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Highlights

Compared with initial values, final SOC stock increased by 24–74% under manure.

SOC_r was positive for all regions but were negative under CK and NPK in Northeast.

A decrease in SOC stock was reported in some trials in Northeast under manure.

SOC_r has no significant differences between the M and NPKM in Northeast and South.

SOC_r was main controlled by initial SOC, soil BD and climate.

Spatial changes and driving variables of topsoil organic carbon stocks in Chinese croplands under different fertilization strategies

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Abstract

The effect of different fertilization strategies on changes in soil organic carbon (SOC) largely depends on the current status of a given agricultural region. We analysed the results of 90 long-term field trials (20–37 years) in Chinese croplands to determine the effects of fertilization strategies [i.e., no fertilizer (CK), chemical fertilizer (NPK), manure only (M) and manure plus chemical fertilizers (NPKM)] on soil organic carbon stock (SOC_s) at 0–20 cm depth in the North (NC), Northeast (NEC), Northwest (NWC) and South (SC) China. Compared with initial values, SOC_s increased by 24–68% and 24–74% under NPKM and M applications, respectively, over the experimental periods. Furthermore, final SOC_s under NPKM in NEC and NWC were significantly higher than those under other treatments, but there was no significant difference between NPKM and M in SC and no significant differences among fertiliser treatments in NC. Average SOC stock change rates (SOC_r) were positive under all treatments for all regions except for CK and NPK in NEC, which were negative. There were regional differences in treatment effects: all treatments showed significantly different rates in NC and NWC, whereas there were no significant differences between the M and NPKM in NEC and SC. Random forest (RF) modelling showed that among the selected variables initial SOC_s was the most important in accounting for differences in SOC_r, followed by soil bulk density, mean annual temperature and precipitation for all treatments. Soil total nitrogen content was also an important explanatory variable for SOC_r for CK and NPK, and soil pH for M. This study has highlighted the main driving variables of SOC change which can be of use in optimizing fertilization strategies, by taking account of the baseline SOC_s status and environmental factors for different regions, to minimize soil carbon emissions while maximizing carbon sequestration in soils.

Keywords: soil organic carbon, topsoil, Chinese croplands, long-term fertilization, region

1 Introduction

Soil organic carbon (SOC) in croplands is linked to soil fertility, crop production and food security, and its increase has been considered as a potential measure to reduce or offset the anthropogenic emission of carbon dioxide (CO_2), mitigating global climate change (Lal, 2004; Zhao et al., 2018). About 10% of the total organic carbon (C) reserve on the earth (140–170 Pg, 1 Pg=10^{15}g) is in cropland ecosystems, which is the most active part of world's terrestrial soil C pool (Liang et al., 2016). Furthermore, it has been demonstrated that agricultural soils provide a potential sink for atmospheric CO_2 by sequestering SOC (Smith et al., 2008). Therefore, how to maintain and increase SOC in agricultural ecosystems has become a key question for improving soil quality and mitigating global climate change.

China occupies only 7% of the world's arable land but it feeds nearly 20% of the world population (Fang et al., 2018). To meet the growing demand for food, China's agriculture has greatly intensified, with high inputs of chemical fertilizers and intensive agricultural land use (Huang and Sun, 2006; Ju et al., 2009). Between 1980 and 2014, nitrogen (N) fertilizer consumption nearly tripled in China (Liu et al., 2016). As a result, SOC stock (SOC_S) has been decreasing, along with nutrient loss, raising serious concerns (Gattinger et al., 2012; Lal, 2004). Manure has been widely applied in order to increase C sequestration in agricultural ecosystems (Gattinger et al., 2012; Maillard and Angers, 2014). However, some studies demonstrate no significant increase or even a decrease in SOC_S after manure amendment (Leifeld et al., 2009; Li et al., 2012) because when compared to chemical fertilizer application, the C input from crop biomass is lower. Generally, it is very important to improve the soil fertility and biodiversity in organic agricultural systems (Mader et al., 2002). Several field experiments have proven that long-term chemical fertilizers combined with manure input increases SOC concentration (Guo et al., 2016; Zhang et al., 2015). Chemical fertilizers indirectly affect the

C input to the soil by influencing the additions of plant stubble and root and rhizodeposition; in addition to this, manures directly influence C input to the soil and over the long-term assist the stabilization of SOC (Zhao et al., 2008). Moreover, SOC distribution and storage can be indicators of soil fertility, hydrology and propensity for greenhouse gas emissions (Lal, 2004). However, changes in SOC_s vary under different long-term fertilization strategies and across different regions.

Individual studies have attempted to relate the variability in SOC change to various explanatory factors such as climatic conditions, soil properties and management practices (Huang et al., 2012; Liang et al., 2016; Maillard and Angers, 2014; Tian et al., 2015). Generally, climate conditions are often regarded as the dominant factor driving soil C dynamics accounting for most of the variations in SOC change across larger scales (Carvalhais et al., 2014; Chen et al., 2013). Soil physiochemical properties are also regarded as relevant to the SOC_s change rate, including soil clay content which influences the rate of decomposition and sequestration of organic C due to the stronger adsorption ability of soil clay particles (Zhao et al., 2006). Soil acidity affects soil physical and biological properties, and plant growth and therefore affects soil C (Grybos et al., 2009; Qin et al., 2013). Furthermore, management practices, such as fertilization practices, also exert significant impacts on SOC dynamics by influencing C inputs; in particular, SOC changes are usually closely related to soil N (Qin et al., 2013). Few studies have attempted to comprehensively quantify the effects of these explanatory factors from all the available data for China. It is therefore important to understand the key driving factors of SOC change under different fertilization strategies which will contribute to the understanding of the underlying processes of SOC stabilization in soils as well as the development of potential strategies for the mitigation of soil degradation and climate change.

