Soil organic carbon saturation deficit under primary agricultural managements across major croplands in China

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ABSTRACT

Introduction: To generate information for the effective management of soil organic carbon (SOC) sequestration in Chinese croplands, we compared the additional organic carbon (C) that can be stabilized by fine soil particles (<20 μm) with typical fertilization practices across soil types and climate zones. Using data from 30 long-term experimental study sites across the major agricultural zones in China, we estimated stable SOC saturation deficit (SOC deficit) under a variety of management practices (Smith 2007; Yan et al. 2007; Cheng et al. 2013). Although previous studies have compared SOC stocks under a variety of agricultural practices to identify the optimal management practices that maximize soil C sequestration (Yan et al. 2007; Lu et al. 2009; Qin and Huang 2010; Wang et al. 2010; Cheng et al. 2013), most of these studies focused on C accumulation in bulk soil. However, SOC is heterogeneous in terms of C turnover time (Bernoux et al. 1998). Labile SOC, such as particulate organic matter or macro-aggregate C, has a turnover time of days or months; in addition, it is vulnerable to agricultural practices and can be easily lost (Blanco-Canqui and Lal 2004). Stable SOC, which is associated with fine soil particles, has turnover times ranging from years to millennia (Trumbore 2000; Feng et al. 2016) and accounts for at least half of the total SOC stock (Christensen 1998; Wiesmeier et al. 2014; Cai et al. 2016). Thus, measures to

Introduction

Enhancing soil organic carbon (SOC) is important for improving soil quality and mitigating climate change, particularly for croplands, where soils are always SOC depleted, and thus have considerable potential to sequester carbon (C) (Lal 2004). Many agricultural practices, such as fertilization, manure amendments, and straw return, are commonly used to increase C inputs or to reduce organic matter decomposition (Lal 2004; Chung, Grove, and Six 2008; Banger et al. 2009; Zhang et al. 2010, 2012; Maillard and Angers 2014). Therefore, SOC is a result of the balance between C inputs and C loss primarily through decomposition. Estimating SOC sequestration potential is thus the key for managing soil C sequestration, because it indicates the amount of C that can be stabilized in soils.

Two approaches are commonly used to estimate SOC sequestration potential (Sun et al. 2008; Cheng et al. 2013). One method is calculating the difference between SOC stocks in arable (cropland) and native ecosystem or best cropland management practices (Smith 2004; Yan et al. 2007; Cheng et al. 2013). Although previous studies have compared SOC stocks under a variety of agricultural practices to identify the optimal management practices that maximize soil C sequestration (Yan et al. 2007; Lu et al. 2009; Qin and Huang 2010; Wang et al. 2010; Cheng et al. 2013), most of these studies focused on C accumulation in bulk soil. However, SOC is heterogeneous in terms of C turnover time (Bernoux et al. 1998). Labile SOC, such as particulate organic matter or macro-aggregate C, has a turnover time of days or months; in addition, it is vulnerable to agricultural practices and can be easily lost (Blanco-Canqui and Lal 2004). Stable SOC, which is associated with fine soil particles, has turnover times ranging from years to millennia (Trumbore 2000; Feng et al. 2016) and accounts for at least half of the total SOC stock (Christensen 1998; Wiesmeier et al. 2014; Cai et al. 2016). Thus, measures to
mitigate climate change impact should focus on the C sequestration potential of stable SOC.

The other method for estimating SOC sequestration potential is based on model simulations, which assumes a linear increase in SOC with increasing C inputs, and SOC accumulation is not limited and its decomposition rate does not change (Parton et al. 1987). However, several studies have suggested the existence of SOC saturation, particularly for stable SOC (Chung, Grove, and Six 2008; Stewart et al. 2008; Di et al. 2014; Du et al. 2014), because it is absorbed by the mineral surfaces of soil particles or trapped in pore spaces, which are finite and must have an upper limit to hold SOC (Hassink 1997; Six et al. 2002). Soil C saturation suggests that, with increasing C inputs, SOC stock will reach a maximum and SOC accumulation rate will decrease in this process (West and Six 2007; Stewart et al. 2007, 2008). Hence, SOC model simulations might generate large uncertainties, if SOC saturation is not accounted for.

