Empirical model predictions and field measurements manifested divergent changes of SOC and SIC in calcareous soils in China

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Background. Considerable amounts of calcareous soils exist in China and various management practices are used for improving their productivity; however, no holistic view is currently available of their effects on soil organic carbon (SOC) and soil inorganic carbon (SIC) pools. Our study aims to define co-current changes of SOC and SIC by using empirical model predictions and reviewing analysis of actual fielddata.

Methods. Three datasets were compiled for the present study; the first was obtained from 9 soil survey reports from China and included data for SOC and SIC concentrations and various soil fertility parameters of soil N and available N; soil P and available P; soil K and available K; and soil pH and cation exchange capacity (CEC). These data were used for empirical prediction of SIC and SOC changes with changes in other soil properties via regression analysis. The second dataset comprised 111 data points from concurrent measurements of SOC and SIC from long-term fixed sites and paired sampling sites (long-term fertilization, tillage treatment, paired land-uses, degraded farmland afforestation etc), which were used to confirm the empirical predication. The third dataset comprised separated measurements of rates of changes in SIC (36 data points) and SOC (74 data points), and frequent distribution and averages were analyzed for finding changing rate differences. These datasets were used to determine the relative magnitude of rates of changes in SIC and SOC to identify the importance of co-inclusion of the two components for soil carbon budget estimation.

Results. Empirical relationships between soil fertility parameters (total N and available N; total P and available P; total K and available K; and pH and CEC) and SOC were generally opposite to relationships between soil fertility parameters and SIC ($p < 0.001$), indicating that soil physicochemical changes as a result of management strategies may affect SOC and SIC in a divergent direction. A total of 111 concurrent measurements of SIC and SOC revealed that soil fertilization and tillage practices could increase SOC andlower SIC by 18% and 11%, respectively, compared to control practices. Similarly, the dataset comprising separated measurements showed that SOC changing rate averaged at 37.3 g m^{-2} yr^{-1}(SOC accrual), and SIC changing rate averaged at -17.1 g m^{-2} yr^{-1}(SIC loss), counteracting the SOC accumulation.

Discussion. Changes in SIC are more complicated than those of SOC. In a semiarid region with abundant CO_2 and Ca^{2+}, pedogenic formation of SIC was observed, while in a moist region (such as a karst land region) with sufficient water supply, dissolution-induced SIC loss in surface soils was frequently observed. Our findings highlight that SOC and SIC should be simultaneously included in the computation of soil carbon budgets to avoid false estimation of carbon changes as a result of using either SOC or SIC alone.

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Empirical Model Predictions and Field Measurements Manifested Divergent Changes of SOC and SIC in Calcareous Soils in China

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Abstract

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Keywords: soil physicochemical properties, soil organic carbon, soil inorganic carbon, soil carbon sequestration and depletion

Introduction

Although a better estimate of regional and global soil organic carbon (SOC) stocks is now becoming available (Lal, 2004, Lorenz et al., 2011), this is not the case for soil inorganic carbon (SIC) (Mi et al., 2008; Zamanian et al., 2016). In China, soils with SIC cover about 3.44 x 10^6 km^2 of China (Mi et al., 2008; Wu et al., 2009). Hence, SIC may have played an important contribution to C balance (Feng et al., 2002; Yang et al., 2012; Wu et al., 2003, 2009), and concurrent study of SIC and SOC dynamics can give a more holistic view of soil carbon (C) balances (Wang et al., 2012; Xu et al., 2012; Zhang et al., 2013), particularly in the wide semiarid region of calcareous soil (Yang et al., 2016; Li et al., 2016). However, some basic questions are still waiting for exact answers, e.g., did inclusion of both SIC and SOC over- or under-estimate the C budget compared with SOC alone, and by how much percentage? The importance of both SOC and SIC inclusion in carbon budget calculation can be evaluated by a systematic response to these questions.

The most important soil management strategy in China is the use of various fertilization practices including chemical addition and biological rehabilitation for securing food production (Song et al., 2007; Sun & Suo, 2011; Wang et al., 2011; Wei et al., 2012). China has ranked as the top consumer of N fertilizer. Consumption of K fertilizer is as high as 4~5.5 M ton K_2O, while utilization of P fertilizer is as high as 11 M ton, surpassing the per hectare anthropogenic fertilizer additions in the United States and Northern Europe (Guo et al., 2010) by far. This overuse of fertilizers has maintained crop production (Wang & Wang, 2006; Song et al., 2007; Suo & Han, 2009; Sun & Suo, 2011), but also resulted in soil degradation and soil acidification (Guo et al., 2010). Furthermore, returning farmland to forest or grassland is a national policy to rehabilitate the environment and soil erosion, and implementation of this policy over 10 years has improved soil fertility and soil quality (Wang et al., 2011; Wei et al., 2012; Wang et al., 2014a). In addition to soil physiochemical changes, a lot of papers, books, and thesis papers have focused on the changes of SOC, SIC, or both (as listed in references). A meta-data analysis of these reference data can provide a holistic view of soil C balance as affected by soil management practices in calcareous soils of China.

Regression analysis for soil C content and soil fertility could help in predicting the consequences of soil carbon changes due to intensive cultivation practices (Zu et al., 2011), while field data of actual measurements of SIC and SOC are useful in verifying any empirical predictions (Li et al., 2016; Yang et al., 2016). A divergent response of SIC and SOC to soil fertility alternation indicates that any single component may overestimate the soil carbon changes owing to human activity, while a consistent response of SIC and SOC to soil fertility changes means that any single component may underestimate the size of C sink or source (Zu et al., 2011). Analysis of data from previous concurrent measurements and separated measurements (as shown in reference lists) could help to verify the empirical predictions from linear regression models, and define the importance of SIC and SOC inclusion in soil C budget estimation.

In this paper, we hypothesize that the inclusion of SOC or SIC alone in soil C budget estimation can result in a large bias in soil C studies in calcareous soils in China, and the significance of this bias may be demonstrated via empirical relationships and field datasets from concurrent or separated measurements of SOC and SIC in well-designed experiments.

Materials & Methods

Data collection

The data used for regression analysis in this paper were obtained from nine soil survey reports with calcareous soils, including Soil of Heilongjiang Province (HLJTR editorial committee, 1992), Soil of Jilin Provinces (JLTR editorial committee, 1992), Soil of Liaoning Province (Jia, 1992), Soil of Inner Mongolia (Wang et al., 1994), Soil of Shandong Province (Yan et al., 1994), Agricultural Soil of Shaanxi Province (ASDIS, 1982), Soil of Henan Province (Wei, 1979), Soil of Qinghai Province (ARZOQ, 1997), and Soil of China (Institute of Soil Science CAS, 1978). The dataset includes SOC concentration, SIC concentration, and variable soil fertility parameters (soil N and available N, soil P and available P, soil K and available K, soil pH, and cation exchange capacity (CEC)). All soils, including loess soil, chestnut soil, dark brown earth soil, brown earth soil, gray earth soil, and chernozem soil, etc., are typical calcareous soils.

Field data included two datasets. One dataset was concurrent measurement data (111 data) of SOC and SIC from long-term fixed sites and paired sampling sites (Huang et al., 2006; Jin, 2006; Yang et al., 2007; Geng et al. 2008; Li, 2008; Zeng et al., 2008; An et al. 2012a; An et al. 2012b). In this dataset, concentration or storage of SIC and SOC under variable treatments (chemical fertilizer addition, straw covering and return, manure addition, returning farmland to forest or grassland, etc.) and control were compiled. Considering that most studies have measured SIC or SOC separately, another dataset was compiled based on these separated measurements. SIC data (36 data) included leaching rates of pedogenic carbonates in surface soils and accumulation rates of lithogenic carbonates in sub-surface soils (Sheng & Wang, 1989; Yuan, 1994; Liu & Dreybrodt, 1998; Pan, 1999; Duan et al., 1999; Wang et al., 2012). SOC data (74 data) included the changing rate of SOC during chemical fertilizer addition, straw covering and return, manure addition, and returning farmland to forest practices (Ma et al., 1994; Zhu et al., 1995; Zhang et al., 2003; Meng et al., 2005; Yin & Cai, 2006; Zhang et al., 2006; Wang & Wang, 2006; Zhou et al., 2006; Li, 2007; Song et al. 2007; Guo et al., 2009; Suo & Han, 2009; Sun & Suo, 2011; Wang et al., 2011). The changing rates of SIC and SOC were recalculated as g C m^{-2} yr^{-1}. For the data without soil bulk density, the soil bulk density was calculated using their SOC concentration by using the following equation, $\gamma = 1.3770 \times e^{-0.0048 \times SOC}$(Song et al. 2005).