Previous observations have shown that fertilizer applications can have a big impact on SOC change but most studies have been at the local or limited regional scale (Guo et al., 2016; Li et al., 2010; Zhu et al., 2010). However, SOC can vary greatly at a wider regional scale (Gattinger et al., 2012; Zhang et al., 2016a). Very few studies have estimated SOC changes at the national scale, and the results of those were highly variable or even contrasting (Pan et al., 2010; Yan et al., 2011; Yu et al., 2009; Zhao et al., 2018). For example, Yu et al. (2009) reported reductions in SOC_s of 41% in North (NC), 35% in Northwest (NWC), 26% in South (SC) and 37% in Northeast (NEC) China, respectively in agricultural soils. However, Pan et al. (2010) report that SOC_s have increased by 1%, 12%, 11% and 6% in NEC, NC, NWC and SC, respectively. While Wang et al. (2013) showed that in NEC and NWC, SOC_s in agricultural soils was still decreasing. However, previous studies have usually focused on the short-term SOC change for a single site or a small area, and there is still lacking long-term comparative studies on SOC changes at a national scale in China. In this context, the assessment of regional or national SOC_s dynamics has recently become a very important topic (Gattinger et al., 2012; Carvalhais et al., 2014; Liu et al., 2013).

The long-term experiments across China provide a good opportunity to investigate regional variation in SOC and the driving factors for SOC change under different fertilization strategies. Therefore, in this study a series of factors known to affect soil C dynamics were selected and analysed statistically using data from 90 long-term field trials (>20 years) with the following aims: 1) to characterize the regional variability in SOC change in the topsoil (0–20 cm) in response to different agricultural fertilization strategies in China; 2) to identify the main driving factors of SOC change for these agricultural soils.

2 Materials and methods

2.1 Data sources

In China, most of long-term field experiments were established since the 1970s. Thus, to investigate the effects of long-term fertilization strategies on SOC changes, we only selected data from long-term field trials that had been running for at least 20 years (started from 1973 to 1995 and ended before 2019). Two sources were used to obtain data on cropland SOC_s, soil and climate characteristics (Appendix S1.): i) the Long-term National Soil Fertility Monitoring Network (NSFMN, Xu et al., 2015), which was established during the 1970s and 1980s in typical agricultural areas of China; ii) published peer-reviewed journal articles. We selected 38 long-term experiments representing different climates, soil types and cropping systems along a latitudinal transect of China from the NSFMN and added a further 52 sites from the published journal articles. Publications up until October 2019 were searched through the Web of Science (http://apps.webofknowledge.com/) and the China Knowledge Resource Integrated Database (http://www.cnki.net/) in which measured SOC from 20 cm topsoil was presented for Chinese croplands grown with either wheat, maize, rice or soybean. Field experimental treatments had to include: no fertilizers (CK), chemical fertilizers (N, phosphorus [P] and potassium [K]) - NPK, manure applied only (M) or M plus chemical fertilizers (NPKM). China has a wide variation in mean annual temperature (MAT) and mean annual precipitation (MAP) (i.e. -1.5-19.5 °C in MAT, 127-1975 mm in MAP) based on the average over the experimental periods using data from the Chinese Meteorological Administration (http://cdc.cma.gov.cn). To assess regional variations, we divided the mainland China into four regions with distinct climate, soil types and cropping systems: NEC (humid and cool temperate monsoon climate with a single cropping system with maize, wheat, soybean or rice), NC (semi-arid and temperate monsoon climate predominantly with a double cropping wheat-maize rotation), NWC (arid and temperate continental climate predominantly with a single cropping winter wheat or summer maize rotation), and SC (humid and sub-tropical or tropical monsoon climate predominantly with a double cropping (either wheat-rice or rice-rice) rotation. In total, we assembled a database of 762 observations of 0-20 cm SOC change for 23 counties from 90 long-term field trials covering diverse climate, soil and agricultural management conditions (Fig. 1).

2.2 Data analysis

2.2.1 SOC stock

Soil organic carbon stock (SOC_s stock, Mg C ha^{-1}) was estimated from:

$$SOC_{s} = BD \times SOC \times H \times 0.1 \qquad (1)$$

where H is soil depth (cm, it was set to 20 cm in this study), SOC is organic carbon content (g kg^{-1}within soil depth H), and BD is soil bulk density (g cm^{-3}).

2.2.2 SOC change rate

A mean annual SOC change rate (SOC_r, Mg C ha^{-1} yr^{-1}) was calculated as:

$$SOC_r = \left(\frac{SOC_t - SOC_0}{t}\right) \qquad (2)$$

where t is the experimental duration; and SOC_0 and SOC_t are SOC stocks in the beginning and final reported year of the experiment, respectively.

2.3 Statistical analysis

To assess the different fertilization treatment effects on SOC_s and SOC_r, one-way ANOVA was implemented using the SPSS 19.0 software package. Graphs were prepared using Origin profession version 8.5.

2.3.1 Data analysis

We used a Random Forest (RF) model to analyse impact factors on SOC_r. This is an ensemble approach to regression in which the classification algorithm builds a large number of decision trees with the output as the mean prediction of the individual trees (Breiman, 2001; Liaw and Wiener, 2002). The RF classifier consists of tree classifiers and each classifier is generated using a randomly selected subset of the variables that are independent of the input variable sampling, and each tree votes for the most popular projective units to classify the input variables (Breiman, 2001). The RF classifier has been shown to be more successful than other classifiers in assembling the ensemble (Dietterich, 2002).

We first selected dependent variables related to SOC_r under the different treatments, using correlation analysis (SPSS 19.0 for Windows). In total, fourteen variables (Table 2) were chosen for the RF model. To run the RF model, three parameters to generate forest trees have to be determined: the number of trees to be generated in the forest (ntree), the number of variables to be selected and tested for the best split when growing the trees (mtry) and the minimum number of nodes of the trees and below which the tree is not split (nodesize) in a RF. According to previous published articles, ntree was set to 1,000 to stabilise the errors (Colditz., 2015; Reese et al., 2014) and mtry was the square root of the number of input variables (Gislason et al., 2006) and nodesize was set at 5. RF uses the bootstrap repeated sampling method; two thirds of the samples (called in-bag samples) are used to train the trees, with the remaining one-third of the samples (called out-of-the bag samples: OOB) used as a test sample for RF (Breiman, 2001). RF uses an extension of cross-validation, where each OOB sample is predicted by its corresponding bootstrap training tree. The Mean Square Error (MSE) was used to estimate the accuracy of OOB predictions (Liaw and Wiener, 2002). The RF model was validated by calculating the differences between the observed and predicted value of the mean percentage error (MPE), root mean square error of prediction (RMSEP) and R^2 (Liaw and Wiener, 2002; Wiesmeier et al., 2011):

