Distribution of soil inorganic carbon storage and its changes due to agricultural land use activity in China

Haibin Wu [a,b], Zhengtang Guo [a], Qiong Gao [c], Changhui Peng [b,d,*]

[a] Institute of Geology and Geophysics, Chinese Academy of Science, P.O. Box 9825, Beijing 100029, China
[b] Institut des sciences de Íenvironnement, Département des sciences biologiques, Université du Québec à Montréal, Montréal, Canada H3C 3P8
[c] College of Resources Science and Technology, Beijing Normal University, Beijing 100875, China
[d] Ecology Research Section, Central-South University of Forestry & Technology, Changsha, Hunan 410004, China

ABSTRACT

The accurate estimation of soil carbon (C) stocks is a necessary component for understanding the global C budget. Although the importance of soil organic C (SOC) within the C cycle is well established, the quantity of soil inorganic C (SIC, including lithogenic and pedogenic inorganic C) pool, another important soil C pool component, has been poorly studied to date. In this study, soil profile data compiled by China's second national soil survey conducted in the 1980s was used to investigate the spatial distribution of SIC for the entire country under present day conditions as well as changes in SIC under historical land use. Results showed that the total SIC storage in China was approximately 55.3 ± 10.7 Pg C with a current average content of 6.3 ± 1.2 kg C m^{-2}, representing 5.8% of the global SIC pool. Land use has significantly affected SIC levels in cultivated soils. Approximately 51% of total cultivated soil surfaces in China have experienced C loss where the most significant loss has been observed in the eastern part of northern China in dry farmlands as well as irrigated soils and paddy soils. On the contrary, SIC has increased ($\sim$10%) in irrigated soils in northwestern China. No significant change ($\sim$39%) has been observed in soils in southern and the eastern part of northeast China. The total loss of SIC in China was approximately 1.6 Pg C due to extensive human activity. Results of this study indicate that human activity may have had a great impact on SIC as well as SOC pools.

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1. Introduction

The growth rate of atmospheric CO_2, the largest human contribution to climate change, is increasing rapidly while the long-term storage capacity and removal rates of terrestrial and ocean sinks appear to be slowing down (Canadell et al., 2007), highlighting a need for improved data on individual sinks and their relationship to other fluxes (Mayorga, 2008). Soil organic and inorganic carbon (C) is by far the largest terrestrial C pool, storing more than double the quantity of C in vegetation or the atmosphere (Batjes, 1996; Eswaran et al., 2000). Changes in soil organic and inorganic C content could have a great effect on the global C budget. Moreover, a potential increase in the C storage

capacity of soil is a recognized option for mitigating the buildup of atmospheric CO_2 in the future (Watson et al., 2000; Lal and Kimble, 2000).
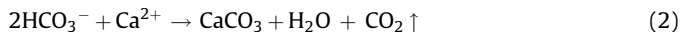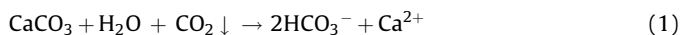
The role of soil organic C in the greenhouse effect has received considerable attention (Bouwman, 1990; Lal et al., 1998; Lal, 2004). The soil C pool, however, has two principal components: soil organic C (SOC) and soil inorganic C (SIC). In arid and semiarid regions that cover as much as one-third of the surface of the planet, the SIC pool is approximately two to ten times larger than SOC storage (Schlesinger, 1982; Eswaran et al., 2000; Batjes, 2004b) while the SIC rate of accumulation is generally higher than that of SOC (Landi et al., 2003).

The SIC pool can be classified as lithogenic inorganic C (LIC) and pedogenic inorganic C (PIC). The former is inherited from parent material of the soil, with no change in soil inorganic C content. The latter is formed through the dissolution and precipitation of carbonate parent material (including water table fluctuations contributing), consumes a mole of atmospheric CO_2 during carbonate dissolution but liberates an equal amount
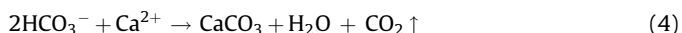
during pedogenic carbonate precipitation.

$$CaCO_3 + H_2O + CO_2 \downarrow \rightarrow 2HCO_3^- + Ca^{2+} \tag{1}$$

$$2HCO_3^- + Ca^{2+} \rightarrow CaCO_3 + H_2O + CO_2 \uparrow \tag{2}$$

PIC may also be formed from weathering of Ca/Mg-bearing silicates (Lal and Kimble, 2000; Emmerich, 2003; Goddard et al., 2007).

$$2CO_2 \downarrow + 3H_2O + CaSiO_3 \rightarrow H_4SiO_4 + 2HCO_3^- + Ca^{2+} \tag{3}$$

$$2HCO_3^- + Ca^{2+} \rightarrow CaCO_3 + H_2O + CO_2 \uparrow \tag{4}$$

The process of calcrete reservoir weathering consumes 2 moles of atmospheric CO_2 for every mole released during the precipitation of pedogenic carbonate (Schlesinger, 1982). These can lead to the sequestration of atmospheric CO_2 on land (Adams, 1993; Lal and Kimble, 2000), representing the transfer of atmospheric C and the buffering capacity to oceans at timescales from hundreds to thousands of years (Raymond and Cole, 2003; Mayorga, 2008). Recent studies have shown the occurrence of significant changes in SIC pools of different soil types linked to climatic variation (Adams and Post, 1999; Landi et al., 2003), land use (Emmerich, 2003; Entry et al., 2004; Mikhailova and Post, 2006; Papiernik et al., 2007; Sartori et al., 2007), and atmospheric nutrient deposition (Goddard et al., 2007). The SIC pool (e.g., PIC), therefore, plays an important role in the global C cycle.

To date, studies on SIC storage and dynamics have primarily focused on local or regional assessments (Schlesinger, 1982; Grossman et al., 1995; Monger and Matrinez-Rios, 2000; Rasmussen, 2006; Singh et al., 2007). SIC pool estimates at a national or global scale have been poorly documented in comparison to estimates of SOC pools (Lal et al., 1998; Batjes, 2004a,b, 2006; Guo et al., 2006). Since the C sequestration potential of soils depends partly on the C stock under present-day conditions, an accurate quantitative assessment of soil C storage is needed as a baseline to estimate the overall C budget and to assess the impact of land use change on the inventory in order to identify regions where C sequestration effort should be concentrated (Sleutel et al., 2003).

