06\text{--}0.64$ and RMSE: $32\text{--}83\%$; Begum et al., 2017) and Roth-CNP (R^2 : $0.20\text{--}0.76$ and RMSE: $52\text{--}70\%$; Muhammed et al., 2018) when using the same dataset, indicating that SPACSYS is a competitive tool for predicting wheat yield. Moreover, SPACSYS has also a good performance for modelling SOC and TN stocks, which was comparable to the performance of DayCent, C-TOOL and Roth-C with R^2 of $0.23\text{--}0.85$ and RMSE of $7\text{--}9\%$ (Begum et al., 2017; Falloon and Smith, 2000; Taghizadeh-Toosi et al., 2014), but was better than the performance of CENTURY and Roth-CNP with R^2 of $0.18\text{--}0.86$ and RMSE of $6\text{--}17\%$ (Falloon and Smith, 2000; Muhammed et al., 2018; Taghizadeh-Toosi et al., 2014).

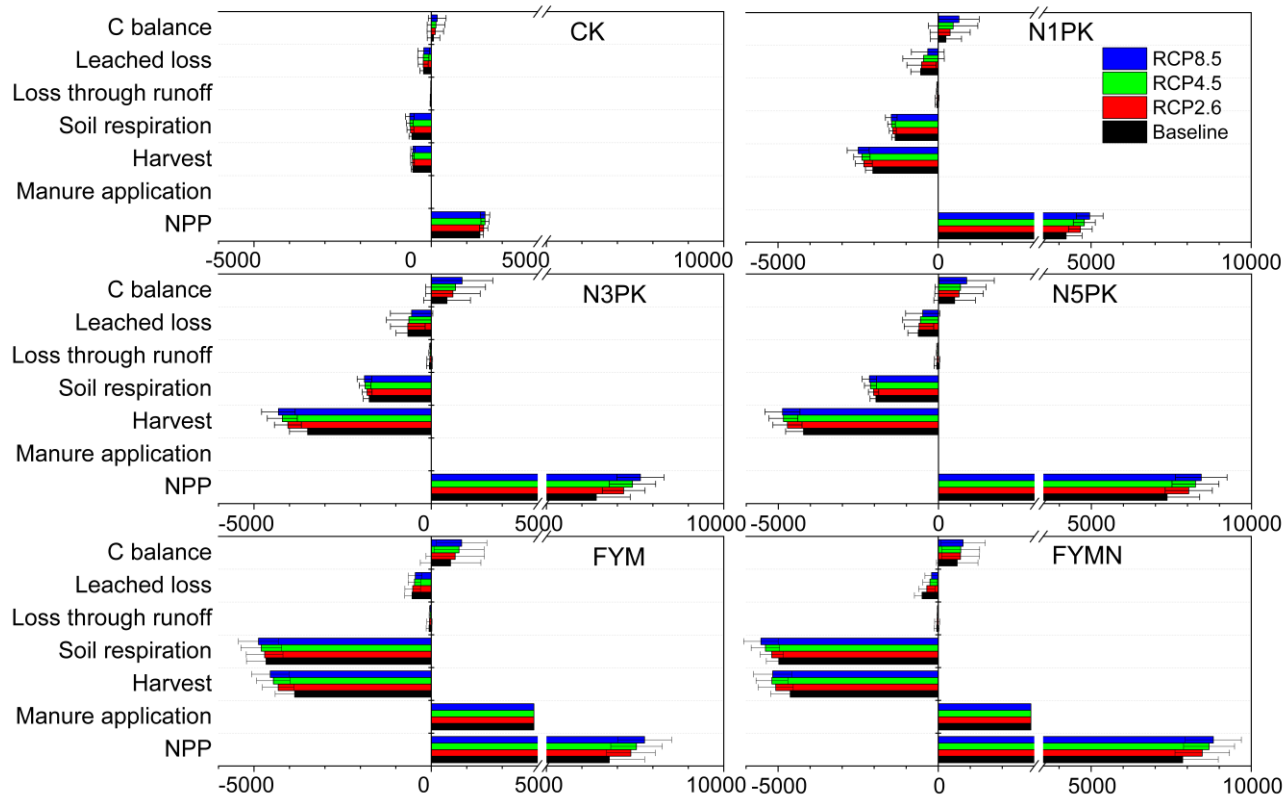


Figure 4-6 Simulated average annual carbon inputs and outputs ($\text{kg C ha}^{-1} \text{ yr}^{-1}$) with different fertiliser practises under the baseline and RCP climate scenarios. The bar is the standard deviation of annual values from 2021 to 2100.

Although the simulations turned up to be better performance, the discrepancies between simulated and observed values still existed over a century period, especially for some specific growing seasons. The reasons may be due to model simplification on the interactions between plant growth and driving variables, and sampling errors. The model was assumed that plant growth is not affected by either weeds, diseases or insects (Liang et al., 2018; Zhang et al., 2016c), which would be impossible for long-term field experiments especially before herbicide and insecticides were introduced. In reality, any adverse events happened in a growing season, the model would overestimate crop growth and grain yield. For example, an overestimation of 41% of grain yield and 69% of straw yield for cv. Red Standard during 1918 to 1925, which is in accordance with the record that “a decline set in during the first World War, when labour for hand weeding became scarce and weeds got out of control, and following the field one year in five was adopted to control weeds after 1925” (Jenkinson, 1991). Furthermore, extreme weather events occurred much more frequently recently. Heavy and persistent precipitation in the spring and/or summer of 1979, 2007 and 2012, a prolonged drought in 1989 and 2011, and a cold winter in 1991 and 2013 were recorded in the studied region (Addy et al., 2020; Harkness et al., 2020). The influence of the events, especially waterlogging, on crop growth and development has not been given enough attention in the model, which should be improved in the future.

4.2. Wheat yield under different climate change scenarios

Our prediction showed that future climate change had positive effects on the wheat yields for all the N application practises. It was reported that grain yield of winter wheat decreased by approximately 10% in South-East England if only precipitation or temperature changes relative to the baseline without considering the CO₂-enrichment effect (Cho et al., 2012). Apparently, an increasing CO₂ concentration plays a critical role in crop growth. Our results confirmed no significant difference in grain yield under the baseline and the RCP scenarios (N1PK, N3PK except RCP8.5) or a significantly decreased yield under the RCP scenarios (CK, N5PK, FYM and FYMN) when the CO₂ concentration was kept unchanged (Figure 4-S5). The contribution of the concentration to the grain yield was in the order of RCP8.5 > RCP4.5 > RCP2.6 (Table 4-S5). Controlled experiments on winter wheat in the studied region showed that the photosynthetic rate can increase 10% at a doubled CO₂ concentration (i.e. 700 $\mu\text{mol mol}^{-1}$) compared to that at the ambient concentration (Delgado et al., 1994), and a doubled CO₂ concentration (i.e. 684 $\mu\text{mol mol}^{-1}$) also increased the partitioning of assimilates to grain by 8.0 mg DM ear⁻¹ d⁻¹ as compared with that under 380 $\mu\text{mol mol}^{-1}$ CO₂ (Wheeler et al., 1996). Furthermore, it was reported by a summary of both field and laboratory results that the wheat yield would keep increasing until the CO₂ elevated to around 900 ppm (Amthor, 2001), which is very close to the CO₂ concentration around 2090s under the RCP8.5. Meanwhile, we found that grain yields under all the fertiliser practises could increase more under the high-emissions scenario (RCP8.5) compared with those under the low- or medium-emissions scenario (Tables 4-3 and 4-5; Figure 4-4), which is in agreement with the previous reports for the studied region (Cho et al., 2012; Semenov and Shewry, 2011;

William et al., 2018).

For the impact of the N application practises on the yield, there was no significant difference in grain yields with N5PK or FYMN among three RCP scenarios and the highest yield was achieved with the FYMN practise (Figure 4-4; Table 4-5). Those indicated that high N supplication might favour crop growth under the future climate change. Further, combined inorganic N and manure application could effectively increase soil organic matter and quantity of soil microbial community that release nutrients persistently (Yang et al., 2020). As a consequent, those are much favourable for wheat growth and final grain yield (Dhaliwal et al., 2020). Annual N application amounts between N5PK and FYM were very close (Table 4-1). However, the relative increase of the yield for FYM was higher than that for N5PK under the RCP scenarios (Table 4-5). This may be support by the increase of the soil's water-holding capacity through manure application. Under future increased temperature and reduced precipitation, manure application significantly mitigated the decline in soil water content during the wheat growth period, except in July, compared to NPK fertiliser (Figure 4-S6). This allows the soil to store and supply water more effectively to wheat roots (Rasool et al., 2008; Yang et al., 2011). Thus, with the similar application rate, manure application should be more beneficial to keep or increase the yield under climate change than that with chemical fertilisers applied only.

4.3. Climate change and fertilisation effects on SOC stock and C balance

The response of SOC stock to future climate change varied among the RCP scenarios and fertiliser practises. Although wheat yields for each NPK fertiliser treatment increased under the future climate change scenarios (Table 4-5, Figure 4-4), a significant decrease of the SOC stock was found under the RCP2.6 compared with the baseline (Table 4-5, Figure 4-5). The remarkable decrease in the fresh litter pool (Table 4-S6), transferred from above-ground residues and dead roots, could explain the SOC stock decrease. More precisely, the shortened grow period under the RCP scenarios resulted in a lower accumulation of dead materials (Figure 4-S7), despite a significant increase in crop biomass. For example, significant decreases in transferring plant biomass to the fresh litter pool for N5PK and FYMN under the climate scenarios compared to that under the baseline (Table 4-S7). Increasing temperature under the RCP scenarios would shorten the growing season of wheat in study region, which could provide less time to accumulate dead leaf, stem and root to above- and below-ground litter under future climate change (Balkovič et al., 2014; Senapati et al., 2019), especially during the reproductive stage in our study (Figure 4-S7). This has also been clarified by other modelling studies in the UK (Harkness et al., 2020; Richter and Semenov, 2005). In addition, our predictions proved that the enhanced biomass under the RCP scenarios increased the dissolved organic matter (DOM) pool (Table 4-S6) that mainly originates from fresh organic material, fresh litter and root exudates. This principally results in the SOC stock increase under higher atmospheric CO₂ concentration and intensified climate change situations (i.e., RCP4.5 and RCP8.5) in our study. C3 crops, like wheat, tend to produce more total exudation under higher

CO₂ concentrations (Drigo et al., 2008; Phillips et al., 2006). Further, an experiment with labelled ¹⁴C wheat showed that increases in CO₂ concentration quantitatively increased rhizosphere soluble C by 60% because of the significantly increased substrate input to the rhizosphere due to both increased root biomass and root activities per unit of roots under the elevated atmospheric CO₂ (Cheng and Johnson, 1998).

Furthermore, C balance analyses showed that climate change resulted in a net C sink and an average increase of 50–424 kg C ha⁻¹ yr⁻¹ as compared with the baseline (Table 4-5). An increase in NPP under the RCP scenarios supports the hypothesis that continuous wheat cropping in south-east England could result in a larger C sink (i.e. 112–881 kg C ha⁻¹ yr⁻¹) in the future as compared with the baseline (i.e. 62–593 kg C ha⁻¹ yr⁻¹) (Figures 4-6 and 4-S7; Table 4-5). The increase in NPP due to CO₂-fertilisation could be a dominant factor in determining whether SOC stocks continue to act as a sink of C in the future (Wieder et al., 2015b). Studies have indicated that the rising CO₂ concentration could ease climatic constraints to plant growth by decreasing stomata conductance and hence reducing transpiration, which could improve water use efficiency and inhibit drought stress, resulting in global NPP increases throughout the next few decades (Pan et al., 2014; Wieder et al., 2015b). Our simulations indicated there was an average increase of 524, 690 and 841 kg C ha⁻¹ yr⁻¹ for NPP under RCP2.6, RCP4.5 and RCP8.5 (Figure 4-6), respectively, which is close to previous study reporting increases in the range of 0–500 kg C ha⁻¹ yr⁻¹ under a low emission scenario, and 500–1000 kg C ha⁻¹ yr⁻¹ under a high emission scenario for the South-East of England (Pan et al., 2014). In addition, our predictions also showed significantly high soil respiration rates, especially from the microbial pool, under all the RCP scenarios compared with the baseline (Figure 4-S8; Table 4-S8). This is corroborated by the previous simulation reports that warming future climates in Europe would accelerate microbial respiration predicted by CENTURY (Lugato et al., 2018) and Roth-C (Gutierrez et al., 2023; Wiesmeier et al., 2016) both of which have the similar respiration-temperature relationship as SPACSYS. Therefore, increased C input through the CO₂ concentration effect can compensate C losses caused by the increased temperatures under the climate scenarios in the region.

4.4. Adaptation strategies to future climate change

Our simulations suggested that the practise of manure application only (FYM) or combined chemical N fertiliser and manure application (FYMN) can realise higher grain yields of winter wheat and SOC sequestration rates among the investigated practises under the future climate scenarios. Despite of higher soil respiration rates under the two practises (Fig. S7), the SOC sequestration rates are higher than those for N3PK and N5PK (Table 6). Thus, manure application practises seem to be sustainable for the continuous winter wheat cropping system in southeastern England to adapt to climate change.

However, in our prediction, the N application rates keep unchanged under future climate change. It has been reported that increasing the N application rate enhanced both crop yield and plant residues that was incorporated in the soil (Johnston et al.,

2009). Furthermore, the contribution of “CO₂-fertilisation effects” to vegetation photosynthesis has been declined at the global scale, owing to nutrient supply limitations (Wang et al., 2021). Thus, further investigation should be made to explore an appropriate N applied rate to enhance both crop yield and C sequestration in the study region under future climate change. Additionally, although planting the current variety of winter wheat under future climate change would not result in a yield decline due to the fertilising effect of increased CO₂, it is important to note that increased temperatures significantly reduce the growth period (Figure 4-S7). New cultivars, better adapted to the anticipated warmer and drier climates, could be introduced in the study region to achieve a greater yield increase and lower GHG emissions compared to the current variety (Ainsworth and Long, 2021; Shi et al., 2021).

5. Conclusion

Verified by the Broadbalk continuous wheat experiment over a period of more than 100 years, the SPACSYS model was able to simulate grain yield of different tall and modern short-strawed varieties of winter wheat, and the dynamics of SOC and TN stocks in the plough layer (i.e. 0–23 cm). Future climate change in the studied region had positive impacts on wheat grain yield for both chemical and manure applied practises because of a gradual CO₂ concentration increase. However, the effects of future climate change on the SOC stock varied among the RCP scenarios and fertiliser practises. Compared to the baseline, the medium- and high- emissions scenarios would cause significantly increase the SOC stock for all the N treatments by 2100 whilst the very stringent pathway (RCP2.6) resulted in reduction of the SOC stock for the practises of chemical fertiliser application only because of the least residue return among the scenarios. Further, manure application practises are effective to sustain the winter wheat production and promote soil C sequestration under future climate change in southeastern England. In conclusion, our results provide in-depth insights into the response of wheat yield and cropland SOC dynamics under long-term fertilisation as well as future climate change conditions, especially for the regions with long history of fertilisations or approaching the SOC equilibrium level.

6. Supplementary Figures and Tables

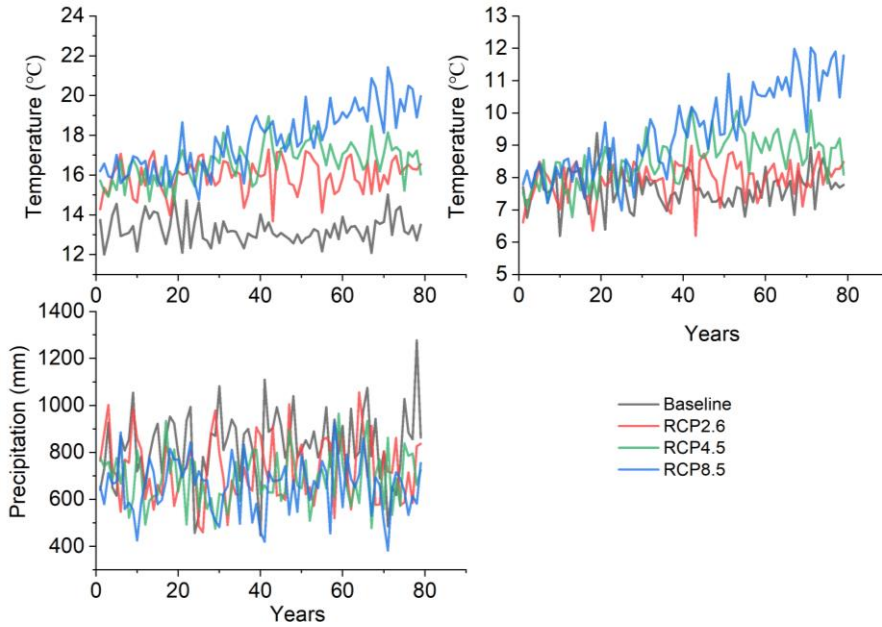


Figure 4-S1 Annual maximum and minimum temperatures and precipitation under future climate scenarios and the baseline.

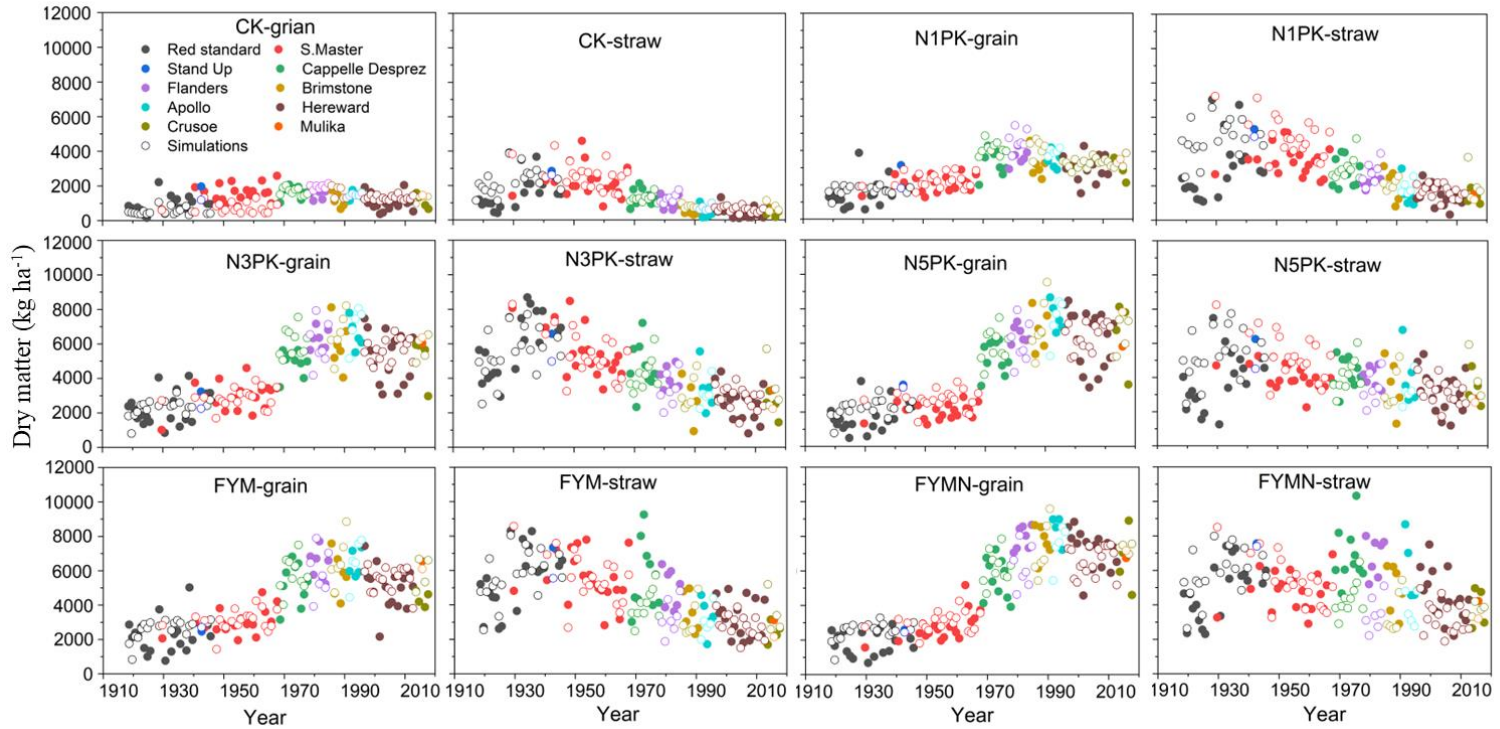


Figure 4-S2 Simulated and grain yield and straw dry matter of winter wheat over the simulated period for different fertiliser application treatments. Open circles represent simulated results.