The concept of SOC saturation provides a way to estimate the C sequestration potential of fine soil particles under different land use or soil groups at different spatiotemporal scales (Angers et al. 2011; Wiesmeier et al. 2014; Cao et al. 2016). According to this view, the difference between the upper C limit ($C_{f-max}$) and current C concentration ($C_{f-cur}$) of fine soil particles is defined as the stable SOC saturation deficit (SOC$_{deficit}$, i.e., C sequestration potential), indicating the amount of additional C that can be stabilized by fine soil particles. Thus, identifying soils with largest SOC$_{deficit}$ values depends on estimating $C_{f-max}$ and $C_{f-cur}$. The former is primarily regulated by soil texture (Hassink 1997; Six et al. 2002; Liang et al. 2009) and mineralogy (Six et al. 2002; Feng, Plante, and Six 2013). The commonly used Hassink model might underestimate SOC$_{deficit}$ (Angers et al. 2011; Feng, Plante, and Six 2013; Wiesmeier et al. 2014) by using the linear regression models (Feng, Plante, and Six 2013), and ignore the influence of soil mineralogy on stable SOC. Feng, Plante, and Six (2013) improved the estimate of $C_{f-max}$ by separating soils into 2:1 and 1:1 clay-mineral-dominated soils, and assessed the individual $C_{f-max}$ by using the boundary line analysis, which is a better method to estimate $C_{f-max}$, because it only uses data representing soils that have reached the maximum organic C concentration of fine particles.

The factors that influence current SOC concentration can affect stable SOC$_{deficit}$. SOC could be regulated by the quantity and quality of C inputs (Maillard and Angers 2014), cropland use (e.g., paddy and upland) (Kögel-Knabner et al. 2010; Yan et al. 2013), and climatic conditions (Davidson and Janssens 2006; Zhang et al. 2010). After decades of improved fertilization management, most agricultural soils in China can still sequester C (Zhang et al. 2010; Feng et al. 2014; Hua et al. 2014), but the amount of additional organic C that can be stabilized under fertilization treatments requires optimization of agricultural management practices for stabilizing the maximum C inputs in cropland soils.

A total of 30 long-term soil fertility experimental sites, representing 5 typical cropland regions in China, were analyzed in the present study: northeast, north, northwest, south upland, and middle and lower Yangtze River. These sites represent different agro-climatic zones, from cold-tempperate to southern subtropics (north–south gradient), and from arid to wet zones (west–east gradient). Thus, this study aimed to (1) compare stable SOC$_{deficit}$ in Chinese croplands after decades of application of different types of fertilizers, in order to identify the fertilization and cropland use that has greater stable SOC$_{deficit}$, and (2) explore the influence of soil texture and climate on stable SOC$_{deficit}$.

Materials and methods

Study sites

This study involved 30 long-term experimental sites across the major agricultural zones in China (Figure 1). These sites covered a wide range of climates, soil types, crop types, crop rotations, and land uses (Table 1) (Jiang et al. 2014; Xu et al. 2015b). Among them, 19 sites were uplands in northern China with soil minerals dominated by 2:1 clay minerals. The climate at these 19 sites varied from arid to semi-humid, and from mild- to warm-temperate, with mean annual temperature (MAT) ranging from −1.5°C in the northeast to 14.8°C in the central region, and mean annual precipitation (MAP) from 150 mm in the northwest to 872 mm in the central eastern region. The remaining 11 sites consisted of upland, paddy-upland, and paddy in southern China, where soil minerals were dominated by 1:1 clay minerals. The MAT of these 11 sites ranged from 15.7°C to 19.5°C, and the MAP ranged from 927 mm to 1553 mm (Xu et al. 2015b).

Fertilization treatments

Fertilization has been widely used to maintain and increase crop yields and SOC sequestration. In recent years, the addition of organic waste and straw return has been widely used in agriculture to maintain soil fertility and increase SOC (Xu et al. 2015a). These fertilization practices increase crop growth and, consequently, C inputs to soil derived from plant residues increase. SOC sequestration in cropland is primarily achieved by the input of organic C originating from manure and compost. In this study, each
experiment comprised treatments aiming to induce different amounts of soil C inputs, including multiple levels of plant residues and manure amendment. According to the type and amount of organic C inputs, we grouped fertilization treatments applied to the 30 study sites into four categories: (1) unfertilized (CK); (2) chemical fertilization (CF), i.e., inorganic nitrogen (N), and/or phosphorus (P) and/or (potassium) K fertilizer combined; (3) crop straw return combined with CF (S + CF); (4) and manure amendment combined with chemical fertilization (M + CF), i.e., livestock or farmyard combined with chemical fertilization. The treatments used are presented in Table S1. In this study, annual fertilizer application rate for different treatments was not shown because the fertilizer treatments, amount of

Table 1. Mean annual precipitation (MAP), mean annual temperature (MAT), soil and clay types, cropping system, land use, and number of years during which the 30 long-term study sites in China’s croplands were fertilized.