China has an expansive terrestrial surface with strong spatial climatic and topographic variability. Approximately 40% of the total land surface is arid and/or semiarid in northern and northwestern China (NSSO, 1998) where soils contain significant amounts of SIC. Currently, China retains approximately 137.5 million hectares of cropland (NSSO, 1998) from the southern tropical-humid zone to the northern temperate-arid zone. Its long history of agricultural exploitation and changes in land use suggest that the terrestrial ecosystem of China probably played an important role in the global C cycle (Fang et al., 1996; Peng and Apps, 1997; Zhao et al., 1997; Li and Zhao, 2001; Wu et al., 2003; Pan et al., 2003). An attempt was made to estimate the SIC pool (from 60 to 234.2 Pg C) in China by Pan (1999) and Li et al. (2007) based on approximately 2500 soil profile data. Using 136 soil profile data, Feng et al. (2001) estimated approximately 14.9 Pg C of SIC storage held in desertified lands in northern China. Since processes governing the dynamics of the SIC pool differ among ecoregions and strongly interact with land use activities (Lal and Kimble, 2000), considerable uncertainties remain for C stock inventories and for projected changes at a national scale. Consequently, there is an urgent need to accurately estimate the SIC pool quantity and its changes due to agricultural activity in China as a whole.

In this study, China's second national soil survey (NSSO, 1993, 1994a,b, 1995a,b, 1996, 1998) conducted in 1980s was employed, being the most accurate and comprehensive soil database available

in China, to investigate spatial distributions of SIC storage under present-day conditions (for both cultivated and non-cultivated soils). Since the data acquired during this investigation has a great spatial density than previous national soil survey, it is expected to more accurately represent present-day conditions of SIC in China. At the same time, changes in SIC due to land use activity was also investigated by comparing the differences in C between cultivated and non-cultivated soils to provide a primary budget of SIC by means of the human activity factor.

2. Methods

Data used in this study was collected from 34,411 soil profiles analyzed during the second national soil survey (NSSO, 1993, 1994a,b, 1995a,b, 1996, 1998) in China. Each soil profile contains both organic (A horizon) and mineral horizons (B and C horizons) and the following types of soil characteristics: taxonomic classification, thickness, soil organic matter, inorganic C concentration, soil bulk density, volume percentage of rock fragment >2 mm, and other parameters, that were sampled at the midpoint of each horizon. Measurements of calcium carbonate concentration, organic matter, soil bulk density, and fractions >2 mm were reported using standard techniques (NSSO, 1998). Calcium carbonate was measured by manometric collection of CO_2 evolved during an HCl treatment process. Soil organic matter concentrations were measured by means of wet combustion with $K_2Cr_2O_7$. Soil bulk density was determined by applying the oven-dry method to the soil mass and the core volume. Fractions >2 mm were sieved and separated using a 2-mm screen. The database includes 100% of the soil component data for thickness and organic C concentration, 36% for calcium carbonate concentration, 25% for bulk density, and 7% for the quantity of fragments.

Based on geomorphological units, hydrothermal conditions, morphological peculiarities, and physicochemical properties, 2553 profiles were evaluated and selected as the most representative of soil local types occurring in different regions in China (NSSO, 1993, 1994a,b, 1995a,b, 1996) (Fig. 1), although it represents only a small spatial profile density in the northwestern region and the Tibetan Plateau. These selected profiles were described in greater detail in features such as land use conditions, aboveground vegetation, and profile location, while the information was not completed for the other soil profiles. The representative profiles can be divided into two basic parts according to land use conditions. The first part consists of 923 profiles (Fig. 1) that were uncultivated throughout their land use history, because the profiles had not experienced any human activity disturbance, and where the current vegetation is ecologically consistent with present climatic conditions (NSSO, 1993, 1994a,b, 1995a,b, 1996). The selected profiles are regarded as non-cultivated soil profiles in this study. Taken together, they provide a good spatial coverage and include all soil types. Although some soil samples were still more or less subject to possible indirect human activity at some point in the past, such as from human-induced vegetation burning and global atmospheric CO_2 concentration increasing, they nonetheless represent the best soil type samples in an undisturbed condition. Accordingly, the selected data samples were used to reconstruct the C content and storage capacity under non-cultivated conditions.

The second part consisting of 1630 profiles (Fig. 1) are regarded as cultivated profiles and include the present-day cultivated soils and cultivated soils in the past. These samples were used to estimate SIC changes by means of land use activity compared to their non-cultivated counterparts, because we did not examine the processes or mechanisms of SIC acumination or loess in this study. Although the different numbers of soil profiles under two conditions might increase errors for the comparison, this is the
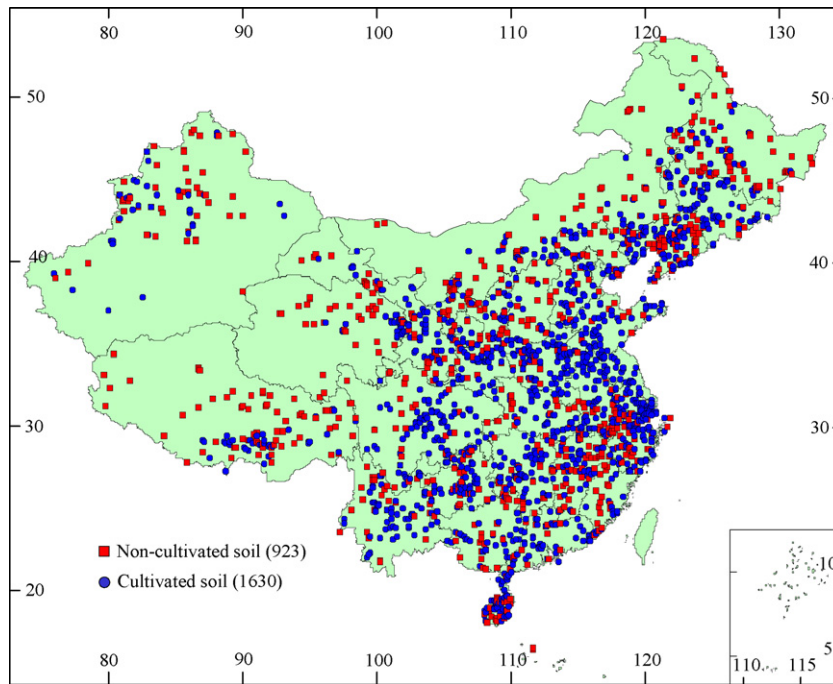
Fig. 1. Distribution of the 2553 representative soil profiles from China's second national soil survey (NSSO, 1998).