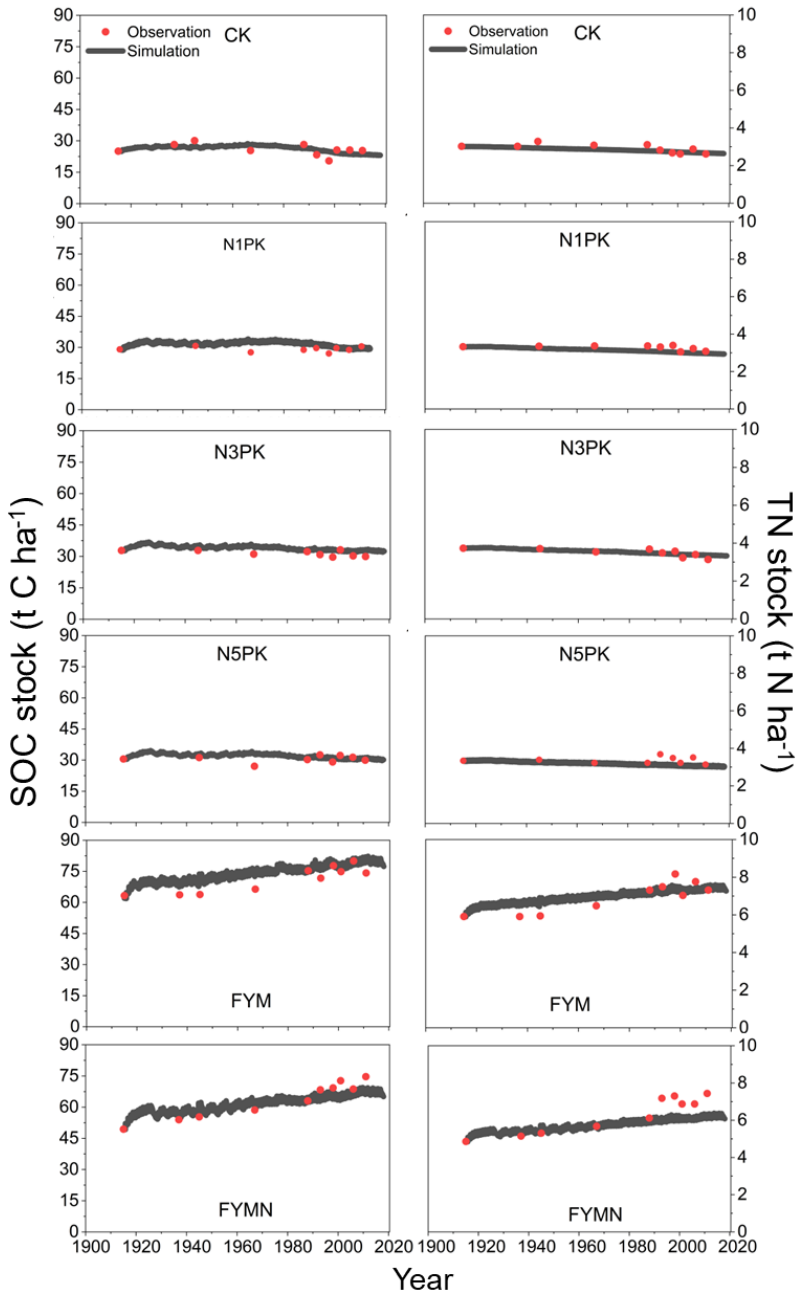


Figure 4-S3 Simulated and measured SOC stock and TN stock over the simulated period for different fertiliser application treatments.

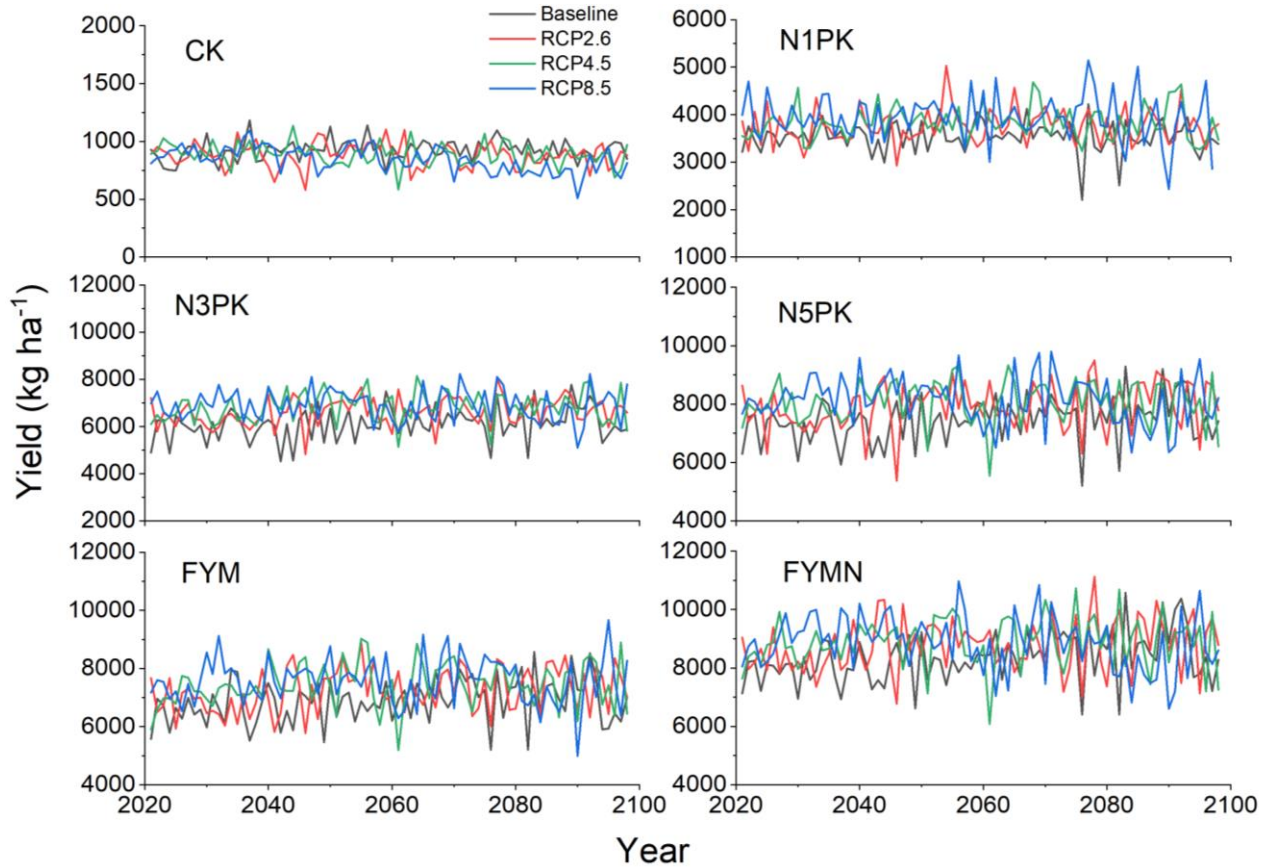


Figure 4-S4 Predicted wheat yield for different fertiliser practises under different climate scenarios.

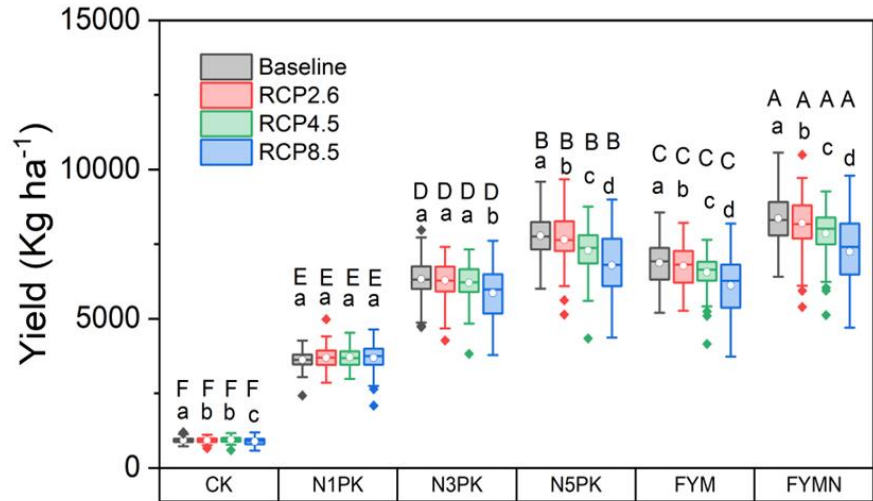


Figure 4-S5 Wheat grain yield during 2021–2100 with different fertiliser practises under future climate scenarios without considering CO₂ concentration change. The lines and circles within the boxes are the median and mean values. The spots are outliers. Columns with different lowercase letters indicate significant differences among climate scenario with an individual treatment ($P < 0.05$). Columns with different capital letters indicate significant differences among different treatments under the same climate scenario ($P < 0.05$).

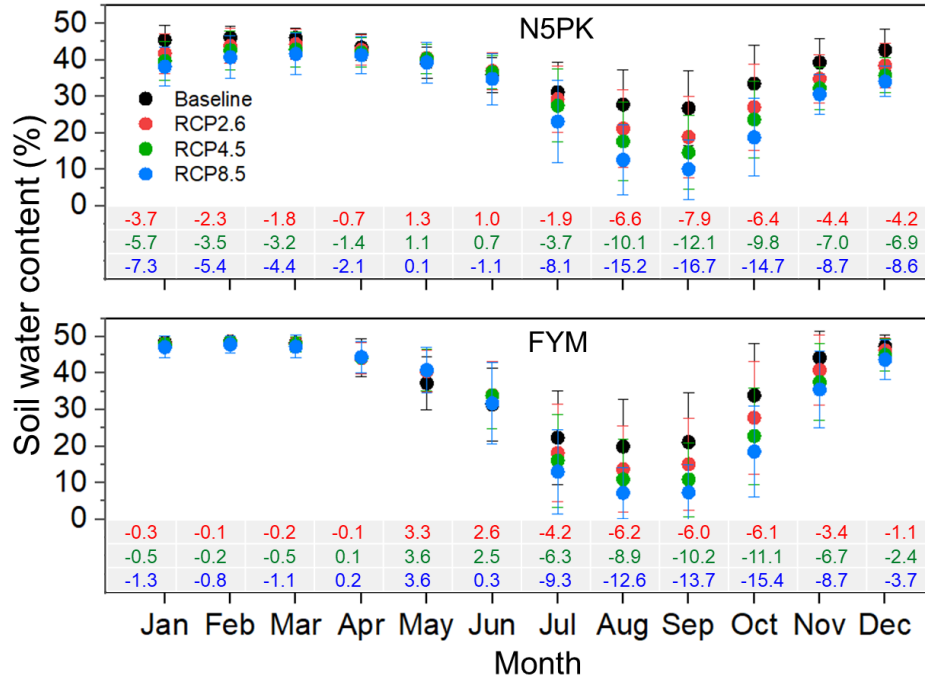


Figure 4-S6 Simulated monthly volumetric soil water content (%) with standard deviation in the plough layer from 2021 to 2100 with N5PK and FYM under future climate scenarios at Broadbalk. The values located at the bottom of the figures are the average changes of soil water content (%) under future climate scenarios compared to the baseline.

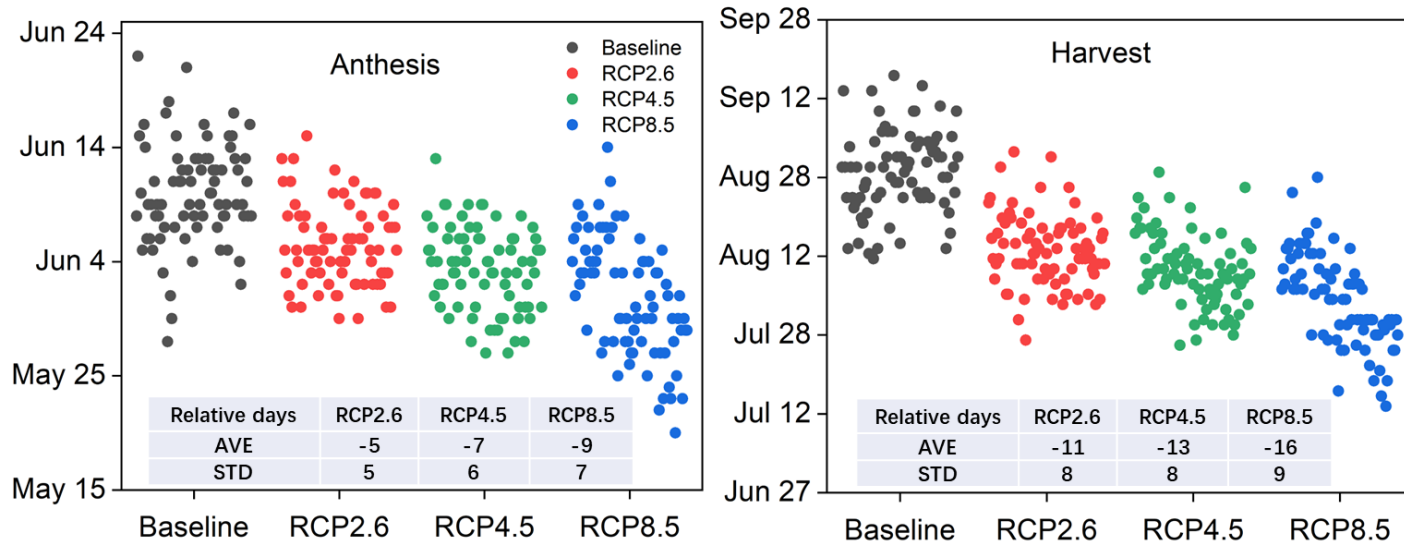


Figure 4-S7 Anthesis and harvest dates under future climate scenarios from 2021 to 2100, and average length changes and standard deviation for the periods from emergence to anthesis and from anthesis to maturity under the climate scenarios compared with those under the baseline.

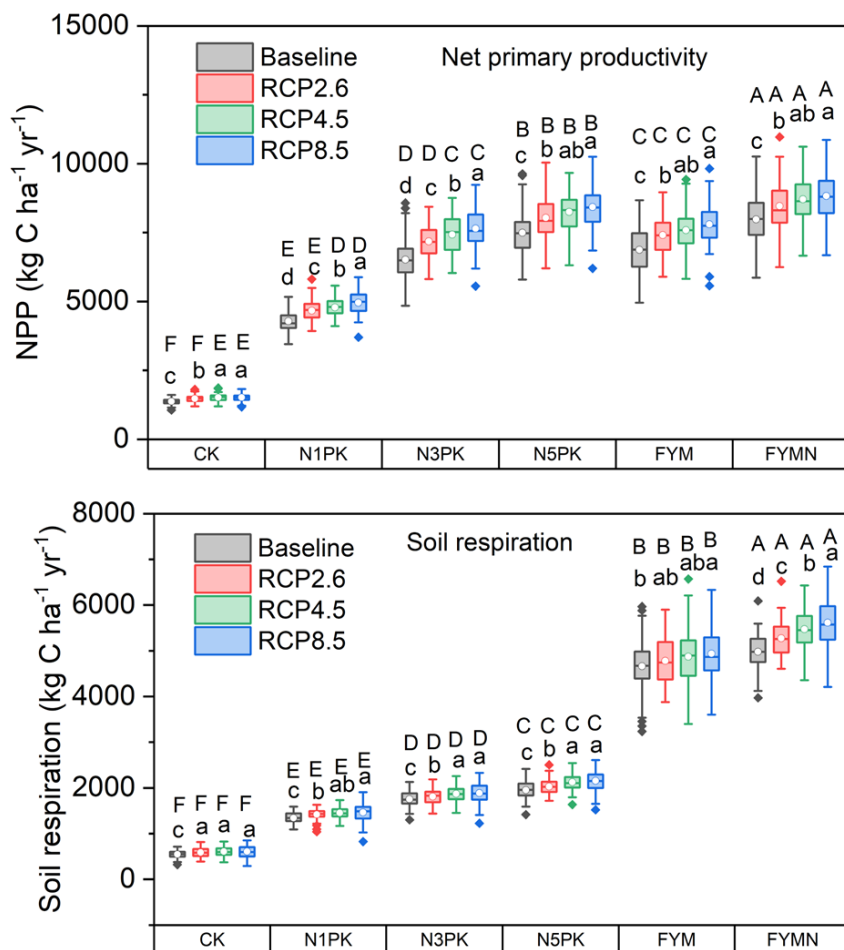


Figure 4-S8 Simulated average annual net primary productivity (NPP) (kg C ha⁻¹ yr⁻¹) and soil respiration (kg C ha⁻¹ yr⁻¹) during 2021 to 2100 with all the fertiliser management practises under future climate scenarios at Broadbalk. The lines and open circles within the boxes are the median and mean values. The spots are outliers. Columns with different lowercase letters indicate significant differences among climate scenario with an individual treatment ($P < 0.05$). Columns with different capital letters indicate significant differences among different treatments under the same climate scenario ($P < 0.05$).

Table 4-S1 Wheat cultivars in the Broadbalk continuous winter wheat experiment over the simulated years at Rothamsted Research, Harpenden, UK

Year	Cultivars
Tall-strawed	
1915-1916	S. Master
1917-1928, 1930-1939, 1944, 1945	Red standard
1929, 1940, 1941, 1943, 1946-1967	S. Master
1942	Stand up
Short-strawed	
1968-1978	Cappelle Desprez
1979-1984	Flanders
1985-1990	Brimstone
1991-1995	Apollo
1996-2012	Hereward
2015	Mulika [†]
2013-2018	Crusoe

[†] In 2015, a spring wheat variety was sown, because the bad weather conditions delayed the sowing from autumn until early spring.

Table 4-S2 Optimized parameters related to wheat growth and development for different cultivars used in simulations

Parameter	Unit	Tall-strawed variety	Short-strawed varieties	
		Red Standard	Cappelle Desprez	Hereward
Accumulated temperatures required from sowing to emergence	°C·d	120	120	120
Accumulated temperatures required from emergence to flowering	°C·d	680	750	750
Accumulated temperatures required from emergence to flag leaf fully expansion	°C·d	880	900	900
Accumulated temperatures required from flowering to maturity	°C·d	1260	1500	1400
Maximum plant height	m	1.55	1.05	1.05
Critical photoperiod for vegetative stage below or over which plant development will not be affected by light	hr	14.5	14.5	14.5
Critical photoperiod for vegetative stage below or over which plant development will stop	hr	8.0	8.0	8.0
Threshold temperature for emergence	°C	3.0	4.0	4.0
Threshold temperature for vegetative stage	°C	-4.0	0.0	-4.0
Threshold temperature for reproductive stage	°C	10.0	10.0	10.0
Specific leaf area	m ² ·g ⁻¹ ·DM	0.023	0.02	0.02
Specific green ear area	m ² ·g ⁻¹ ·DM	0.00637	0.00637	0.00637

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Specific green stem area	$\text{m}^2 \cdot \text{g}^{-1} \cdot \text{DM}$	0.006	0.006	0.006
Rate of carbon translocation from leaf to grain during reproductive stage	d^{-1}	0.0005	0.003	0.003
Rate of carbon translocation from stem to grain during reproductive stage	d^{-1}	0.001	0.01	0.013
Rate of N translocation from leaf to grain during reproductive stage	d^{-1}	0.018	0.02	0.025
Rate of N translocation from stem to grain during reproductive stage	d^{-1}	0.012	0.023	0.028

Table 4-S3 Optimized parameters related to soil C and N cycling in SPACSYS

Parameter	unit	Values
Ammonium immobilised fraction	-	0.45
Humus fraction from dissolved	-	0.03
Humus fraction from fresh litter	-	0.05
Humus fraction from fresh organic matter	-	0.09
Potential decomposition rate for humus	d^{-1}	0.000025
Potential decomposition rate for fresh litter organic matter	d^{-1}	0.008
Potential decomposition rate for fresh organic matter	d^{-1}	0.009
Potential decomposition rate for dissolved organic matter	d^{-1}	0.012
Critical C/N ratio favourable to immobilization	-	10
Microbial maintenance respiration rate	d^{-1}	0.678
Maximum autotrophic nitrification rate	d^{-1}	0.08
Potential heterotrophic nitrification rate	d^{-1}	0.04

Maximum nitrifier growth rate	d^{-1}	9.74
Precipitation N concentration	$g\ N\ m^{-3}$	0.80
Relative activity at saturated water content	-	0.60
Dry deposition mineral N to soil	$g\ N\ m^{-2}\ day^{-1}$	0.004
Base temperature at which temperature function is in unity for denitrification process	$^{\circ}C$	25
Base temperature at which temperature function is in unity for mineralization-immobilisation process	$^{\circ}C$	28
Base temperature at which temperature function is in unity for nitrification process	$^{\circ}C$	20

