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Spatial distributions and sequestrations of organic carbon and black carbon in soils from the Chinese loess plateau

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HIGHLIGHTS

- ► There was a wide variability of SOC, BC, char and soot in the soils from the CLP.
- ► The highest SOC concentrations were found in the clayey loess zone.
- ► There was far more char than soot when BC was partitioned into char and soot.
- ► There was a strong correlation between char and BC.

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ABSTRACT

Concentrations of soil organic carbon (SOC), black carbon (BC), char, and soot in topsoils (0–20 cm) and vertical soil profiles (0–100 cm) from the Chinese Loess Plateau (CLP) were investigated. Objectives of the study were to establish the spatial distributions and estimate the sequestrations of these substances. The SOC, BC, char and soot concentrations were higher in the eastern and southeastern parts of the plateau and lower in the north, which is consistent with the patterns of economic development and energy consumption. The highest average SOC concentration was found in the clayey loess zone, followed by the loess and sandy loess zones. Similar trends were observed for BC, char and soot, suggesting interactions with clay and silt are potentially important influences on OC and BC. The SOC contents in topsoils varied from 0.31 to 51.81 $g\ kg^{-1}$, with a mean value of 6.54 $g\ kg^{-1}$, while BC and char concentrations were 0.02 to 5.5 $g\ kg^{-1}$ and 0.003 to 4.19 $g\ kg^{-1}$, respectively, and soot ranged from 0.01 to 1.32 $g\ kg^{-1}$. Unlike SOC, both BC and char decreased with soil depth, whereas soot showed little variation with depth. BC and char were correlated in the topsoils, and both correlated moderately well with SOC ($R^2=0.60$) and soot ($R^2=0.53$). The SOC pools sequestered in the 0 to 20 cm and 0 to 100 cm depths were estimated to be 0.741 and 3.63 Pg, respectively, and the BC pools sequestered in the 0 to 20 cm and 0 to 100 cm depths were 0.073 and 0.456 Pg, respectively. Therefore the quantity of carbon stored in the sediments of the CLP evidently exceeds 10^9 tons. The char contained in the upper 20 cm layer was 0.053 Pg, which amounted to 72.5% of the BC in that layer.

1. Introduction

Concerns over global climate change have fueled interest in the carbon cycle, and this has impelled scientists to study the distributions of various forms of carbon in terrestrial ecosystems, and to estimate the amounts of carbon stored in various reservoirs. The distribution and storage of soil organic carbon (SOC) are highly relevant to studies of climate and climate change, because this material can influence the mixing ratios of atmospheric carbon dioxide and other gases, such as methane, that affect the earth's radiative balance. Owing to the large quantities of C sequestered in terrestrial ecosystems, changes in the sizes of the SOC pools could affect the amounts of CO_2 and other gases in the atmosphere, and potentially influence global climate.

Black carbon (BC) is an important component of organic carbon (OC), and it is generally defined as the carbonaceous material that forms during the incomplete combustion of fossil fuels and biomass burning. In reality, BC consists of a continuum of compounds ranging from slightly-charred biomass to highly-condensed and refractory soot. This is important from an environmental standpoint because these various compounds possess different physical and chemical properties and, therefore, can affect climate and be affected by climate in different ways (Goldberg, 1985; Schmidt and Noack, 2000). Studies have shown that BC can make up a significant proportion of the organic carbon in soils, but the amounts have been found to differ considerably

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among soil types. For example, BC has been reported to account for 1.6 to 4.5% of the SOC in Siberian boreal forest soils (Czimczik et al., 2003), but it contributed up to 45% of the SOC in German chernozemic soils (Schmidt et al., 1999). Due to its chemical and biological stability, it has been suggested that the BC sequestered in soils and sediments may represent a significant sink for carbon. The storage and release of BC thus have major implications for biogeochemical cycles over the short-term and the carbon cycle over the long-term (Kuhlbusch and Crutzen, 1995; Schmidt and Noack, 2000).