best comprehensive soil database available in China now. Present-day SIC was calculated using data from the 34,411 soil profiles obtained from the second national soil survey due to the great density of profiles that typify soil types throughout the whole country including all cultivated and non-cultivated soil profiles. The difference between reconstructed SIC under non-cultivated and cultivated conditions provides an estimation of SIC changes attributable to historical agricultural land use activity in China.

Since Chinese soil taxonomy (NSSO, 1998) was used in the soil survey, which contains 61 soil groups and 228 soil subgroups, the same terminology was used in this study. They were tentatively compared to the FAO-UNESCO (1988) soil classification system (Table 1). The base soil distribution map used for this study is from Tian et al. (1996a). Soil taxonomy in the second national soil survey was not completely consistent with the soil map legend, making it necessary to merge soil types especially at the subgroup level based on soil properties and distribution (Li et al., 2001). This merging just affects the spatial distribution of SIC content in some soil types of Fig. 2a, but does not affect the SIC storage in China because for this study, soil subgroup areas were taken from the second national soil survey in which calculations were based on 1:50,000 topography maps, excluding water areas, glacial and

permanent snow covered areas, and rock mountain areas (NSSO, 1998).

There are three stages to undertake when determining the size of the SIC pool for the country as a whole. First, the SIC content is calculated for individual soil profiles. Second, this information is analyzed on the basis of the 228 soil taxonomy subgroups. Finally, results (e.g., mean value, standard error, and coefficient of variation) of the analyses are combined with information on the spatial extent of various soil subgroups in China to estimate the total SIC pool.

Since SIC concentration varies among the soil profile, the SIC content (SICC) of each profile was calculated as follows:

$$\text{SICC} = \sum_{i=1}^{n} 0.12 \times T_i \times \rho_i \times \frac{IM_i}{100} \times \left(1 - \frac{C_i}{100}\right) \div 10 \qquad (5)$$

where n is the number of pedogenic horizons defined in the soil survey (NSSO, 1998) and T_i, ρ_i, IM_i, and C_i represent thickness (cm), bulk density (g cm^{-3}), inorganic matter concentration (%), and the volumetric percentage of the fractions >2 mm (rock fragments) in the layer i, respectively. The division by 10 at the equation end is for unit transition from g cm^{-2} to kg m^{-2}.

Table 1
Inorganic carbon content and storage of soil groups under present-day conditions in China.

Soil group in Chinese soil taxonomy	FAO/UNESCO taxonomy	Climatic zone[a]	Number of subgroups (Chinese soil taxonomy)	Area (millions of hectare)	SIC in soil profile to actual depth of sampling			Total SIC storage in soil group within China (10^{10} kg C)
					Carbon content (kg C m^{-2})	CV[b] (%)	Number of profiles	
Latosols	Haplic Acrisols	1	2	4.27[c]	0.00 ± 0.00[d]	0	864	0.00 ± 0.00
Latosolic red earths	Haplic Acrisols/Alisols	1	3	18.13	0.00 ± 0.00	0	193	0.00 ± 0.00
Red earths	Haplic Alisols/Haplic Acrisols	1	5	57.85	0.00 ± 0.00	0	2008	0.00 ± 0.00
Yellow earths	Haplic Alisols	1	3	23.93	0.00 ± 0.00	0	638	0.00 ± 0.00
Yellow-brown earths	Ferric/Haplic Luvisols	1	3	18.42	0.20 ± 0.11	125	273	2.88 ± 2.11
Yellow-cinnamon soils	Eutric Cambisols	1	4	3.81	0.20 ± 0.06	2	221	0.61 ± 0.22
Torrid red soils	Ferralic Cambisols	1	2	0.71	0.09 ± 0.09	41	99	0.06 ± 0.06
Limestone soils	Regosols/Leptisols	1	4	10.77	0.61 ± 0.52	219	463	6.61 ± 5.61
Purplish soils	Calcaric Regosols	1	3	18.90	1.94 ± 0.44	347	1027	36.61 ± 8.30

Table 1 (Continued)