$$\mathrm{MSE}=\frac{\sum_{i=1}^{n}(Zi-Z)^2}{n} \tag{3}$$

$$\mathrm{MPE}=\frac{\sum_{i=1}^{n}(Pi-Oi)}{n} \tag{4}$$

$$\mathrm{RMSEP}=\sqrt{\frac{\sum_{i=1}^{n}(Oi-Pi)^2}{n}} \tag{5}$$

$$R^2=1-\sum_{i=1}^{n}\frac{(Oi-Pi)^2}{(Oi-O)^2} \tag{6}$$

where *Z* is the average value of all OOB predictions, Zi is the *i*th OOB, Oi is the *i*th observed value, Pi is the predicted by RF models for the *i*th value, O is the average observed values and n is the total number of observations. The RF models was implemented using the “RandomForest” package in R program (R version 3.2.2, 2015) (Liaw and Wiener, 2002).

3 Results

3.1 SOC stock changes under long-term fertilization strategies in different regions

Final SOC_s under NPKM in NEC and NWC were significantly higher than those under other treatments; there was no significant difference between NPKM (46.00 Mg ha^{-1}) and M (46.17 Mg ha^{-1}) in SC and no significant differences among NPK (46.00 Mg ha^{-1}), M (32.53 Mg ha^{-1}) and NPKM (33.40 Mg ha^{-1}) in NC (Fig. 2). On average, SOC_r was positive under all treatments for all regions but negative rates were observed under CK (-0.12 Mg ha^{-1} yr^{-1}) and NPK (-0.03 Mg ha^{-1} yr^{-1}) in NEC (Fig. 3). There were significant differences between all treatments in NC (with a significant order of M > NPKM > NPK > CK from 0.03 to 0.58 Mg ha^{-1} yr^{-1}) and NWC (with a significant order of NPKM > M > NPK > CK from 0.07 to 0.59 Mg ha^{-1} yr^{-1}) but no significant differences between the M and NPKM in NEC and SC. Although there was an increase in SOC stocks under M and NPKM on average in all the regions, a decrease in SOC was observed in some trails, especially in NEC where there was no significant difference between NPK (-0.03 Mg ha^{-1} yr^{-1}) and CK (-0.12 Mg ha^{-1} yr^{-1}) (Table 1).

3.2 Influencing variables of SOCr

Selected independent variables from the RF models explained 21, 59, 56 and 22% of the total variance in SOC_r under the CK, NPK, NPKM and M, respectively (Fig 4 a – d). Initial SOC_s was the most important explanatory variable, followed by BD, MAT and MAP (Fig. 4 e – h). In addition, soil total nitrogen (TN) was an important driving variable for the CK and NPK . However, pH was an important driving variable for NPKM and M. Overall, RF analysis suggests that SOC_r in the topsoil under different long-term fertilization treatments was mostly controlled by edaphic characteristics and environmental factors, and the influence of fertilization-related factors were moderate.

To assess whether the influence of the driving factors were significantly different, correlation coefficients (r) were calculated. Results confirmed significant negative correlations between initial SOC or BD and SOC_r of for all fertilization treatments (all $P < 0.05$; Table 2). There was a significant positive correlation between soil microbial biomass carbon (SMBC), MAP or MAT and SOC_r across all treatments. For the fertilization treatments, there were significant positive relationships of SOC_r with TN for the NPKM (r=0.20) and M (r=0.40) but a negative relationship for the NPK (r=-0.12). Moreover, in NPKM and M, there was a significant effect on SOC_r of manure-C and N (r =0.11 and 0.21 for NPKM and r =0.25 and 0.43 for M, respectively).

4 Discussion

Our analysis clearly showed that compared with initial SOC values, NPK increased SOC_s in NC, NWC and SC but not in NEC. The relative increase for SOC_s under NPK in NC, NWC and SC implies that adequate NPK application can result in a greater return of crop residues and root-related C associated with a higher crop productivity (Mandal et al., 2007), which is necessary to stabilize and maintain SOC levels in most intensive cropping systems. However, in NEC which has a naturally high initial SOC_s, the accumulated SOC under NPK could not compensate for the C loss. Khan et al. (2007) also found that long-term (40 to 50 years) application of chemical N fertilizer caused a net decline in soil SOC and a negative C balance in the Morrow Plots located in USA, the soils of which are similar to those of NEC. Manure application, especially in addition to chemical NPK, has a great potential in improving C sequestration for major agricultural systems in China, consistent with some previous studies (Li et al., 2010; Wang et al., 2015; Yan et al., 2013). The positive SOC response following manure application can probably be mainly attributed to direct C input by manure and indirect C inputs through the promoted crop biomass return as roots and crop residues (Gattinger et al., 2012; Gabriel and Kellman, 2011).