| Site No. | Site       | MAP (mm) | MAT (°C) | FAO/UNESCO     | Clay types     | Cropping system^ | Land use^ | Fertilization years |
|---------|------------|----------|----------|----------------|----------------|------------------|-----------|---------------------|
| 1       | Heihe      | 510      | −1.5     | HaplicLuvisol  | 2:1            | MC–WS           | U         | 34                  |
| 2       | Haerbin    | 533      | 3.5      | LuvisPhtaeozems| 2:1            | MC–WSC          | U         | 32                  |
| 3       | Congzhuang | 525      | 4.5      | LuvisPhtaeozems| 2:1            | MC–CCC          | U         | 23                  |
| 4       | Shenyang   | 547      | 7.7      | HaplicCalcisol | 2:1            | MC–CCS          | U         | 34                  |
| 5       | Urumqi     | 310      | 7.7      | HaplicCalcisol | 2:1            | MC–CWU          | U         | 23                  |
| 6       | Pingliang   | 540      | 8.0      | CalcKastanozem | 2:1            | MC–CW           | U         | 34                  |
| 7       | Wuxui      | 150      | 7.7      | Anthrosol      | 2:1            | MC–CW           | U         | 25                  |
| 8       | Tianzhui   | 500      | 11.5     | CalcarCambisol | 2:1            | MC–W            | U         | 29                  |
| 9       | Yangling   | 575      | 13.0     | CumulicAnthrosol| 2:1            | DC–CW           | U         | 23                  |
| 10      | Tianjin    | 607      | 11.6     | CalcarCambisol | 2:1            | DC–CW           | U         | 34                  |
| 11      | Hengshui   | 550      | 13.0     | CalcarCambisol | 2:1            | DC–CW           | U         | 32                  |
| 12      | Yucheng    | 570      | 13.4     | CalcarCambisol | 2:1            | DC–CW           | U         | 23                  |
| 13      | Jinxian1   | 693      | 14.8     | Luvisol        | 2:1            | DC–CW           | U         | 29                  |
| 14      | Jixinan2   | 693      | 14.8     | Luvisol        | 2:1            | DC–CW           | U         | 30                  |
| 15      | Jinxian3   | 693      | 14.8     | Luvisol        | 2:1            | DC–CW           | U         | 23                  |
| 16      | Zhengzhou  | 615      | 14.5     | CalcarCambisol | 2:1            | DC–CW           | U         | 23                  |
| 17      | Xuzhou     | 860      | 14.0     | CalcarCambisol | 2:1            | DC–CW           | U         | 22                  |
| 18      | Mengcheng | 872      | 14.8     | EutricVertisol | 2:1            | DC–CW           | U         | 32                  |
| 19      | Qiyang1    | 1255     | 18.0     | HaplicAcrisol  | 1:1            | DC–CW           | U         | 31                  |
| 20      | Qiyang2    | 1255     | 18.0     | HaplicAcrisol  | 1:1            | DC–RR           | P         | 31                  |
| 21      | Jinxian1   | 1537     | 18.1     | HaplicAcrisol  | 1:1            | DC–CW           | U         | 27                  |
| 22      | Jinxian2   | 1537     | 18.1     | HaplicAcrisol  | 1:1            | DC–RR           | P         | 31                  |
| 23      | Jinxian3   | 1537     | 18.1     | HaplicAcrisol  | 1:1            | DC–RR           | P         | 31                  |
| 24      | Suzhou     | 1100     | 15.7     | HaplicLuvisol  | 1:1            | DC–WR           | P–U       | 32                  |
| 25      | Wuchang    | 1300     | 17.5     | HaplicLuvisol  | 1:1            | DC–WR           | P–U       | 32                  |
| 26      | Chongqing  | 1087     | 18.3     | Regosols       | 1:1            | DC–WR           | P–U       | 23                  |
| 27      | Suining    | 927      | 18.5     | Regosols       | 1:1            | DC–WR           | P–U       | 30                  |
| 28      | Wangcheng  | 1370     | 17.0     | HaplicAcrisol  | 1:1            | DC–RR           | P         | 33                  |
| 29      | Ningxiang  | 1553     | 17.2     | HaplicAcrisol  | 1:1            | DC–RR           | P         | 24                  |
| 30      | Fuzhou     | 1403     | 19.5     | HaplicAcrisol  | 1:1            | DC–RR           | P         | 28                  |

*MC: mono–cropping; DC: double–cropping; WS: wheat/soybean; WSC: wheat/soybean/corn; CCC: corn–corn–corn; CCS: corn–corn–soybean; CWW: com–wheat–wheat; CW: corn–wheat; RR: rice–rice; WR: wheat–rice; U: upland; P–U: paddy and upland; ^Three experiments were performed in Jinxian (13, 14, and 15), two in Qiyang (19 and 20), and three in Jinan (21, 22, and 23).
chemical fertilizer applied, and crop straw and manure usage differed among sites, as did the types of fertilizers and fertilizer application times. Specific information on fertilizer application in the different treatments at each site is available in the work by Xu et al. (2015a, 2015b), Jiang et al. (2014), and Zhang et al. (2016).