Char and soot are two of the most abundant and geochemically important components of the BC continuum (Elmquist et al., 2006). However, owing to limitations in the most commonly used analytical methods, many of the existing studies have focused on only one component or an operationally-defined fraction of the BC continuum (Schmidt et al., 2001). Recently, Han et al. (2007) suggested that a thermal optical reflectance (TOR) method could be used to distinguish char from soot by oxidizing the various carbon fractions in a stepwise manner, that is, different temperatures and mixtures of gases could be used to separate the materials of interest prior to instrumental analysis. Indeed, this technique has been successfully used to determine char and soot concentrations in sediments (Han et al., 2011) and aerosol particles (Han et al., 2009c, 2010b; Lim et al., 2012).

Studies of SOC storage on global to regional scales (Batjes, 1996; Bhatti et al., 2002; Eswaran et al., 1993) and investigations of BC concentrations and fluxes (Ohlson et al., 2009; Preston and Schmidt, 2006; Schmidt and Noack, 2000) have been undertaken for many years. However, the quantities of BC in terrestrial soils are still poorly unknown (Schmidt and Noack, 2000). In China, a series of studies has focused on SOC, including some investigations of SOC in relation to soil types and vegetation; and some preliminary assessments of the effects of cultivation and land use on SOC concentrations also have been made (Chen et al., 2007; Fu et al., 2010; Lal, 2002; Pan et al., 2004; Wang et al., 2002; Wu et al., 2003b; Yu et al., 2009). Chinese Loess Plateau, which is overlain by a mantle of fine-grained, wind-deposited, yellowish alluvium known as loess, covers areas of 440,000 km^2 and tens to hundreds of meters thick (Liu, 1985). Due to natural drought conditions and intensive human activities such as deforestation, overgrazing and land reclamation, soil erosion and desertification in these regions are very serious. With the implementation of "Grain-for-Green" project (Uchida et al., 2005), estimation of SOC distributions and sequestration on the CLP is of great importance to evaluate the environmental effect of policy of grain for green. Research on distribution and storage of SOC on the Chinese Loess Plateau (CLP) has been limited to a few studies by Xu et al. (2003), Han et al. (2010a) and Wang et al. (2010). However, less is known about the spatial distributions and stocks of BC, char, and soot in soils.

Therefore, the main objectives of this study were to apply the thermal optical reflectance (TOR) method to study the spatial distributions and storages of SOC, BC, char and soot in topsoils (0 to 20 cm) and in loess-paleosol sections (0 to 100 cm) from the CLP. In addition, we examined the data for quantitative relationships among SOC, BC, char and soot, and we used the data to identify the most probable sources for these substances. As this is the first in-depth investigation of BC, char and soot in soils from the CLP, the basic data will contribute to our general understanding of their biogeochemical cycling and relationships to global climate change.

2. Materials and methods

2.1. Study area

Located in the upper and middle reaches of the Yellow River in the northern China (100°–114°E, 34°–41°N), the CLP is the largest region on earth covered by loess, and it also holds the world's thickest loess deposits. What is more, the CLP was the cradle of Chinese civilization, and even today it is one of China's most important agricultural areas. Meanwhile, it is also among the areas most susceptible to soil erosion, and its ecology is highly vulnerable to perturbation and change. The CLP extends over eight provinces and autonomous regions: Shanxi, central and northern Shaanxi, southeastern Gansu, southern Ningxia, northeastern Qinghai, northern Henan, southern Inner Mongolia and a small part of northwestern Hebei. The average altitude varies from 1000 to 2000 m. The climate of the CLP ranges from temperate arid to semi-arid, and the weather is generally warm and rainy in summer but cold and dry in winter. Annual average precipitation ranges from 185 to 750 mm but is typically 300 to 600 mm. The rainfall from May to September accounts for approximately 68 to 87% of the total annual precipitation. Annual average temperature ranges from 3.6 to 14.3 °C, with extreme low temperatures from −13.9 to −38.2 °C. The main soil types on the CLP have been classified as cinnamon, Heilu, chestnut, brown calcic, sierozem, gray desert, loessal, and sandy. The loess of the middle reaches of Yellow River can be divided into three zones on the basis of particle size, these are: sandy loess, loess and clayey loess (Liu, 1985).