Furthermore, all SOC data in references are in unit of C, while all SIC data in references are in unit of $CaCO_3$. For inter-comparison, the SIC was transformed from $CaCO_3$ to C by a factor of 0.12 (molecular weight for $CaCO_3$ is 100, while that of C is 12). Although some of the C may still be in soil in the forms of CO_3^{2-} or HCO_3^-, this transformation into the unit of C should be feasible because the turnover time of soil CO_3^{2-} or HCO_3^- is much shorter than the long turnover time of SIC (about 85 000 years) (Lal & Kimble, 2000; Ding et al., 2010).

Data analysis

Linear regression analyses among SOC, SIC, and soil fertility parameters (soil N and available N, soil P and available P, soil K and available K, soil pH, and CEC) were performed. Significant slope values were used to describe the rate of change in SOC or SIC with changes in soil fertility. If slopes for SOC are opposite to those for SIC, they may indicate that soil physicochemical changes may contrarily affect SOC and SIC (i.e., inducing SOC accumulation accompany SIC loss, or *vice versa*), and the inclusion of both components could moderate the C sink or source intensity. On the other hand, when the slopes for SOC are in the same direction with those of SIC, they indicate that soil physicochemical changes may consistently affect SOC and SIC, and therefore the inclusion of both parameters may increase the sink or source size of C.

Multivariate analysis of covariance (MANCOVA) was used to check the homogeneity of the linear regression slope among SIC, SOC, and variable soil parameters. The fixed factor of MANCOVA was The C form (SIC or SOC), the dependent variables were soil parameters, and the covariate was the concentration of soil C (SIC or SOC). Detailed description of this analysis method can be found in Wang et al. (2014a).

In the case of the external reference datasets of concurrent measurements of SIC and SOC, the ratios under various management practices (long term fertilization, tillage treatment, afforestation treatment, etc. as shown in Table S1 and Table S3) to their control treatments were used to describe the effects of management practices and various treatments on SOC and SIC. Frequency distribution and average of these ratios were performed. When the ratio is higher than 1, these treatments could increase soil C; when the ratio is less than 1, these treatments could decrease soil C. The relationships between soil depth and SOC or SIC changes with reference to control treatments were also checked by regression analysis between average soil depth and the ratio between treatments and controls. A significant increase in the ratio of soil depth indicates that treatment-induced changes of SIC or SOC increased with soil depth, while a non-significant increase in the ratio of soil depth indicates treatment-induced changes decreased with soil depth.

The datasets of separated measurement of rates of SIC and SOC changes were also analyzed by frequency distribution and average analysis. Given that similar rates of changes in SOC and SIC accumulation or depletion were observed, inclusion of SOC and SIC into soil carbon budgets could overestimate rates of change; or else, when rates of changes in SOC and SIC were contrary each other, inclusion of SOC and SIC could underestimate the rates of change. The effect of SIC inclusion in C budget estimation was calculated by using linear regression analysis, concurrent measurement dataset, and separated measurement datasets by assuming a rate of change of SOC alone of 100%.

Linear regression analysis and all statistical analyses were performed using JMP 5.0 (SAS, USA) and SPSS 17.0 (SPSS, USA).

Results

Linear gradients of nutrient-carbon relationships manifest the tradeoff between SIC and SOC

Relationships among SIC, SOC, total N, and available N were significant at the $p < 0.0001$ (Table 1, Fig. S1). The relationships between SIC concentration and total soil N and available N were negative, while the relationships between SOC and total soil N and available N were positive. The rates of change represented by the regression gradients show that SIC concentration decreased by 1.249 g for 1 g kg^{-1} increase in total soil N, while the same rate of increase in soil N was equivalent to 12.97 g SOC increase.

All the correlations between SIC or SOC and soil P or available P were statistically significant ($p < 0.01$) (Table 1, Fig. S2). The relationships between SIC and available P were opposite to those of SOC, i.e., positive correlations between SOC and available P were found (slope >0), while negative relationships were found in SIC (slope <0). However, similar positive correlations between SIC or SOC and soil P were found (Fig. S2).

The relationships between SIC and total K were positive and significant (slope >0, $p < 0.01$), and the rates of change represented by the regression gradients show that SIC concentration increased by 0.232 g for 1 g kg^{-1} increase in total soil K (Table 1, Fig. S3). The corresponding relationship between SOC and total K was negative but not statistically significant. Similarly, the relationships between SOC and available K were positive and significant ($p < 0.0001$), and the rate of change represented by the regression gradients was 0.0646 g kg^{-1} increase for 1 mg kg^{-1} increase in available K (Table 1).

All relationships between SIC or SOC and soil pH or CEC were significant ($p < 0.0001$) (Table 1, Fig. S4). However, SIC-relationships were generally contrary to those of SOC, i.e., positive correlations between SIC and pH and SOC and CEC were observed, while those of SOC were negative. The rate of change represented by the regression gradients showed that SIC concentration increased by 4.80 g kg^{-1} for 1 unit increase in soil pH, while the same pH change was equivalent to 5.72 g kg^{-1} decrease in SOC. Similarly, increases of 1 cmol kg^{-1} in CEC could produce 0.205 g kg^{-1} decrease in SIC and 1.35 g kg^{-1} increase in SOC.

The empirical offsetting effects between SIC and SOC were analyzed by using the rate of changes represented by the regression gradients (Fig. 1). Among all 8 pairs of correlations between SIC or SOC and variable soil fertility parameters, seven of them showed opposite linear gradients. Taking the SOC changing rate at 100%, the offsetting effects ranged from -7% to -84% with an exceptional high value in total K of -314%. For N, the offsetting effects ranged from -10% to -18%. For soil pH and CEC, the offsetting effects ranged from -15% to -84%. For available K and available P, the offsetting effects ranged from -7% to -23%. These contrasting gradients showed the changes in SOC levels in soil can be offset by changes in SIC levels, diminishing the change in total soil C (SIC + SOC) (Fig. 1).

Stepwise regression analysis also showed different responses of SIC and SOC to soil physicochemical changes (Table 2). The magnitudes of standardized coefficients of stepwise regression can manifest the impact of soil fertility (x) on SOC or SIC (y). We observed negative relationships between pH, total P, total K, available P, CEC, and SOC, while positive relationships were found in the corresponding for SIC (Table 2). Of all significant coefficients, SOC changes were mainly related with total N, available N, and available P ($p < 0.05$), while the SIC changes were mainly related with pH, total P, and available N ($p < 0.05$).

Field concurrent measurements manifest the opposite changes between SOC and SIC

Based on an external paired measurement dataset of designed experiments (long-term fertilization, different tillage treatment, paired land uses), changes in SIC and SOC were crosschecked here (Fig. 2; Table S1). In the case of SIC, 51% of the 111 data were distributed ranging from 0.9 to 1.0, and 85% of the data showed that the ratio was lower than 1.0. Thus, variable management practices could decrease SIC in most cases. In the case of SOC, 67% of the data were distributed ranging from 1.0 to 1.3 (Fig. 2), indicating that variable treatments increased SOC concentration in most cases. In the case of SIC + SOC, 28% of the data showed ratios lower than 1.0, and the peak distribution region was 1.0-1.1 (37%). On average, variable management practices increased SOC content by 18% compared with the control, while these management practices decreased SIC content by 11%. The contrasting changes between SIC and SOC significantly neutralized the changes in total soil C (SIC + SOC) (3% increases) (Fig. 2).

The relationships between the ratio (between treatment and control for SOC or SIC) and soil depth were also checked (Fig. 3). With the depth deepening, the SIC ratio between treatment and control decreased ($r^2 = 0.1866$), while the SOC ratio increased ($r^2 = 0.3436$) (Fig. 3). This opposite change resulted in a moderated decrease of SIC + SOC ratio between treatment and control, indicating that the C sink (SIC + SOC) from various soil management practices may change to C source when deeper soil is included in soil C budget calculation (slope = -0.0024, $r^2 = 0.3309$) (Fig. 3).

Separated measurement datasets manifest contrasting changes between SOC and SIC

In the study of soil C balance, there were more separated measurements on the changes of SOC or SIC compared with the concurrent measurements of both components. Therefore, we also pooled the separated data for frequency and average analyses to check their offsetting effects between the two components (Fig. 4, Tables S2 and S3).