4.1 Changes in the SOC stock dynamics with treatments and geographical regions

There was great regional variation in cropland soil C dynamics under different long-term fertilization strategies across China. In this study, there was high variation in the frequency (37–100%) of SOC_s increase and SOC_r varied between –0.12 and 0.59 Mg ha^{-1} yr^{-1} on average across the different regions of China. In general, climate conditions, especially MAP and MAT, are acknowledged as important environmental factors influencing the distribution of SOC at the large geographical scale (Mao et al., 2015; Wiesmeier et al., 2013) which could affect the quantity and quality of crop residue inputs and the SOC turnover rates (Hevia et al., 2003). The observed correlations between the SOC change and climate variables (MAP and MAT) in this study are in agreement with other research results (Chen et al., 2018; Jobbagy and Jackson, 2000), showing a strong positive correlation with MAP and MAT. Our results show a relatively higher SOC increase after fertilization in tropical SC than that in cool temperate NEC, which was also supported by Han et al. (2016) and Huang et al. (2012). In NEC, SOC significantly decreased without manure amendments, and other studies have also reported that cropland soils in NEC have been losing C (Huang and Sun, 2006; Zhang et al., 2016a). The NEC is one of the few world regions characterized by black soil and a cold climate (Duan et al., 2011) with a short growth period and low rates of C inputs to soils. The overall SOC decline in NEC croplands may also be related to the significant drying trend under climate change (Ding et al., 2006). The more favourable MAT and MAP in SC for aboveground plant growth is therefore likely to increase C input and SOC stabilization (Pan et al., 2009). Although higher MAT and MAP conditions accelerate the decomposition of organic C, the excess C inputs are converted to soil C and offset the C loss from soil respiration (Yu et al., 2013). The relatively higher SOC increase in croplands from NWC may be attributed to the enhanced crop production favoured, in part, by the increasing rainfall due to climate change over the last decades (Ding et al., 2006). NWC is a vast semi-arid area with MAP ranging from 127 to 632 mm and more than 90% of the cropland depends on rainfall. Thus, MAP is a dominant factor for production, and even a small increase could significantly enhance the bio-productivity and thus contribute to the accumulation of SOC (Jobbagy and Jackson, 2000). Interestingly, a high frequency (78%) of increase in SOC_S was observed under the CK after more than 20 years in the NWC. In this region, with low MAP, the crop residue and manure C decomposition and soil respiration rates would be slower, so the C input supplied through the incorporation of only crop roots and residues was sufficient to maintain or even enhance the C stock (Tewksbury and Van Miegroet, 2007).

China has a vast territory and diverse natural conditions which includes most kinds of agricultural systems in the world (Liu et al., 2013). NEC and NWC are mainly associated with a mono-cropping system and NC with double cropping systems, whereas double or triple-cropping systems are applied in SC (Yu et al., 2013). The multiple cropping index together with greater incidence of manure application in SC result in C inputs in that region approximately twice those for NC, NEC and NWC under the same agricultural management scenario (Wang et al., 2013; Yu et al., 2013). Moreover, SC has a large area of paddy fields and prolonged water-logging may induce anaerobic soil conditions, slowing microbial decomposition of SOC, resulting in a higher SOC content (Sahrawat, 2004). Our analysis also showed that in SC, SOC_r was not significantly different between the NPKM and M. Generally, C input is the major factor controlling the C stocks change (Huang and Sun, 2006), so this indicates that the magnitude of C inputs from the crop residues, exudates and manure-C and the outputs from decomposition may be roughly equal among the treatments. Additionally, some studies have shown that crop production did not differ significantly between the NPKM and M from long-term studies in SC (Zhang et al., 2009; Zhou et al., 2016).

We speculate that sites with inherently high and low fertility differ in the turnover, formation and accumulation of SOC (Sollins et al., 2007). A negative relationship was derived between SOC_r and initial SOC_S, as has been shown previously (Huang et al., 2012; Maillard and Angers, 2014; Tian et al., 2015). The initial SOC content varied widely across the different regions of China (i.e., 33.47–38.24, 18.71–23.83, 19.53–20.46 and 32.96–34.53 t C ha^{-1} in NEC, NC, NWC and SC, respectively). A greater SOC_r was observed in NWC and NC soils with lower initial SOC content than that in NEC. The NEC is generally characterized by high SOC content and fertility (Liu et al., 2003) but over recent decades has experienced soil erosion, intensive cultivation, low C return, and a significant warming and drying trend under climate change (Xie et al., 2011; Zhang et al., 2016b) which have resulted in an initial rapid SOC loss despite the inherently high content of SOC (Sun et al., 2010; Wang et al., 2010). Our observation of a decrease in SOC in the NEC when relying solely on the native soil fertility (CK treatment) or using only NPK is supported by others (Wang et al., 2013; Zhang et al., 2016a). In addition, in NEC there was no significant difference in SOC_r between the NPKM and M which could also be partly attributed to the relatively low average initial SOC content in M (Table 1). In NC, the higher SOC_r under M may be attributed to the low initial SOC content (Table 1) and to the fact that the NC has one of the highest N deposition rates both nationally and globally (Jandl et al., 2007) which enhances crop production. This would result in more C input to the soil via crop stubble and root biomass, and manure application would supply exogenous organic C, elevating the relatively low SOC content of the soil in this region. However, for NWC with lower productivity and nutrient retention, NPK application improves soil nutrient conditions to meet crop nutrient demands and maintain a high level of crop production, leading to greater C input from biomass (Campbell et al., 2001), but NPKM additionally directly adds C input and so SOC_r was largest under NPKM.

4.2 Factors influencing SOC_r under different fertilization treatments

From the RF model, initial SOC_s was the most important factor driving SOC change for all the treatments, even within the same region (e.g. SC, Appendix S2, combined NEC, NC and NWC regions, Appendix S3). The negative relationship between SOC_r and initial SOC_s

suggests that soils with low C contents have the greatest potential and efficiency to sequester C because they are further from their saturation level (Stewart et al., 2008; Yan et al., 2013). Moreover, soil with a lower C content has insufficient substrate to maintain an active decomposer population, resulting in conditions less favourable for C chemicalization compared to soils with a higher C content (Mann, 1986). Some studies have shown little or no increase in SOC for high C content soils, even with two- or three-fold increases in extraneous C inputs (Campbell et al., 1991).

Soil BD has been identified as another important variable explaining differences in SOC dynamics (Don et al., 2011; Liang et al., 2016), and this soil property varied across the cropland soils in the different regions of China (being much higher in NEC). Increases in soil BD under CK and NPK have been reported, especially the highest soil BD under CK (Guo et al., 2016). Loss of SOC often results in soil BD increase because organic matter stabilizes soil aggregates prevent soil from slaking, dispersion and collapse (Logsdon and Karlen, 2004). Soil BD data showed a highly significant negative correlation with SOC_r ($P<0.01$, Table 2), in accordance with results reported by Guo et al. (2016). Soil BD is an important indicator of soil compaction, influencing not only the soil-air-water interactions, microbial activity, nutrient uptake, moisture and temperature retention (Martinez and Zinck, 2004; Whalley et al., 1995) but also nutrient availability, indirectly affecting soil quality and productivity and SOC turnover (Reichert et al., 2009).