Soil group in Chinese soil taxonomy	FAO/UNESCO taxonomy	Climatic zone[a]	Number of subgroups (Chinese soil taxonomy)	Area (millions of hectare)	SIC in soil profile to actual depth of sampling			Total SIC storage in soil group within China (10^{10} kg C)
					Carbon content (kg C m^{-2})	CV[b] (%)	Number of profiles	
Phospho-calcic soils	Calcaric Regosols	1	2	0.00	0.00 ± 0.00	0	4	0.00 ± 0.00
Volcanic soils	Andosols	1, 2	3	0.19	0.00 ± 0.00	0	47	0.00 ± 0.00
Coastal solonchaks	Solonchaks	1, 2	3	2.12	1.90 ± 1.01	156	146	4.04 ± 2.14
Skeletal soils	Regosols/Leptisols	1, 2	4	26.11	3.52 ± 0.67	19	589	91.89 ± 17.49
Mountain meadow soils	Umbric Leptisols/ Dystric Cambisols	1, 2	3	4.22	7.60 ± 0.78	7	143	32.09 ± 3.30
Paddy soils	Fluvisols/Cambisols	1, 2	8	30.68	0.20 ± 0.12	545	8993	6.27 ± 3.66
Neo-alluvial soils	Fluvisols	1, 2	3	4.29	6.16 ± 1.44	114	872	26.45 ± 6.16
Brown earths	Haplic/Albic Luvisols or Eutric/Dystric Cambisols	2	4	20.16	0.00 ± 0.00	0	1510	0.00 ± 0.00
Dark-brown earths	Haplic Luvisosl/Eutric Cambisols	2	5	40.11	0.00 ± 0.00	0	275	0.00 ± 0.00
Bleached Beijing soils	Albic Luvisols	2	3	5.27	0.00 ± 0.00	0	282	0.00 ± 0.00
Brown coniferous forest soils	Humic Cambisols	2	3	11.66	0.00 ± 0.00	0	49	0.00 ± 0.00
Podzolic soils	Haplic Podzols	2	1	0.00	0.00 ± 0.00	0	3	0.00 ± 0.00
Cinnamon soils	Eutric Cambisols	2	7	25.17	8.64 ± 1.30	81	1828	217.49 ± 32.81
Black soils	Phaeozems	2	4	7.36	0.00 ± 0.00	0	435	0.00 ± 0.00
Chernozems	Chernozems	2	6	13.22	8.49 ± 1.56	67	612	112.20 ± 20.59
Castano-cinnamon soils	Kastanozems	2	3	4.82	19.01 ± 0.94	14	282	91.62 ± 4.55
Dark loessial soils	Calcisols	2	3	2.55	21.93 ± 2.66	25	860	55.91 ± 5.77
Loessial soils	Calcaric Regosols	2	1	12.29	22.33 ± 0.52	14	1368	274.41 ± 6.35
Red primitive soils	Lixisols	2	3	2.28	4.39 ± 1.16	113	449	10.01 ± 2.63
Lithosols	Regosols/Leptisols	2	3	18.53	1.07 ± 0.19	32	179	19.77 ± 3.60
Meadow soils	Umbric Gleysols/ Haplic Phaeozem	2	6	25.09	3.64 ± 0.94	32	940	91.33 ± 12.42
Saijiang black soils	Eutric Vertisols/ Gleyic Cambisol	2	5	3.77	7.41 ± 2.14	101	271	27.97 ± 8.06
Acid sulphate soils	Solonchaks	2	2	0.02	0.00 ± 0.00	0	13	0.00 ± 0.00
Fluvi-aquic soils	Fluvisols	2, 3	7	25.68	14.31 ± 0.81	39	4206	367.36 ± 20.83
Meadow solonchaks	Solonchaks	2, 3	4	10.44	6.88 ± 1.16	41	57	71.82 ± 12.11
Solonetzs	Solonchaks	2, 3	5	0.87	9.46 ± 0.90	23	85	8.23 ± 0.79
Bog soils	Gleysols	2, 4	5	12.62	2.70 ± 1.84	151	245	34.12 ± 23.25
Gray-cinnamon soils	Haplic/Capcic Luvisol	2, 4	5	6.18	8.45 ± 2.39	112	105	52.24 ± 14.79
Gray forest soils	Albic Luvisosls	3	2	3.15	1.70 ± 0.00	0	10	5.35 ± 0.00
Castanozems	Kastanozems	3	7	37.50	17.16 ± 2.42	48	918	643.42 ± 9.90
Brown caliche soils	Haplic/Haplic Calcisols	3	6	26.51	12.56 ± 1.09	19	68	332.93 ± 28.77
Sierozems	Calcaric Cambisols	3	4	5.38	19.86 ± 1.53	26	506	106.83 ± 8.21
Gray desert soils	Haplic Calcisols	3	6	4.60	13.28 ± 1.23	23	14	61.10 ± 5.64
Gray-brown desert soils	Haplic Calcisols	3	4	30.73	8.41 ± 3.27	67	99	258.48 ± 100.56
Brown desert soils	Soloncnaks	3	5	24.30	10.36 ± 4.61	66	19	251.72 ± 112.01
Takyr	Solonchaks	3	1	0.68	14.96 ± 2.31	0	2	10.17 ± 1.57
Aeolian soils	Arenosols	3	4	67.57	16.58 ± 2.19	50	287	1120.05 ± 147.94
Shruby meadow soils	Calcaric Cambisols	3	2	2.48	7.62 ± 0.22	1	3	18.90 ± 0.54
Desert solonchaks	Solonchaks	3	3	2.87	1.68 ± 1.07	139	15	4.81 ± 3.07
Irrigated silting soils	Calcaric Fluvisols	3	4	1.52	17.13 ± 2.26	25	570	26.10 ± 3.45
Irrigated desert soils	Calcaric Fluvisols	3	4	0.91	16.59 ± 1.47	27	262	15.10 ± 1.33
Peat soils	Histosols	4	3	1.48	0.00 ± 0.00	0	67	0.00 ± 0.00
Frigid plateau solonchaks	Solonchaks	4	3	0.69	20.71 ± 2.60	27	8	14.29 ± 1.79
Felty soils (alpine meadow soils)	Cambisols	4	4	53.54	1.10 ± 0.60	250	187	58.91 ± 32.38
Dark felty soils (subalpine meadow soils)	Cambisols	4	4	19.44	2.13 ± 1.59	165	135	41.44 ± 31.00
Frigid calcic soils (alpine steppe soils)	Cambisols	4	4	68.85	10.98 ± 3.22	52	106	756.22 ± 221.80
Cold calcic soils (subalpine steppe soils)	Cambisols	4	4	11.29	4.76 ± 0.94	66	162	53.73 ± 10.62
Cold brown calcic soils (mountain shrub steppe soils)	Cambisols	4	2	0.96	5.18 ± 1.44	91	306	4.97 ± 1.39
Frigid desert soils (mountain shrub steppe soils)	Gelic Arenosols	4	1	8.96	7.56 ± 4.09	94	3	67.70 ± 36.61
Cold desert soils (subalpine desert soils)	Gelic Arenosols	4	1	5.22	5.89 ± 2.92	70	2	30.74 ± 15.27
Frigid frozen soils (alpine frozen soils)	Gelic Regosols	4	1	30.65	0.04 ± 0.02	137	28	1.17 ± 0.72
Total			219[e]	881.81				5526.15 ± 1073.18

[a] The four major climatic zones shown in Fig. 2a, Zone 1: the monsoonal tropical–subtropical zone; Zone 2: the monsoonal temperate zone; Zone 3: the arid zone in northwestern China, and Zone 4: the frigid zone in the Tibetan Plateau region.