As shown in Fig. 4, 83% of the SOC data showed SOC accumulation (positive changing rate), and the most frequent region was from 0 to 30 g m^{-2} yr^{-1} (45%). The mean value of these data was 37.3 g m^{-2} yr^{-1} (Fig. 4). In the case of SIC, over 60% of the data showed SIC loss (negative changing rate) averaged at -29.4 g m^{-2} yr^{-1}, while the other 39% showed accumulation of SIC with an average of 2.2 g m^{-2} yr^{-1}. The most frequent region for SIC changes ranged from -10 to 0 g m^{-2} yr^{-1}. The mean value of these data was -17.1 g m^{-2} yr^{-1} (Fig. 4). Thus, part of the SOC accumulation were counteracted by the SIC depletion in soil (Fig. 4).

Comprehensive comparison of the offsetting effects

Linear regression analysis, field concurrent measurements, and separated measurements showed that inclusion of SIC and SOC into soil carbon budget calculation will counteract the size of carbon budgets estimated by SOC alone.

Fig. 5 shows average offsetting percentage assuming the change of SOC as 100%. Predication from linear regression gradients between SIC, SOC, and variable soil fertility parameters showed that the inclusion of SIC could moderate 51.5% of SOC accumulation, i.e., C change calculated using SOC alone was 2-fold greater than that calculated using SOC and SIC combined. The concurrent measurements showed the offsetting percentage of SIC of 73.8%, while the separated measurements showed a 45% offsetting percentage. Therefore, inclusion of SIC into carbon budget estimation could offset 56.8% of the SOC changes, and the C change calculated from SOC + SIC was nearly 40% of that calculated from SOC alone, on average of 3 independent datasets (Fig. 5).

Discussion

Higher amounts of SOC and SIC have always been recognized as a major sink of the global C pool (Lal & Kimble, 2000; Schlesinger, 2002; Mi et al., 2008; Zamanian et al., 2016). In arid and semiarid regions that cover as much as one-third of the surface of the planet, the SIC pool is approximately two to ten times larger than the SOC stock (Post et al., 1982; Schlesinger, 1982; Eswaran et al., 2000). In China, near half of soil subgroups contain SIC of pedogenic and lithogenic carbonates, and cover 30%-40% of the total land area (Mi et al., 2008; Wu et al., 2009). Although studies have highlighted the potential role of SIC in soil C sequestration (Mi et al., 2008; Wu et al., 2009), few studies have concurrently considered SIC and SOC dynamics (Wang et al., 2012) and different measurements tended to lead to different conclusions (Zu et al., 2010). In this paper, this topic is assessed by using empirical prediction using regression analysis and verification using field data observation.

Opposite changes of SIC and SOC during soil management: overestimation of C changes by one component alone

Contrasting relationships between soil fertility parameters, SOC, and SIC (Figs. S1-S4) clearly indicate that soil fertility alterations can result in divergent changes in SOC and SIC (Table 1, Fig. 1). Similar contrasting relationships have been reported by previous studies. Using a data-reanalysis, Zu et al. (2012) observed that soil fertilizer addition for crop production may increase SOC but decrease SIC at the same time. A chronosequence measurement manifested that larch forest growth could increase SOC at the expense of SIC loss (Wang et al., 2013). Pan et al. (1999) observed significant negative relationships between SOC and SIC, and the data in this paper also showed a negative logarithmic relationship between SOC and SIC (SOC = -6.679ln (SIC) +38.874, $r^2 = 0.2037$, $p < 0.001$, Fig. 6). Changes in SIC and SOC may variably affect soil physicochemical properties, i.e., SIC mainly influences soil acidity (pH), while SOC mainly affects soil fertility parameters of N and P (Wang et al., 2013). Stepwise regression in this paper also supported these findings (Table 2). However, the auto-correlations among soil fertility parameters were not excluded in the regression analysis. Thus, so the divergent relationships between SIC, SOC, and soil properties of N, P, K, pH, and CEC (Table 1, Fig. 1, and Figs. S1-S4) were just empirical model for predicating soil C changes with soil fertility (without any hint for their mechanic relationships).

Opposite changes in SIC and SOC determined by using linear regression analysis should be cross-checked by well-designed field measurements for making a reliable conclusion. Present study found that both field concurrent measurements and separated measurements strongly support the tradeoff effects between SIC and SOC (Fig. 2, Table S2). Concurrent measurements of SOC and SIC showed that soil treatments have improved SOC sinks by about 18%, while the reduction in SIC (by 89% compared with the control) significantly counteracted the sink size of total C in soil (3% increase) (Fig. 2). Similar to the concurrent measurements, separated measurements of SOC and SIC also showed that fertilization and tillage practices could improve SOC accumulation with an average rate of 37.3 g m^{-2} yr^{-1} (Fig. 4), and the increase in SOC is due to the fact that fertilization practices could return more organic C from roots, litter, and organic manure to soils (Meng et al., 2005; Yin & Cai, 2006; Zhang et al., 2006; Pan et al., 2006; Wei et al., 2012). Counteracting depletion of SIC averaged at a rate of 17.1 g m^{-2} yr^{-1} (Fig. 4). Recent studies showed similar patterns of SIC and SOC changes. For example, in the upper Yellow River Delta of North China Plain, mean SOC content decreased from 9.30 g kg^{-1} near the surface to 2.36 g kg^{-1} in 80-100 cm soil layer, whereas mean SIC content increased from 10.48 to 12.72 g kg^{-1}, with significant positive relationships between SIC and Ca^{2+} or Mg^{2+} ($p < 0.05$) (Yang et al., 2016). In Northwest China, the natural alpine grassland had the highest SOC (96.0 Mg C ha^{-1}) and lowest SIC (19.8 Mg C ha^{-1}) stocks, while the farmland showed the opposite result, with average of SOC and SIC stocks of 65.0 and 57.7 Mg C ha^{-1}, respectively. Moreover, after 10 years of restoration, SOC stock increased to 73.2 Mg C ha^{-1}, while SIC decreased to 47.8 Mg C ha^{-1} in the restored farmland (Li et al., 2016). In the Songnen Plain, Northeast China, grassland reclamation to farmland decreased SOC at a rate of 3.63 Mg C ha^{-1} yr^{-1}, with SIC stock increasing at a rate of 0.53 Mg C ha^{-1} yr^{-1}, underlining the importance of including SIC and SOC in soil C budget estimations (Yu et al., 2014).

Complexity of SIC dynamics: concurrent observation of C deposition and leaching

Compared with SOC, the change of SIC in soil is more complicated during inclusion in C budget estimation (Wohlfahrt et al., 2008; Xie et al., 2008; Yang et al., 2012); one reason is that SIC is depleted during soil managing practices. Approximately 51% of the total cultivated soil surface in China has experienced SIC loss (Wu et al., 2009), and leaching is the main pathway of SIC loss in China and takes place in any region with calcicolous soil and carbonate rocks (Yuan, 1994; Liu & Dreybrodt, 1998; Duan et al., 1999; Pan, 1999; Wang et al., 2012). The SIC lost due to carbonate leaching during weathering processes may be released to the atmosphere through reactions with additional inputs of hydrogen ions from acid deposition (Yang et al., 2012), fertilizer N additions (Havlin et al., 2005; Guo et al., 2010), and organic acids released from roots and litter decomposition. Plant Ca absorption can be increased by increasing NO_3^-, but can be depressed by NH_4^+ and other cations of K^+, Mg^+, Mn^{2+}, and Al^{2+} (Havlin et al., 2005). Absorption of Ca in the biomass might be an important biological process promoting SIC depletion. In the calcareous soils, biomass productivities are 3.2 ton ha^{-1} yr^{-1} for grassland (Ma et al., 2010), 12.0 ton ha^{-1} yr^{-1} for various forests (Fang et al., 1996), and 8.3~18.5 ton ha^{-1} yr^{-1} for crops such as rice (Wang et al., 2008), maize (Yan et al., 2013), soybean (Bi et al., 1999), and wheat (Zhu et al., 2011). By using the average of these data (10.4 ton ha^{-1} yr^{-1}) and averaging Ca concentration in different plants (0.8% to 10.4% with a mean 4.1%) (Zhou, 1997; Ji et al., 2009; Wang et al., 2011), the absorption of Ca by biomass stands accounts for a SIC loss of 12.8 g m^{-2} yr^{-1} given that all Ca^{2+} in biomass is from calcite leaches (Table S4).