Table 4-S4 ANOVA test for the effects of fertilisation treatments and climate scenarios on wheat yield, SOC stock, C budget, net primary productivity and soil respiration between 2021 to 2100 ($P < 0.05$)

	df	F	P
Yield (considering CO ₂ concentration)			
Treatment	5	5375	<0.001
Climate scenario	3	60	<0.001
Treatment*Climate scenario	15	4	<0.001
SOC stock			
Treatment	5	4708483	<0.001
Climate scenario	3	1229	<0.001
Treatment*Climate scenario	15	41	<0.001
C budget			
Treatment	5	49	<0.001
Climate scenario	3	11	<0.001
Treatment*Climate scenario	15	0.4	0.98
Yield (without considering CO ₂ concentration)			
Treatment	5	4388	<0.001
Climate scenario	3	62	<0.001
Treatment*Climate scenario	15	11	<0.001
Net primary productivity			
Treatment	5	4632	<0.001
Climate scenario	3	141	<0.001
Treatment*Climate scenario	15	5	<0.001
Soil respiration			
Treatment	5	11650	<0.001
Climate scenario	3	45	<0.001
Treatment*Climate scenario	15	5	<0.001

Table 4-S5 The relative change (%) of wheat yield with and without considering CO₂ concentration compared to those under the baseline during 2021–2100[‡]

Treatments	Scenarios	Relative changes
CK	RCP2.6	0.06
	RCP4.5	0.10
	RCP8.5	0.62
N1PK	RCP2.6	0.11
	RCP4.5	0.34
	RCP8.5	3.77
N3PK	RCP2.6	0.80
	RCP4.5	5.53
	RCP8.5	13.44
N5PK	RCP2.6	3.84
	RCP4.5	10.88
	RCP8.5	20.78
FYM	RCP2.6	7.43
	RCP4.5	13.82
	RCP8.5	24.02
FYMN	RCP2.6	6.15
	RCP4.5	13.23
	RCP8.5	22.76

[‡] It is calculated as: $\frac{Yield_{RCP\ with\ CO_2} - Yield_{RCP\ without\ CO_2}}{Yield_{baseline}} \times 100$

Table 4-S6 Average relative changes ($\text{kg C ha}^{-1} \text{ yr}^{-1}$) of main carbon pools under the RCP scenarios compared with the baseline during 2021–2100

Treatments	Scenarios	Fresh litter pool	Dissolved organic matter pool	Humus pool
CK	RCP2.6	-47	151	-93
	RCP4.5	7	298	-98
	RCP8.5	45	352	-19
N1PK	RCP2.6	-552	612	-175
	RCP4.5	-508	1353	-174
	RCP8.5	-449	1886	-52
N3PK	RCP2.6	-649	650	-42
	RCP4.5	-561	1293	-17
	RCP8.5	-541	1753	69
N5PK	RCP2.6	-798	556	-123
	RCP4.5	-767	1050	-53
	RCP8.5	-745	1572	34
FYM	RCP2.6	-384	254	925
	RCP4.5	-348	327	1093
	RCP8.5	-360	507	1294
FYMN	RCP2.6	-848	171	1676
	RCP4.5	-859	236	1740
	RCP8.5	-871	395	1905

Table 4-S7 Average annual transferred C ($\text{kg C ha}^{-1} \text{ yr}^{-1}$) from dead leaves and stems to residue and from roots to the fresh litter pool with N5PK and FYMN under each climate scenario during 2021–2100

Treatment	Climate scenarios	Leaves to residue	Stems to residue	Roots to litter
N5PK	Baseline	571a	1170a	420b
	RCP2.6	507b	1069b	439b
	RCP4.5	513b	1061b	457ab
	RCP8.5	511b	1009b	488a
FYMN	Baseline	579a	1233a	430b
	RCP2.6	513b	1104b	445b
	RCP4.5	520b	1095b	464ab
	RCP8.5	521b	1040b	498a

Numbers with different lowercase letters indicate significant differences among climate scenarios for a treatment ($P < 0.05$).

Table 4-S8 Average CO_2 release rate ($\text{kg C ha}^{-1} \text{ yr}^{-1}$) from different soil carbon pools with N5PK and FYMN under four climate scenarios during 2021–2100

Treatment	SOM pools	Baseline	RCP2.6	RCP4.5	RCP8.5
N5PK	Dissolved organic matter pool	44	67	91	93
	Humus pool	106	115	119	122
	Microbial pool	876	937	986	1026
	Fresh litter pool	921	927	932	936
FYMN	Dissolved organic matter pool	354	439	515	577
	Humus pool	186	165	170	171
	Microbial pool	2241	2389	2479	2593
	Fresh litter pool	953	958	985	999
	Fresh OM pool	1145	1147	1149	1149

Chapter V

Both soil productivity and carbon sequestration of Mollisols decrease under future climate change

From: Liang, S., Sun, N., Longdoz, B., Meersmans, J., Ma, X., Gao, H., Zhang, X., Qiao, L., Colinet, G., Xu, M., Wu, L., 2024. Both yields of maize and soybean and soil carbon sequestration in typical Mollisols cropland decrease under future climate change: SPACSYS simulation. *Frontiers In Sustainable Food Systems* 8, 1332483. <https://doi.org/10.3389/fsufs.2024.1332483>

Abstract

Although Mollisols are renowned for their fertility and high-productivity, high C losses pose a substantial challenge to the sustainable provision of ecosystem services, including food security and climate regulation. Protecting these soils with a specific focus on revitalizing their C sequestration potential emerges as a crucial measure to address various threats associated with climate change. In this study, we employed a modelling approach to assess the impact of different fertilisation strategies on crop yield, soil organic carbon (SOC) stock, and C sequestration efficiency (CSE) under various climate change scenarios (baseline, RCP2.6, RCP4.5, and RCP8.5). The process-based SPACSYS model was calibrated and validated using data from two representative Mollisol long-term experiments in Northeast China, including three crops (wheat, corn and soyabean) and four fertilisations (no-fertiliser (CK), mineral nitrogen, phosphorus and potassium (NPK), manure only (FYM), and chemical fertilisers plus FYM (NPKM or FYMN)). SPACSYS effectively simulated crop yields and the dynamics of SOC stock. According to SPACSYS projections, climate change is anticipated to reduce corn yield by an average of 14.5% in Harbin and 13.3% in Gongzhuling, and soybean yield by an average of 10.6%, across all the treatments and climatic scenarios. Conversely, a slight but not statistically significant average yield increase of 2.5% was predicted for spring wheat. SOC stock showed a decrease of 8.2% for Harbin and 7.6% for Gongzhuling by 2100 under the RCP scenarios. Future climates also led to a reduction in CSE by an average of 6.3% in Harbin (except NPK) and 12.1% in Gongzhuling. In addition, the higher average crop yields, annual SOC stocks, and annual CSE (10.64–14.56%) were found when manure amendments were performed under all climate scenarios compared with the chemical fertilisation. Soil CSE displayed an exponential decrease with the C accumulated input, asymptotically approaching a constant. Importantly, the CSE asymptote associated with manure application was higher than that of other treatments. Our findings emphasize the consequences of climate change on crop yields, SOC stock, and CSE in the Mollisol regions, identifying manure application as a targeted fertiliser practise for effective climate change mitigation.

Key words: Mollisols, yield, SOC stock, carbon sequestration efficiency, SPACSYS model, long-term fertilization, climate change

1. Introduction

Mollisols, characterized by their thick, dark and well-structured surface horizon, stand out as the most fertile and productive soils on Earth (Durán et al., 2011). Comprising approximately 7% of the world's ice-free land surface, these soils account for 28.6% of global farmland across all soil types (Durán et al., 2011; Liu et al., 2012; Xu et al., 2020). Their unique characteristics make Mollisols exceptionally suitable for cultivating cereal crops and establishing pasture and forage systems (Durán et al., 2011; Xu et al., 2020), playing a vital role in agricultural productivity and global food security (Liu et al., 2012).

Nearly 50% of China's corn yield and approximately one-third of its soybean yield are cultivated on the fertile Mollisols located in the Northeastern provinces of Heilongjiang, Jilin, Liaoning, and the Inner Mongolian Autonomous Region (Liu et al., 2013; Xu et al., 2020). However, the crop yield growth rate has significantly decreased since 2000, despite the ongoing rise in fertiliser application and the expansion of irrigated area (Tong et al., 2017). Among the numerous challenges faced in this region, the declining SOC is the most notable. Reports indicated Mollisols in Northeast China have experienced a decline of around 50% in SOC (from 103 to 43.2 g kg⁻¹) between the 1950s and the 1980s, and a further 50% decrease (to an average of 21.9 g kg⁻¹ from 2016 to 2021) occurred over the subsequent four decades (Tong et al., 2017; Meng et al., 2024). This decline is attributed to active agricultural utilization, low organic matter return, and intensive tillage practises (Durán et al., 2011; Liu et al., 2012; Xu et al., 2020). In addition, even a slight reduction in C from the substantial SOC pool (approximately 4 Pg) in Mollisols could lead to significant CO₂ emissions within these regions (Cheng et al., 2010). For instance, the average C release rate from Mollisols in Northeast China ranged from 0.17 to 2.17 Tg C yr⁻¹, significantly surpassing the global average of 0.09–0.78 Tg C yr⁻¹ from the pedological C pool (Lal et al., 2007; Li et al., 2013). Therefore, strategic C sequestration in Mollisols is crucial not only for ensuring food security and promoting soil stability but also for effectively mitigating global climate change.

The potential to sequester C in soils depends not only on the quantity of C input but also on the retention coefficient of the applied C, a concept referred to as C sequestration efficiency (i.e., CSE, $\Delta\text{SOC}/\Delta\text{C input}$) (Maillard and Angers, 2014; Stewart et al., 2007). Stewart et al. (2007) established an asymptotically increasing relationship between C inputs and changes in SOC, based on data from 14 long-term experiments conducted over 12 to 96 years of fertilisation. This highlighted a decreasing CSE as the soil approached C saturation (Hassink and Whitmore, 1997; Six et al., 2002). However, our study in Chapter II indicated CSE remained nearly constant within four decades of fertilisation. This was accompanied by a linear relationship between C inputs and SOC changes, as validated by eight extensive long-term experimental datasets, including the Mollisol site at Gongzhuling. Therefore, a narrow range of C input levels, corresponding to shorter fertilisation durations, did not necessarily reflect the full spectrum of linear-to-asymptotic behaviours for SOC

content (Stewart et al., 2007), and, consequently, the CSE characteristics. Further, examining SOC dynamics that encompasses Mollisols across a broad range of C input levels is essential and urgent to comprehensively elucidate their CSE characteristics.

Optimal fertiliser practises play a critical role in promoting SOC restoration in degraded Mollisols (Xu et al., 2020). Manure application, especially when combined with chemical fertilisers, has demonstrated the potential to enhance CSE due to the beneficial effects of additional organic C and nutrients on soil fertility and soil structure in Chapter II. However, the sustainability of this benefit under long-term fertilisation conditions as soil C approaches saturation requires further investigation. In addition, most Mollisol regions experience cool, moist, and semi-humid climates (Xu et al., 2020), and the warmer and wetter future climates in these regions might significantly impact SOC sequestration by influencing crop production and soil ecological processes (Chu et al., 2017; Lin et al., 2017). Thus, predicting SOC changes and crop yields (associated with C input) in Mollisols is necessary to explore the dynamics of CSE and its response to future climate change under different fertilisation strategies.

Process-based models have become essential decision support tools for addressing climate change issues in agriculture management and production, as these models enable the integration of diverse data sources and knowledge concerning the impacts of phenological and environmental variables allowing for valuable evaluations of specific hypotheses and scenarios (Smith et al., 2005). Among the widely used process-based models, the SPACSYS model stands out for its comprehensive consideration of processes in C, N and P cycling. Numerous studies have demonstrated its robust capabilities in accurately simulating plant growth, N and P uptake, SOC and TN stocks, as well as CO₂ and N₂O emissions across cropland and grassland in diverse regions of Europe and China (Li et al., 2017; Liang et al., 2018; Perego et al., 2016; Wu et al., 2015). In addition, Furthermore, validated using data from the Broadbalk continuous wheat experiment over a century of fertilisation, the SPACSYS model has successfully captured the dynamics of SOC stock in the plough layer (Chapter V). These results confirm the SPACSYS model's reliability in simulating SOC changes and crop yields in response to different long-term fertilisation strategies and various climatic conditions. The Mollisol long-term experiments established in the 1980 s in Northeast China offer a valuable resource with well-documented and comprehensive records, detailing fertilisation practises, cultivation methods, crop management, climate conditions, grain and straw yields, and soil properties. These detailed records provide a solid foundation for the modelling study and enable accurate predictions of the impacts of fertiliser management and climate change on CSE.

In this study, we aimed to elucidate the responses of crop yield and CSE to climate change in the typical Mollisol regions of China, while proposing a sustainable strategy to enhance both crop productivity and C sequestration. Specifically, firstly we initially calibrated and validated the SPACSYS model using data from two Mollisol long-term experiments in Northeast China. Subsequently, we quantified the impacts of various future climate scenarios and fertiliser application practises on crop yield, SOC stock,

and CSE within the plough layer (0–20 cm) of Mollisols. Finally, we elucidated the comprehensive CSE characteristics when a substantial amount of C has accumulated in the future.

2. Materials and Methods

2.1. Study sites background information

2.1.1. Study sites and experimental design

Two typical Mollisols long-term experiments located in Northeast China at Harbin (45°40' N, 126°35' E) and Gongzhuling (43°30' N, 124°48' E) have been selected for this study. The essential background information, including the locations, initial soil properties, cropping systems and climatic conditions, is given in Table 5-1. For both experiments, four fertiliser treatments common were chosen, namely (1) no fertiliser (CK), (2) combinations of chemical nitrogen (N), phosphorus (P) and potassium (K) fertilisers (NPK), (3) manure application (FYM), and (4) chemical fertilisers (N or NPK) plus manure (FYMN/NPKM) (refer to Table 5-S1 for details). Detail information can be found in the works of Jiang et al. (2014, 2018a).

Table 5-1 Information of location, climate type and initial soil properties of two experimental sites

Site	Harbin	Gongzhuling
Starting year	1979	1989
Location	45°40' N, 126°35' E	43°30' N, 124°48' E
Climate type	Mild temperate; Semi-humid	Mild temperate; Semi-humid
Annual temperature, °C	3.5	4.5
Annual precipitation, mm	533	595
Cropping system	Single cropping Corn-soybean-wheat	Single cropping Corn
Initial soil properties		
Initial SOC ¹ , g kg ⁻¹	15.66	13.23
Total N ² , g kg ⁻¹	1.47	1.40
Available N, mg kg ⁻¹	151	114
Total P ³ , g kg ⁻¹	1.07	1.39
Olsen P, mg kg ⁻¹	51	27.0
Total K ⁴ , g kg ⁻¹	25.16	22.1
Available K, mg kg ⁻¹	200	190

pH	7.2	7.6
Bulk density, g cm ⁻³	1.37	1.19
Clay, %	30	32

¹ SOC means soil organic carbon; ² N means nitrogen; ³ P means phosphorus; ⁴ K means potassium.

2.1.2. Soil sampling and analysis

The soil samples of Gongzhuling in the plough layer (0–20 cm) were collected from 1989 to 2018 annually in approximately 15 days after harvest (n=28 for each treatment). The soil samples of Harbin in the plough layer (0–20 cm) were collected annually from 1979 to 1988 and almost every three years from 1988 to 2018 in approximately 15 days after harvest (n=23 for each treatment). The SOC content was determined using dichromate oxidation following the modified Walkley & Black method (i.e., heated for 12 minutes at 220°C) with a correction factor of 1.1 (Walkley and Black, 1934; Xu et al., 2016). While the factor may vary between samples with and without manure applications, a very high heating temperature of 220°C results in a nearly complete oxidation of the organic carbon, and as such, also the more recalcitrant fractions. This strongly limits the potential error introduced to the application of it across all treatments. In this context, we assumed that the error introduced in SOC measurements due to variations in the correction factor among the different treatments could be considered negligible.

2.2. Model parameterisation and prediction

2.2.1. Model description

The SPACSYS model is a field-scale, weather-driven with flexible time steps (ranging from minutes to daily), employing a process-based approach that quantifies the biogeochemical processes related to C, N, and P cycling, as well as the water and heat budgets within the soil-plants-and atmosphere continuum. In the current version (6.0) the model incorporates the Farquhar method for photosynthesis (Yin and Struik, 2009) coupled with a modified stomatal conductance sub-model based on Leuning et al. (1995) and Tuzet et al. (2003). This integration enables simulation of the plant photosynthesis rates accounting for the impacts of atmospheric CO₂ concentration on photosynthesis and transpiration.

Compared to other widely cited process-based simulation models, SPACSYS stands out due to its detailed representation biogeochemical processes (refer to Table 1-1) and a substantial number of organic matter pools (five). Additionally, SPACSYS includes a detailed root architecture that significantly influences nutrients and water capture, comprehensive descriptions of above- and below-ground plant growth and a nutrient recycling (Wu et al., 2007). More information about SPACSYS can be found in previous articles (Bingham and Wu, 2011; Wu et al., 2007, 2015).

2.2.2. Calibration, validation, and prediction

SPACSYS was operated from the starting year of the experiments (1979 for Harbin and 1989 for Gongzhuling) until 2020 using a daily time-step. Daily weather data (maximum and minimum temperatures, precipitation, wind speed, relative humidity, and solar radiation) were obtained as inputs from the National Meteorological Information Center ([http:// data.cma.cn/](http://data.cma.cn/)). Agronomic management details including soil work (ploughing depth and date), plant management (seeding and harvesting dates), and fertiliser application (date, amount and type) for each treatment in the Harbin and Gongzhuling long-term experiments, were sourced from records maintained by the Heilongjiang Academy of Black Soil Conservation and Utilization and Jilin Academy of Agricultural Sciences, respectively. Soil properties in the top 20 cm deep at each site (Table 5-1) were acquired through field surveys. Other missing properties that the model required were estimated from soil texture using pedo transfer functions (PTFs) implemented in the model. The atmospheric CO₂ concentration was set from 330 to 380 ppm for Harbin and from 348 to 380 ppm for Gongzhuling, following a linear increase over the experimental period, as per IPCC (2021) guidelines. Grain yields and straw dry matter (if recorded) of maize, wheat and soybean, as well as SOC and TN stocks from the NPK and NPKM/FYMN treatments, were randomly chosen for the model calibration. Data from the other treatments served for model validation. Parameters related to C and N cycling, soil water movement, heat transformation, and plant photosynthesis were adopted from previous studies (Bingham and Wu, 2011; Wu et al., 2015; Liu et al., 2020). The parameters related to the manure decomposition and transformation were from our previous reports (Zhang et al., 2016c; Liang et al., 2018). The built-in Multi-Objective Shuffled Complex Evolution Metropolis algorithm (MOSCEM-UA) (Vrugt et al., 2003) was applied to determine the other parameters, primarily those linked to phenology and vegetation characteristics. The optimized parameters and their values for corn, wheat, soybean, and soil C and N cycling are listed in Tables 5-S3 and 5-S4.

For crop yield and SOC predictions, four climate scenarios were considered, encompassing the baseline and three representative concentration pathways (RCP2.6, RCP4.5, and RCP8.5; Riahi et al., 2011; van Vuuren et al., 2011). The baseline scenario involved historic meteorological data rotated between 1979 and 2020 for Harbin and between 1989 and 2020 for Gongzhuling with a constant CO₂ concentration of 380 ppm. Weather data for the RCP scenarios between 2021 and 2100 were extracted from the HadGEM2-ES model with a spatial resolution of 0.5° × 0.5° (Collins et al., 2011; Jones et al., 2011). The annual average of maximum and minimum temperatures, precipitation, and CO₂ concentration under the three RCP scenarios are shown in Figure 5-S1 and Table 5-S2. The climate projections indicated warmer and wetter future conditions in the studied regions, with average increases of 0.8–3.5°C and 0.4–2.5°C observed in Harbin and Gongzhuling, respectively, when compared to the baseline. The mean annual precipitation (MAP) was projected to rise by 43–121 mm in Harbin and 137–207 mm in Gongzhuling. The RCP8.5 scenario demonstrated the most significant alteration, nearly doubling CO₂ concentrations by 2100.

Crop varieties and field management practises were fixed as identical to those before 2021 for both sites and scenario. To simplify the assessment of future climate change on crop phenology and yield, the sowing dates were fixed on 30th April for maize, 23rd April for wheat, and 29th April for soybean in Harbin and 26th April in Gongzhuling each year in this study. Harvest dates were determined based on simulated physiological maturity. Simulations were run for each of the four treatments under the various proposed climate scenarios.