4.2.1 Factors influencing SOC_r under the control treatment

Soil TN content was identified as another main influencing factor of SOC_r under the CK, consistent with the observations of other studies (Duete et al., 2008; Liang et al., 2016). Generally, soil C and N cycles are linked (Song et al., 2015) and N is a key to ecosystem functioning and used frequently to evaluate soil fertility, i.e. the balance between plant demand and soil supply of nutrients in croplands (Vitousek et al., 2009). Soil N tends to be a limiting factor on crop growth and belowground C transformations (Duete et al., 2008) and it is required to support C sequestration. For the CK, with no fertilizer input for more than 20 years, the nutrient input depends on the natural soil fertility and crop residue input (Zhang et al., 2009). A significant positive correlation between soil TN and SOC change was found in this study of long-term experiments, which is consistent with other studies (Chen et al., 2013; Yu et al., 2014).

The SMBC was another important variable related to the SOC_r under the CK. Wang et al. (2015) also reported a significant positive linear correlation between SMBC and SOC. The SOC provides substrates for soil microorganisms, thus stimulating their growth; in turn, SMBC can have important effects on belowground C and N cycles and plant nutrient availability (Sabahi et al., 2010; Zhang et al., 2015) thus increasing the crop biomass C input. Moreover, SMBC contributes to C sequestration by producing polysaccharides and fungal hyphae that improve soil aggregation and by directly contributing to stable soil C pools (Kindler et al., 2009).

4.2.2 Factors influencing SOC_r under inorganic fertilization

The soil TN were identified as the most important edaphic factors explaining the variance in SOC_r under the NPK. The results suggested a negative relationship between SOC_r and TN content. These effects of N fertilizer input on crop C inputs to soils represent an important mechanism via soil N on the SOC_r under these fertiliser treatments without external manure inputs (Liang et al., 2016). Generally, N addition is understood to increase plant biomass and thereby organic C inputs to the soil by enhancing soil TN content and promoting N availability, resulting in a positive effect on SOC (Huang et al., 2010; Zhang et al., 2010). However, Tong et al. (2009) observed that both SOC and TN contents tended to be stable, having reached an equilibrium under the present conditions after long-term fertilization. Although N is required to support C sequestration in soils (Lal, 2004), it was not a limiting factor for crop growth under long-term chemical fertilizer application (Han et al., 2017). Under long-term chemical fertilizer application alone, an N-induced decrease in soil pH and deterioration of soil properties may depress root and microbial activity, thereby decreasing SOC (Tian and Niu, 2015). In addition, under long-term intensive use of chemical N fertilizer, the C/N ratio also reduces which accelerates SOC decomposition (Tong et al., 2009).

4.2.3 Factors influencing SOC_r under manure fertilization

Under the M, soil pH was a key explanatory variable for SOC_r, preceded only by the initial SOC content. This agrees with previous observations that SOC concentration significantly increased with decreasing pH (Kemmitt et al., 2006). Soil pH has a direct impact on SOC decomposition by affecting its solubility (Kemmitt et al., 2006) and an indirect impact by affecting plant and microorganism growth (Pietri et al., 2009). At high soil pH, the solubility of iron, zinc, and manganese decrease, restricting plant growth, resulting in lower indirect C inputs (Grybos et al., 2009). In addition, manure contains dissolved organic C which is more readily released under high pH conditions (Andersson et al., 2000). Abundance and activity of microorganisms are lower in low pH soils, resulting in a lower SOC quality and bioavailability and increased recalcitrance because SOC forms complexes with aluminium (Al) (Kemmitt et al., 2006).

4.2.4 Factors influencing SOC_r under combined inorganic fertilizer and manure

For the NPKM treatment, the soil TN was not identified as the most important explanatory variable for SOC_r, suggesting that when compared with CK and NPK treatments, manure application in the NPKM treatment most likely reduced the N constraints on crop growth and its subsequent influence on SOC turnover (Duan et al., 2014; He et al., 2015). Our results indicated that clay content was a relatively weak variable for predicting SOC dynamics. This is inconsistent with some studies that have highlighted soil clay content as a key factor controlling SOC dynamics, because, under short-term observations, SOC is generally protected by the soil clay particles (Haynes and Naidu, 1998). However, this has not been shown for Chinese cropland soils, where clay content has been only weakly related to long-term SOC accumulation (Percival et al., 2000). These apparently contradictory observations may be due to differences in clay chemically in different soils and under different land use (Li et al., 2016). Further research is needed to explore the influence of clay content on SOC dynamics for different soils.

The assessment of multiple influential factors on SOC change in this study suggests that the effects of fertilization-related factors may be overshadowed by soil factors. However, the inherent soil properties are difficult to change and the costs of change are too high. This study informs the needs to develop strategic soil management plans to promote soil carbon sequestration under long-term intensive fertilization. Our analysis showed that in NEC, SOC in agricultural soils is still decreasing without manure amendment and future efforts should be made to control this through increased use of livestock manure. We suggest that increased productivity is most likely to be the main driver of SOC increase and conclude that the regular use of manure with chemical fertilizers is essential for the long-term SOC increase in NWC. In NC, however, with a relatively low SOC state, soils have a greater potential to sequester C, so the M with more C input also served as a pathway for substantial soil C sequestration. In addition, our analysis highlights the need to take account of the baseline status to assess the net soil C balance over time and space. On average, the initial SOC_S is higher in SC, therefore, high manure amendments as substitution or additional nutrient input are not recommended for sustainable SOC sequestration, and that moderate manure application is a conducive practice for the SC.

5 Conclusions

Compared with initial values, final SOC_S increased by 24–68% and 24–74% under NPKM and M fertilization strategies, respectively, over the experimental periods. It is suggested that

Chinese croplands have great potential to sequester SOC and manure application can be a highly important choice to enhance SOC_s and mitigate climate change. We found regional differences in SOC_s changes and treatment effects. In NEC and NWC, the final SOC_s under NPKM was significantly higher than under other treatments; in SC there was no significant difference between NPKM and M, and in NC, there were no significant differences between NPK, M and NPKM. Averaged across all treatments, SOC_r was positive for all regions but treatment-specific rates were negative under CK and NPK in NEC. There were significant differences in SOC_r between all treatments in NC and NWC, but there was no significant difference between M and NPKM in NEC and SC. Although SOC_s increased on average under treatments M and NPKM in all the regions, a decrease in SOC was reported in some trials, especially in NEC. Among the selected variables, initial SOC_s had the strongest influence on SOC_r, followed by soil BD and climate factors across all the treatments. Under specific treatments, soil TN was the most important driving variable for CK and NPK, and soil pH was for M. This study has demonstrated differential mechanisms in control of long-term SOC change over large regions of China and informed the need to develop different soil management plans to promote SOC_s. The responses of SOC to fertilizer managements as revealed by this synthesis can be used in cropland models for better understanding and prediction of the agro-ecosystem C cycle feedbacks under different fertilizer management strategies.