[b] The coefficient of variation under present-day conditions.

[c] Each group soil surface on the mainland was calculated by summing provincial data (NSSO, 1998) and that in Taiwan was computed by the map of soils (Tian et al., 1996a).

[d] Mean ± standard error.

[e] One soil group and nine soil subgroups of very small surface area were not included.
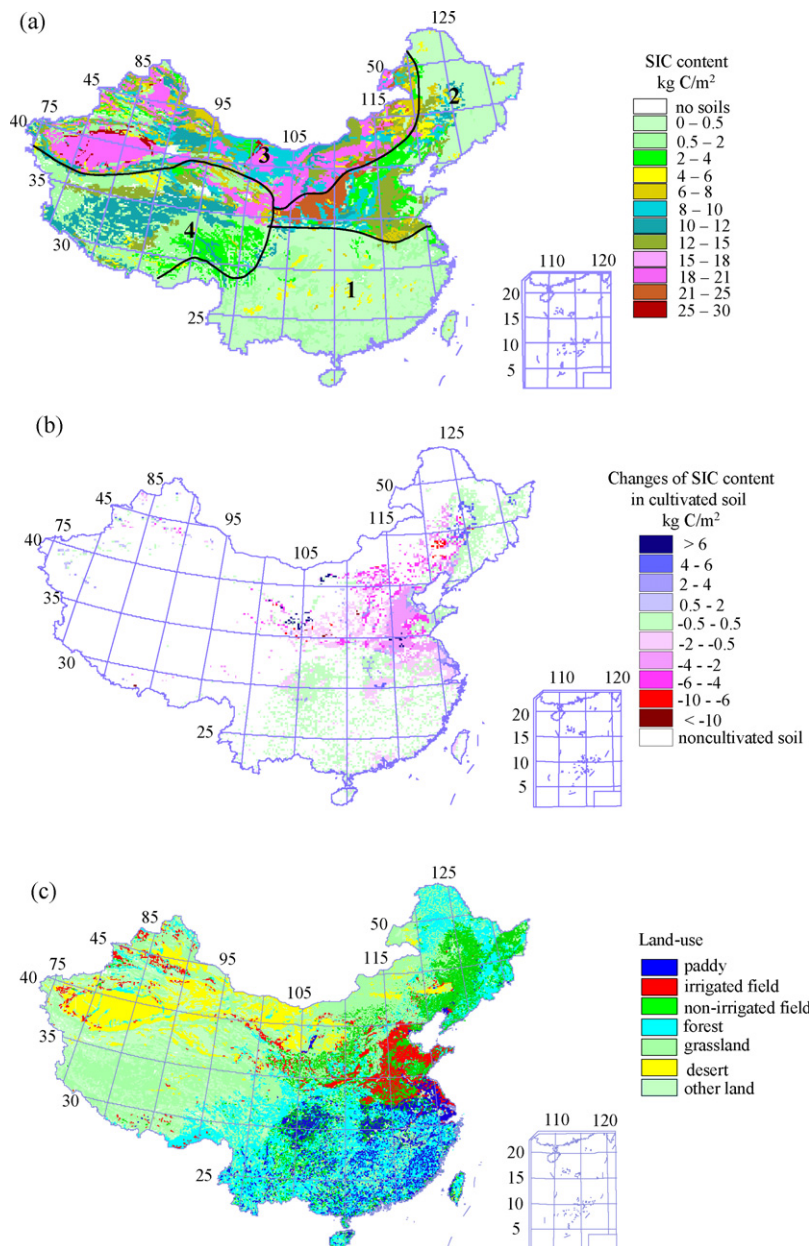
Fig. 2. Spatial distribution of soil inorganic carbon (SIC) contents and the changes in SIC content and land use activity in China. (a) Distribution in SIC content under present-day conditions; (b) distribution of SIC content changes; and (c) distribution of land use activity (Tian et al., 1996b). The four major climatic zones shown in (a) refer to the monsoonal tropical-subtropical zone (Zone 1), the monsoonal temperate zone (Zone 2), the arid zone in northwestern China (Zone 3), and the frigid zone in the Tibetan Plateau region (Zone 4), respectively.

In this study, the term carbonate refers to the sum of $CaCO_3$, $MgCO_3$ and other carbonate minerals in soils, of which $CaCO_3$ is dominant and dolomite and $MgCO_3$ are usually of minor components (Doner and Lynn, 1977). Calcium carbonate equivalents were converted to C content by multiplying them by 0.12, the mole fraction of C in $CaCO_3$. In China's second national soil survey, SIC content includes all C from both LIC and PIC since the SIC concentration measured by the HCl method. The SIC is, in fact, approximately 90% of the total soil carbonate classified as pedogenic carbonate reported by Weng (1989) based on mineralogy micromorphological analysis and the HCl method in the Loess Plateau region that extensively distributed in northern China. Pan and Guo (2000) also suggested that most SIC in arid and semiarid regions in China belongs to the pedogenic carbonate class,

based on correlations between SOC and SIC, the formation of $CaCO_3$ of field observation, soil micromorphological evidence, and SIC $\delta^{13}C$ tracing. These studies have indicated that a significant portion of Chinese SIC storage is, in fact, a PIC pool, which may play an important role in the global C cycle. A more reliable estimation concerning PIC storage levels in soils in China needs to be further developed and the database needs to be improved in order to quantitatively separate PIC and LIC in the future.

Since the thickness of soils is highly variable in spatial distribution, in this study, the C content of the soil profiles was calculated based on their actual thickness in the Chinese second national soil survey for a more accurate estimation of the SIC storage in China. The thickness ranged from 0.08 to 2.63 m where the mean value in China was 0.98 m. This data represents the SIC

storage capacity of the soil profile at a depth of approximately 1 m for the soil subgroups listed in Table 1. An area-weighted approach was used to calculate the regional SICC.