2.3. Soil carbon sequestration efficiency calculation

Since SOC predominately undergoes changes in the plough layer (0–20 cm) in Mollisol (Liang et al., 2009), our research concentrates on SOC within this layer. The calculations for SOC stock, carbon input, SOC sequestration rate, and soil carbon sequestration efficiency within the plough layer at each study site were conducted utilizing the consistent equations presented in Section 2.4 of Chapter III. The C content of shoots and roots, the RSR and HI for the studied crops, as well as the C content of manure, were detailed in Table 5-S5.

2.4. Statistical analysis

We used three statistical criteria to evaluate the model performance: (1) the coefficient of determination (R^2) that measures the degree of agreement between simulation and observation; (2) the root mean squared error (RMSE), providing a measure of the average deviation of the estimates from observed values; and (3) the relative error (RE) that reflects the overall difference between simulated and observed data.

Two-way analysis of variance (ANOVA) and Tukey tests ($P < 0.05$) were used to compare the effects of fertiliser practises and future climate scenarios on grain yield, SOC stock, C input, and CSE. All statistical analyses were conducted using SPSS 24.0 (SPSS, Inc., 2017, Chicago, USA). Graphs were performed by Origin Profession version 2019 (OriginLab, 2019, Northampton, USA).

3. Results

3.1. Model calibration and validation

The simulated crop yields for different treatments at both sites closely aligned with observations, demonstrating an RMSE below 24% (Table 5-2, Figures 5-S2 – 5-S5). Linear regression analysis further revealed a satisfactory correlation between the observed and simulated values ($R^2 > 0.54$) (Figure 5-1), despite a tendency for the model to overestimate low observed yields and underestimate high observed ones. It was noted that SPACSYS underestimated corn grain and straw yields at Gongzhuling by an average of 16% and 20%, respectively, from 2010 to 2012 (Figure 5-S3).

Moreover, SPACSYS effectively simulated SOC and TN stocks, with R^2 values ranging from 0.60 to 0.79 and RMSE ranging from 6.8 to 8.2% (Table 5-3; Figure 5-1). This favourable agreement with measurements is particularly evident in the first

decade of the simulation period (Figures 5-S4 and 5-S5). In general, the model exhibited a 20% underestimation of SOC stock and a 37% underestimation of TN stock during model validation (Figure 5-1). However, it overestimated TN stock by an average of 7.3% for CK, 10.2% for NPK, 8.5% for FYMN, and 9.0% for FYM after 2000 in Harbin (Figure 5-S4).

3.2. Crop yield and SOC stock

Average yields of wheat, corn, and soybean from 2021 to 2100 under each climate scenario with different fertiliser practises are shown in Figure 5-2. Notably, a slight but not statistically significant average yield increase of 2.5% was predicted for spring wheat ($P < 0.05$). Under all climate scenarios, there was also no significant difference ($P < 0.05$) between the NPK and NPKM/FYMN fertiliser treatments for corn (1005–12101 vs 1112–12573 kg ha⁻¹), wheat (2729–5548 vs 2727–5287 kg ha⁻¹), and soybean (975–4314 vs 1024–4350 kg ha⁻¹) at both sites. However, the yields for these treatments were significantly higher than those for the FYM and CK treatments ($P < 0.05$). Compared with the baseline, the corn yield decreased by an average of 14.5% and 13.3% for Harbin and Gongzhuling, respectively, across all RCP scenarios and under all considered fertiliser practises (Table 5-3). The decreases in corn yield under the RCP scenarios were ranked as follows: RCP8.5 (20.6–40.8%) > RCP4.5 (5.5–25.4%) > RCP2.6 (0.1–7.8%). The soybean yield decreased by 10.6% on average in Harbin, with the highest decline under RCP8.5 ($P < 0.05$) (Table 5-3).

SOC stock decreased by 8.2% in Harbin and 7.6% in Gongzhuling by 2100 across all RCP scenarios and fertiliser treatments when compared with the baseline (Figure 5-3, Table 5-3). The decline under RCP8.5 (10.5–15.6%) was significantly higher than those under RCP4.5 (5.3–8.5%) and RCP2.6 (3.9–6.8%). The ranking of SOC stocks in 2100 was generally as follows: baseline > RCP2.6 > RCP4.5 > RCP8.5. In addition, manure amendment resulted in the highest SOC stock among the treatments, with the values of 38.9–50.3 t C ha⁻¹ in Harbin and 60.6–69.2 t C ha⁻¹ in Gongzhuling, respectively (Table 5-3).

Table 5-2 Statistical criteria of model performance for crop grain yield, straw dry matter, and SOC and TN stocks in the Harbin and Gongzhuling long-term experiments

Index	Calibration	Validation	Calibration	Validation
	<u>Corn grain yield</u>		<u>Corn straw dry matter</u>	
R ²	0.58**	0.58**	0.72**	0.60**
RMSE (%)	15	23	16	15
RE (%)	-12	-11	9	4
n	81	54	56	28
	<u>Wheat grain yield[†]</u>		<u>Soybean grain yield[†]</u>	
R ²	0.66**	0.60**	0.54**	0.56**
RMSE (%)	21	17	23	18
RE (%)	-7	-6	-24	-11
n	26	26	26	26
	<u>SOC stock</u>		<u>TN stock</u>	
R ²	0.79**	0.71**	0.67**	0.60**
RMSE (%)	8.2	6.8	8.6	7.5
RE (%)	-0.8	0.4	-2.0	-3.3
n	128	94	128	94

[†] There was no record about the wheat and soybean straw biomass productions at Harbin long-term experiment;

** means $P < 0.01$.

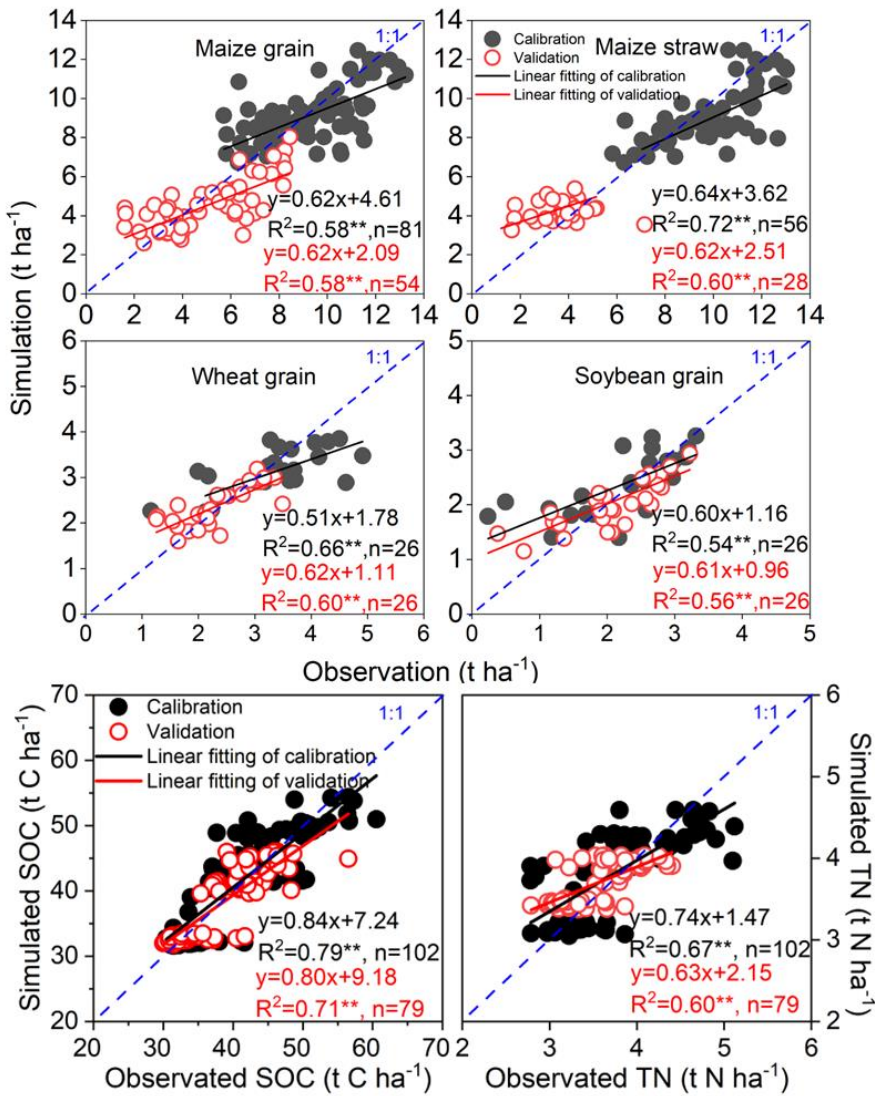


Figure 5-1 Calibration and Validation of corn, wheat and soybean grain yields and straw dry matters with data from Harbin and Gongzhuling combined. Note: wheat and soybean were only planted in Harbin, and data on straw dry matter for all crops in the Harbin long-term experiment are unavailable.

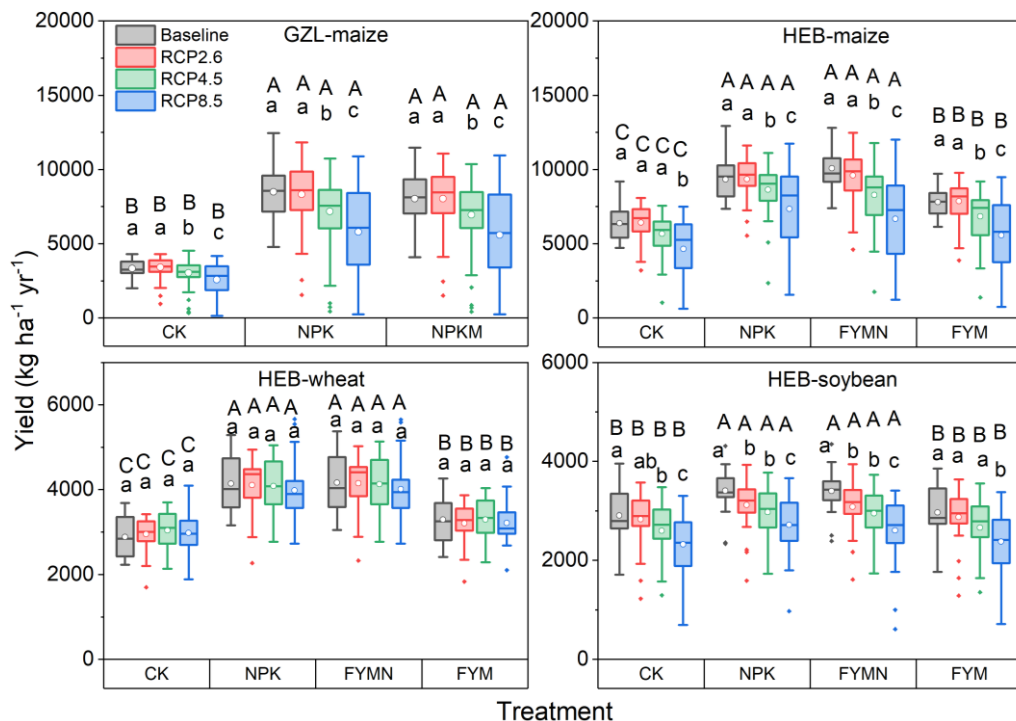


Figure 5-2 Corn, wheat and soybean grain yields with all fertiliser practises under future climate scenarios between 2021 and 2100 in the Harbin (HEB) and Gongzhuling (GZL) long-term experiments. Numbers with different lowercase letters indicate significant difference ($P < 0.05$) under different climate scenarios in an individual fertiliser practise. Numbers with different capital letters indicate significant difference ($P < 0.05$) in different fertiliser practises under an individual climate scenario.

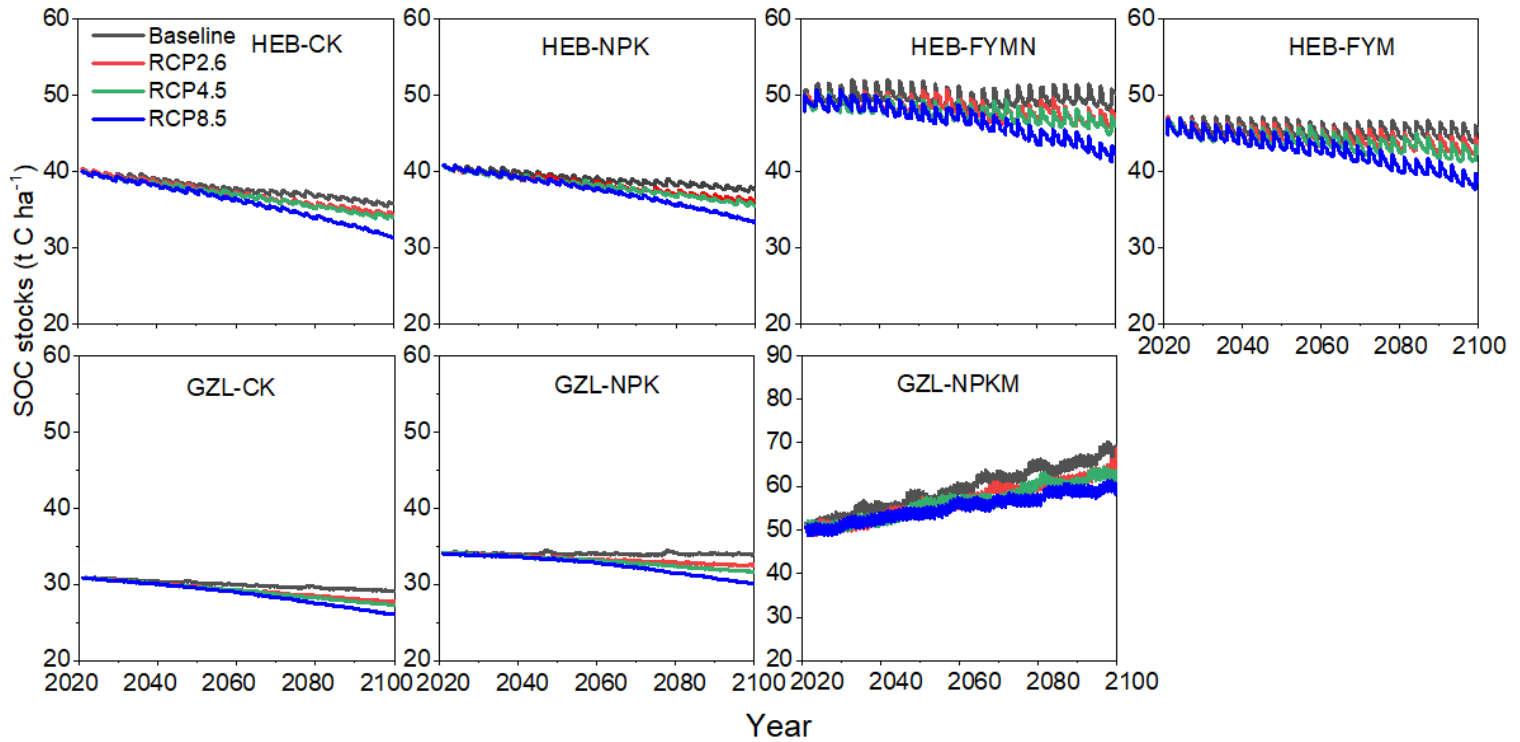


Figure 5-3 Dynamics of soil organic carbon (SOC) stocks with all fertiliser practises under future climate scenarios from 2021 and 2100 in the Harbin (HEB) and Gongzhuling (GZL) long-term experiments.

Table 5-3 Relative change of crop yield (%) and average annual CSE (%) between 2021 and 2100, annual SOC stock in 2100 (t C ha⁻¹), and their relative changes (%) with each fertiliser treatments under RCP climate scenarios compared with the baseline.

Treatment	Harbin						Gongzhuling						
	Climate scenario	Corn Yield _c ¹	Wheat Yield _c	Soybean Yield _c	SOC	SOCc ²	CSE	CSEc ³	Corn Yield _c	SOC	SOCc	CSE	CSEc
CK	Baseline				35.88					29.12			
	RCP2.6	3.76	5.39	1.26	34.43	-4.02			5.27	27.82	-4.46		
	RCP4.5	-8.83	8.58	-8.72	33.99	-5.26			-6.16	27.31	-6.23		
	RCP8.5	-26.79	6.46	-19.89	31.36	-12.59			-20.60	26.06	-10.51		
NPK	Baseline				37.77		4.27			33.92		5.92	
	RCP2.6	2.42	2.14	-6.62	36.14	-4.32	4.32	0.83	1.98	32.57	-3.98	5.04	-17.49
	RCP4.5	-5.53	1.65	-11.61	35.64	-5.65	4.47	1.46	-11.64	31.65	-6.69	5.09	-16.40
	RCP8.5	-20.66	-1.12	-21.13	33.37	-11.65	5.08	8.98	-28.46	30.07	-11.35	4.95	-19.17
FYMN/ NPKM	Baseline				50.31		12.60			69.19		14.56	
	RCP2.6	-7.82	2.48	-4.13	46.91	-6.76	12.00	-4.41	3.97	66.01	-4.60	13.79	-6.36
	RCP4.5	-25.41	2.44	-8.15	46.03	-8.51	11.85	-5.69	-9.55	63.51	-8.20	13.87	-5.87
	RCP8.5	-40.77	-1.44	-20.04	42.45	-15.63	11.66	-7.89	-27.10	60.56	-12.46	13.68	-7.21

Difference and Simulation of Soil Carbon Sequestration Efficiency of Typical Cropland in China, UK and USA

FYM	Baseline				45.34		11.91	
							aB	
	RCP2.6	-0.14	0.41	0.55	43.57	-3.90	11.50	-3.10
							bB	
	RCP4.5	-13.32	1.95	-8.65	42.47	-6.33	11.33	-4.91
							bB	
	RCP8.5	-30.87	0.42	-20.00	38.85	-14.31	10.64	-11.34
							cA	

¹ It is defined as the relative change of crop yield under an individual RCP scenario to the baseline (%);

² SOCc means the relative change of SOC stock under an individual RCP scenario to the baseline (%);

³ CSEc means the relative change of CSE under an individual RCP scenario to the baseline (%);

Numbers with different lowercase letters indicate significant difference ($P < 0.05$) in an individual fertilisation treatment under different climate scenarios and those with different capital letters indicate significant difference ($P < 0.05$) in the different fertilisation treatments under an individual climate scenario.

3.3. Carbon sequestration efficiency

Compared with the baseline, the annual total C input from crops, including stubble, roots, and rhizospheres, decreased by an average of 5.7% in Harbin and 9.3% in Gongzhuling under all the RCP scenarios (Figure 5-4). The rates for manure-amended treatments exceeded those without manure ranging from 62 to 154 kg C ha⁻¹ yr⁻¹ in Harbin and 311 to 429 kg C ha⁻¹ yr⁻¹ in Gongzhuling (Table 5-4). Additionally, the relative change rate under RCP8.5 was lower than those under RCP4.5 and RCP2.6.

The future climate change and fertilisation effects on the CSE are shown in Table 5-3. For most treatments, the annual CSE averaged over the simulation period showed significantly reduction compared to the baseline across RCP scenarios at both sites (except those for NPK in Harbin). The average decreases of this variable due to climate change were 6.3% in Harbin (except NPK) and 12.1% in Gongzhuling. The CSE values for treatments excluding NPK in Harbin were ranked as follows: baseline > RCP2.6, RCP4.5 > RCP8.5 ($P < 0.05$). Among the treatments, those with manure amendment (10.64–14.56%) were higher than those for NPK (4.27–5.92%).

The relationship between CSE and cumulative C input (C_{input}) was accurately captured by an exponential decreasing function ($CSE = b + a * e^{-k * C_{input}}$, $P < 0.001$) for all the treatments (Figure 5-5). The CSE asymptotic value, denoted by the parameter ‘b’ in the function, consistently exhibited higher values for FYM (Harbin: 8.19–10.56%) and FYMN/NPKM (Harbin: 9.90–11.36%, Gongzhuling: 8.19–9.29%) under all climate scenarios compared to NPK (Harbin: 3.78–5.00%, Gongzhuling: 4.20–5.51%). In addition, the asymptotic CSE values under all RCP scenarios (4.20–10.86%) generally were lower than those under the baseline (5.51–11.36%) for the majority of treatments in both Harbin (except NPK in Harbin) and Gongzhuling long-term experiments (Table 5-5).