2.2. Soil sampling and sample preparation

A total of 260 topsoil samples (0 to 20 cm) were collected with a steel shovel from Shanxi, Shaanxi, Gansu Provinces, the Inner Mongolian Autonomous Region, and the Ningxia Hui Autonomous Region between October 2009 and July 2010. Sampling locations were chosen to have minimal human disturbance, to be 1 to 3 km away from the towns and residential areas, and to be approximately 0.5 km away from roads (Fig. 1). Using an "S-shaped" design within a radius of approximately 30 cm, five separate sub-samples were taken from the topsoil, and then composited to form a representative sample for each site. In addition to the topsoil samples, 18 vertical soil cores were collected from the surface to a depth of 100 cm with a soil auger (4 cm diameter × 100 cm). Each soil core was cut into six layers (0 to 10, 10 to 20, 20 to 40, 40 to 60, 60 to 80 and 80 to 100 cm).

Before analysis, all the samples were air dried at ambient temperature for one week. Then they were processed through a 2 mm sieve to remove gravel and coarse organic debris, grounded with an agate mortar, and finally passed through a 63 μm sieve to homogenize the particles. The bulk density of each layer in the soil profiles was determined by taking three to four replicate samples for each soil horizon.

2.3. Organic carbon analysis

Soil samples were first treated with 1 M HCl to remove inorganic carbon. The residues were then centrifuged (4500 rpm, 12 min) and rinsed three times with deionized (DI) water and air dried overnight at 60 °C. Organic carbon content was determined by CuO-catalyzed dry combustion with the use of a CHN elemental analyzer (ELEMENTAR Vario EL, Hanau, Germany). The minimum detection level for C was 0.4 μg. Each prepared sample was analyzed in triplicate with the CHN analyzer, and an average value was adopted for this study.

2.4. Black carbon measurement

The BC in the samples was chemically separated from other materials prior to the instrumental analysis. The pretreatment procedures involved the following four steps: (1) removal of inorganic carbonates via acidification; (2) destruction of silicates and any carbonaceous materials trapped between the silicate sheets (Song et al., 2002); (3) elimination of minerals such as fluorite that formed during the demineralization procedure; (4) filtration with a quartz membrane filter. Briefly, the loess samples were decarbonated using 2 M HCl; this reaction was allowed to proceed overnight. The residue was washed with DI water, centrifuged, and then treated with a mixture of 46% HF and 6 M HCl (Volume$_{HF}$:Volume$_{HCl}$ = 1:1) for 24 h to remove silicates. The mixture was centrifuged, resuspended and rinsed three times with DI water and then treated with 4 M HCl overnight to remove residual fluorite and carbonate. The remaining sample material was rinsed with DI water until the pH of the aqueous phase