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Figure legends

Fig 1. Distribution of long-term field trials in China as used in this study. Northeast (NEC), North (NC), Northwest (NEC) and South (SC) China.

Fig 2. The soil organic carbon stock (0-20 cm) in different treatments (no fertilizers - CK; chemical fertilizers (nitrogen [N], phosphorus [P] and potassium [K]) - NPK; manure applied only – M; and manure plus chemical fertilizers - NPKM) in the final report year of experiments in Northeast (NEC), North (NC), Northwest (NEC) and South (SC) China. Black and red dashed lines within the boxes are the median and mean values. The lowercase letters above the bars signify differences at $P<0.05$.

Fig 3. The soil organic carbon stock change rate (0–20 cm) in different treatments (no fertilizers – CK; chemical fertilizers (nitrogen [N], phosphorus [P] and potassium [K]) - NPK; manure applied only – M; and manure plus chemical fertilizers – NPKM) in Northeast (NEC), North (NC), Northwest (NEC) and South (SC) China. Black and red dashed lines within the boxes are the median and mean values. The lowercase letters above the bars signify differences at $P<0.05$.

Fig 4. Performance of random forests (RF) models for identifying the controlling factors of soil organic carbon stock change rate (0–20 cm) and the relative importance of independent variables for controlling soil organic carbon stock changes rate in different treatments (no fertilizers – (a) and (e); chemical fertilizers (nitrogen [N], phosphorus [P] and potassium [K])

– (b) and (f); manure applied only – (c) and (g); and manure plus chemical fertilizers – (d) and (h) on croplands in China. MAT: mean annual temperature, MAP: mean annual precipitation, pH: soil pH, BD: soil bulk density, TN: soil total nitrogen content, SMBC: soil microbial biomass carbon.

Supplementary material

Appendix S1 Basic information of the long-term experimental sites for this study.

Appendix S2 Performance of random forests (RF) models for identifying controlling factors of soil organic carbon stock change rate (0–20 cm) and the relative importance of independent variables for controlling soil organic carbon stock change rate for different treatments in croplands in the South China.

Appendix S3. Performance of random forests (RF) models for identifying controlling factors of soil organic carbon stock change rate (0–20 cm) and the relative importance of independent variables for controlling soil organic carbon stock change rate for different regions in China (Northeast, North and Northwest).

Table 1

Variation in SOC dynamics across geographical regions of China

Geographical region	Treatment	Sample size	Initial SOC stock (Mg ha^{-1})		Final SOC stock (Mg ha^{-1})		Frequency of increase (%)	Mean SOC stock change rate (Mg ha^{-1}yr^{-1})	
			Mean	95% CI	Mean	95% CI		Mean	95% CI
NEC	CK	20	38.24	34.35-42.14	37.12c	34.01-41.13	36.54	-0.12c	-0.20--0.04
	NPK	52	38.07	34.30-41.84	37.35c	34.60-40.11	59.72	-0.03b	-0.10-0.03
	M	28	33.47	29.23-37.71	41.49b	37.16-45.82	76.32	0.29a	0.15-0.43
	NPKM	84	38.07	34.30-41.85	47.25a	44.34-49.52	83.33	0.32a	0.24-0.40
NC	CK	33	22.98	19.58-26.37	25.00b	20.64-29.36	57.58	0.03d	-0.03-0.09
	NPK	52	23.83	20.53-27.13	31.40a	27.69-35.11	86.54	0.19c	0.13-0.25
	M	17	18.71	13.92-23.51	32.53a	25.76-39.30	100	0.58a	0.39-0.77
	NPKM	47	22.21	19.24-25.17	33.40a	29.80-37.01	93.62	0.40b	0.32-0.47
NWC	CK	20	19.73	18.07-21.38	22.24c	20.20-24.27	77.5	0.07d	-0.004-0.15
	NPK	41	19.53	17.88-21.19	25.72b	23.98-27.46	86.88	0.27c	0.19-0.36
	M	18	20.46	17.44-23.48	26.96b	24.42-29.51	100	0.30b	0.23-0.37
	NPKM	40	19.70	18.03-21.37	33.08a	30.80-35.37	98.36	0.59a	0.49-0.68
SC	CK	42	34.53	32.19-36.87	35.69b	33.30-38.09	55	0.03c	-0.04-0.11
	NPK	94	32.96	30.86-35.06	37.88b	36.08-39.68	76.12	0.21b	0.14-0.29
	M	40	32.96	29.19-36.72	46.17a	41.59-50.76	91.67	0.52a	0.39-0.65
	NPKM	134	33.35	31.33-35.36	46.00a	44.07-47.93	89.08	0.51a	0.44-0.58

Note: NEC: Northeast China, NC: North China, NEC: Northwest China and SC: South China. CK- no fertilizers; NPK- chemical fertilizers (nitrogen [N], phosphorus [P] and potassium [K]); M - manure applied only and NPKM- manure plus chemical fertilizers. 95% CI: 95% confidence intervals.

Table 2

Pearson’s correlation coefficients (r) between soil organic carbon stock change rate and controlling factors.