Table 5-4 Average annual change rates (kg C ha⁻¹ yr⁻¹) in plough layer (0–20 cm) of the Harbin and Gongzhuling long-term experiments

Treatment	Climate scenario	Harbin		Gongzhuling	
		2021-2060	2061-2100	2021-2060	2061-2020
NPK	Baseline	15.7	15.7	57.1	43.2
	RCP2.6	15.4	14.1	57.0	42.8
	RCP4.5	15.1	13.7	55.3	39.1
	RCP8.5	16.8	16.6	54.5	36.1
FYMN/NPKM	Baseline	154.0	119.3	429.0	361.0
	RCP2.6	147.3	103.1	400.3	344.0
	RCP4.5	138.0	99.5	401.4	326.2
	RCP8.5	141.8	91.6	397.1	310.8

Difference and Simulation of Soil Carbon Sequestration Efficiency of Typical Cropland in China, UK and USA

FYM	Baseline	98.5	78.2
	RCP2.6	95.7	75.5
	RCP4.5	96.3	70.1
	RCP8.5	88.2	61.9

Table 5-5 Relation between carbon sequestration efficiency (CSE) and cumulative carbon (C) input with data from beginning to 2100 in plough layer (0–20 cm) of the Harbin (HEB) and Gongzhuling (GZL) long-term experiments

Site	Cropping systems	Treatments	Climate Scenarios	Model ($y=b+a*e^{-kCinput}$) [‡]
HEB	Corn-wheat-soybean	NPK	Baseline	$CSE=3.78+49.12e^{-0.049Cinput}$
			RCP2.6	$CSE=3.89+48.87e^{-0.049Cinput}$
			RCP4.5	$CSE=4.04+48.88e^{-0.050Cinput}$
			RCP8.5	$CSE=5.00+48.13e^{-0.052Cinput}$
		FYMN	Baseline	$CSE=11.36+46.18e^{-0.028Cinput}$
			RCP2.6	$CSE=10.86+47.96e^{-0.029Cinput}$
			RCP4.5	$CSE=10.60+34.55e^{-0.028Cinput}$
			RCP8.5	$CSE=9.90+45.00e^{-0.024Cinput}$
	FYM	Baseline	$CSE=10.56+24.55e^{-0.024Cinput}$	
		RCP2.6	$CSE=9.84+24.84e^{-0.022Cinput}$	
		RCP4.5	$CSE=9.95+25.75e^{-0.024Cinput}$	
		RCP8.5	$CSE=8.19+25.42e^{-0.018Cinput}$	
GZL	Corn	NPK	Baseline	$CSE=5.51+16.04e^{-0.038Cinput}$
			RCP2.6	$CSE=4.22+15.93e^{-0.027Cinput}$
			RCP4.5	$CSE=4.30+16.02e^{-0.028Cinput}$
			RCP8.5	$CSE=4.20+16.49e^{-0.029Cinput}$
		NPKM	Baseline	$CSE=9.29+27.37e^{-0.006Cinput}$
			RCP2.6	$CSE=8.74+28.84e^{-0.007Cinput}$
			RCP4.5	$CSE=8.66+28.89e^{-0.007Cinput}$
			RCP8.5	$CSE=8.19+29.31e^{-0.007Cinput}$

[‡] Where b is the CSE asymptotic value (lower value of asymptotic line).

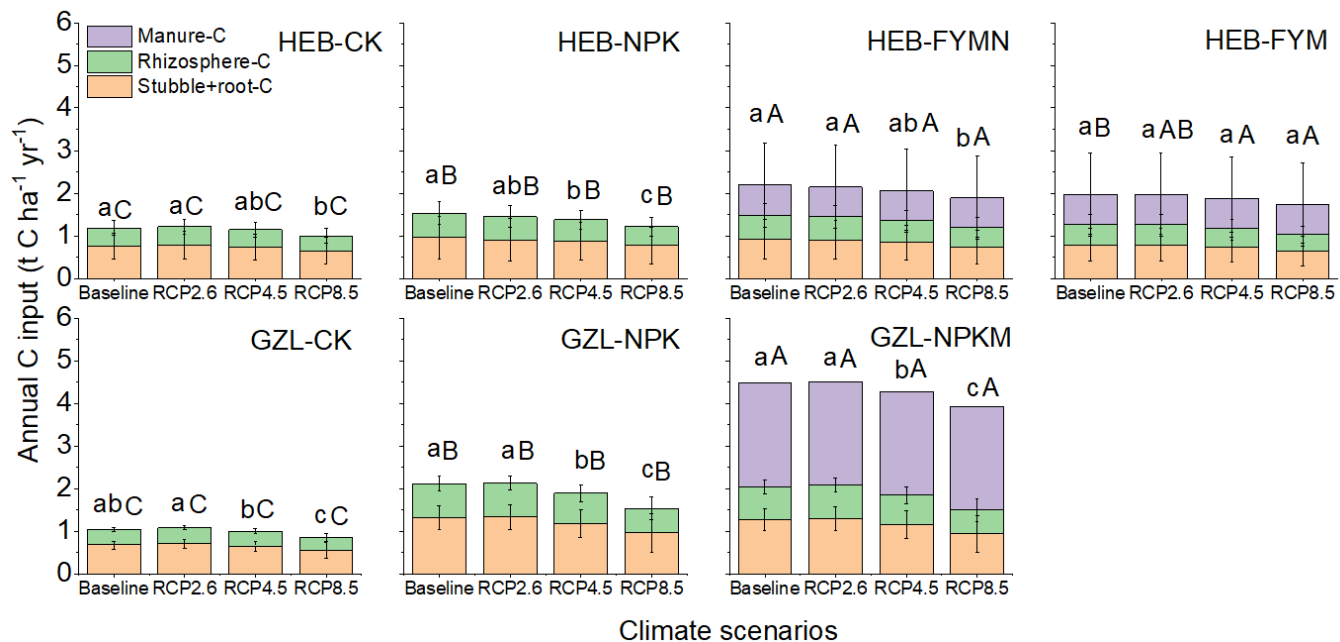


Figure 5-4 Average annual C input from different sources under different fertiliser treatments in Harbin (HEB) and Gongzhuling (GZL) long-term experimental sites. Numbers with different lowercase letters indicate significant difference ($P < 0.05$) among different climate scenarios under an individual fertiliser practise within an individual site. Numbers with different capital letters indicate significant difference ($P < 0.05$) in different fertiliser practises under an individual climate scenario within an individual site.

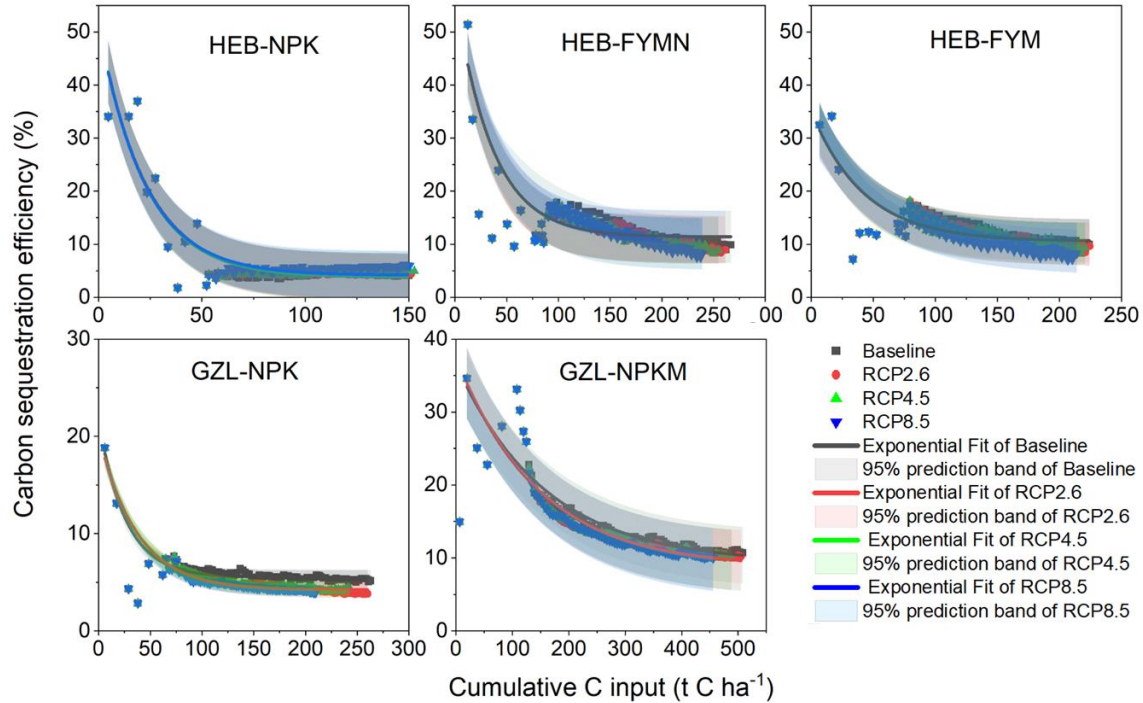


Figure 5-5 Dynamics of carbon sequestration efficiency (CSE) with carbon input cumulation for all fertiliser practises from the beginning to 2100 under future climate scenarios in the Harbin (HEB) and Gongzhuling (GZL) long-term experiments. Note: Due to the limited amount of measurement data, the value is plotted every three years.

4. Discussion

4.1. Model performance

Our results highlight that SPACSYS emerges as a competitive model for simulating corn yield in typical Mollisol regions of Northeast China achieving R^2 values of 0.58–0.72 (Table 5-2, Figure 5-1). This performance compares favourably with the PRYM-Maize model ($R^2 = 0.57$) and the CERES-Maize ($R^2 = 0.80$) model (Jiang et al., 2021; Zhang et al., 2021). Similarly, SPACSYS demonstrated a competitive soybean yield simulation with an RMSE of 18–23%, comparable to the performance of the DSSAT model with an RMSE of 15–22% in Northeast China (Liu et al., 2013b). It should be noted that SPACSYS has previously employed for simulating winter wheat yield (Liang et al., 2018; Liu et al., 2020), and our study further validates its effectiveness in simulating spring wheat, expanding the application scope of SPACSYS. Furthermore, SPACSYS adequately simulated SOC stocks in the study regions for all treatments (Table 5-2, Figures 5-1, 5-S3 and 5-S4), despite underestimating SOC stocks by 20% during validation. Jiang et al. (2014) applied the Roth-C model to the same sites, our study demonstrated superior performance with R^2 values of 0.79 for calibration and 0.71 for validation, while Roth-C achieved R^2 values of 0.25 for Harbin and 0.77 for Gongzhuling. In terms of TN stock, SPACSYS also performed well with R^2 values of 0.60–0.67 and RMSE of 7.5–8.6% (Table 5-2), surpassing the GWRK model ($R^2 = 0.50$ and RMSE = 29%) when applied to data from the central Northeast China, including Gongzhuling (Li et al., 2020b).

Nevertheless, the model still exhibited certain inevitable errors. For example, it consistently overestimated TN stock for all treatments after 2000 in Harbin. This discrepancy could be partially due to the relocation of the long-term experiment in December 2010, despite the soil at a depth of 1.5 m being transferred under frozen soil conditions. Moreover, the model's excessive reliance on stomatal resistance to soil water could lead to underestimations of corn grain and straw yields (Figure 5-S3) during the prolonged droughts from April to June in 2010, 2011 and 2012 in Gongzhuling. In addition, the extreme drought experienced after flowering in 2008, with the precipitation about 80–100 mm lower than those of adjacent years in Harbin, might have contributed to the overestimation of soybean yield by 2.5 times (Figure 5-S2). Improvements are needed in the model's representation of how drought impacts crop growth and development in future iterations.

4.2. Climate and fertiliser impacts on crop yield

Future climate change was projected to decrease corn yield in the typical Mollisol regions of Northeast China. Previous studies have indicated that increasing precipitation during the corn growing season would have no discernible influences on corn yield in Northeast China under future climate change (Lin et al., 2017; Xiong et al., 2007). By contrast, it seemed that the increased temperature could be the leading factor contributing the decline in corn yield, as it led to early development in the crop (i.e., anthesis precocity) and accelerated the reproductive stage, relative to the cooler conditions observed under the baseline (Figure 5-S6). Although a warming climate

might mitigate the effects of chilling damage on corn growth and production in Harbin and Gongzhuling, our study states that the effect of future temperatures would be more negative rather than positive for corn crop growth (Lin et al., 2017). Furthermore, several studies have suggested that the corn yield increase resulting from the “CO₂ fertilisation effect” could not offset the reduction caused by the increased temperature in study regions of Northeast China (Jiang et al., 2021; Lin et al., 2017; Wang et al., 2011). Consequently, future climate was expected to significantly decrease corn yield in the Mollisol regions of Northeastern China, especially at the end of the simulation period (2090–2100) under RCP8.5 (57.6–69.2% for Harbin and 54.8–67.1% for Gongzhuling), coinciding with the largest increments in both CO₂ concentration and temperature (Table 5-S6).

Climate change did not significantly affect spring wheat yield in the study regions, being consistent with the earlier reports (Jin and Zhu, 2008; Yin et al., 2015). The anticipated warmer climate is expected to result in a shortened growing period, leading to a diminished accumulation of photosynthate (Figure 5-S6). However, under the RCP scenarios, CO₂ concentrations are projected to increase greatly (Table 5-S2), which could enhance the thermotolerance of crop photosynthesis, especially in the context of higher temperatures compared with the baseline. This effect is more favourable for C3 (e.g., wheat) rather than C4 (e.g., corn) species (Jin et al., 2017; Taub et al., 2000). For major C3 crops, increased CO₂ concentration could improve the radiation use efficiency and net photosynthesis of crops by increasing the availability of intercellular CO₂ as a substrate and restraining competition with photorespiration for the Rubisco enzyme (Dermody et al., 2008). Similarly, the reduction in soybean yield in Harbin was less pronounced (10.6%) compared to corn (14.5%). According to Chen et al. (2017), the effect of enhanced CO₂ concentration on offsetting yield gaps caused by temperature and precipitation changes is more substantial for soybean than for corn. On the other hand, previous studies showed that soybean yield in Northeast China could increase with rising precipitation and temperature, considering the effects of “CO₂ fertilisation” (Lin et al., 2017; Yin et al., 2015). However, when temperatures reach high levels (e.g., a 4°C increase), other studies corroborate our findings (Guo et al., 2022; Jin and Zhu, 2008). They suggested that soybean yield would decline without cultivar adaption in response to temperature extremes, which could disrupt the pollination stage and reduce the number of grains by shortening the grain-filling period.

4.3. Response of the carbon sequestration to future climate change under long-term fertilisation

Our simulations revealed a significant decrease in SOC stock under all future climate scenarios, consistent with previous studies conducted in Mollisol regions (Gao et al., 2008; Ramírez et al., 2019; Bao et al., 2023). The decline in C input for each fertiliser practise, due to the reduction of corn and soybean productions under the RCP scenarios, likely played a predominant role in the SOC decline. Consequently, SOC stock under RCP8.5 was significantly lower than those under RCP4.5 and RCP 2.6 ($P < 0.05$) Furthermore, the combined application of manure with chemical N treatments

simultaneously showed the highest SOC stock and crop yield at both sites under each climate scenario (Table 5-3, Figures 5-2 and 5-3). It has been reported that manure combined with chemical fertiliser treatment has the beneficial effects of both C inputs and readily available nutrients for plant growth, enhancing C inputs from crop residues and sequestering a greater amount of C compared to other treatments (Triberti et al., 2016; Jiang et al., 2018a; Gross and Glaser, 2021). Notably, the impact of fertilisation on SOC stock was more pronounced with the manure-only treatment compared to NPK. The significantly higher annual C input could be the primary reason for the observed difference, along with the very close amount of C inputs from crop residues for both treatments (Figure 5-4). However, a contrary conclusion was found primarily when the C input to croplands from manure was from other ecosystems within the study region, rather than relying on consistent C inputs from fertilisation (Schmidt et al., 2011).

Our results showed that fields with manure application had greater efficiencies in C sequestration than those receiving chemical fertiliser alone. Applied manure creates a favourable growth environment for plants and microorganisms (Mandal et al., 2007). Our simulations demonstrated that manure amendments led to increased C inputs and microbial biomass. Those might be beneficial for retaining massive amounts of total C in the Mollisol regions of Northeast China (Ding et al., 2016; Ma et al., 2010). Consistently, compared to the practise using chemical fertiliser alone, manure application exhibited significantly higher SOC stocks and change rates (Figure 5-3 and Table 5-4), supporting a higher value of CSE.

Our investigation revealed a pronounced negative exponential correlation between CSE and the cumulative C input. Specifically, CSE showed an initial rapid decline following the onset of C input, then a decelerated decrease, ultimately settling into a slow decay until reaching a stable asymptotic value with a substantial accumulation of C input. This dynamics pattern has been directly or indirectly supported by previous findings (Jiang et al., 2018a; Maillard and Angers, 2014; Stewart et al., 2007, 2008; Yan et al., 2013). Stewart et al. (2007), in their synthesis of data from 14 long-term experiments in the USA and Canada, identified an asymptotic increase in the relationship between C inputs and SOC content, ascribable to the soil C saturation phenomena at high C inputs. Additionally, Stewart et al. (2008) suggested the stabilization efficiency of added C would decrease as C input level increased. These studies collectively supported our observation of a negative exponential relationship between soil CSE and C inputs. Warmer and wetter environmental conditions are likely to enhance the mineralization rate of added C (Fang et al., 2022; Bao et al., 2023), leading to a reduction in CSE. Our predictions indicated that the soil respiration rate under the RCP scenarios which soil temperature is a primary control factor increased 3.10–9.36% in Gongzhuling and 1.67–16.18% in Harbin compared to the baseline (Table S5-7). Similar findings have been reported (Wang et al., 2022).

Notably, the average annual CSE for NPK under future climate scenarios in Harbin increased, contrasting with the absence of such a trend in Gongzhuling (Table 5-3). The incorporation of soybean into the crop rotation in Harbin likely contributed to the divergent responses observed in CSE. Biologically fixed N by soyabean can be

available for subsequent crops, which can increase biomass accumulation in non-leguminous crops within the rotation (Stagnari et al., 2017). This results in an increase of C input to the soils. Additionally, root exudation and senescence of legume roots and nodules further contribute to the C input to the soils, offering benefits for SOC sequestration (Virk et al., 2022). The findings suggested that integrating leguminous crop in a rotation cropping system could contribute to improved C retention in soils, especially in situations where manure is not readily available.

4.4. Adaptation strategies for climate change

In our study, the average soil CSE for NPKM/FYMN/FYM treatments ranged from 10.6% to 14.6%, aligning closely with the reported global manure C conversion ratio of $12\% \pm 4\%$, derived from a meta-analysis of 130 observations considering 4–82 years of manure applications (Maillard and Angers, 2014). Furthermore, predictions regarding the CSE of manure application in the soils of 20 long-term experiments in China, including the two Mollisols examined in our research, indicated that CSE approaching the stable stage ranged from 8.8% to 10.4% under baseline and RCP 4.5 conditions (Jiang et al., 2018a). Our findings, ranging from 8.7% to 11.4%, closely resembled these results (Table 5-5). This suggested that our results can serve as a useful reference for investigating the characteristics of CSE under long-term fertilisation conditions in Chinese Mollisols, especially with a substantial amount of C input. Despite the presence of several quite common uncertainties in long-term experimental studies, such as discrepancies in C input assessment methods and the estimated crop straw and root biomass in the absence of measured data, our results remain informative.

Our predictions indicated a worse situation for both food security and soil fertility for Mollisols in the future, in comparison to the current condition. In addition to spring wheat, the production of corn and soybean, which are the primary crops in the world's Mollisol regions (Durán et al., 2011; Xu et al., 2020), would decline under future climate change. Notably, the RCP8.5 scenario anticipated the largest decreases, with approximately 30–40% for corn and 20% for soybeans (Figure 5-2). To address this, our predictions suggest the following interventions: (1) the development of new cultivars that better adapt to the anticipated warmer and wetter climates; (2) an earlier planting of crops to prevent the negative effects of heat and drought during the growing season; (3) the adoption of conservation tillage techniques such as no-tillage and crop rotation to reduce soil erosion, maintain soil moisture and nutrients, and increase SOC content. In addition, the combination of manure and NPK is recommended based on our prediction. However, the goal of sequestering more C in Mollisols to mitigate global warming cannot be achieved as both the SOC stocks and CSE decreased under RCP scenarios compared to the Baseline. Further study can focus on the optimization of nutrient application rates and the ratio of chemical fertilisers to manure to maximize crop yield and SOC sequestration while minimizing environmental risks.

5. Conclusion

Our study successfully validated the SPACSYS model for simulating yields of corn, spring wheat and soybean, and the SOC stock in typical Mollisol regions in China. SPACSYS predicted a decline in both yields and SOC stock under future climate scenarios compared with the baseline. The anticipated reduction in corn and soybean yields was primarily attributed to increased temperatures with corn being more significantly affected than soybean. Interestingly, the warmer and wetter future had no significant effects on spring wheat yield in this region. Future climate conditions led to a decrease in SOC stocks and CSE at the two typical Mollisol sites for fertilisation treatments. Our prediction confirmed that CSE initially decreased rapidly following C inputs, exhibited a decelerated decline, and eventually decayed slowly until reaching a stable asymptotic value due to substantial C accumulation. Notably, the combined application of manure and chemical fertilisers demonstrated the highest potential to mitigate the negative impacts of climate change on crop yields and CSE. Our findings suggest that future research should prioritize improving crop cultivars, optimizing planting timings, and establishing effective fertiliser management strategies to mitigate climate change risks in Mollisol regions.