**Laboratory analyses**

Soil samples were collected from the topsoil layer (0–20 cm) of the 30 long-term experimental sites from September to November after crop harvest, to analyze the concentration of bulk soil ($C_{bs\text{-cur}}$), soil texture, and soil bulk density (BD), after decades of fertilization practices (Table 1). Although there was no field replication of experimental plots in some sites, the area of each experimental plot was sufficiently large to represent the variation of local soil properties, owing to the following soil sampling procedure described below. Five soil cores (0–20 cm) were collected from random points within a plot and thoroughly mixed as one pseudo replicate; three pseudo replicates were collected from each plot in the sites where replication was not possible. Except for BD, core samples from each plot were mixed thoroughly, air-dried for seven days, and sieved through a 2-mm mesh. A subset of the samples collected from the CK treatment at each site was used for soil texture analysis using the pipette method (Gee and Or 2002), to obtain the mass proportion of fine soil particles (silt and clay particles <20 μm). We measured C concentration in bulk soils from all fertilization treatments (each containing three replicates) in the 30 long-term study sites. A portion of the air-dried bulk soil (<2 mm) was further ground into particles (<250 μm) for organic C content determination, using the vitriol acid–potassium dichromate oxidation method (Walkley and Black 1934).

BD of 188 soil samples was measured for 188 of the 258 treatments by using a steel ring of diameter 5 cm, and soil samples were taken at 10 cm depth, dried at 105°C for 8 h, and weighed (Blake and Hartge 1986). For the remaining 70 treatments, soil BD was estimated based on the regression between the measured BD and the $C_{bs\text{-cur}}$ concentration of the 188 samples (Pan et al. 2004), which was calculated according to the following equation:

$$BD = 1.530 \times e^{-0.011 \times C_{bs\text{-cur}}} \quad (1)$$

where BD is the bulk density (g cm$^{-3}$) and $C_{bs\text{-cur}}$ is the current SOC concentration of bulk soil (g kg$^{-1}$).

**Estimation of C inputs**

C inputs to soils included organic materials from plant residues (i.e., plant roots and stubbles) for all the treatments, additional crop straw return in the S + CF treatment, and manure input in the M + CF treatment. The C inputs from plant roots were estimated as the ratio of plant roots to aboveground biomass, with aboveground biomass determined according to Zhang et al. (2012). The proportion of plant roots to aboveground biomass was considered 30% for wheat, 26% for maize, and 30% for rice (Li, Frohling, and Harriss 1994; Jiang et al. 2014). We assumed that 75.3% of wheat roots and 85.1% of maize roots were in the top 20 cm of the soil (Jiang et al. 2014) and all rice roots were in the top 20 cm of the soil, because most of the roots of rice are in the topsoil layer. Stubble C inputs were estimated using the ratio of stubble biomass to straw biomass, which was 13.1% for fertilized wheat plots and 18.3% for control wheat plots (Jiang et al. 2014), and 3.0% for all maize plots (Xu et al. 2006). For rice, we used the stubble to straw biomass ratios estimated by Zhang et al. (2012). The national averages of C concentration in soils were used for converting biomass inputs to C inputs, which were 399, 444, and 418 g C kg$^{-1}$ for wheat, maize, and rice residues (or straw), respectively (NCATS 1994). Similarly, the national averages of C concentration known for several manures were used to estimate the C inputs from manure application: 414, 368, 360, 336, 135, and 174 g C kg$^{-1}$ for pig, cow, horse, sheep, farmyard manure–soil mixture, and farmyard manure–crop residue mixture, respectively (NCATS 1994). Total organic C inputs from straw return or manure were calculated by multiplying the frequency of straw return or annual manure application rates by the average C concentrations of straw or manure applied at the study sites.

**Calculation of stable SOC$_\text{deficit}$**

The stable SOC$_\text{deficit}$ was estimated as the difference between the $C_{f\text{-max}}$ and $C_{f\text{-cur}}$. Feng, Plante, and Six (2013) classified soils into 2:1 and 1:1 clay-minerals-dominated soils, and estimated $C_{f\text{-max}}$ using boundary line analysis, which is a better method than the linear regression approach for estimating of maximal organic carbon stabilization by fine soil particles (Hassink 1997; Six et al. 2002). Following Feng’s equations, we calculated $C_{f\text{-max}}$ as follows:

$$C_{f\text{-max}} = 0.84x \quad \text{for} \quad 2 : 1 \text{ clay mineral soils, and}$$

$$C_{f\text{-max}} = 0.43x \quad \text{for} \quad 1 : 1 \text{ clay mineral soils,}$$

where $x$ is the mass proportion of fine soil particles (%), $C_{f\text{-max}}$ and $C_{f\text{-cur}}$ is the maximal C concentration and the current C concentration of fine soil particles (g kg$^{-1}$), respectively. We estimated C concentration on fine soil particles as 77% of bulk SOC concentration, as reported by Wiesmeier et al. (2014).