The second complexity for SIC dynamics is the secondary SIC deposition via pedogenic carbonate re-formation in soils (Fig. 4). In China, this kind of pedogenic deposition region is observed in about 10% of total farmland area and is mainly distributed in the irrigated soils in northwestern China (Wu et al., 2009). Carbonate ions are mainly derived from root and microbial respiration, while calcium and magnesium ions are usually derived from weathering reactions (Lal & Kimble, 2000) or from atmospheric deposition, irrigation water, or fertilization (Schlesinger et al., 2009; Wang et al., 2014b). The process of carbonate deposition functions to re-capture C in the soil system (Wohlfahrt et al., 2008; Xie et al., 2008), and the important premise is the abundant Ca^{2+} or Mg^{2+} together with enough biological CO_3^{2+}. In this paper, pooled separated measurements gave approximate estimates of C changes, i.e., the loss rate averaged at -29.4 g m^{-2} yr^{-1}, while the secondary SIC deposition rate (accumulation) averaged at 2.2 g m^{-2} yr^{-1} (Table S2; Fig. 4). The SIC secondary deposition was much lower than the loss due to leaching, which resulted in the net loss of SIC (Fig. 4). Although the dissolution of carbonates may absorb CO_2 from the atmosphere via the reaction of $CaCO_3 + H_2O + CO_2 = Ca^{2+} + 2HCO_3^{-1}$ (Liu & Dreybrodt, 1998; Xie et al., 2008), the turnover time decreases from thousands of years for calcite SIC (Lal & Kimble, 2000) to month-levels for soil CO_2 solution (HCO_3^{-1} and CO_3^{2-}) (Ding et al., 2010), which may sharply diminish the C sink size and efficiency. Thus, the net loss of SIC can effectively counteract the accumulation of SOC owing to soil fertilization and tillage practices (Fig. 4).

Soils comprise the largest terrestrial carbon pool of organic and inorganic C, with SIC frequently neglected. Reasons are that SIC is partly derived from soil parent material, with very slow formation processes as well as slow exchange with atmospheric CO_2. However, recent evidence highlighted the global importance of SIC owing to its links with the long-term geological C cycle with the fast-biotic C cycle (Zamanian et al., 2016). At present, little attention has been paid to the role of SOC as well as management of soil in the formation of pedogenic carbonates of SIC, and our result provide hints on this. SOC accumulation and SIC depletion caused by various management practices could extend to 80 cm soil depth (Fig. 3). The SIC deposition or loss through leaching should be related with climatic data of rainfall, and distinct differences have been reported in arid and semi-arid regions (Su et al., 2010; Wang et al., 2004) and in other regions with enough rainfall (Yuan, 1994; Liu et al., 1998; Pan, 1999). The traditional view of SIC deposition is generally in deeper soil layers in the occasion of water deficiency (Sheng and Wang, 1989; Duan et al., 1999; Pan, 1999). In this paper, most of the data are from Hebei, Beijing, Shaanxi, Shanxi, Heilongjiang, and Gansu (irrigated soil), etc. (Table S1), which usually have a humid climate with >400 mm rainfall. SIC deposition in this region may be deeper than 80 cm. Extra-calcium additions from fertilizers, irrigation, and atmospheric deposition may markedly increase SIC deposition (Wang et al., 2014b), indicating that intensive farming practices (as shown in Table S1) may also strengthen the observation of SOC and SIC dynamics in a vertical profile (Fig. 3). Furthermore, soil dehydrogenase activity (DHA) and $CaCO_3$ positively correlated with SOC content, indicating that abundant OM content contributed to the pedogenic formation of $CaCO_3$ in desert soil (Zhang et al., 2010). Isotope techniques are often used to differentiate between pedogenic carbonate and lithogenic carbonate because of their distinct isotopic signatures (Wang et al., 2014b). Pedogenic carbonate formation in the USA is 0.12 to 0.42 g C m^{-2} yr^{-1} (Schlesinger et al., 2009). More detailed studies of SOC and SIC in combination with enzymatic data, isotope data, and weather data may improve our understanding of their relationships (Yuan, 1994; Zhang et al., 2010; Yang et al., 2012; Wang et al., 2014b).

Implications for soil carbon sequestration evaluation under soil management practices

In China, to meet the increasing food demand to feed 22% of the world's population from limited arable farmland (about 7% of world farmland), intensive management practices of these lands have long been practiced (e.g., chemical fertilizer and manure addition) for improving productivity of the soil (Song et al., 2007; Sun & Suo, 2011). Recently, some other measures, such as straw returning to field, non-tillage cropping systems, and returning farmland to forest or pasture, have been advocated and practiced for soil quality recovery, soil erosion control, and ecological rehabilitation (Guo et al., 2010; Wang et al., 2011) (also as shown in Table S1 and Table S3).

At present, annual C change owing to these intensive management practices at national level is a hot topic and some studies have been carried out (Piao et al., 2009). However, no studies have considered the SIC changes in combination with SOC in a large-scale survey. Thus, an estimate of the offsetting effect resulting from SIC inclusion compared with SOC inclusion alone is useful. Here, the offsetting percentage was defined by linear regression prediction, concurrent measurements, and separated measurements (from 3 independent datasets) (Fig. 5). The offsetting percentage ranged from 45.0% to 73.8%, and averaged at 56.8% (Fig. 5). This numeration is important for re-evaluation of the estimation of regional C balances. For example, the C balance of terrestrial ecosystems in China has been estimated as net C sink of 0.19–0.26 Pg C yr^{-1} by Piao et al. (2009) with inclusion of SOC accumulation rate (0.069 Pg C yr^{-1}). By including SIC as described in this article, the SIC + SOC accumulation rate will decrease to 0.030 Pg C yr^{-1}. This offsetting effect resulting from SIC changes (0.039 Pg C yr^{-1}) could 15%~21% decrease the net C sink of whole China terrestrial ecosystems, accordingly. Being an abundant component of soil carbon stocks in China, SIC dynamics and the processes involved in its accumulation or loss from soils require a better understanding; our study may provide a possible insight of the interaction between SOC and SIC changes.

In future studies, quantification of the sources of uncertainty in studies of SIC need to be informed. For example, in gravelly sites with more calcitrates, a portion of soil greater than 2 mm may store a large percentage of SIC. As shown by examination of 26 soil sites, an average of 13% of the total SIC is stored as carbonate coasts within in the gravel fraction (Stanbery, 2016). The compensation relationship between SOC and SIC stocks with land-use and soil fertilization treatments suggested that the changes of soil properties and plant above- and below-ground biomass resulting from cultivation and restoration were potentially responsible for the transformation of soil C forms (Li et al., 2016). Pedogenic carbonates are an important part of SIC that reflect the time periods and formation processes in soils, and greater accumulation of pedogenic carbonate (21–49 g C m^{-2} year^{-1}) than SOC (10–39 g C m^{-2} year^{-1}) over 0–20 cm soil depth exceeding organic C has been reported (Zhang et al., 2013). Thus, the most important future research directions on pedogenic carbonates should include the anthropogenic effects of fertilization, soil management, and land use changes (Zamanian et al., 2016), particularly with special attentions to precipitation of the region, the abundance of soil Ca^{2+} and isotopic mechanism clarifications.

Conclusion

The opposite linear correlations between SIC or SOC and variable soil parameters indicates that soil physicochemical changes caused by soil management practices may divergently affect SOC and SIC in calculation of total soil C (SIC + SOC) budgets. Field data both from concurrent measurements and separated measurements in China strongly support this conclusion. On average, the offsetting percentage was 56.8%. Thus, SOC and SIC should be simultaneously included in assessments of soil carbon budgets, and that reliance on any one component alone may overestimate the sizes of C sinks or sources.

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Figure 1

Changing rates of SIC, SOC responding to soil N, P, K, pH and CEC estimated from linear regression analysis.