6. Supplementary Figures and Tables

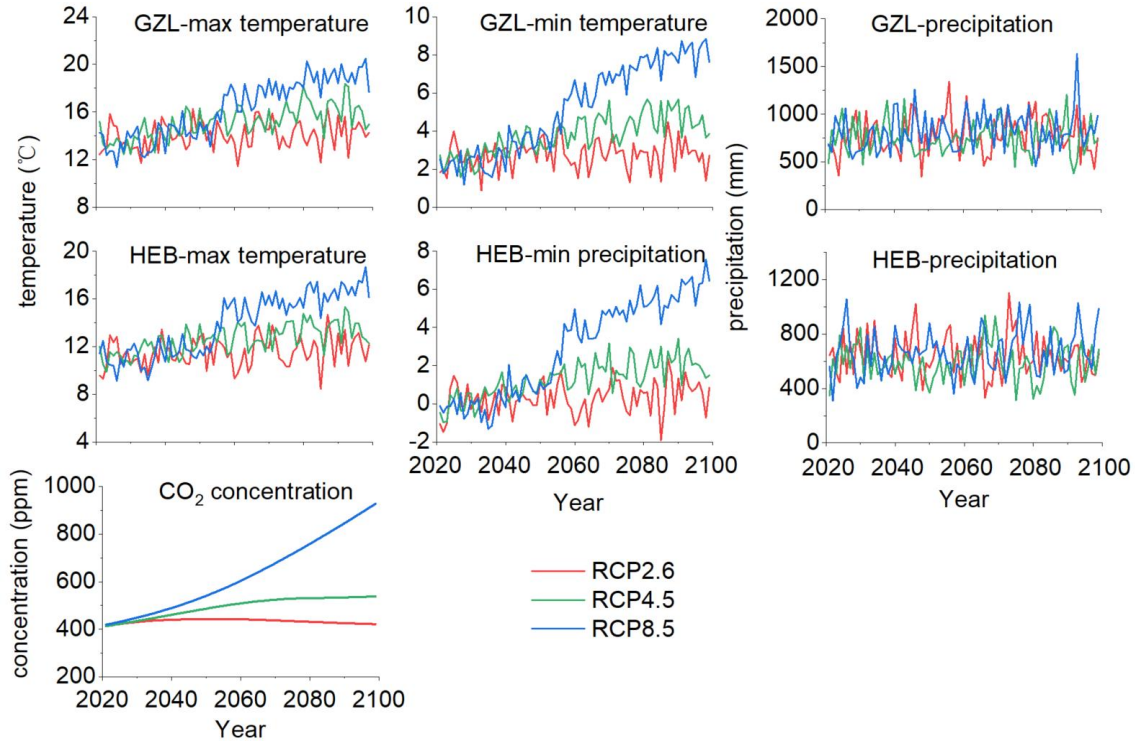


Figure 5-S1 Dynamics of annual average maximum and minimum temperature (°C), precipitation (mm) and CO₂ concentration (ppm) under RCP climate scenarios between 2021 and 2100 in the Harbin (HEB) and Gongzhuling (GZL) long-term experiments.

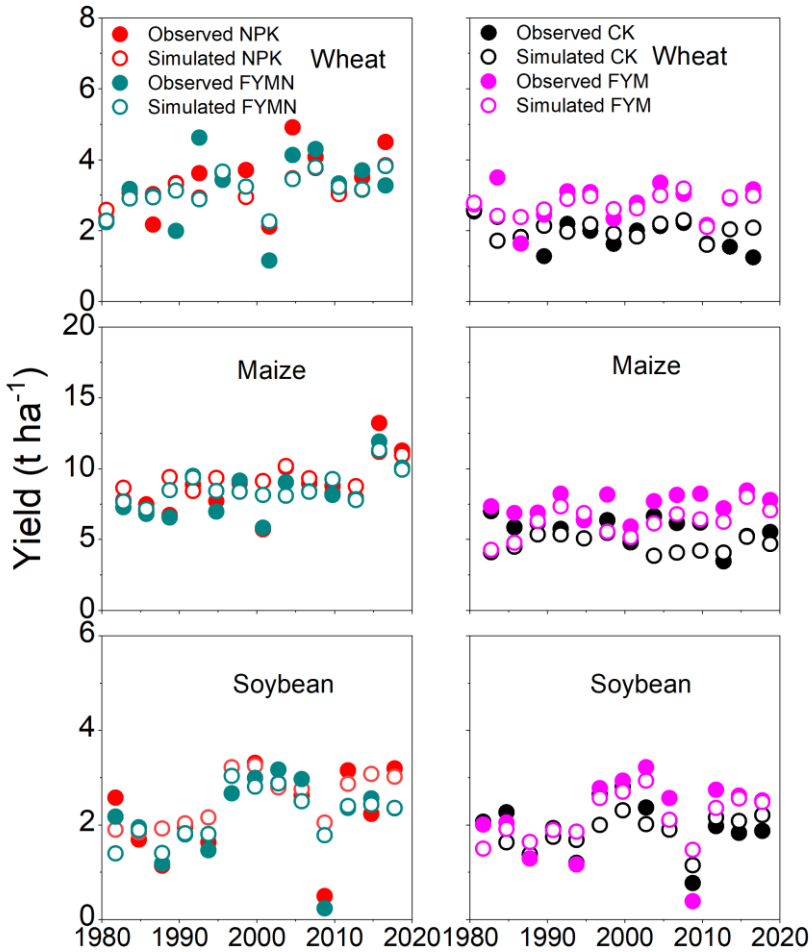


Figure 5-S2 Simulated and observed yields of wheat, corn and soybean over the simulated period in the Harbin long-term experiment. No straw dry matter record in this experiment.

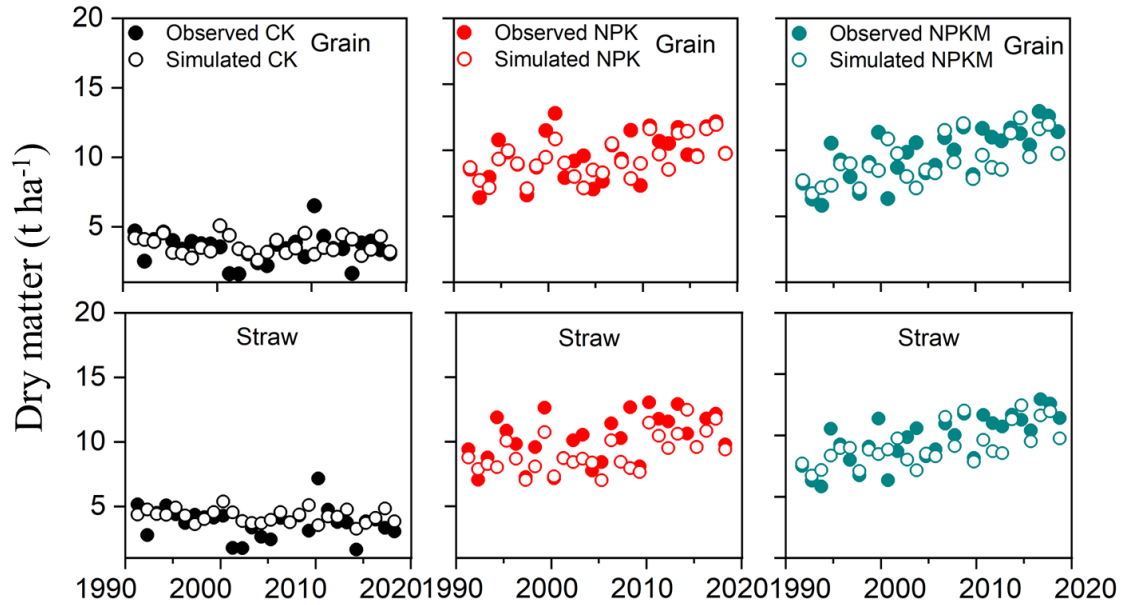


Figure 5-S3 Simulated and observed corn grain yields and straw dry matter over the simulated period in the Gongzhuling long-term experiment.

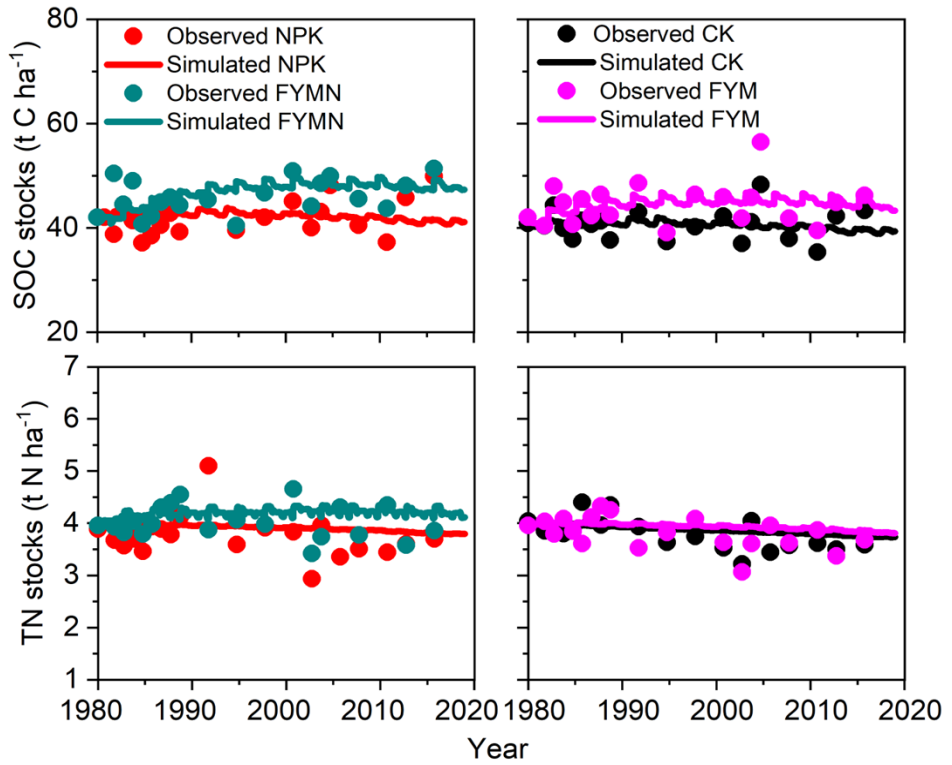


Figure 5-S4 Simulated and observed SOC and TN stocks over the simulated period in the Harbin long-term experiment.

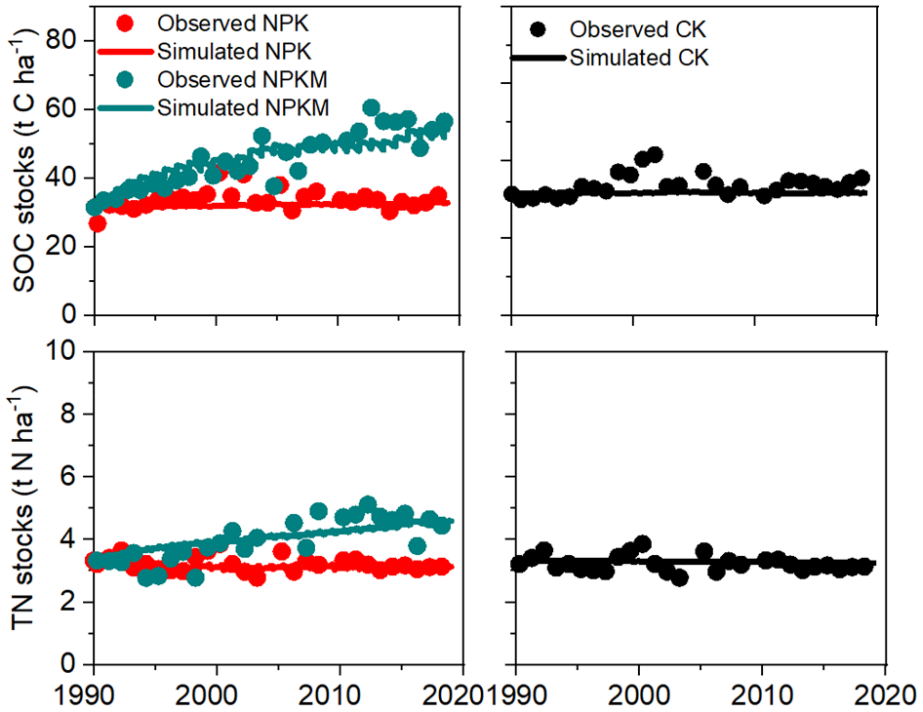


Figure 5-S5 Simulated and observed SOC and TN stocks over the simulated period in the Gongzhuling long-term experiment.

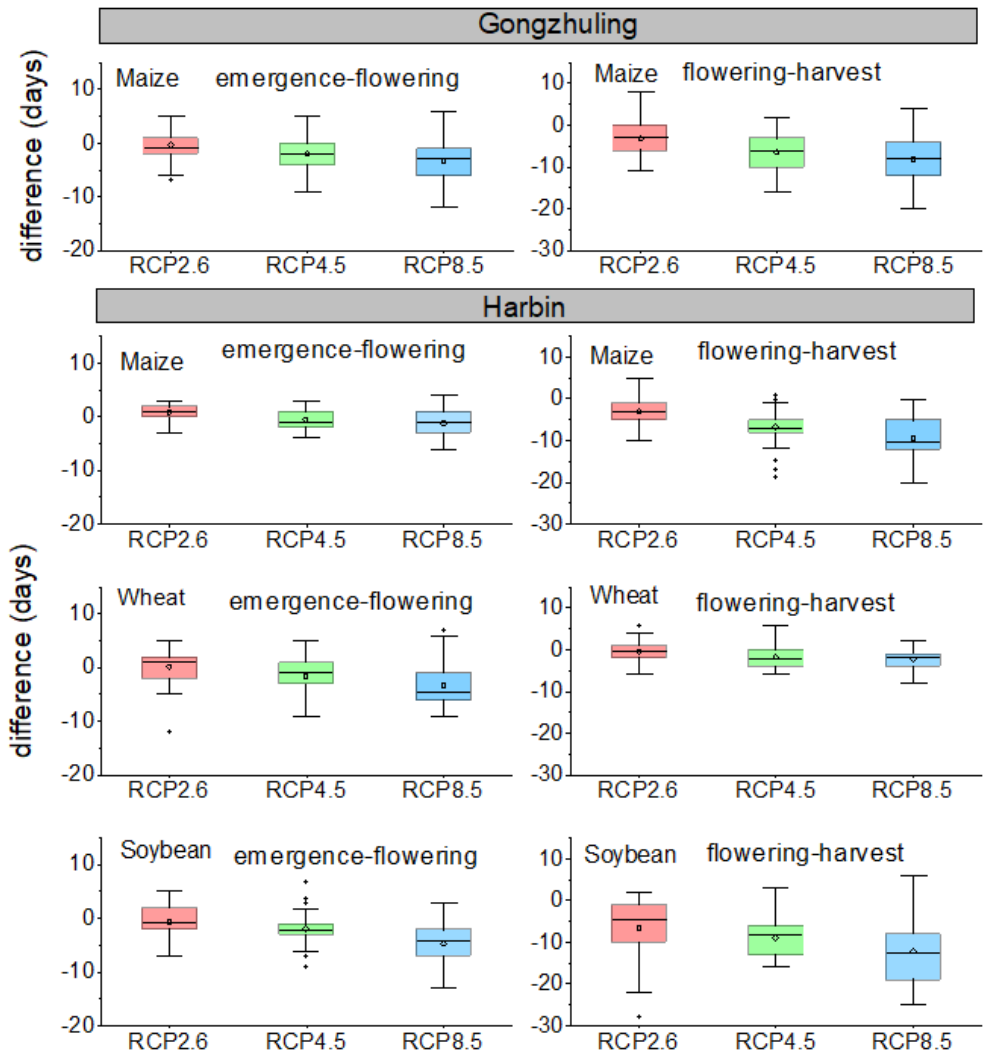


Figure 5-S6 Changes of the lengths from emergence to anthesis and from anthesis to maturity under future climate scenarios from 2021 to 2100 compared with those under the baseline in the Gongzhuling and Harbin long-term experiments.

Table 5-S1 Nitrogen, phosphorus and potassium application rates in different fertiliser treatments (kg ha⁻¹)

Treatment	Harbin			Gongzhuling
	Wheat	Soybean	Corn	Corn
Nitrogen (kg N ha ⁻¹)				
NPK	150	75	150	165
NPKM/FYMN	150+33*	75	150	50+115*
FYM	33*	-	-	-
Phosphorus (kg P ha ⁻¹)				
NPK	75	150	75	36
NPKM/FYMN	36*	-	-	36+39*
FYM	36*	-	-	-
Potassium (kg K ha ⁻¹)				
NPK	75	75	75	68
NPKM/FYMN	50*	-	-	68+77*
FYM	50*	-	-	-
Manure type	Horse			Cow, pig

*The amount of nitrogen, phosphorus and potassium from manure or straw; - means no application.

Table 5-S2 Average annual temperature, precipitation and rain events, CO₂ concentration of the Harbin and Gongzhuling long-term experiments from 2021 to 2100

Site	Climate scenario	Max temperature (°C)	Min temperature (°C)	Annual Precipitation (mm)	CO ₂ [#] Concentration (ppm)	Precipitation events	Precipitation events [‡]	
							>10mm	>50mm
Harbin	Baseline	10.4 (0.8) [†]	-0.5 (1.0)	549 (108)	380.0	11317	1237	60
	RCP2.6	11.5 (1.2)	0.3 (0.8)	641 (150)	421.4	10706	1359	22
	RCP4.5	12.6 (1.3)	1.3 (1.0)	592 (138)	537.9	10576	1245	22
	RCP8.5	13.9 (2.5)	2.9 (2.5)	670 (160)	926.7	10364	1441	46
Gongzhuling	Baseline	12.9 (0.9)	2.3 (0.7)	615 (138)	380.0	11206	1423	99
	RCP2.6	13.7 (1.1)	2.7 (0.8)	784 (200)	421.4	13121	1908	93
	RCP4.5	14.5 (1.3)	3.5 (1.0)	752 (174)	537.9	13059	1821	98
	RCP8.5	15.4 (2.5)	4.9 (2.4)	822 (203)	926.7	13033	1990	118

[†] Numbers in parentheses are standard deviation;

[#] The CO₂ concentration in 2100;

[‡] means the total amount of precipitation events from 2021 to 2100.

Table 5-S3 Optimized parameters related to wheat growth and development for different cultivars used in simulations

Parameter	Unit	Harbin			Gongzhuling
		Wheat (Longfumai 19)	Soybean (Heinong 42)	Corn (Sidan 19)	Corn (Zhengdan 958)
Accumulated temperatures from sowing to emergence	°C·d	140	160	100	120
Accumulated temperatures from emergence to flowering	°C·d	900	1150	650	800
Accumulated temperatures from emergence to flag leaf fully expansion	°C·d	650	1600	520	700
Accumulated temperatures from flowering to maturity	°C·d	800	1570	910	1000
Maximum plant height	m	0.8	1.0	2.4	2.4
Critical photoperiod for vegetative stage below or over which plant development will not be affected by light	hr	10	8.0	14.5	14.5
Critical photoperiod for vegetative stage below or over which plant development will stop	hr	8.0	16.0	19.0	19.0
Threshold temperature for emergence	°C	3.0	6.0	4.0	5.0
Threshold temperature for vegetative stage	°C	0.0	9.0	0.0	0.0
Threshold temperature for reproductive stage	°C	10.0	10.0	10.0	10.0
Specific leaf area	m ² ·g ⁻¹ ·DM	0.035	0.03	0.035	0.04
Specific green ear area	m ² ·g ⁻¹ ·DM	0.00637	-	0.00637	0.00637
Specific green stem area	m ² ·g ⁻¹ ·DM	0.006	0.006	0.006	0.006

Rate of N translocation from leaf to grain during reproductive stage	d ⁻¹	0.035	0.07	0.018	0.05
Rate of N translocation from stem to grain during reproductive stage	d ⁻¹	0.042	0.01	0.032	0.025
The lower threshold of optimal temperature for N fixation	°C	-	13	-	-
The maximum temperature over which N fixation stops	°C	-	30	-	-
The minimum temperature below which N fixation ceases	°C	-	9	-	-
The upper threshold of optimal temperature for N fixation	°C	-	26	-	-
Potential N fixation rate for legume plant (based on belowground biomass)	g N·g ⁻¹ DM·d ⁻¹	-	106	-	-

Table 5-S4 Optimized parameters related to soil C and N cycling in SPACSYS

Parameter	unit	Harbin	Gongzhuling
Ammonium immobilised fraction	-	0.45	0.45
Humus fraction from dissolved	-	0.15	0.15
Humus fraction from fresh litter	-	0.25	0.25
Humus fraction from fresh organic matter	-	0.07	0.10
Potential decomposition rate for humus	d ⁻¹	0.000015	0.000015
Potential decomposition rate for fresh litter organic matter	d ⁻¹	0.012	0.008
Potential decomposition rate for fresh organic matter	d ⁻¹	0.013	0.009
Potential decomposition rate for dissolved organic matter	d ⁻¹	0.02	0.04
Critical C/N ratio favourable to immobilization	-	15	15
Microbial maintenance respiration rate	d ⁻¹	0.678	0.678

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Maximum autotrophic nitrification rate	d ⁻¹	0.08	0.08
Potential heterotrophic nitrification rate	d ⁻¹	0.04	0.04
Maximum nitrifier growth rate	d ⁻¹	9.74	9.74
Precipitation N concentration	g N m ⁻³	0.80	0.80
Relative activity at saturated water content	-	0.60	0.60
Dry deposition mineral N to soil	g N m ⁻² day ⁻¹	0.0045	0.0045
Base temperature at which temperature function is in unity for denitrification	°C	20	20
Base temperature at which temperature function is in unity for mineralization/immobilisation	°C	22	25
Base temperature at which temperature function is in unity for nitrification	°C	20	20

Table 5-S5 Carbon (C) content in manure and cereals shoot and root, and root: shoot ratio (RSR) and harvest index (HI) of studied crops.