Stable and absolute stable SOC$_\text{deficit}$ were calculated as shown in Equations (2) and (3), respectively:
Stable SOCl deficit (%) = \left( 1 - \frac{C_{bs-cur} \times 0.77}{C_{f-max}} \right) \times 100 \quad (2)

Absolute stable SOCl deficit (Mg ha\(^{-1}\))
= \left( C_{f-cur} - C_{bs-cur} \times 0.77 \right) \times BD \times depth \times 0.1 \quad (3)

where depth is 20 cm, 0.1 is a conversion coefficient, 0.77 is the ratio of \(C_{f-cur}\) to \(C_{bs-cur}\).

**Statistical analysis**

One-way analyses of variance (ANOVA) were used to compare stable SOCl deficit among the several fertilization treatments between soils dominated by 2:1 and 1:1 clay minerals, and to test whether stable SOCl deficit of 1:1 clay-mineral-dominated soils were different among upland, paddy-upland, and paddy, subject to CK, CF, and M + CF treatments. Prior to analysis, residuals were checked for homogeneity of variance and normality to ensure that they satisfied the assumptions of parametric tests. If the data were not normally distributed before ANOVA, we used the nonparametric test to compare stable SOCl deficit among the several fertilization treatments. All statistical analyses were performed in SPSS 20. Significant differences were evaluated using the least significant difference test, considering at the \(p < 0.05\) level.

Figure 1 was prepared in ArcGIS 10.2, using a free map obtained from http://www.cehui8.com/3S/GIS/20130702/205.html. Graphics were prepared using SigmaPlot 12.0.

**Results**

**Stable SOCl deficit in fertilization treatments**

After decades of soil fertilization practices, mean \(C_{f-max}\) in soils dominated by 2:1 clay minerals was significantly higher than that in soils dominated by 1:1 clay minerals (Figure 2(a,b)). In contrast, the averages of \(C_{f-cur}\) in soils dominated by 2:1 clay minerals were lower than soils dominated by 1:1 clay minerals \((p < 0.05, \text{ Table 2})\).

Stable SOCl deficit in soils dominated by 2:1 clay minerals was significantly higher than that in soils dominated by 1:1 clay minerals in all the fertilization treatments \((p < 0.05, \text{ Figure 3})\), indicating that soils dominated by 2:1 clay minerals had greater C sequestration potentials than that of soils dominated by 1:1 clay minerals.

**Table 2.** Current soil organic carbon (SOC) density and absolute stable SOC saturation deficit in soils dominated by 1:1 and 2:1 clay minerals in four treatments.

| Site No. | CK | CF | S + CF | M + CF | CK | CF | S + CF | M + CF |
|---------|----|----|--------|--------|----|----|--------|--------|
| 1       | 37.0 | 38.0 | 37.5 | 36.4 | 95.1 | 94.8 | 93.7 | 96.4 |
| 2       | 27.3 | 29.1 | 30.9 | 97.0 | 95.5 | 92.5 | 87.0 |
| 3       | 20.6 | 20.2 | 21.8 | 103.4 | 97.7 | 93.9 | 70.7 |
| 4       | 14.5 | 16.6 | 13.7 | 78.5 | 76.9 | 74.6 |
| 5       | 16.3 | 20.2 | 22.3 | 94.6 | 92.1 | 93.7 | 86.2 |
| 6       | 17.1 | 18.0 | 21.2 | 117.7 | 115.4 | 112.2 | 108.6 |
| 7       | 23.3 | 25.1 | 27.6 | 30.3 | 88.4 | 80.9 | 79.7 |
| 8       | 23.9 | 21.8 | 25.0 | 117.3 | 98.6 | 90.3 |
| 9       | 17.1 | 20.4 | 26.3 | 147.5 | 140.4 | 123.9 | 120.7 |
| 10      | 24.9 | 24.8 | 36.5 | 95.1 | 84.6 | 78.3 |
| 11      | 13.4 | 18.6 | 17.6 | 104.1 | 111.9 | 108.5 |
| 12      | 16.1 | 20.3 | 27.8 | 99.1 | 92.4 | 80.5 |
| 13      | 20.8 | 21.8 | 25.7 | 42.0 | 40.6 | 35.4 |
| 14      | 17.0 | 18.0 | 22.0 | 46.9 | 45.6 | 40.3 |
| 15      | 12.2 | 15.4 | 21.1 | 53.2 | 49.0 | 41.5 |
| 16      | 14.7 | 15.7 | 22.3 | 54.3 | 50.6 | 43.6 | 41.4 |
| 17      | 11.3 | 13.9 | 24.9 | 64.2 | 61.5 | 50.5 |
| 18      | 12.1 | 19.3 | 24.0 | 125.1 | 120.3 | 100.4 | 87 |
| Average | 23.2 | 38.5 | 105.3 | 108.5 | 80.5 |

CK: control; CF: chemical fertilization; S + CF: crop straw return combined with chemical fertilization; M + CF: manure combined with chemical fertilization.