Because of the lack of bulk density data in some soil profiles and since it is critical to convert the inorganic C percentage by weight to content by volume, empirical relationships between organic C content (x) and bulk density (Y) were established based on 784 analytical data that were measured in parallel at the same time in the second national soil survey (Wu et al., 2003).

$$Y = -0.1229 \, Ln(x) + 1.2901 \qquad (6)$$

$$(R = -0.58, \quad F = 357.61 > F_{0.01} = 6.69)$$

$$Y = 1.3774 \, e^{-0.0413x} \qquad (7)$$

$$(R = -0.62, \quad F = 474.72 > F_{0.01} = 6.69)$$

Eq. (6) above is suitable for samples with SOC content <6% while Eq. (7) is suitable for samples with SOC content >6%. This method was frequently used in earlier studies (Grigal et al., 1989; Siltanen et al., 1997). The bulk density of soils without a measured value was then obtained using these established empirical relationships. The mean value of the same soil subgroup was used for soil horizons without a measured rock fragment volume (C_i).

SIC storage (SICS) was then computed using:

$$SICS = \sum_{i=1}^{n} area_i \times SICC_i \qquad (8)$$

where $area_i$ and $SICC_i$ are the surface area and the SIC content of the soil subgroup i, respectively. The established soil surface area for the entire country amounted to approximately 881.81 million hectares.

3. Results and discussions

3.1. Distribution and storage of SIC under present-day conditions

Table 1 lists the present-day SIC contents and storage capacities of different soil groups in China. The results presented in this paper are primarily at the soil group level regardless of the fact that the original analyses made for this study were carried out for the subgroups. This was done largely to enhance the legibility of the tabular output. SIC contents ranged from 0.0 to 22.3 kg C m^{-2} for soil groups where the majority of values fall into the range from 0.0 to 10.0 kg C m^{-2}. Fig. 2a illustrates the distribution of SIC contents at the soil subgroup level. In general, SIC contents in China increased from east to west and decreased from north to south. SIC contents were the highest, ranging from 12.0 to 25.0 kg C m^{-2}, in loessial soils (Calcaric Regosols) and Castano-cinnamon soils (Kastanozems) in northern China while SIC contents were generally less than 1.0 kg C m^{-2} in tropical and subtropical soils in southern China.

Calcic horizons form through the process of dissolution of carbonates in the upper sections of soils and their subsequent precipitation into lower sections, closely related to climatic activities. Higher humidity levels prevail in southern China due to the influence of the Asian summer monsoon (Zhang, 1991) while the barrier effect of the Tibetan Plateau and the distance from ocean influence on moisture levels in northwestern China results in an arid and semiarid climate. The latter climate condition favors the formation and accumulation of calcic and petrocalcic horizons due to receiving little effective precipitation that leaches carbonates from soil systems (Lal and Kimble, 2000). SIC contents generally increase in correspondence with the increase of aridity

from east to west and from south to north. The lowest SIC contents were found in southern and northeastern China where strong carbonate leaching under humid climatic conditions is not in favor to its precipitation. The highest SIC contents, found in the Loess Plateau region, are also associated with the primary eolian deposit CaCO$_3$ source, containing high amounts of CaCO$_3$ (~10%) in dust transported from desert regions in northern China (Liu, 1985; Weng, 1989; Li et al., 1999).

The empirical relationship between SIC contents and climatic variables was established based on 798 analytical data that were measured in parallel at the same time for the non-cultivated soil profiles from the second national soil survey. The relationship was expressed as follows:

$$SICC = 13.51843 + 0.45263T - 0.02127P + 0.02127P + 1.12955$$
$$\times 10^{-4}T \times P - 0.01607T^2 + 5.47857 \times 10^{-6}P^2$$

$$(N = 798, \quad R = 0.58, \quad F = 80.10972)$$

where T is the mean annual temperature in degrees Celsius (degree), and P is the annual precipitation in millimeters (mm). The F test value ($F > F_{0.01} = 4.65$) indicates that the relationship is statistically significant. In general, SIC content in China is positively correlated with temperature and negatively correlated with precipitation. Results of this study are qualitatively consistent with those studies reported in Arizona in the USA (Schlesinger, 1982; Rasmussen, 2006) and those in northern China (Feng et al., 2001).

Four major climatic zones were delineated and calculated for SIC storage in different regions in China (Fig. 2a). These zones are the tropical-subtropical regions (Zone 1), the temperate monsoon regions in eastern China (Zone 2), the arid region in northwestern China (Zone 3), and the frigid region of the Tibetan Plateau (Zone 4).
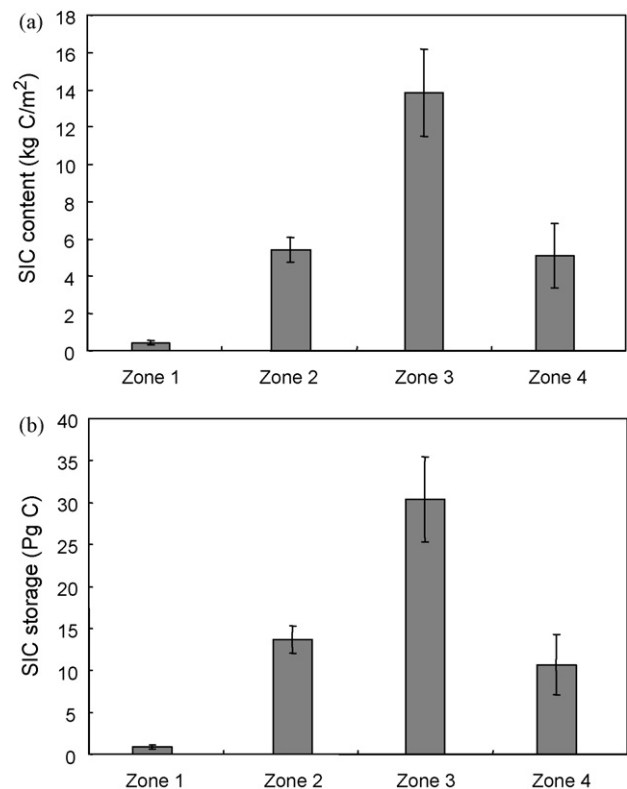

Fig. 3. SIC contents (a) and storages (b) under present-day conditions (including both cultivated and non-cultivated soils) in the four major climatic zones (seen in Fig. 2a) in China. The bars represent the standard errors.