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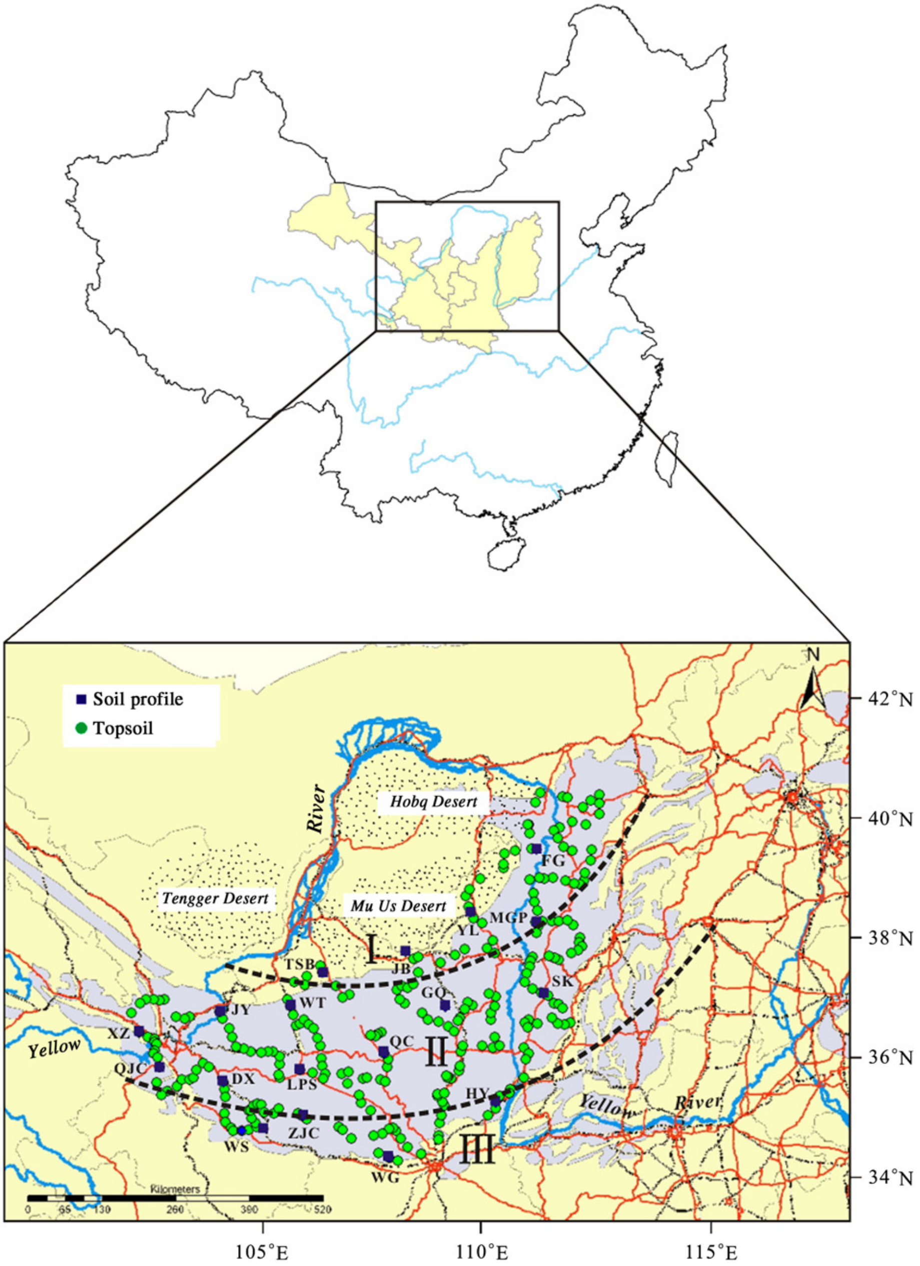

Fig. 1. Locations of the sampling sites. The dashed lines separate the regions into zones (I) sandy loess zone; (II) loess zone; (III) clay loess zone (after Liu, 1985). Abbreviations for the sites are as follows: Gaoqiao (GQ); Jingbian (JB); Yulin (YL); Fugu (FG); Muguaping (MGP); Shikou (SK); Heyang (HY); Qingcheng (QC); Tianshuibao (TSB); Wangtuan (WT); Liupanshan (LPS); Jingyuan (JY); Xinzhuang (XZ); Qianjincun (QJC); Dingxi (DX); Wushan (WS); Zhangjiachuan (ZJC); Wugong (WG).

rose to pH 4 to 5—this was to avoid the formation of a colloidal suspension of the BC (Lim and Cachier, 1996). Then it was filtered through a 47 mm quartz filter (Whatman International Ltd., Maidstone, England). Finally, the filter sample was dried in an oven at 40 °C for 6 to 8 h and then put in a circular petri dish and stored in a refrigerator.

A DRI Model 2001 Thermal/Optical Carbon Analyzer (Atmoslytic Inc., Calabasas, CA) was used for black carbon analysis following the IMPROVE_A protocol (Chow et al., 2007; Han et al., 2009a). Detailed information on the procedure is available in Han et al. (2009b). Data for four fractions of organic carbon (OC1, OC2, OC3 and OC4 at 140 °C, 280 °C, 480 °C and 580 °C in a He atmosphere, respectively) and three elemental carbon fractions (EC1, EC2 and EC3 at 580 °C, 740 °C and 840 °C in a 2% O_2/98% He atmosphere, respectively) were produced using this procedure. A pyrolized carbon (PC) fraction, produced in an inert atmosphere, also was obtained, which was used to correct for charred OC. Char and soot are operationally defined as EC1 − PC and EC2 + EC3, respectively, according to the study of Han et al. (2007). The quantification limit is 0.45 μg cm^{-2} for OC, and 0.06 μg cm^{-2} for EC.