![Figure 2](image-url) Maximum and current carbon (C) density of fine soil particles in soils dominated by 2:1 (a) and 1:1 (b) clay minerals, within the 30 long-term fertilization experiments in China.
clay minerals. Therefore, the former can sequester more C. Stable SOC deficit of soils dominated by 2:1 clay minerals in the M + CF treatment (69%) was significantly lower than that of soils subjected to the CK (82%), CF (78%), or S + CF (80%) treatments ($p < 0.05$, Figure 3(a)). In soils dominated by 1:1 clay minerals, S + CF (51%) and M + CF (52%), stable SOC deficit was significantly lower than that of soils subjected to the CK (65%) and CF (62%) treatments ($p < 0.05$, Figure 3(b)).

**Effect of management, texture, and climate on stable SOC deficit**

Upland soils dominated by 1:1 clay minerals and in the CK, CF, and M + CF treatments, presented significantly higher stable SOC deficit values (82%, 78%, and 68%, respectively) than that of paddy and paddy-upland soils subjected to the same treatments, whereas paddy-upland soils showed similar levels of stable SOC deficit as that of paddy soils (Figure 4).

Mean annual C inputs in the fertilization treatments varied according to the following order: $\text{CK} < \text{CF} < \text{S + CF} < \text{M + CF}$. In CK, CF, and S + CF treatments, no significant relationship was observed between stable SOC deficit and C inputs (Figure 5(a)), but stable SOC deficit decreased significantly with increasing C inputs in soils dominated by 1:1 clay minerals (Figure 5(b)). In the M + CF treatment, no clear trends of stable SOC deficit in response to C inputs were found in both soil types.

Stable SOC deficit showed dependence on soil texture. It increased with increasing mass proportion of fine soil particles in soils dominated by 2:1 clay minerals under CK, CF, and M + CF treatments (Figure 6(a)), but showed no apparent relationship with the mass proportion of fine soil particles in soils dominated by 1:1 clay minerals (Figure 6(b)).

Climate factors and clay mineralogy influenced stable SOC deficit interactively. In soils dominated by 2:1 clay minerals, stable SOC deficit increased with increasing MAT at temperatures lower than 10°C ($-1.5°C$ to $8.0°C$), particularly in soils subjected to CK and CF treatments. For MAT above 10°C, stable SOC deficit in soils dominated by 2:1 clay minerals showed a strong variation (Figure 7(a)). In soils dominated by 1:1 clay minerals, where MAT was high and within a narrow range ($15.7°C$–$19.5°C$), stable SOC deficit varied greatly among the four fertilization treatments, showing no significant trends according to MAT (Figure 7(b)). In both 2:1 and 1:1 clay-mineral-
dominated soils, stable SOC$_{\text{deficit}}$ displayed no relationship with MAP (Figure 7(c,d)).

**Discussion**

Our results showed that changes in stable SOC$_{\text{deficit}}$ depended on the amount of C inputs and on the current stable SOC$_{\text{deficit}}$ level. Across the 30 studied sites, stable SOC$_{\text{deficit}}$ was significantly lower in the treatments using manure than in the treatments without manure, which might be due to the large organic C inputs in the treatments using manure. A series of field experiments have been demonstrated that a higher annual C addition with manure could increase SOC concentration (Wang et al. 2010; Zhang et al. 2010, 2012, 2016). Straw return provides the greatest SOC sequestration in both current and potential scenarios (Lu et al. 2009). However, we found that the stable SOC$_{\text{deficit}}$ of S+CF was similar to that of CK or inorganic fertilizer treatments (Figure 3), although there was a large amount of straw–C return to soils. To promote C sequestration of stable SOC, straw C conversion efficiency needs to be improved. In addition, the results suggest that, when the values of stable SOC are far from C saturation values (i.e., soil has a large stable SOC$_{\text{deficit}}$), small C inputs do not significantly alter stable SOC$_{\text{deficit}}$. In contrast, when stable SOC values are close to C saturation values (i.e., soil has a small stable SOC$_{\text{deficit}}$), C inputs might remarkably reduce stable SOC$_{\text{deficit}}$ (Figure 5(a,b)). Indeed, the current stable SOC$_{\text{deficit}}$ values of the soils dominated by 1:1 clay minerals were significantly lower than that of soils dominated by 2:1 clay minerals (Figure 3(a)), which could explain the changes in stable SOC$_{\text{deficit}}$ due to C inputs observed in soils dominated by 1:1 clay minerals.