The zones were mainly defined based on temperature and the appropriate degree of dryness (Zhang, 1991). The four zones are dominated by allitic, siallitic, arid, and alpine soils, respectively (Xiong and Li, 1987). In the arid zone (Zone 3) of northwestern China (Fig. 3), soils had the highest SIC content ($\sim$13.6 $\pm$ 2.3 kg C m^{-2}) and storage ($\sim$30.1 $\pm$ 5.1 Pg C). SIC content in the tropical-subtropical zone (Zone 1) was approximately 0.4($\pm$0.1) kg C m^{-2} and the C storage amounts to 0.8($\pm$0.3) Pg C. Moderate content and storage were observed for soils in the temperate zone (Zone 2) as well as in the Tibetan Plateau region (Zone 4) with contents of approximately 5.5($\pm$0.7) kg C m^{-2} for the former and 5.1($\pm$1.7) kg C m^{-2} for the latter. SIC storage of the two zones amounted to approximately 13.7($\pm$1.6) Pg C and 10.7($\pm$3.6) Pg C, respectively. Overall, the total current SIC storage in China was approximately 55.3($\pm$10.7) Pg C with an average SIC content of 6.3($\pm$1.2) kg C m^{-2}.

The measurement of approximately 55.3 $\pm$ 10.7 Pg C obtained by means of this study for the present-day SIC storage in China is consistent with the estimate of 60 Pg C obtained by Pan (1999) and Pan and Guo (2000). It is, however, lower than the estimate of 77.9–234.2 Pg C obtained by Li et al. (2007) that was also based on the data from the second national soil survey. The discrepancy may be related to two factors. First, the data of 34,411 profiles used in this study was much greater than the number of samples used in the study of Li et al. (2007) that was based on approximately 2456 profiles. Second, this study used the actual soil thickness measurement data obtained from the soil survey for calculating the profile C density while Li et al. (2007) assumed soil at a standard depth of 1 m (for 77.9 Pg C) and 3 m (for 234.2 Pg C).

The global SIC pool was estimated at approximately 700–1700 Pg C in upper 1 m depth using the similar method (Sombroek et al., 1993; Batjes, 1996; Eswaran et al., 1995, 2000) with a mean value of 950 Pg C (Lal, 2004). The SIC storage level in China therefore represents approximately 5.8% of the world's SIC pool, contrast with the approximate 6.4% share of the world's surface area. This similarity may be due to the extended terrestrial surface with widely spatial climatic and topographic variability in China that leads to a great diversity of soils, including latosols in the tropical zone, podzolic soils in the frigid-temperate zone, desert soils in the northwestern inland, solonchaks on the coastal plains, and alpine soils on the Tibetan Plateau (NSSO, 1998). Our result (5.8%) was relatively lower than the earlier estimate of world SIC (8.3%) reported by Li et al. (2007) for China, because the SIC pool in this study is only about 55.3 Pg C, while it is 77.9 Pg C estimated by Li et al. (2007).

3.2. Impact of agricultural land use on SIC

3.2.1. Spatial changes in SIC due to cultivation

Changes in SIC contents throughout cultivated soils in China due to historical land use activity was calculated by comparing the differences in SIC content under non-cultivated and cultivated conditions in the same subgroups, based on SIC data of 1630 cultivated and 923 non-cultivated soil profiles. Fig. 2b illustrates the spatial distribution for the difference in SIC content. Overall, approximately 51% of the total cultivated soil area experienced SIC loss. The regions that experienced significant C loss were located in eastern part of northern China. SIC had generally decreased by approximately 0.5–4.0 kg C m^{-2} where most C loss occurred in cultivated soils that include irrigated fields, dry farmland, and paddy fields.

SIC loss in these areas may be attributed to irrigation practices that increase carbonate leaching in soil profile (Magaritz and Amiel, 1981; Nordt et al., 2000; Sartori et al., 2007), particularly in the presence of acidifying fertilizers that increase nitrogen and sulphur inputs (Batjes and Sombroek, 1997). The loss may also be

attributed to agricultural practices that expose the calciferous horizon to the soil surface, increasing carbonate weathering and erosion (NSSO, 1998; Lal and Kimble, 2000). In paddy soils, SIC submerged for long periods each year favors carbonate dissolution (NSSO, 1998). The existing data makes it impossible to point out where the missing C has gone in terms of SIC loss. Certain amounts, for example, may have leached into deeper soil layers or may have weathered into bicarbonate that was transported into groundwater systems (Nordt et al., 2000; Kessler and Harvey, 2001) or, for that matter, washed down river surfaces and lakes where it was ultimately output into ocean systems (Probst, 1994; Raymond and Cole, 2003). The latter scenario is consistent with observed increases in dissolved inorganic C and Ca^{2+} in the Yellow River across northern China over the past 40 years (Chen and He, 2003). Because carbonate minerals only donate a second mole of C for every mole of CO$_2$ sequestered during carbonate weathering, therefore, a diminishing SIC reservoir needs an additional C from atmospheric CO$_2$ in order for it to be weathered into dissolved bicarbonate. For the leached C, some might be taken up by aquatic ecosystem and sequestered as organic/inorganic matter; others might release back to the atmosphere as CO$_2$ or converted to CH$_4$ by methanogens in aquatic environment (Cole et al., 2007).

On the other hand, an obvious SIC increase was observed in approximately 10% of the total cultivated surfaces (Fig. 2b). The increase in SIC density in these soils was between 0.5 and 5.0 kg C m^{-2} where the most significant increases were found in irrigated silty soils (Calcaric Fluvisols), irrigated desert soils (Calcaric Fluvisols), sierozems (Calcaric Cambisols) in northwestern China, and black soils (Phaeozems) in northeastern China. This phenomenon was also recorded in arid and semiarid soils in the USA (Cihacek and Ulmer, 2002; Entry et al., 2004), India (Pal et al., 2000), and in Chernozem soils in Russia (Mikhailova and Post, 2006).