Site	Cereal	Shoot C (g/kg)	Root C g/kg	RSR	HI
Harbin	Wheat ¹⁻⁴	434	425	0.29*	0.41-0.45*
	Corn ¹⁻⁵	433	412	0.12*	0.46-0.52*
	Soybean ⁴	473	447	0.14	0.46
	Manure	C: 17.45-41.38% (fresh)*			
Gongzhuling	Corn ¹⁻⁵	433	412	0.12*	0.46-0.52*
	Manure	C: 17.45-41.38% (fresh)*			

*Measured data; 1-5 are references, and the value in this table is the average value from the references (Cai et al., 2015; E et al., 2018; Li et al, 2015; Wang et al., 1989; Wang et al., 1991).

Table 5-S6 Crop yield (t ha⁻¹), average annual SOC stock (t C ha⁻¹) and CSE (%) and their relative changes (%) with each fertiliser treatment under RCP climate scenarios compared with the baseline between 2090 and 2100

Treatments	Climate scenarios	Harbin				Gongzhuling			
		Corn Yield	Wheat Yield	Soybean Yield	SOC	CSE	Corn Yield	SOC	CSE
CK	Baseline	5.48aB	2.49aB	2.37abB	36.0aD	-	2.98ab	29.2aC	-
	RCP2.6	6.94aB (20.4) [‡]	2.74aB (9.5)	2.78aB (16.4)	34.8bD (-3.4)	-	3.25a (11.3)	27.9bC (-4.5)	-
	RCP4.5	5.47aC (0.6)	3.24aB (19.8)	2.23abA (-2.4)	34.3bD (-4.7)	-	2.48b (-12.6)	27.5bC (-5.9)	-
	RCP8.5	2.02bB (-65.2)	2.83aB (15.6)	1.60bA (-27.1)	32.1cD (-10.8)	-	1.38c (-54.8)	26.4cC (-9.8)	-
NPK	Baseline	8.67aA	3.48aA	3.05aA	37.9aC	4.6cC	7.72ab	34.0aB	5.2aB
	RCP2.6	9.51aA (11.6)	3.83aAB (9.0)	3.20aA (8.2)	36.4bC (-3.7)	5.0bC (10.8)	8.10a (7.8)	32.6bB (-4.1)	3.8dB (-26.2)
	RCP4.5	8.72aA (1.7)	4.25aAB (21.6)	2.74aA (-7.6)	36.0bC (-5.0)	5.0bC (11.1)	5.95b (-18.0)	31.8cB (-6.4)	4.2bB (-17.8)
	RCP8.5	3.91bA (-57.6)	4.05aAB (18.6)	2.14bA (-25.4)	34.0cC (-10.2)	5.5aB (32.3)	2.72c (-66.7)	30.4dB (-10.6)	4.0bB (-22.1)
FYMN/NPKM	Baseline	8.89abA	3.48aA	3.09aA	49.6aA	9.7aB	7.33ab	68.2aA	10.8aA
	RCP2.6	10.09aA (10.1)	3.87aA (8.4)	3.23aA (4.2)	46.7bA (-5.8)	9.1abB (-1.8)	7.78a (9.0)	64.2bA (-5.8)	10.1cA (-6.5)
	RCP4.5	7.59bAB (-27.2)	4.30aA (19.9)	2.77aA (-10.9)	46.2bA (-6.7)	8.9bB (-4.9)	5.78b (-16.0)	63.3bA (-7.1)	10.3bA (-5.0)
	RCP8.5	2.86cAB (-67.8)	4.04aA (17.8)	2.21bA (-27.2)	42.7cA (-13.8)	8.2cA (-12.7)	2.54c (-67.1)	60.3cA (-11.5)	10.2bcA (-6.2)

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FYM	Baseline	6.97abB	2.84aB	2.44abAB	44.9aB	9.8aA
	RCP2.6	8.39aAB (12.6)	2.98aAB (3.8)	2.87aAB (12.0)	43.2bB (-3.8)	9.4abA (-3.3)
	RCP4.5	6.39bBC (-7.6)	3.48aAB (20.7)	2.31abA (-2.5)	42.1cB (-6.1)	9.0bA (-8.1)
	RCP8.5	2.29cB (-69.2)	3.32aAB (16.2)	1.65bA (-27.1)	39.0dB (-13.1)	7.9cA (-19.0)

‡ The values in parentheses are relative changes of the studied indexes.

Numbers with different lowercase letters indicate significant difference ($P < 0.05$) under different climate scenarios in an individual fertiliser treatment; Numbers with different capital letters indicate significant difference ($P < 0.05$) in different fertiliser treatments under an individual climate scenario

Table 5-S7 Relative change (%) of the soil respiration rate under RCP climate scenarios compared with the baseline during 2021-2100

Site	Fertiliser practise	RCP2.6	RCP4.5	RCP8.5
Gongzhuling	CK	8.11	8.16	7.30
	NPK	9.36	7.00	3.10
	NPKM	7.33	6.01	6.28
Harbin	CK	2.23	1.90	2.28
	NPK	16.18	11.28	8.37
	FYMN	12.45	8.59	7.03
	FYM	1.82	1.67	3.35

Table 5-S8 ANOVA test for the effects of fertiliser treatments and climate scenarios on crop yield, soil organic carbon (SOC) stock, carbon (C) input, and carbon sequestration efficiency (CSE) between 2021 to 2100 ($P < 0.05$)

	df	F	P	df	F	P
	Corn yield (Harbin)			Corn yield (Gongzhuling)		
Treatment	3	60	<0.001	2	542	<0.001
Climate scenario	3	33	<0.001	3	59	<0.001
Treatment*Climate scenario	9	1	0.87	6	5	<0.001
	Spring wheat (Harbin)			Soybean (Harbin)		
Treatment	3	100	<0.001	3	18	<0.001
Climate scenario	3	0.4	0.73	3	25	<0.001
Treatment*Climate scenario	9	0.2	0.98	9	0.2	0.99
	SOC stock (Harbin)			SOC stock (Gongzhuling)		
Treatment	3	2074	<0.001	2	12350	<0.001
Climate scenario	3	135	<0.001	3	38	<0.001
Treatment*Climate scenario	9	3	0.001	6	7	<0.001
	Annual C input (Harbin)			Annual C input (Gongzhuling)		
Treatment	3	37	<0.001	2	4260	<0.001
Climate scenario	3	3	0.02	3	52	<0.001
Treatment*Climate scenario	9	0.05	0.99	6	4	0.001
	CSE (Harbin)			CSE (Gongzhuling)		
Treatment	2	1352	<0.001	1	981	<0.001
Climate scenario	3	8	<0.001	3	2	<0.001
Treatment*Climate scenario	6	1	0.30	3	0.01	0.98

Chapter VI

**General discussion, conclusions, and
perspectives**

1. General discussion

1.1. Soil carbon sequestration efficiency characteristics under long-term fertilisation

Soil C is the largest component of the terrestrial C pool (Lettens et al., 2005). It varies with climate, geography, biological activity, and land management, leading to large uncertainties in the soil C budget, especially in agroecosystems (Smith and Conen, 2006; Yadav and Malanson, 2009). Changes in soil C content within agricultural areas have the potential to influence soil fertility, food security, and climate through the sequestration or release of CO₂. Soil C sequestration can be characterized by several aspects, including storage, sequestration rate, change stage, potential, and efficiency. Recently, CSE, as a dimensionless parameter, has been recognized as a critical indicator of soil C sequestration capacity in field and regional studies (Batjes and Sombroek, 1997; Follett, 2001; Hua et al., 2014; Liang et al., 2019; Yan et al., 2013).

Based on the dryland long-term experiments established in 1979 or 1989/1990 in China, where SOC linearly increased with C input, we observed that soil CSE remained nearly constant within a limited range of C input (Luvisols: 27–120 t C ha⁻¹, Calcisols: 25–280 t C ha⁻¹, Anthrosol: 29–167 t C ha⁻¹) (Figures 2-3, 2-S1 and 2-S4). However, the soil from four classical long-term experiments in the UK and USA exhibited an exponential decrease in CSE, which was associated with nonlinear changes in SOC, as a large amount of C input accumulated (Chromic Luvisols: 187–650 t C ha⁻¹, Mollisols: 268–502 t C ha⁻¹) (Figures 3-6, 3-7 and 5-5). In addition, the CSE in the Chromic Luvisol of southeast England experienced a rapid decrease until C input reached 123–315 t C ha⁻¹, and a similar trend was observed in the Mollisols of the Central Great Plains of America before C input levels of 93–145 t C ha⁻¹. Then, the CSE decreased more slowly until it approached a more stable, equilibrium level. As proved by previous studies (Bhattacharyya et al., 2010; Liu et al., 2019; Maillard and Angers, 2014; Zhang et al., 2010), soil CSE could be a constant value or decrease with C input accumulation (Hassink and Whitmore, 1997; Six et al., 2002; Stewart et al., 2007). This depends on how far a soil is from the C saturation (Hassink and Whitmore, 1997).

The constant CSE was typically derived from a linear relationship between SOC stock and C input, with no evidence of a C saturation level. However, the logarithmic relation emphasized a decrease of CSE when the SOC approached the saturation level (Yan et al., 2013). Similarly, Stewart et al. (2007) reported a reduction in SOC accumulation efficiency with increasing C input levels in a study centred on the C saturation model. This observation was supported by data compiled from 14 agroecosystems with varying durations ranging from 12 to 96 years. The decrease in CSE was attributed to the limited capacity of high-C soils to retain newly applied OC (Six et al., 2002). As the soil approached C saturation, the proportion of newly added C sequestered within the soil C pool was diminished due to inherent physicochemical

constraints, particularly when a majority of protective sites were occupied (Hassink and Whitmore, 1997). For example, soils with greater chemical stability (e.g., oxalate-soluble Fe) and physical structure exhibited a higher CSE compared to those with lower physicochemical stability (Yan et al., 2013). Additionally, our findings highlighted that within a narrow range of C inputs, the variability in observed CSE was constrained, and the asymptotic curve might not be evident.

Our findings, supported by observations from four classical long-term experiments and predictions from two Mollisol sites in Northeast China, demonstrate an exponential decrease in CSE with C input accumulation, aligning well with a negative exponential function ($CSE = b + a * e^{-k * C_{input}}$, $P < 0.01$). Hence, our results suggest that soil CSE could approach an asymptotically stable value, which corresponds to the maximum increment in soil carbon sequestration capacity attributable to a specific fertilisation strategy, as compared to the non-fertilised condition. This can account for the observed differences in SOC levels under various fertiliser practises within the same soil type.

1.2. Main drivers of carbon sequestration efficiency at different stages

Climate, soil properties, and management factors that influence C input and SOM decomposition consequently affect SOC stocks and their relationship with C input (Barbera et al., 2012; Follett, 2001; Paustian et al., 2000). In our study, soil CSE remains constant across fertiliser treatments when SOC linearly increases with C input. Edaphic factors were more important drivers of CSE than C input and climate factors when SOC accumulated at a constant efficiency (Figure 2-5). However, when SOC shows non-linear changes with the accumulation of a large amount of C input, the CSE exponentially decreases and eventually approaches an asymptote, with the contribution of C input and climate factors promoting ahead of soil properties (Figure 3-10).

A meta-analysis of globally published studies indicated that the quality of the organic amendment was the main driver determining CSE (Maillard and Angers, 2014), which could be a major reason explaining the difference in soil C/N ratios and their impacts on CSE. The source of C input varied greatly among the different treatments, leading to significant differences in soil C/N ratios after 30–40 years of fertilisation in long-term experiments of China. They affected the activity and structure of microorganisms, thereby affecting the rates of organic matter decomposition (Marschner et al., 2003; Maillard and Angers, 2014; Zhang et al., 2015). Three decades of fertilisation in Gongzhuling demonstrated that the soil C/N ratio was positively correlated with the microbial N limitation, which controlled the ratio of bacterial functional genes (i.e., Cellulase/Amylase) involved in the degradation of the recalcitrant SOC (Cui et al., 2022). In our study, soil C/N ratios were higher for manure amendment treatments than for NPKS and NPK (Figure 2-S3). This indicated a larger microbial N limitation due to manure than that caused by straw (Cui et al., 2022), resulting in a lower rate of SOC decomposition (Maillard and Angers, 2014). In addition, the lower soil C/N ratio observed for NPK in our study

suggested a potential decrease in SOC stocks and GHG emissions due to higher mineralization rates (Xu et al., 2016). This could be related to the microbial C limitation caused by the long-term intensified N fertiliser application (Wang et al., 2019). Consequently, soil C/N ratio mainly controlled soil CSE by limiting nutrient availability and affecting microbial activity when SOC was linearly increased within a limited range C inputs (e. g., 58–275 t C ha⁻¹ for the main dryland of China). However, a study has summarised that the “high-quality” organic matter, characterized by high N content, low C/N ratio, and low phenol/lignin concentration, did not stabilize in SOC with a greater efficiency compared to the “low-quality” organic matter, which has low N content, high C/N ratio, and high phenol/lignin concentration (Castellano et al., 2015). The impact of the added organic material quality on the SOC pool size and CSE was modulated by the extent of soil C saturation and was eventually controlled by physicochemical protection mechanisms within the mineral soil matrix (Castellano et al., 2015).

In our study, the influence of clay content on soil CSE is more pronounced when C inputs are limited compared to when a substantial amount of carbon is accumulated. A study based on 10 soils found that the clay fraction predominantly determined the maximum capacity of each soil to protect organic matter (Hassink and Whitmore, 1997). However, soil physical properties and their protection capacity seemed to be limited by their characteristics, such as the availability of free space on the surface, which was consistent with the phenomenon of C saturation during long-term soil C accumulation (Hassink, 1997). The eight studied long-term experiments in China were all characterized by rapid SOC accumulation in association with C inputs (Jiang et al., 2018a; Jiang et al., 2014). In such cases, the soil could provide more protective sites for stabilizing organic carbon than when most of those sites were occupied (Boyle et al., 1989; Jenkinson, 1990).

The C input level determined the duration and SOC content when a soil approached its steady state (West and Six, 2007). However, its relative importance was ranked behind the soil C/N ratio and clay content in eight long-term experiments within four-decades of fertilisations (Figure 2-5). This might be supported by the significantly positive correlation between C input and SOC content (Figure 2-S4), which was generally found in soils that are far from soil C saturation, reflecting an increase in the relatively rapid-turnover SOC pools (Six et al., 2002; West and Six, 2007). This result indicated that the significant effect of C input on CSE at this stage was still controlled by SOC fractions.

Furthermore, the importance of the MAT effect on CSE was heightened with the accumulation of a substantial amount of C input. This could be explained as follows: (1) Long-term climate variations affected C input by controlling crop yield. Utilizing data from the Broadbalk continuous experiment, which was conducted from 1864 to 1967, a study examined the impact of weather on the interannual variability in winter wheat yield in response to N fertiliser, finding that maximum temperature variations in May and June were negatively correlated with crop yields (Chmielewski and Potts, 1995). Furthermore, a separate study focusing on the period from 1968 to 2016 suggested that wheat yield was particularly sensitive to mean temperatures in

November, April and May (Addy et al., 2020). (2) Changes in MAT primarily influenced the decomposition of C inputs, thereby altering soil CSE. The impact of long-term changes in MAT on soil CSE is discussed in Chapter III. These findings highlighted the importance of considering temporal weather variations when assessing the impacts of climate change on crop yield and SOC sequestration, especially under long-term fertilisation (Addy et al., 2020).

1.3. Differences in carbon sequestration efficiency among different regions

We concentrated on the Harbin, Gongzhuling, Morrow and Sanborn long-term experiments, which had identical soil type (i.e., Mollisol) and similar agricultural systems (i.e., continuous corn cropping and its rotation with legumes) across the four classical long-term experiments in the UK and the USA, as well as all those conducted in China. In addition, all four Mollisol experiments were located at approximately 45°N. They fulfilled the prerequisite for comparing CSE across different regions. Based on the characteristics of CSE from the four long-term experiments (Figures 3-6, 3-7 and 5-5), we investigated the differences in typical Mollisols between Northeast China and Central USA during the rapid descent stage (i.e., preceding the inflection point) and the asymptotically stable stage (i.e., as they approached the CSE asymptotic value). The inflection point confirmed by the GAM and segmented regression is presented in Figure 6-1. The C input and CSE at the inflection point (CSE_{IP}), the average CSE before the inflection point, and the asymptotic value of CSE (CSE_{as}) are shown in Table 6-1.

By analysing the C input at the inflection point for each treatment in the Sanborn and Morrow experiments, we determined the corresponding year of the inflection point. The analysis further revealed that each plot was in the phase characterized by manure application and the absence of straw return. The average soil CSE before the inflection points of the Sanborn and Morrow experiments (13.6–27.4%) was slightly lower or close to that observed in the Gongzhuling and Harbin experiments (18.6–27.1%). However, the C inputs at the inflection points (93–141 t C ha⁻¹) in the Sanborn and Morrow experiments were lower than those in the Gongzhuling experiment (234–250 t C ha⁻¹). Additionally, compared to the Sanborn and Morrow experiments, the Gongzhuling experiment took a longer time to approach the IP (Table 6-1). Generally, soils with high initial SOC content show lower C sequestration potential and efficiency due to their proximity to C saturation, in contrast to soils with low initial SOC content. Consequently, a relatively lower C input is required to reach the IP. Moreover, soils with low initial C content have a large saturation deficit, leading to a faster initial rate and a longer total duration of SOC sequestration until reaching a steady-state level (Stewart et al., 2007; West and Six, 2007). Therefore, the high initial SOC content of the Sanborn and Morrow experiments might be the main reason for the differences in C inputs and the duration to reach the inflection points between Mollisols in China and the USA.