# indicates the gradient is from a non-statistical significant linear correlations ($p>0.05$).

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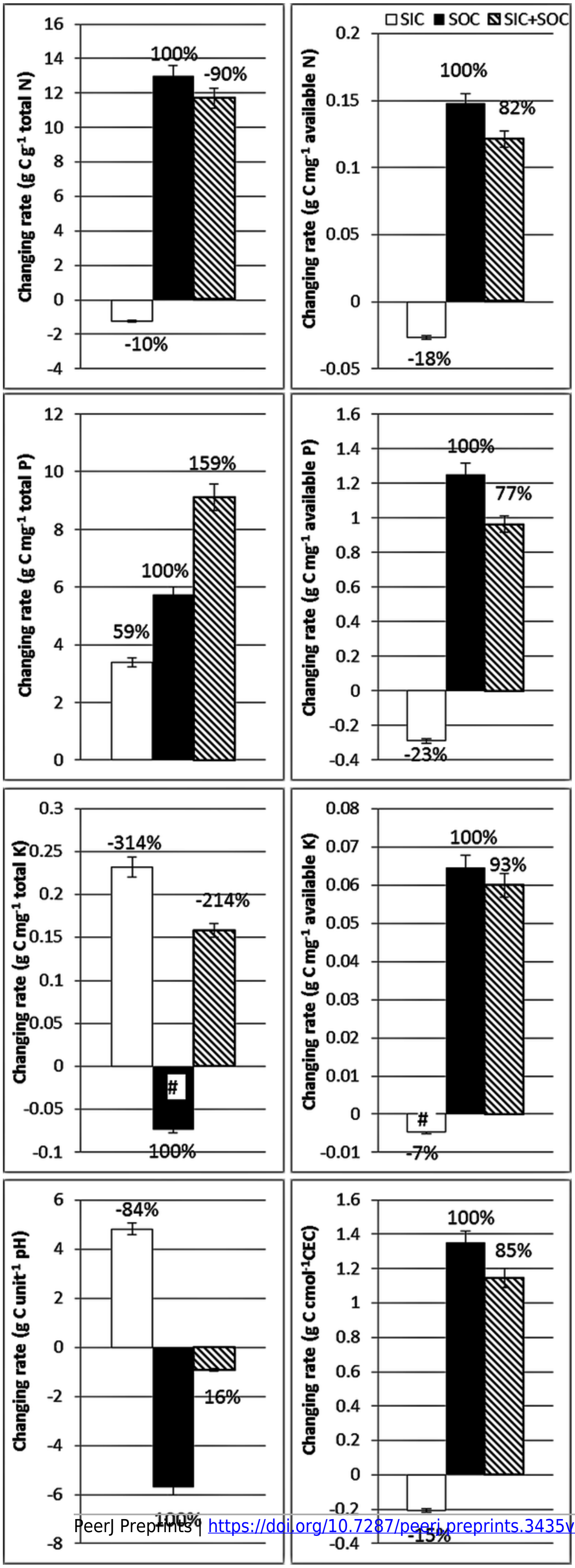
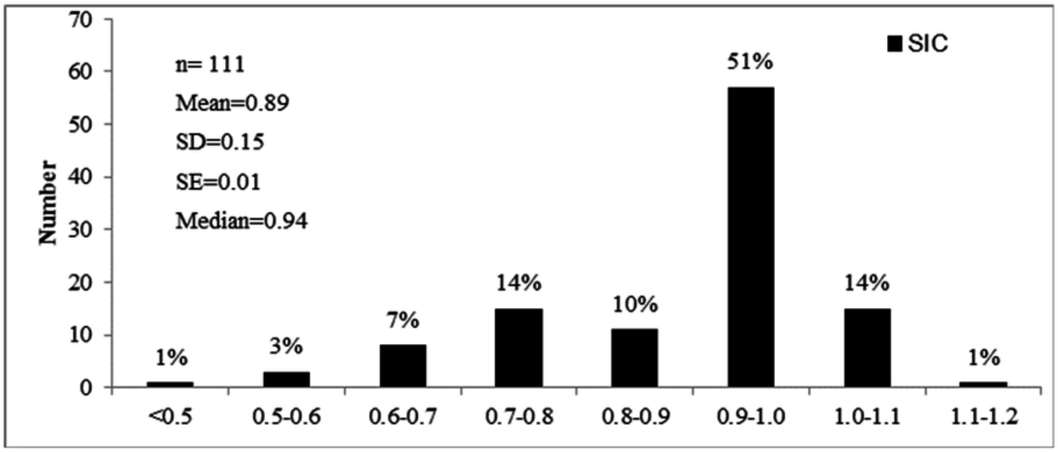
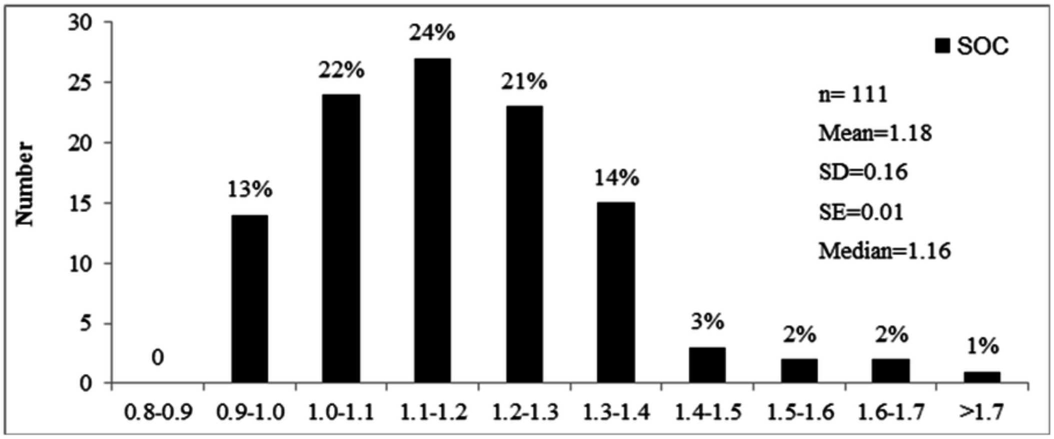

☐ SIC ■ SOC ▨ SIC+SOC
Changing rate (g C g-1 total N)
16
14
12
10
8
6
4
2
0
-2
-4
100%
-90%
-10%
Changing rate (g C mg-1 available N)
0.2
0.15
0.1
0.05
0
-0.05
100%
82%
-18%
Changing rate (g C mg-1 total P)
12
10
8
6
4
2
0
159%
100%
59%
Changing rate (g C mg-1 available P)
1.6
1.4
1.2
1
0.8
0.6
0.4
0.2
0
-0.2
-0.4
100%
77%
-23%
Changing rate (g C mg-1 total K)
0.3
0.25
0.2
0.15
0.1
0.05
0
-0.05
-0.1
-314%
-214%
#
100%
Changing rate (g C mg-1 available K)
0.08
0.07
0.06
0.05
0.04
0.03
0.02
0.01
0
-0.01
100%
93%
#
-7%
Changing rate (g C unit-1 pH)
6
4
2
0
-2
-4
-6
-8
-84%
16%
100%
Changing rate (g C cmol-1CEC)
1.6
1.4
1.2
1
0.8
0.6
0.4
0.2
0
-0.2
-0.4
100%
85%
-15%

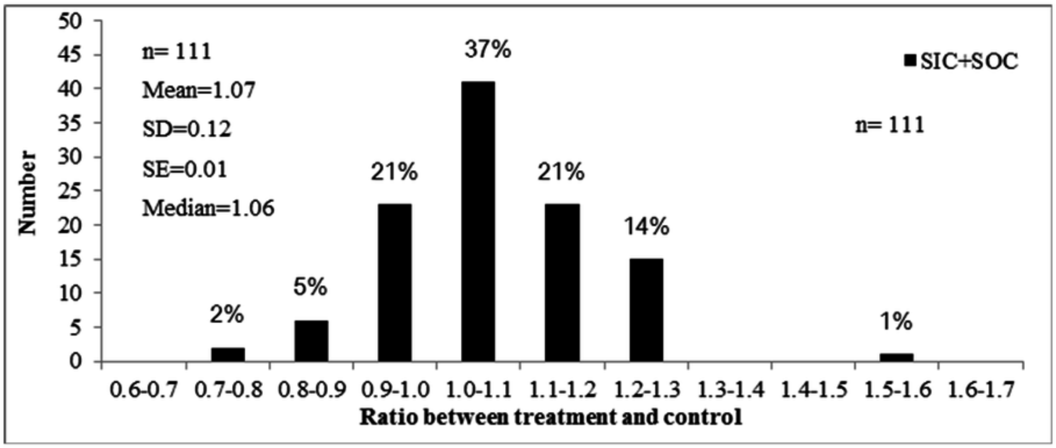
Figure 2

Frequency distribution of the ratio between treatment and control for SIC, SOC, SIC+SOC, and their average analysis result.

Data are from Jin(2006), Zeng et al.(2008),Li(2008), Geng et al.(2008), Yang et al.(2007), An et al.(2012a,b) and Huang et al.(2006).