	Controlling factors										
Treatment	Initial SOC (Mg ha^{-1})	Manure - C (kg C ha^{-1})	Manure - N (kg N ha^{-1})	Chemical N (kg N ha^{-1})	BD (g cm^{-3})	Clay (%)	pH	TN (g kg^{-1})	SMBC (mg kg^{-1})	MAP (mm)	MAT (℃)
CK	-0.29**	N/A	N/A	N/A	-0.18**	0.06	-0.22**	0.10*	0.25**	0.31**	0.32**
NPK	-0.42**	N/A	N/A	0.16**	-0.18**	0.01	-0.07	-0.12*	0.25**	0.18**	0.28**
M	-0.40**	0.25**	0.43**	N/A	-0.42**	0.16	-0.12**	0.40**	0.10*	0.34**	0.29**
NPKM	-0.30**	0.11*	0.21**	0.09	-0.28**	0.04	-0.11**	0.20**	0.25**	0.13**	0.21**

Note: CK- no fertilizers; NPK- chemical fertilizers (nitrogen [N], phosphorus [P] and potassium [K]); M - manure applied only and NPKM-manure plus chemical fertilizer

MAT: mean annual temperature, MAP: mean annual precipitation, pH: soil pH, BD: soil bulk density, TN: soil total nitrogen content, SMBC: soil microbial biomass carbon.

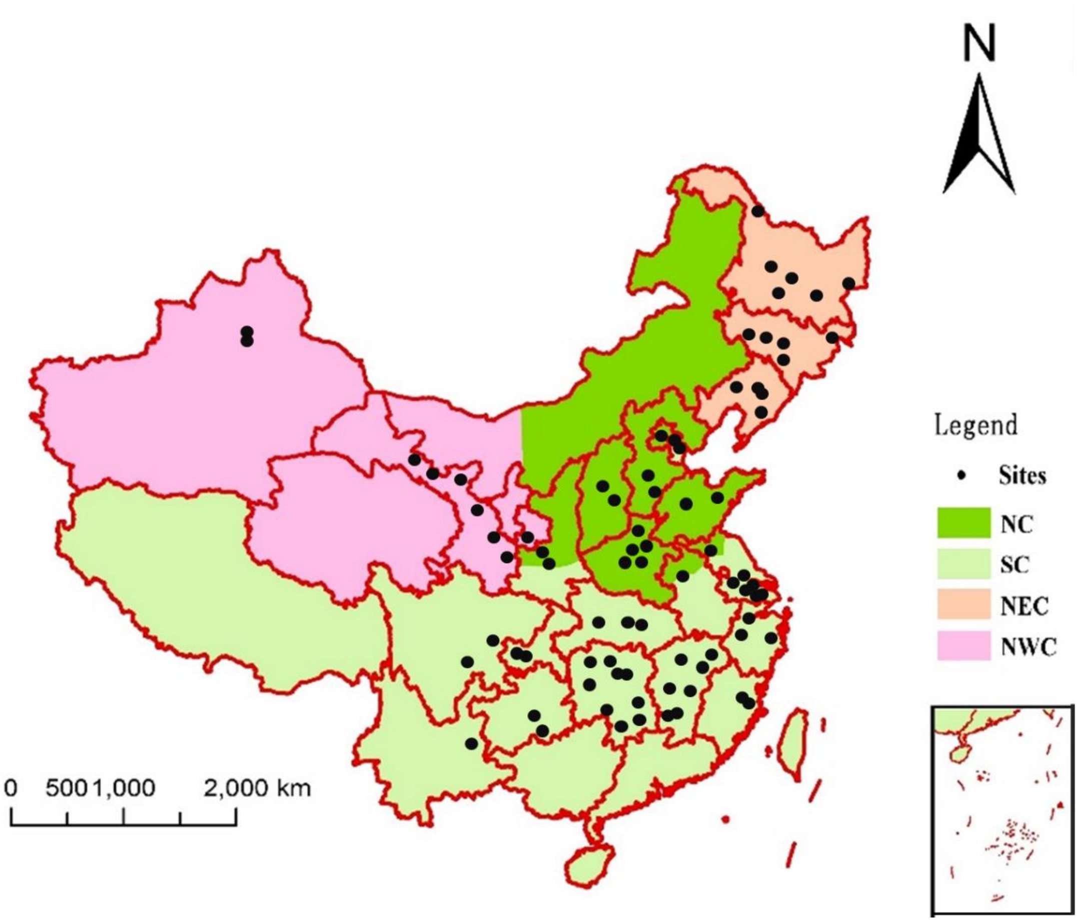

Fig 1. Distribution of long-term field trials in China as used in this study. Northeast (NEC), North (NC), Northwest (NEC) and South (SC) China.