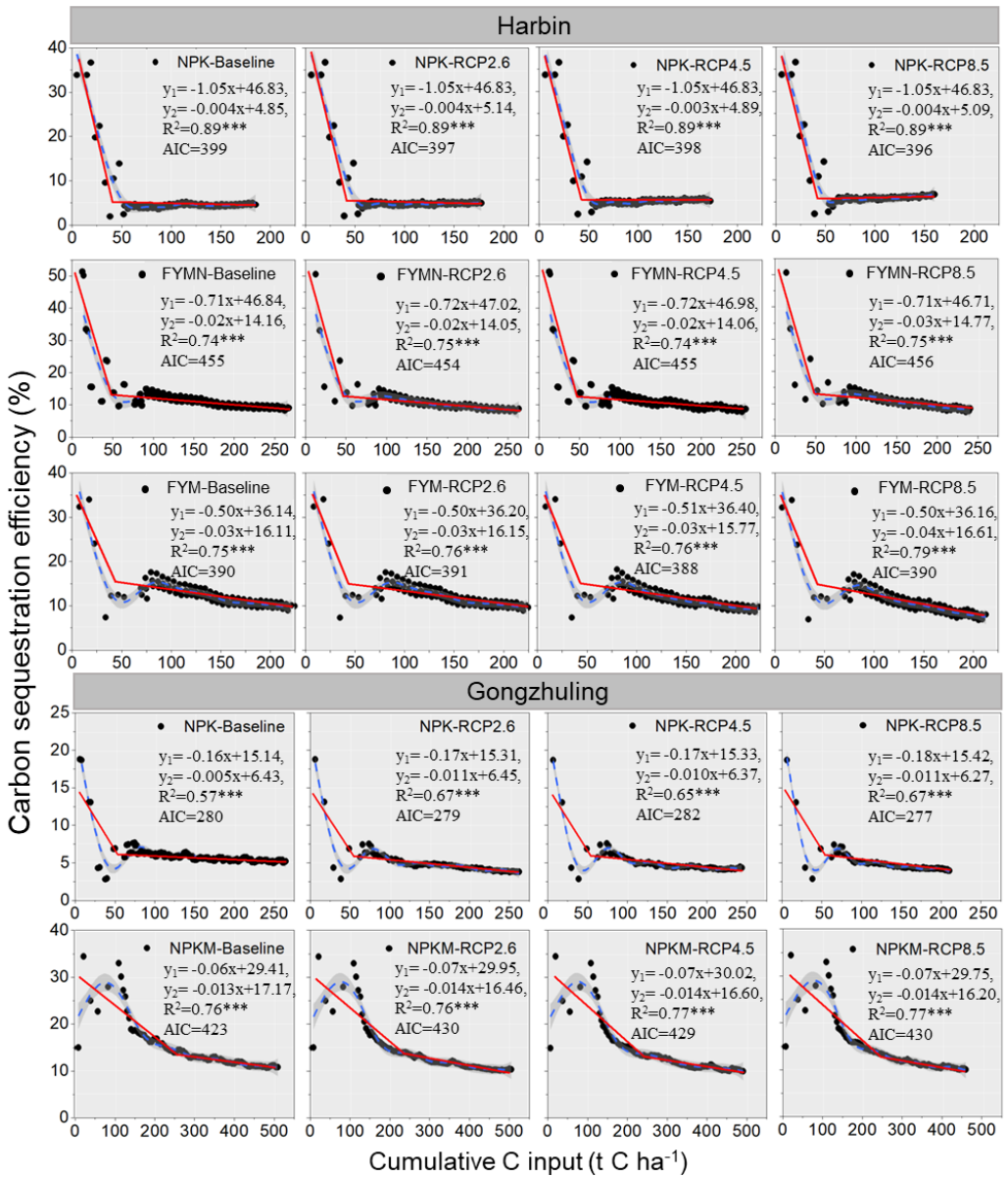


Figure 6-1 The generalized additive model and piecewise linear regression for the relation between carbon sequestration efficiency (%) and cumulative C input (t C ha⁻¹). AIC means Akaike information criterion. *** $P < 0.001$.

Table 6-1 The cumulative C input and CSE at the inflection point (CSE_{IP}), average CSE before the inflection point, and the CSE asymptotic value (CSE_{as})

Site	Cropping system	Treatment	Scenario	Inflection point (IP)			Average CSE before IP	CSE _{as}
				C input	CSE _{IP}	Year		
Sanborn (1888)	Corn	FYM	-	122	14.9	1942	25.4	6.8
		FYM-NPK	-	103	7.1	1944	20.3	3.6
	Rotation	FYM-FYMN	-	133	9.9	1943	14.7	4.5
Morrow (1904)	Corn	FYM-NPK	-	141	7.5	1947	19.3	3.9
		FYM	-	105	12.1	1935	23.8	4.7
	Rotation	FYM-NPK	-	93	8.7	1927	13.6	0.5
		FYM	-	128	13.0	1938	27.4	2.5
HEB (1979)	Corn-wheat-soybean	NPK	Baseline	40.1	5.2	2010	21.1	3.8
			RCP2.6	39.8	5.2	2009	21.1	3.9
			RCP4.5	39.9	5.2	2009	21.1	4.0
			RCP8.5	39.6	5.2	2009	21.1	5.0
		FYMN	Baseline	47.2	13.7	2003	27.1	11.4
			RCP2.6	47.2	13.7	2003	27.1	10.9
			RCP4.5	47.2	13.7	2003	27.1	10.6
			RCP8.5	47.2	13.7	2003	27.1	9.9
		FYM	Baseline	42.7	15.8	2003	22.0	10.6
			RCP2.6	42.7	15.8	2003	22.0	9.8
			RCP4.5	42.7	15.8	2003	22.0	10.0
			RCP8.5	42.7	15.8	2003	22.0	8.2
GZL (1989)	Corn	NPK	Baseline	54.5	6.3	2013	9.2	5.5
			RCP2.6	54.5	6.3	2013	9.2	4.2
			RCP4.5	54.5	6.3	2013	9.2	4.3
			RCP8.5	54.5	6.3	2013	9.2	4.2
		NPKM	Baseline	250	13.3	2046	19.4	9.3
			RCP2.6	234	13.3	2042	18.6	8.7
			RCP4.5	236	13.3	2043	18.7	8.7
			RCP8.5	243	13.3	2044	18.6	8.2

Furthermore, when considering the CSE asymptotic value, both NPK and manure application treatments showed lower CSE in the Sanborn and Morrow experiments (NPK: 0.2–3.9%, NPKM: 2.5–6.8%) as compared to those in the Harbin and Gongzhuling experiments (NPK: 3.8–5.5%, FYM/FYMN/NPKM: 8.2–10.6%). As our results demonstrated, carbon input and MAT were the two main controllers of soil CSE when a large amount of C input was accumulated (Figure 3-7). The differences in C input caused by straw returning in all treatments in the Sanborn and Morrow experiments, as well as the differences in climate between the two Mollisol regions of China and the USA, might be the main reasons. Because microbial communities were more active in areas with higher temperature and precipitation, this resulted in faster C mineralization and thus lower soil CSE in these environments (Zhang et al., 2016a). Compared with Harbin and Gongzhuling, the twofold higher MAT of Sanborn and Morrow (Tables 3-1 and 5-S2) could accelerate the decomposition of SOC, which was not beneficial for achieving high CSE. In addition, the returned straw in all treatments of Sanborn and Morrow, as the primary C source during the asymptotically stable stage of soil CSE, resulted in slow biodegradation and low stabilization of litter-derived C (Berhane et al., 2020; Poeplau et al., 2015). Such straw-derived C sources hindered the achievement of high CSE in Mollisols within these experiments.

In summary, the Mollisol CSE at two sites in the USA, due to the manure application for all fields, was higher than in Northeast China during the rapid decline phase of CSE. During the asymptotically stable stage, the Mollisol CSE at two sites in the USA, with higher temperatures and large C inputs from straw return, was lower than in Northeast China. Notably, in addition to some common uncertainties in long-term experiments, such as estimating C input through roots and stubbles using a fixed ratio of aboveground biomass, the use of different methods for measuring SOC in long-term experiments between two regions—Walkley & Black and dry combustion—may also introduce additional uncertainties for the comparison. Although dry combustion is considered the most accurate method to determine total (organic and inorganic) C (Meersmans et al., 2009), a modified Walkley & Black method (i.e. heated for 12 minutes at 220 °C) with a correction factor of 1.1 was applied in long-term experiments from China to minimize errors (Hu et al., 2016).

1.4. Fertilisation practises beneficial to improve carbon sequestration and mitigate climate change

Studies conducted through both field observations and model projections have indicated that the application of manure serves as an effective strategy to improve SOC storage (Ding et al., 2016; Ma et al., 2010; Li et al., 2018; Powlson et al., 2012). As proved by our study, manure application enhanced SOC sequestration rates and CSE relative to those observed with chemical fertiliser treatments and straw return treatments. Compared to other fertiliser treatments, the higher SOC sequestration rate under FYM or NPKM/FYMN was mainly related to the higher average plant-derived C input plus exogenous OC input from manure (Tables 2-2, 2-3 and Tables 3-5 to 3-12). As discussed in Chapter II and III, the higher CSE from manure application can be attributed to the direct OC input, the formation of stable soil organic complexes, a

favourable environment for microbial activity and subsequent SOC sequestration, as well as the enhancement of macro-aggregate components associated with a higher CSE (Acikgoz et al., 2017; Huggins et al., 1998; Liu et al., 2019).

In our study, the increased temperature and enhanced CO₂ concentration under future climate change primarily impacted crop yields and SOC sequestration. Key effects include: (1) Increasing temperature under the RCP scenarios were anticipated to shorten the crop growing season in the study regions (Figures 4-S8 and 5-S6), potentially leading to reduced yields due to diminished biomass accumulation (Asseng et al., 2004). (2) The abbreviated growing season reduced the time available for the accumulation of dead leaves, stems and roots into above- and below-ground litter under future climate change (Balkovič et al., 2014; Senapati et al., 2019). This restricted the C input from residual and dead roots (Table 4-S6). (3) “CO₂-fertilisation” benefited winter wheat (e.g., C3 species) more than corn (e.g., C4 species) (Figures 4-4 and 5-2; Table 4-5). For C4 species, elevated CO₂ has minimal impact on C fixation since PEP-carboxylase uses HCO₃⁻ as a substrate instead of CO₂ (Jin et al., 2017; Taub et al., 2000). (4) Increased NPP due to “CO₂-fertilisation” could be a dominant factor in determining the sink of C in studied systems in the future (Figure 4-6). The rising CO₂ concentration might alleviate climatic constraints on plant growth by decreasing stomata conductance, which in turn lowers transpiration, enhances water use efficiency, and inhibit drought stress, potentially leading to global NPP increases over the next few decades (Pan et al., 2014; Wieder et al., 2015b).

The current study forecasts that the persistent application of manure will remain a sustainable approach for the Broadbalk long-term experiment moving forward. This sustainability is attributed to its capacity to simultaneously achieve the highest rates of SOC sequestration and C sink capacity within the soil-wheat-atmosphere system (Tables 4-5, 4-6; Figures 4-5, 4-6), as anticipated under RCP scenarios, despite over 170 years of FYM application. In addition, the integration of chemical fertilisers with manure shows the greatest potential for mitigating the adverse impacts of climate change on crop yields and soil C sequestration in typical Mollisol regions of China in the future. This approach was expected to result in the highest average SOC stock and soil CSE by the year 2100 (Tables 5-3 and 5-4). Beyond the reasons outlined in the preceding paragraph, manure amendments facilitated high C inputs from crop residues, with the microbial community significantly retaining more N, P, and K than the CK, NPK, and NPKS treatments. This increase in retention was also conducive to soil C sequestration (Ding et al., 2016; Jiao et al., 2006). Furthermore, manure application was projected to result in high CO₂ emissions from soil respiration in the future (Figure 4-S3; Table 5-S7). Nevertheless, the C sink under manure application suggests that the enhanced C inputs through the ‘CO₂-fertilisation’ effect can offset C losses due to elevated temperatures projected under RCP climate scenarios.

Therefore, regions facing issues similar to those in Southeast England, where soil degradation results from soil compaction, water erosion, and land use changes (Peake et al., 2022), should consider manure amendments to enhance soil C sequestration and mitigate future climate change. Similarly, areas in Northeastern China, where SOC degradation and crop yield decrease is due to wind and water erosion, soil acidification,

and excessive cultivation (Wang et al., 2022), would benefit from such measures. In the Central Plains of the US, where SOC and crop yield decline is caused by wind erosion, salinization, intensive tillage, and overgrazing (Montanarella et al., 2021), manure application is also advisable for regions with similar problems.

1.5. Recommendations for farmers, scholars and politicians

For farmers:

- (1) Increase the application of manure, such as livestock manure and dairy manure from farm, to replace a portion of chemical fertilisers to enhance soil organic matter content and nutrient levels, especially in regions facing SOC loss.
- (2) Learn and adopt scientific manure application techniques to ensure efficient use of manure and reduce environmental pollution. For example, adopt precise manure application amounts through soil testing and crop demand analysis to avoid over-fertilisation and minimize water pollution.
- (3) Introduce crop rotation to improve soil quality and crop resistance. For instance, the introduction of legume crops was found in this study to effectively offset the negative impact of future climate change on the CSE of Mollisols under chemical fertiliser treatment.
- (4) Participate in C farming projects. Farmers can increase their farm's SOC stocks through the manure application and the introduction of crop rotation. These increased C can be certified as C credits, which farmers can sell in the C credit market to gain economic benefits.

For scholars:

- (1) Determine the range of cumulative C input and the duration of rapid decline in soil CSE for various soil types, and issue warnings before reaching the inflection point to remind farmers to timely improve soil CSE by changing field or crop management practises, such as adopting conservation tillage and rotation.
- (2) Develop new crop varieties and provide guidance to farmers on adjusting planting dates in typical crop production regions (e.g., Mollisol regions) to mitigate heat and/or drought stress during the growing season under future climates, thereby protecting long-term food security.
- (3) Determine the optimal ratio and quantity of manure to replace chemical fertilisers to maximize crop yield and SOC sequestration while minimizing environmental risks.
- (4) Develop models and conduct analyses to quantify the C credit potential of various agricultural practises (e.g., manure amendments, crop rotation), informing policy recommendations and guiding farmers in maximizing their C credit earnings.

For governments:

- (1) Support and protect the development of long-term experiments.
- (2) Increase promotional efforts to enhance farmers' awareness of the benefits of

manure application and encourage its acceptance. Additionally, enact policies, such as offering subsidies and tax incentives, to promote its use, particularly in regions experiencing SOC depletion and crop yield decline.

- (3) Invest in research and development related to enhance quality and efficacy of manure, and establish standards and guidelines for safe and effective use in the main production areas of different cereal crops.
- (4) Develop standards and certification programs for C farming practises to validate farmers' C sequestration requests, thereby building trust in the C credit market and encouraging participation.

Overall, developing sustainable agriculture that enhances soil fertility, ensures food security, and mitigates climate change requires collaborative efforts across various sectors of society.

2. General conclusion

This research, based on eight long-term experiments in China where SOC increased linearly with C input, found that soil CSE remained constant across fertiliser treatments. Edaphic characteristics, especially the soil C/N ratio and clay content, significantly influenced CSE. In four classical long-term experiments, where SOC showed nonlinear changes with the accumulation of a large amount of C input, CSE exponentially decreased and eventually approached an asymptote ($CSE = b + a * e^{-k * C_{input}}$, $P < 0.01$). Carbon input, MAT and TN stock were key factors influencing soil CSE. Manure application had a more significant effect on soil CSE than chemical fertilisation across all experiments.

Using data collected from the Broadbalk continuous winter wheat experiment over a century, the SPACSYS model has been validated to simulate grain yield for different tall and modern short-strawed winter wheat varieties, as well as to capture the dynamics of SOC stocks in the plough layer. Winter wheat yields were projected to significantly increase under future climate change relative to the baseline. However, the impact of future climate change on SOC stocks varied among RCP scenarios and fertiliser practises. Compared to the baseline, the medium- and high- emissions scenarios were found to significantly increase SOC stocks for all N treatments by 2100, while the very stringent pathway (RCP2.6) led to a reduction in SOC stocks under chemical fertiliser application. Further, manure application was shown to be effective in sustaining winter wheat production and enhancing soil C sequestration under future climate change in southeastern England.

For two Mollisols in Northeast China, future climate change would reduce both corn and soybean yields due to increased temperatures. The RCP climate scenarios were also found to be responsible for the decrease of SOC stocks and CSE in the two Mollisol experiments, with the most significant decline under the high-emissions scenario (RCP8.5). Similar to the relationship derived from the four classical long-term experiments, Mollisol CSE decreased exponentially with C input accumulation towards approaching an asymptotically stable value. The combined application of

manure and chemical fertilisers demonstrated the highest potential to mitigate the negative impacts of climate change on crop yields and CSE. Additionally, the Mollisol CSE at two sites in the USA, where manure was applied to all fields, was higher than that in Northeast China during the rapid decline phase of CSE. During the asymptotically stable stage, the Mollisol CSE at two sites in the USA, with higher temperatures and large C inputs from straw return, was lower than that in Northeast China.

Overall, this study clarified the constant or exponentially decreasing trend of soil CSE with C input accumulation and identified the key influencing factors based on observations and simulations from the long-term experiments, providing empirical evidence and insights into the optimal fertiliser strategy to enhance CSE and mitigate future climate change under long-term fertilisations.

3. Innovations

This study, based on typical long-term experiments from the UK, USA, and China with different experimental durations, comprehensively characterized the dynamics of CSE with C input accumulation, highlighting the constant or negative exponential trends.

Through the longest experiment, the Broadbalk continuous winter wheat experiment, the performance of the SPACSYS model was assessed for simulating grain yield of various tall and modern short-strawed winter wheat varieties and capturing the long-term dynamics of SOC stocks in the plough layer over a period of more than a century.

Utilizing historical observations and future predictions of soil C sequestration characteristics, this study confirmed that manure application is a sustainable fertiliser practise, enhancing crop yields and promoting the retention of C within the agroecosystem.

4. Perspectives

To better understand the characteristics of soil CSE and address the negative response of SOC sequestration to future climate change under long-term fertilisation, further investigation into the following aspects is recommended:

(1) This study analysed the differences in the characteristics of soil CSE and their main influencing factors under balanced fertilisations. Based on long-term experiments, significant differences in soil CSE were found between long-term imbalanced and balanced fertilisations. Subsequent studies should elucidate the impact and underlying mechanisms of nutrient availability on soil CSE under unbalanced fertilisations.

(2) Manure application provides a greater advantage for soil CSE over chemical fertilisers, as evidenced by analysis from the plough layer. However, the benefits of manure application on SOC sequestration in the subsoil are controversial, given different responses of SOC in the subsoil to long-term fertilisations and the importance of crop growth and nutrient transport between soil layers. A

comprehensive investigation of the soil CSE in deeper layers (e.g., 20–100 cm) is necessary based on both observed and simulated data.

(3) Manure application had the potential to significantly sustain or enhance SOC sequestration rate and efficiency under future climate change. However, the N application rates were assumed to remain unchanged under future climate change. N application rates can influence SOC sequestration, with higher rates correlating with increased crop yields and the subsequent incorporation of plant residues into the soil. Furthermore, the contribution of “CO₂-fertilisation effects” to vegetation photosynthesis has declined at the global scale due to nutrient supply limitations. Therefore, further investigation should be made to explore an appropriate N applied rate to enhance both crop yield and C sequestration in the study region under future climate change.

(4) Mollisols, recognized as the most fertile and productive soils globally, must adopt adaptive strategies to address the predicted worsened situation for both crop production and C sequestration in the typical Mollisol region of Northeast China under future climate change. Our predictions suggest that future research should prioritize: 1) developing cultivars to adapt them to the warmer and wetter future climates; 2) advancing the planting date to mitigate heat and drought stress during the growing season; 3) determining optimal ratios and amounts of chemical fertilisers combined with manure in order to increase crop yield and CSE while minimising environmental risks; and 4) modelling crop yield and CSE under optimal management practises that promote SOC sequestration, such as conservation tillage (e.g., no-till or reduced tillage) and straw return.

Chapter VII

References and appendix

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Appendix

1. Publications

- (1) **Liang, S.**, Sun, N., Wang, S., Colinet, G., Longdoz, B., Meersmans, J., Wu, L., Xu, M., 2023. Manure amendment acts as a recommended fertilization for improving carbon sequestration efficiency in soils of typical drylands of China. *Frontiers in Environmental Science* 11, 1173509. <https://doi.org/10.3389/fenvs.2023.1173509>.
- (2) **Liang, S.**, Sun, N., Meersmans, J., Longdoz, B., Colinet, G., Xu, M., Wu, L., 2024. Impacts of climate change on crop production and soil carbon stock in a continuous wheat cropping system in southeast England. *Agriculture, Ecosystems & Environment* 365, 108909. <https://doi.org/10.1016/j.agee.2024.108909>.
- (3) **Liang, S.**, Sun, N., Longdoz, B., Meersmans, J., Ma, X., Gao, H., Zhang, X., Qiao, L., Colinet, G., Xu, M., Wu, L., 2024. Both yields of maize and soybean and soil carbon sequestration in typical Mollisols cropland decrease under future climate change: SPACSYS simulation. *Frontiers In Sustainable Food Systems* 8,1332483. <https://doi.org/10.3389/fsufs.2024.1332483>.
- (4) Wang, S., Sun, N., **Liang, S.**, Zhang, S., Meersmans, J., Colinet, G., Xu, M., Wu, L., 2023. SOC sequestration affected by fertilization in rice-based cropping systems over the last four decades. *Frontiers in Environmental Science* 11, 1152439. <https://doi.org/10.3389/fenvs.2023.1152439>.

2. Presentations

- (1) Differences and mechanisms of soil carbon sequestration in typical arable land of multi-regions (Differences and Mechanisms of Soil Carbon Sequestration in Typical Arable Land of China, United States and United Kingdom based on Long-term Various Fertilizations, Zhanjiang, 06/11/2018, Oral presentation).
- (2) Climate change impacts on crop production and soil carbon stock in a continuous wheat cropping system in southeast England (Soil Science Society of Belgium Thematic Day 2022, Brussel, 19/12/2022, Poster presentation).
- (3) Climate change impacts on crop production and soil carbon stock in a continuous wheat cropping system in southeast England (The General Assembly 2023 of the European Geosciences Union, Vienna, 26/04/2023, Poster presentation).