SIC
n= 111
Mean=0.89
SD=0.15
SE=0.01
Median=0.94
Number
70
60
50
40
30
20
10
0
1%
3%
7%
14%
10%
51%
14%
1%
<0.5
0.5-0.6
0.6-0.7
0.7-0.8
0.8-0.9
0.9-1.0
1.0-1.1
1.1-1.2


SOC
n= 111
Mean=1.18
SD=0.16
SE=0.01
Median=1.16
Number
30
25
20
15
10
5
0
0
13%
22%
24%
21%
14%
3%
2%
2%
1%
0.8-0.9
0.9-1.0
1.0-1.1
1.1-1.2
1.2-1.3
1.3-1.4
1.4-1.5
1.5-1.6
1.6-1.7
>1.7


SIC+SOC
n= 111
Mean=1.07
SD=0.12
SE=0.01
Median=1.06
n= 111
Number
50
45
40
35
30
25
20
15
10
5
0
2%
5%
21%
37%
21%
14%
1%
0.6-0.7
0.7-0.8
0.8-0.9
0.9-1.0
1.0-1.1
1.1-1.2
1.2-1.3
1.3-1.4
1.4-1.5
1.5-1.6
1.6-1.7
Ratio between treatment and control

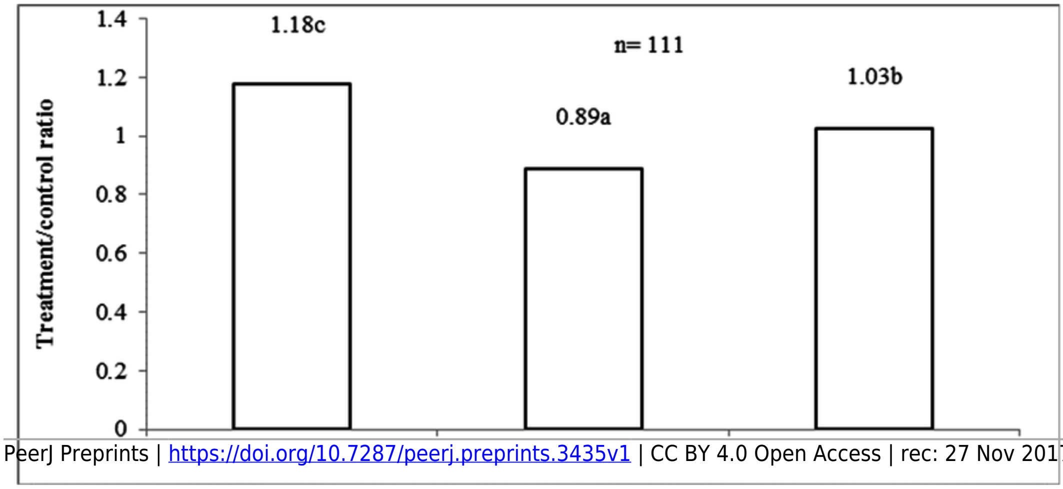

n= 111
Treatment/control ratio
1.4
1.2
1
0.8
0.6
0.4
0.2
0
1.18c
0.89a
1.03b

Figure 3

Relations between soil depth and the ratio between treatment and control for SIC (upper), SOC (middle), SIC+SOC (lower), and their regression analysis result.

Data are the same as Fig. 2.

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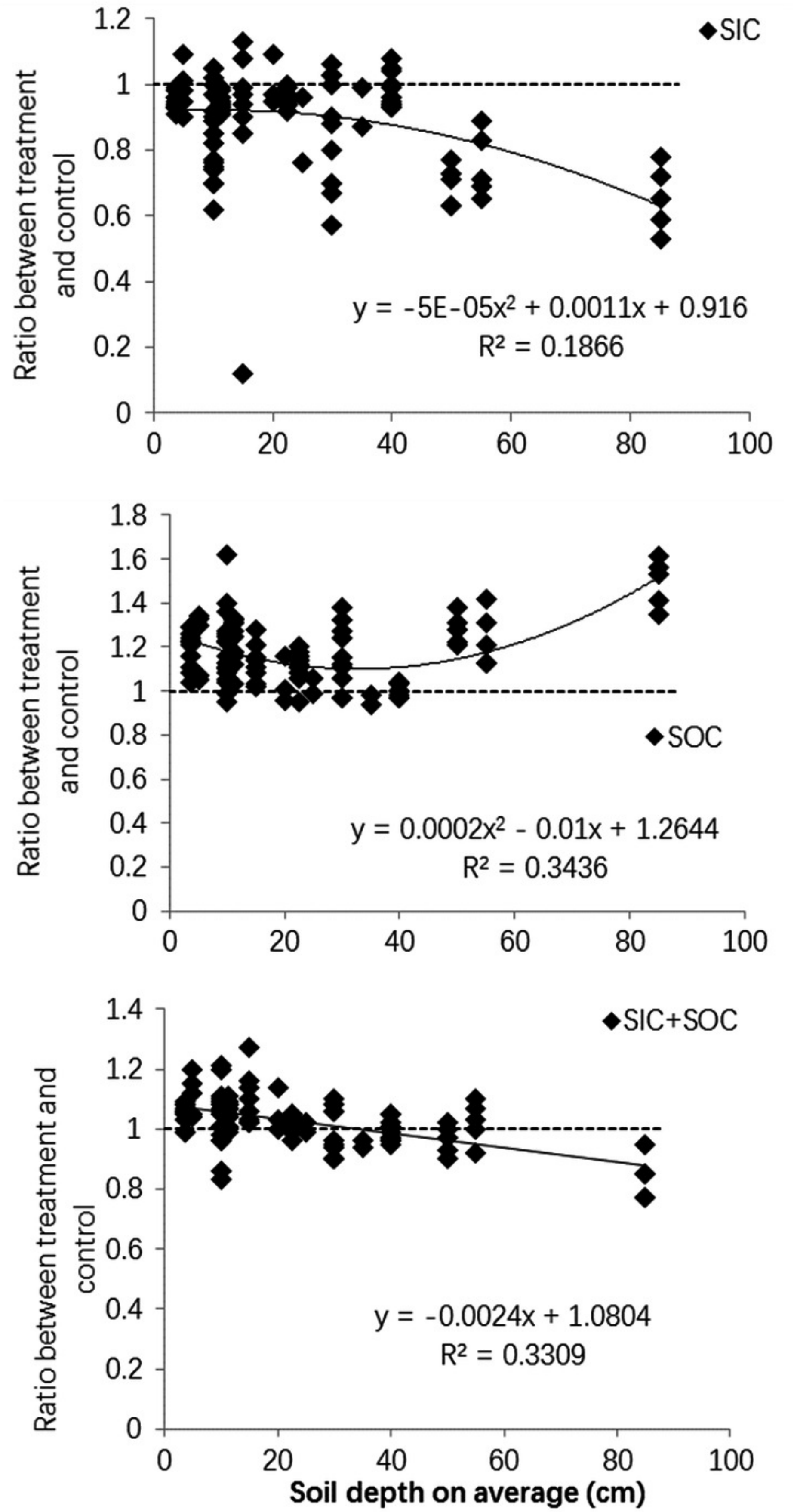
1.2
1
0.8
0.6
0.4
0.2
0
0
20
40
60
80
100
Ratio between treatment and control
◆SIC
y = -5E-05x² + 0.0011x + 0.916
R² = 0.1866
1.8
1.6
1.4
1.2
1
0.8
0.6
0.4
0.2
0
0
20
40
60
80
100
Ratio between treatment and control
◆SOC
y = 0.0002x² - 0.01x + 1.2644
R² = 0.3436
1.4
1.2
1
0.8
0.6
0.4
0.2
0
0
20
40
60
80
100
Ratio between treatment and control
◆SIC+SOC
y = -0.0024x + 1.0804
R² = 0.3309
Soil depth on average (cm)

Figure 4

Frequency distribution of SOC changing rate and SIC changing rate of separated measurements of SOC or SIC from previous studies.

Data for SOC are from Ma et al.1994; Zhu et al.1995; Zhang et al.2003; Meng et al. 2005; Zhang et al. 2006;Wang & Wang, 2006; Zhou et al. 2006;Yin & Cai,2006;Pan et al.2006;Li,2007; Song et al.2007;Guo et al. 2009; Suo & Han,2009;Wang et al. 2011 and Sun & Suo,2011. Data for SIC are from Sheng & Wang, 1989; Yuan, 1994; Liu et al.1998; Pan, 1999; Duan et al. 1999; Yang et al. 2012 and Wang et al.2012.