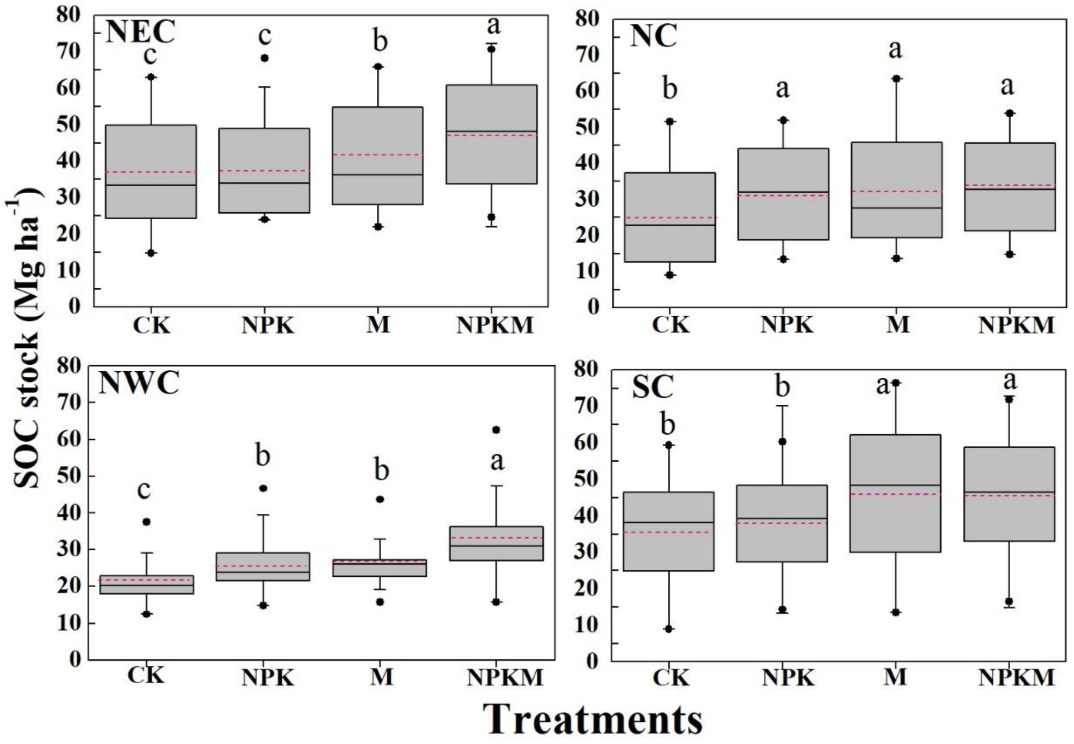

Fig 2. The soil organic carbon stock (0-20 cm) in different treatments (no fertilizers - CK; chemical fertilizers (nitrogen [N], phosphorus [P] and potassium [K]) - NPK; manure applied only – M; and manure plus chemical fertilizers - NPKM) in the final report year of experiments in Northeast (NEC), North (NC), Northwest (NEC) and South (SC) China. Black and red dashed lines within the boxes are the median and mean values. The lowercase letters above the bars signify differences at $P<0.05$.

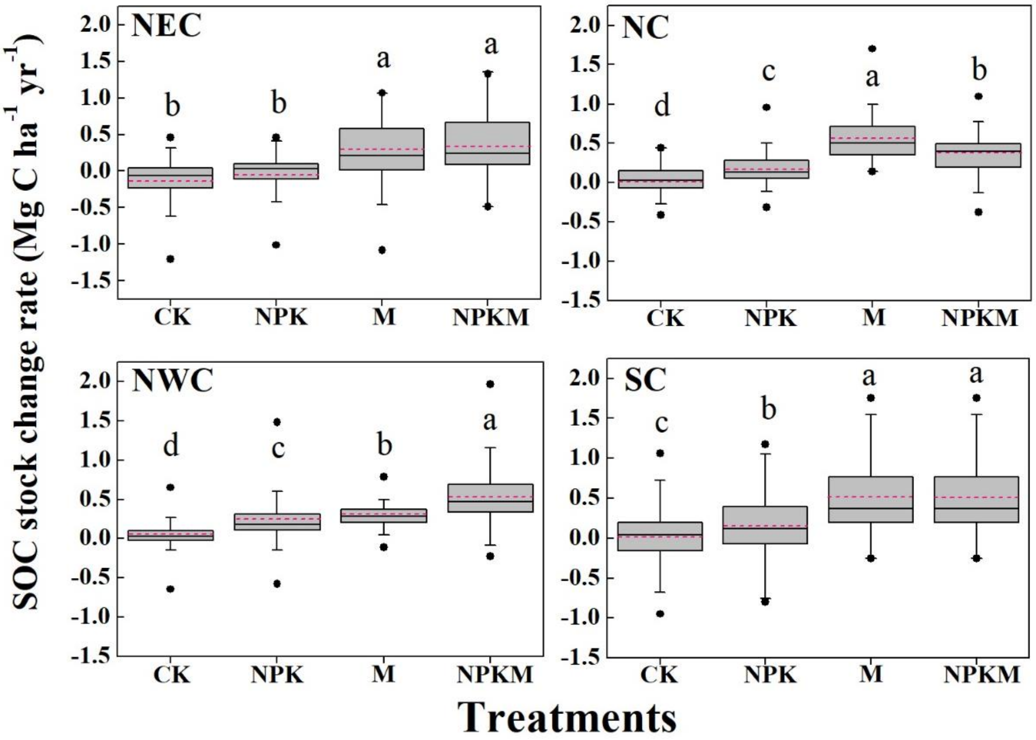

Fig 3. The soil organic carbon stock change rate (0–20 cm) in different treatments (no fertilizers – CK; chemical fertilizers (nitrogen [N], phosphorus [P] and potassium [K]) - NPK; manure applied only – M; and manure plus chemical fertilizers – NPKM) in Northeast (NEC), North (NC), Northwest (NEC) and South (SC) China. Black and red dashed lines within the boxes are the median and mean values. The lowercase letters above the bars signify differences at $P<0.05$.

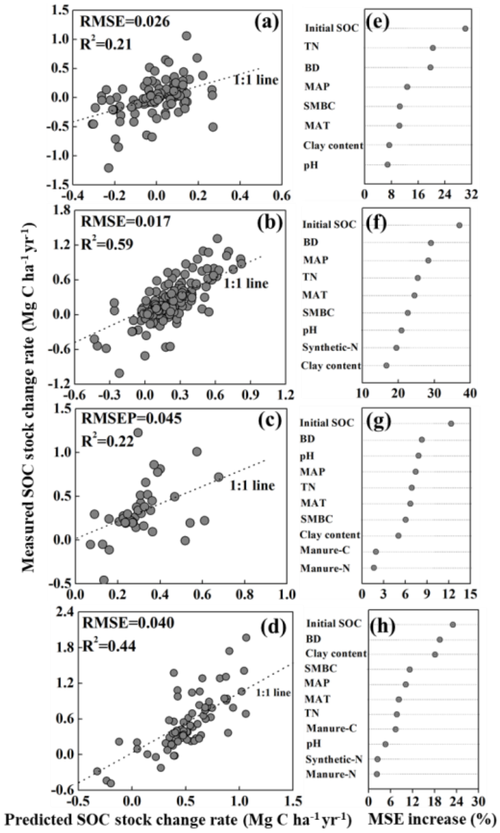

Fig 4. Performance of random forests (RF) models for identifying the controlling factors of soil organic carbon stock change rate (0–20 cm) and the relative importance of independent variables for controlling soil organic carbon stock changes rate in different treatments (no fertilizers – (a) and (e); chemical fertilizers (nitrogen [N], phosphorus [P] and potassium [K]) – (b) and (f); manure applied only – (c) and (g); and manure

plus chemical fertilizers – (d) and (h) on croplands in China. MAT: mean annual temperature, MAP: mean annual precipitation, pH: soil pH, BD: soil bulk density, TN: soil total nitrogen content, SMBC: soil microbial biomass carbon.

Conflict of Interest Statement

The authors declared that there is no conflict of interest either financially or otherwise.

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Declaration of Interest Statement

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