In the present study, different cropland uses led to different stable SOC$_{\text{deficit}}$ values in soils dominated by 1:1 clay minerals (Figure 4(a–c)), suggesting that the factors regulating organo-mineral interactions might affect stable SOC$_{\text{deficit}}$. Mineral availability for C binding critically affects C storage, and mineral availability for further C binding can be affected by management practices (Yu et al. 2017). Yu et al. (2017) found that high organic inputs liberate minerals that can promote C sequestration in soils, and the high concentration of “short-range-ordered” minerals are expected to be key factors that control the storage of soil C. Moreover, similar amounts of C inputs lead to higher C sequestration rates in paddy soils than in
upland soils, which might result from a lower SOC mineralization rate under waterlogged conditions (Yan et al. 2013). The stronger anaerobic conditions in paddy fields than in upland soils suppress microbial activity (Kögel-Knabner et al. 2010; Yan et al. 2013), which might lead to higher C accumulation rates and lower stable SOC deficit in paddy fields than in uplands.

Climate factors, such as temperature and precipitation may modify stable SOC deficit by influencing the decomposition rates of additional C inputs and existing SOC (Carvalhais et al. 2014). In the present study, it is likely that within small MAT ranges (11.5–14.8°C for 2:1 clay-dominated soils and 15.7–19.5°C for 1:1 clay-dominated soils) organic C decomposition rates are similar, and they do not significantly affect soil C accumulation. However, stable SOC deficit of the cropland increased with increasing temperature when MAT was lower than 10°C, thereby suggesting that stable SOC deficit has more temperature sensitivity at low than at high temperatures. Temperature affects the chemical processes of soil organic matter adsorption and desorption onto mineral surfaces, but little is known about the activation energies of these processes (Davidson and Janssens 2006). Huang et al. (2002) found that an increase in temperature within a lower temperature range significantly enhanced decomposition, whereas such an enhancement weakened within a higher temperature range. This observation might be due to the response of SOC decomposition rate of mineral fractions to temperature. In the cropland of southeast Germany, soil SOC deficit was positively correlated with MAT (6–10°C) and negatively correlated with MAP (600–1200 mm) (Wiesmeier et al. 2014), but in the present study a similar relationship was not found for soils dominated by 2:1 and 1:1 clay minerals under each treatment (Figure 7(c,d)). In southern China, where soils are dominated by 1:1 clay minerals, there is abundant rainfall for agricultural use and in northwest China, where precipitation is typically less than 540 mm, irrigation alleviates water shortage effects in the croplands. Therefore, precipitation is not a limiting factor for stable SOC deficit in Chinese croplands after long-term fertilizations.

Soils in the 30 studied sites had a relative stable SOC deficit of 69 ± 0.82%, indicating that current SOC concentrations are far from C saturation levels, despite decades of fertilizers application (Figure 2; Table 2). These results also showed that cropland soils in China have larger C sequestration potentials than agricultural soils in other countries, or soils under other land uses (e.g., forest and grassland) (Angers et al. 2011; Wiesmeier et al. 2014, 2015). Stable SOC deficit of cropland soils was larger in China than in southeast Germany (69% vs. 53%, Wiesmeier et al. 2014). Because the average C concentration of fine soil particles in our study (10.4 g kg⁻¹) was not markedly different from that of southeast Germany (9.6 g kg⁻¹, Wiesmeier et al. 2014), the differences between stable SOC deficit values might be due to the different coefficients used to
estimate C saturation levels of fine soil particles. The stable SOC oversaturation (i.e., higher stable SOC than C saturation) obtained using the Hassink’s equation (Hassink 1997) in coarse-textured agricultural soils in Germany, and in some agricultural soil samples in France seems to indicate an underestimation of the C stabilization potential (Angers et al. 2011; Wiesmeier et al. 2014). In the present study, Chinese cropland soils were classified into soils dominated by 2:1 and 1:1 clay minerals and different coefficients were used in the boundary line method to estimate stable C saturation levels. The coefficients for estimating $C_{f_{-max}}$ of soils dominated by 2:1 clay minerals (0.84) and 1:1 clay minerals (0.43) were larger than the value for German soils (0.37), leading to the estimation of larger C sequestration potentials in Chinese cropland soils than in German soils.