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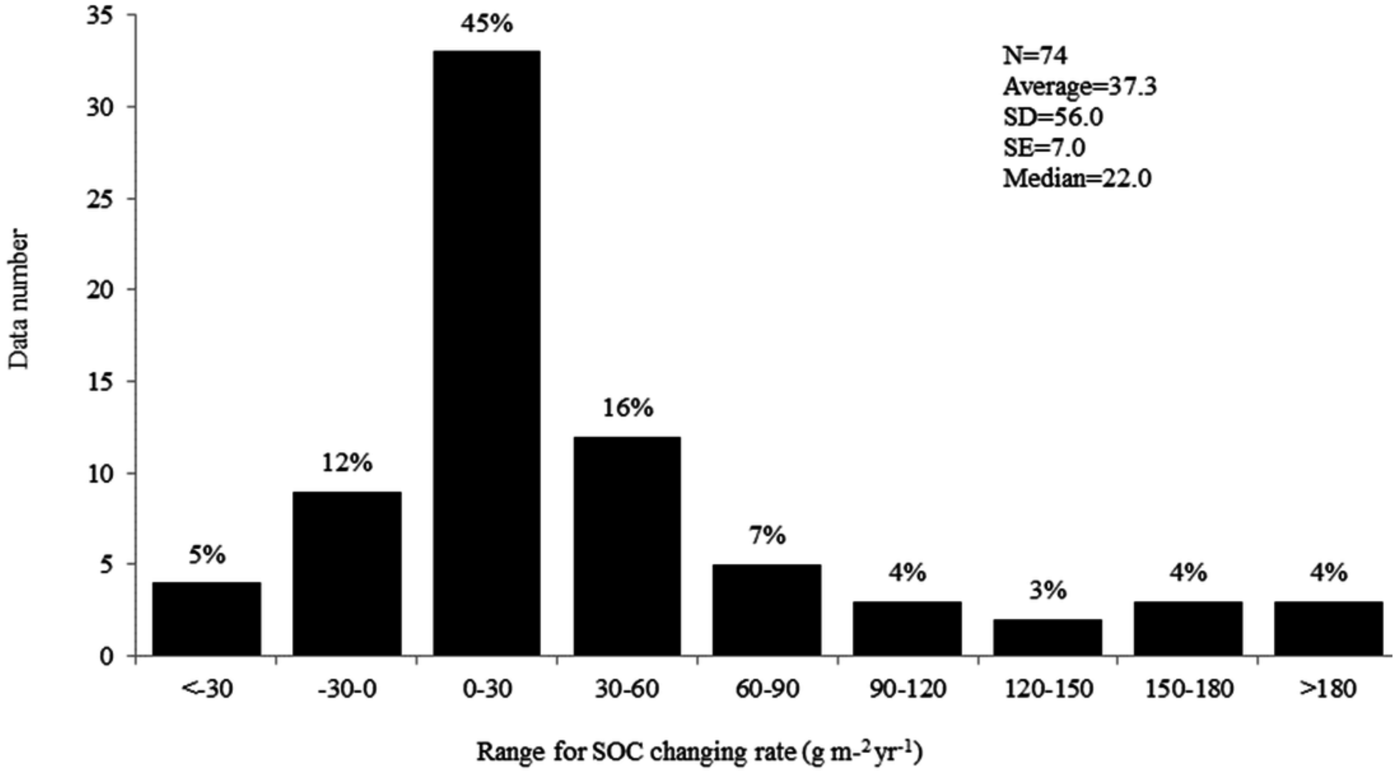

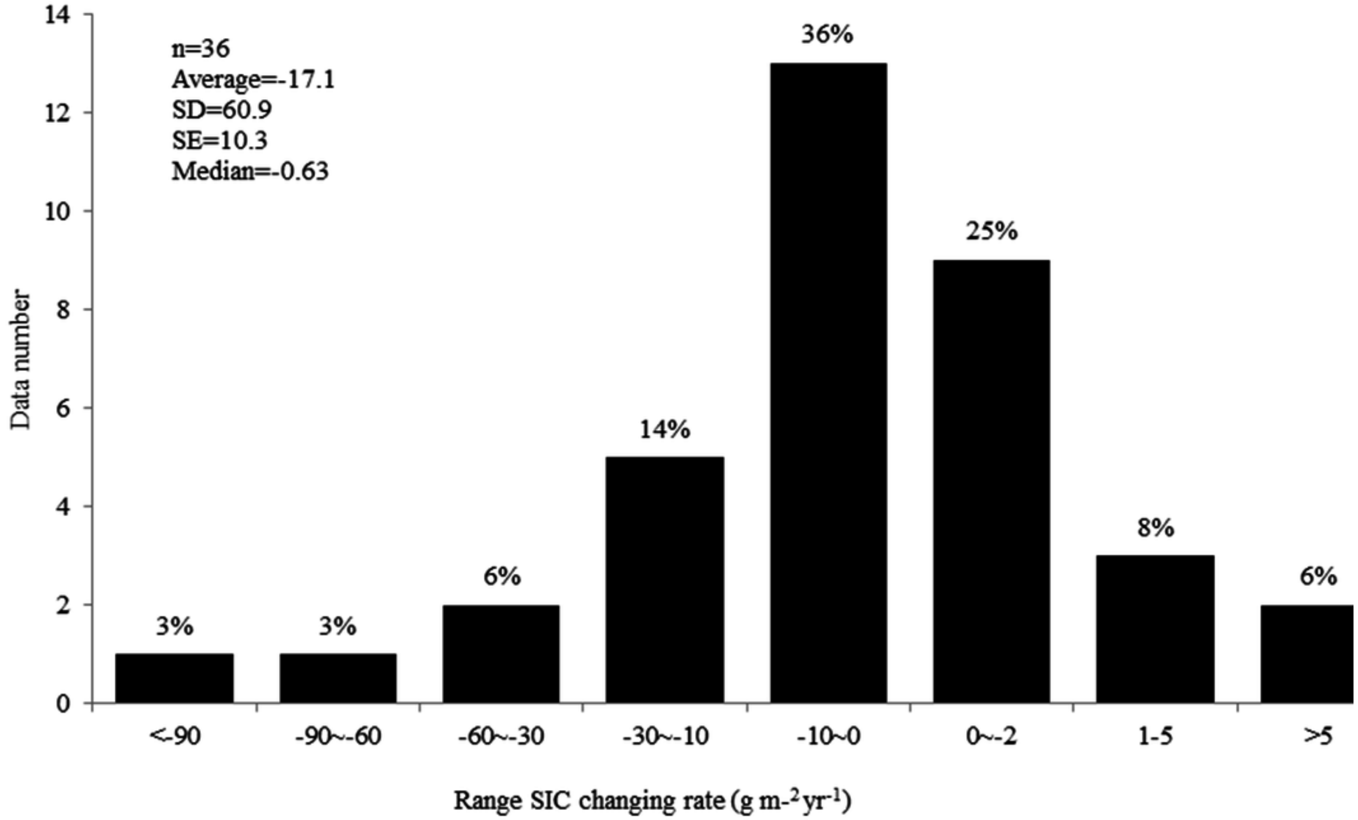

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Figure 5

Offsetting percentage of inclusion of SIC into carbon budget estimation from linear regression, field paired measurement and field separated measurement.