The SIC increase is probably due to irrigation that leads to a dramatic increase in biomass production. This should result in increased plant respiration and microbial decomposition of organic matter and thus lead to increased soil CO$_2$ levels (Lal and Kimble, 2000), which will in turn result in increased weathering (Doner and Lynn, 1977) thus increase the consumption of atmospheric CO$_2$. It may also be the result of external sources of Ca^{2+} and Mg^{2+} as a consequence of an increase in water (Lal and Kimble, 2000) and lime by means of irrigation (West and McBride, 2005), and by fertilizer and manure additions to cropped fields (Mikhailova and Post, 2006) that increases carbonate formation. The total C gain was approximately 0.8 Pg C. On the other hand, potential CO$_2$ emissions may produce from irrigated agricultural land in arid regions based on the study of Schlesinger (1999). Because groundwater in these regions often contains high concentrations of bicarbonates, when such waters are applied to arid lands where are not alkaline soils, CO$_2$ is released into the atmosphere and CaCO$_3$ precipitates.

No significant changes in SIC attributable to cultivation were observed in approximately 39% of the total surface soils analyzed. This may be the result of these soils being located in areas primarily in southern and northeastern China where there is a lack of carbonate material in soils types. But, in fact, atmospheric CO$_2$ could also be consumed where carbonate rock weathering occurred due to the high humidity in these areas (Qiu et al., 2004). The accurate calculations of these weathering rates in future may help to reveal their contributions to the SIC changes.

3.2.2. Changes in SIC by means of different agricultural land use activities

Fig. 4 illustrates changes in SIC by means of three different land use conditions in China, that was, a sequential increasing loss in SIC
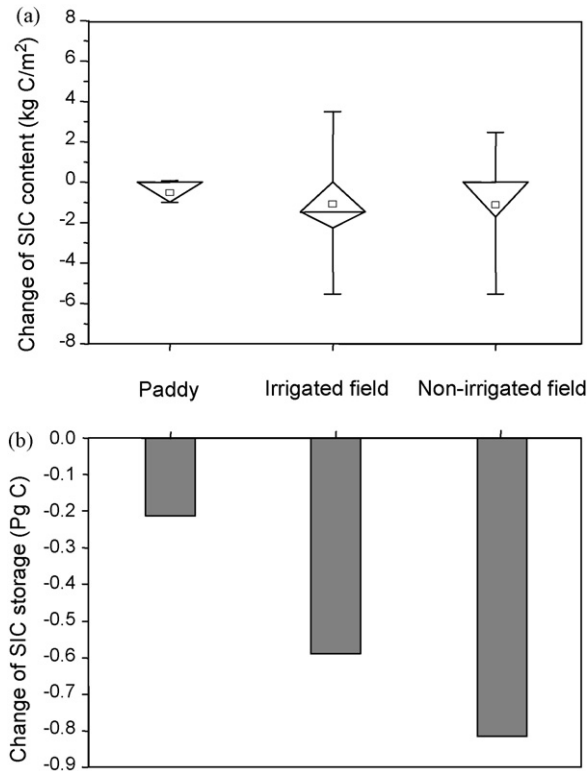
Fig. 4. Changes in SIC contents (a) and storages (b) for different land uses in China. Boxes indicate the interquartile intervals (25th and 75th percentiles), and bars represent 90% intervals (5th and 95th percentiles).

4. Conclusion

Based on the latest available soil profile data investigated by China's second national soil survey, the present-day SIC storage in China was estimated at approximately 55.3 ± 10.7 Pg C where the average C content was 6.3 ± 1.2 kg C m^{-2}. This represents approximately 5.8% of all the estimated SIC in the world. Furthermore, by comparing the difference in SIC densities under non-cultivated and cultivated conditions, approximately 1.6 Pg C of SIC was lost due to agricultural land use activity in China. Regions where significant SIC loss has occurred are located in the expansive cultivated soils in the eastern part of northern China dominated by land use activities such as irrigation, non-irrigation, and paddy fields. Results presented here suggest that human activity has significantly affected the SIC pool that plays an important role in the global C cycle. The estimates contributed by means of this study present a new attempt to provide a realistic picture on the current spatial variation of SIC in China while offering a baseline to estimate potential C sequestration through land use changes for soil types in China under rising atmospheric CO_2 conditions.

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content in paddy soils, non-irrigated fields, and irrigated fields (Fig. 4a). SIC storage loss was approximately 0.2 Pg C for paddy soils, 0.6 Pg C for irrigated fields, and 0.8 Pg C for the non-irrigated fields (Fig. 4b). Non-irrigated fields had the largest SIC loss mainly due to its large expanse in area ($\sim$53% of all cultivated surfaces) (Tian et al., 1996b).

Total SIC loss in China was approximately 1.6 Pg C, representing 3% C loss for present-day conditions. The climate change contribution to SIC loss may be negligible since all data used in this study originated from the same national soil survey that lasted for only a few years. Consequently, SIC loss is primarily attributed to land use activity throughout history.

Compared with SIC change in China, SOC loss was approximately 7.1 Pg C due to land use (Wu et al., 2003). A significant SOC loss was observed within a belt from northeast to southwest China, mainly in non-irrigated soils under semiarid and semihumid conditions with the maximum SOC loss in northeast China. In contract, the paddy soils in southern China and some irrigated soils in northwest China showed a net gain in SOC. No significant changes were observed in other irrigated soils in eastern China. Therefore, the impacts of agricultural land use on SIC and SOC pools in soils were different. For the SOC, the C loss means it is the source of atmospheric CO_2. While for the SIC, if the loss of leached carbonate remains in the lower soil profile or resides in river systems, groundwater, or oceans for a 100 years or more (Probst, 1994; Nordt et al., 2000; Kessler and Harvey, 2001; Raymond and Cole, 2003), current SIC budget accounting suggests that a considerable temporary mechanism that enhances atmospheric CO_2 sequestration with the SIC pool due to historical land use activity will result. Therefore, in the future, a better and integrated understanding of SOC and SIC changes in soils will be critical for reducing the uncertainty in C fluxes from the soils.

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