Further, soil mineral type significantly affects the amount of organic C that can be stabilized by fine soil particles (Six et al. 2002). The larger stable SOC$_{\text{deficit}}$ values found in soils dominated by 2:1 clay minerals than in soils dominated by 1:1 clay minerals suggested that the former are able to stabilize more C, and thus the cropland soils in north China dominated by 2:1 clay minerals might be important C stocks. Moreover, similar to Angers et al. (2011), we found that C saturation deficit generally increased with increasing mass proportion of fine soil particles for soils dominated by 2:1 clay minerals (Figure 6(a)). The conversion rate (i.e., the slope of the linear regression between annual rates of SOC sequestration and C inputs) was much higher in northern Chinese upland soils rich in clay than in soils with low clay content (Zhang et al. 2010). In addition, C sequestration has been shown to be much higher in clay-dominated soils than in sandy or silty soils (Matus, Lusk, and Maire 2007). In addition, our study found that soil C stocks significantly increased with increasing mass proportion of fine particles in soils dominated by 2:1 clay minerals (slope = 0.22). However, in these soils, the slope of the linear regression between the mass proportion of fine particles and $C_{f_{-max}}$ (0.84) was much larger than the slope of the regression between the mass proportion of fine particles and $C_{f_{-cur}}$ (0.22), which could explain their increasing SOC$_{\text{deficit}}$ (C sequestration potential) with increasing mass proportion of fine particles (Figure 6(a)).

In this study, SOC$_{\text{deficit}}$ was determined by the C saturation level and C concentration of fine soil particles. Therefore, the method and data source for obtaining the C saturation level and C concentration of fine soil particles are critical. Feng’s boundary line analysis used to estimate the C saturation level of fine soil particles considered different types of clay minerals (Feng, Plante, and Six 2013). The boundary line assessment used the upper tenth percentile of the samples to predict the maximal organic C stabilized on fine soil particles, which is presumed to better represent which samples have reached the maximal level of organic C stabilization. However, this method will possibly lead to some uncertainties, because it depends on the dataset compiled and lacks a mechanistic underpinning (Feng, Plante, and Six 2013). Therefore, a more mechanistic model which takes into account specific surface area and cation exchange capacity associated with soil clay mineralogy for predicting the maximal level of organic C stabilized by fine soil particles should be developed in future studies.

In addition, another source of limitations was that the values of C concentration of the fine soil particles were not measured, but presumed to be constant at 77% of the total SOC, as evident from other studies (Angers et al. 2011; Cao et al. 2016). Using the constant proportion to estimate the values of C concentration of the fine soil particles can be addressed by the following strategies. In each study site, stable SOC ($<20 \mu m$) is known to be primarily affected by soil texture (Six et al. 2002; Feng, Plante, and Six 2013), which is determined by soil parent materials and rarely changed by agricultural management, such as fertilization treatments. Our main goal was to compare the effects of different fertilization treatments on stable SOC$_{\text{deficit}}$ in multiple sites across China. Although the C concentration in the fine soil particles is affected in various ways (e.g., type and intensity of land management, return of crop residues, tillage, fertilization, climate, and erosion) at regional or global scales, we focused on the difference of stable SOC$_{\text{deficit}}$ among fertilization treatments. All sites included the four fertilization treatments (CK, CF, S + CF, and M + CF), except for a few sites. However, assuming that these sites could be compared, the SOC$_{\text{deficit}}$ between different treatments was estimated, and we explored the factors affecting stable SOC$_{\text{deficit}}$ under different fertilizations. Our results are the rough estimation of stable SOC$_{\text{deficit}}$. To obtain the accurate stable SOC$_{\text{deficit}}$ value, more accurate prediction of the maximal level of organic C stabilized by fine soil particles and measurement of the C concentration of fine soil particles is necessary.

**Conclusions**

Cropland soils in China have large stable SOC$_{\text{deficit}}$ values (average 69%) even after 20–35 years of fertilizer application. Stable SOC$_{\text{deficit}}$ is greater in soils dominated by 2:1 clay minerals (69%) than in soils dominated by 1:1 clay minerals (51%), suggesting that soils in northern China can sequester a higher amount of C inputs, because these soils are dominated by 2:1 clay minerals. However, to promote C sequestration and improve stable SOC in northern China, straw–C conversion efficiency needs to be
improved, because stable SOC\textsubscript{deficit} remains large (80\%) despite the large amount of straw returned to the soils. Compared to straw return, manure amendment was more effective for sequestering C in soils dominated by 2:1 clay minerals. In soils dominated by 1:1 clay minerals, stable SOC\textsubscript{deficit} values were small, and small C inputs reduced SOC\textsubscript{deficit} significantly, probably due to paddy land use facilitating organo-mineral interactions and reducing C decomposition. In contrast, soils dominated by 2:1 clay minerals showed increasing stable SOC\textsubscript{deficit} with increasing mass proportion of fine particles and increasing temperature, when MAT was below 10°C. Overall, Chinese croplands have great potential to sequester additional organic C. However, to maximize C sequestration it is necessary to integrate factors such as soil mineralogy, soil texture, fertilizer management, and cropland use.

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