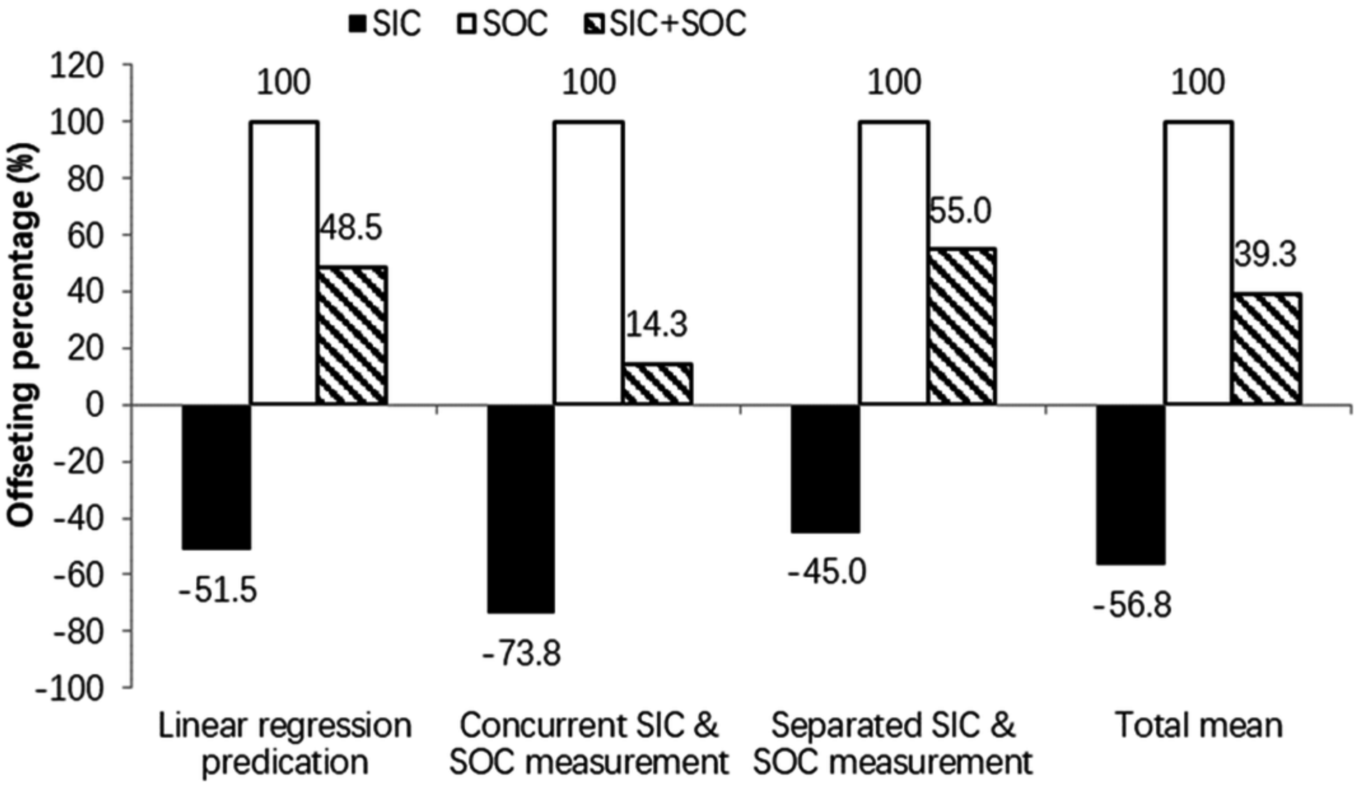

Figure 6

Negative correlation between SOC and SIC.

The same data from Fig. 1, Fig. S1-S4 were used in this figure.

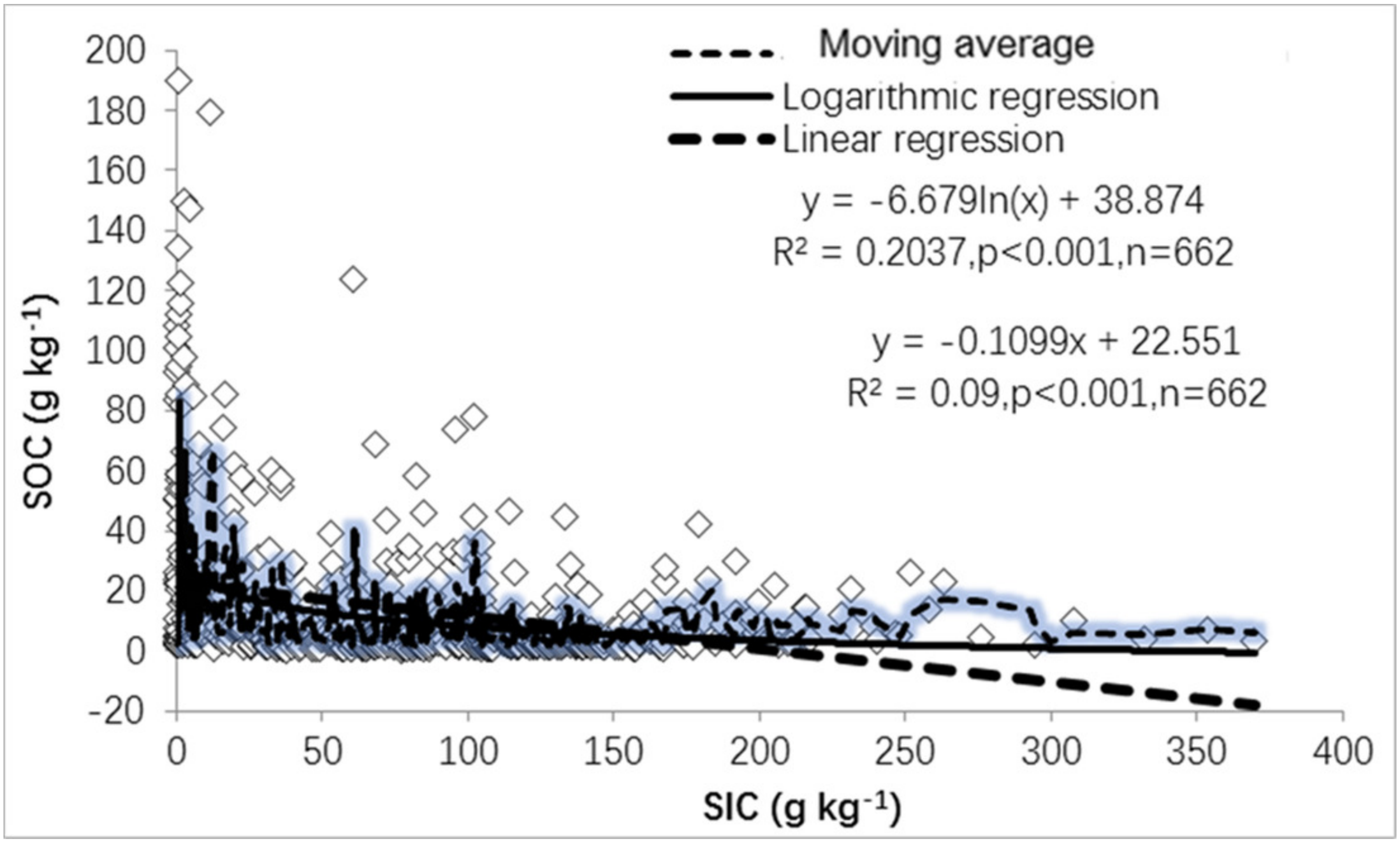

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Table 1(on next page)

Comparison of the linear regressions between SIC((g kg^{-1})), SOC((g kg^{-1})) and variable soil fertility parameters and slope comparisons

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X	y	equations	N	R^2	p	Slope difference
Total N(g kg^{-1})	SIC	Y=-1.249x+11.3	577	0.044	<0.0001	$p < 0.001$
	SOC	Y=12.97x-2.07	902	0.917	0.0000	
Available N (mg kg^{-1})	SIC	Y=-0.027x+13.331	342	0.117	<0.0001	$p < 0.001$
	SOC	Y=0.148x+0.836	511	0.691	<0.0001	
Total P(g kg-1)	SIC	Y=3.386x+6.103	494	0.093	<0.0001	$p < 0.001$
	SOC	Y=5.72x+7.20	698	0.053	<0.0001	
Available P(mg kg^{-1})	SIC	Y=-0.289x+11.69	382	0.026	0.0014	$p < 0.001$
	SOC	Y=1.25x+7.59	554	0.124	<0.0001	
Total K(g kg^{-1})	SIC	Y=0.232x+5.144	467	0.021	0.0015	$p < 0.001$
	SOC	Y=0.074x+14.83	671	0.006	0.5253	
Available K(mg kg^{-1})	SIC	Y=-0.0047x+11.29	386	0.003	0.2595	$p < 0.001$
	SOC	Y=0.0646x+5.13	550	0.114	<0.0001	
pH	SIC	Y=4.80x-29.21	547	0.212	<0.0001	$p < 0.001$
	SOC	Y=-5.72x+57.16	810	0.129	<0.0001	
CEC (cmol kg^{-1})	SIC	Y=-0.205x+14.33	427	0.063	<0.0001	$p < 0.001$
	SOC	Y=1.35x-5.984	418	0.547	<0.0001	

Table 2(on next page)

Stepwise regression comparison between SOC, SIC and soil parameters of pH, total N, total P, total K, available N, available P, available K and CEC

	SOC(g kg-1)			SIC(g kg-1)		
	coefficient	standardized coefficient	Sig.	coefficient	standardized coefficient	Sig.
Intercept	3.391		0.50	-39.885		0.00
pH	-0.303	-0.008	0.62	***5.982***	***0.435***	***0.00***
Total N (g kg^{-1})	***12.220***	***0.926***	***0.00***	0.308	0.064	0.60
Total P (g kg^{-1})	-0.418	-0.014	0.39	***1.143***	***0.108***	***0.06***
Total K (g kg^{-1})	-0.104	-0.025	0.12	0.106	0.069	0.20
Available N (mg kg^{-1})	***0.015***	***0.079***	***0.01***	***-0.024***	***-0.345***	***0.00***
Available P (mg kg^{-1})	***-0.251***	***-0.052***	***0.00***	0.064	0.036	0.50
Available K (mg kg^{-1})	0.002	0.009	0.56	0.000	0.005	0.93
CEC(cmol kg^{-1})	-0.020	-0.011	0.64	0.029	0.043	0.59
R^2	0.947			0.363		
p-level	0.000			0.000		