2.5. Storage estimation

SOC density (SOCD, kg m^{-2}) for each soil profile with a depth of 100 cm was calculated following the approach of Wu et al. (2003a):

$$SOCD=\sum_{i=1}^{n}(1-\delta_i\%)\times T_i\times\rho_i\times C_i\times 10^{-1},$$

where n is the number of layers involved, $\delta_i\%$ represents the volumetric percentage of the fraction >2 mm (rock fragments), ρ_i is the soil bulk density of a soil layer i (g cm^{-3}), C_i is the organic C content (g kg^{-1}), and T_i represents the thickness of the soil layer (cm). As

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Table 1
Soil organic carbon (SOC), black carbon (BC), char and soot concentrations in topsoils from different loess zones.

Analyte	Statistic	Zone			All samples
		Sandy loess	Loess	Clayey loess	
SOC	Range[a]	0.31–3.56	1.16–22.70	1.85–51.81	0.31–51.81
	Mean[a] ± SD[a,b]	3.54 ± 2.70	7.11 ± 4.35	10.02 ± 9.82	6.54 ± 5.36
BC	Range	0.02–1.85	0.06–3.20	0.22–5.50	0.02–5.50
	Mean ± SD	0.37 ± 0.34	0.69 ± 0.55	0.99 ± 1.07	0.65 ± 0.62
Char	Range	0.003–1.74	0.03–2.14	0.04–4.19	0.003–4.19
	Mean ± SD	0.28 ± 0.31	0.49 ± 0.47	0.74 ± 0.91	0.47 ± 0.53
Soot	Range	0.01–0.22	0.04–1.20	0.12–1.32	0.01–1.32
	Mean ± SD	0.09 ± 0.04	0.20 ± 0.14	0.25 ± 0.21	0.18 ± 0.15
BC/SOC	Range	3.23–36.12	2.39–35.55	2.57–36.87	2.39–36.87
	Mean ± SD	10.69 ± 5.61	10.91 ± 6.41	11.57 ± 7.29	10.93 ± 6.31
Number of samples		67	162	31	260

[a] Range, mean and standard deviation in g kg^{-1} dry weight.
[b] SD stands for standard deviation.

the particles in Chinese loess are mostly below 2 mm, the fraction of the total mass of the topsoil > 2 mm is usually negligible.

The total SOC storage (SOCS, Pg) of soils can be estimated as follows:

$$SOCS = \sum_{j=1}^{n} area \times SOCD_j,$$

where *area* and $SOCD_j$ are the surface area of CLP and the organic carbon density of profile *j*, respectively. The BC, char and soot pools can be calculated using the same approach. In this study, only the upper 20 cm were considered for the calculation of the topsoil carbon pools.

3. Results and discussion

3.1. Spatial distribution of SOC, BC, char, and soot in topsoils

The concentration of SOC, expressed on a dry weight basis, ranged from 0.31 to 51.81 g kg^{-1}, with a mean value of 6.54 g kg^{-1} (Table 1). These values are much smaller than those reported for an unburned Siberian forest soil (250 g kg^{-1}, Czimczik et al., 2003) or woodland soils of the UK (249 g kg^{-1}, Nam et al., 2008), but they are comparable to the soils of the Yangjuangou watershed, which is located in the middle of the CLP (6.96 g kg^{-1}, Wang et al., 2010). The average SOC concentration was the highest in the clayey loess zone, followed by the loess and sandy loess zones (Table 1). Spatial distribution plots showed that the SOC concentrations in the western, southern, eastern and central parts of the CLP were higher than those in the north (Fig. 2a). The highest SOC occurred in the southwestern part of the CLP, and that may have been a result of a high litter input to forestland sites. In contrast, the region that had the lowest SOC content is economically underdeveloped and adjacent to the desert.

Generally, the contents of the carbonaceous substances in the clayey loess zone were roughly 3 times those in sandy loess zone and about 1.4 times those in loess zone. One implication of this pattern is that it suggests that SOC is likely to be trapped within or in some other way associated with clay minerals. The proportions of fine particles (i.e. the silt and clay fractions) in the clayey loess are higher than those in sandy loess and loess (Liu, 1985). This is significant from a geochemical perspective because the greater surface area-to-volume ratios of the finer particles provide more potential binding sites for the organics. The zonation in particle size exhibited by the soils is a result of the winnowing of the eolian particle population during transport. Gravitational settling preferentially removes coarse particles from suspension,

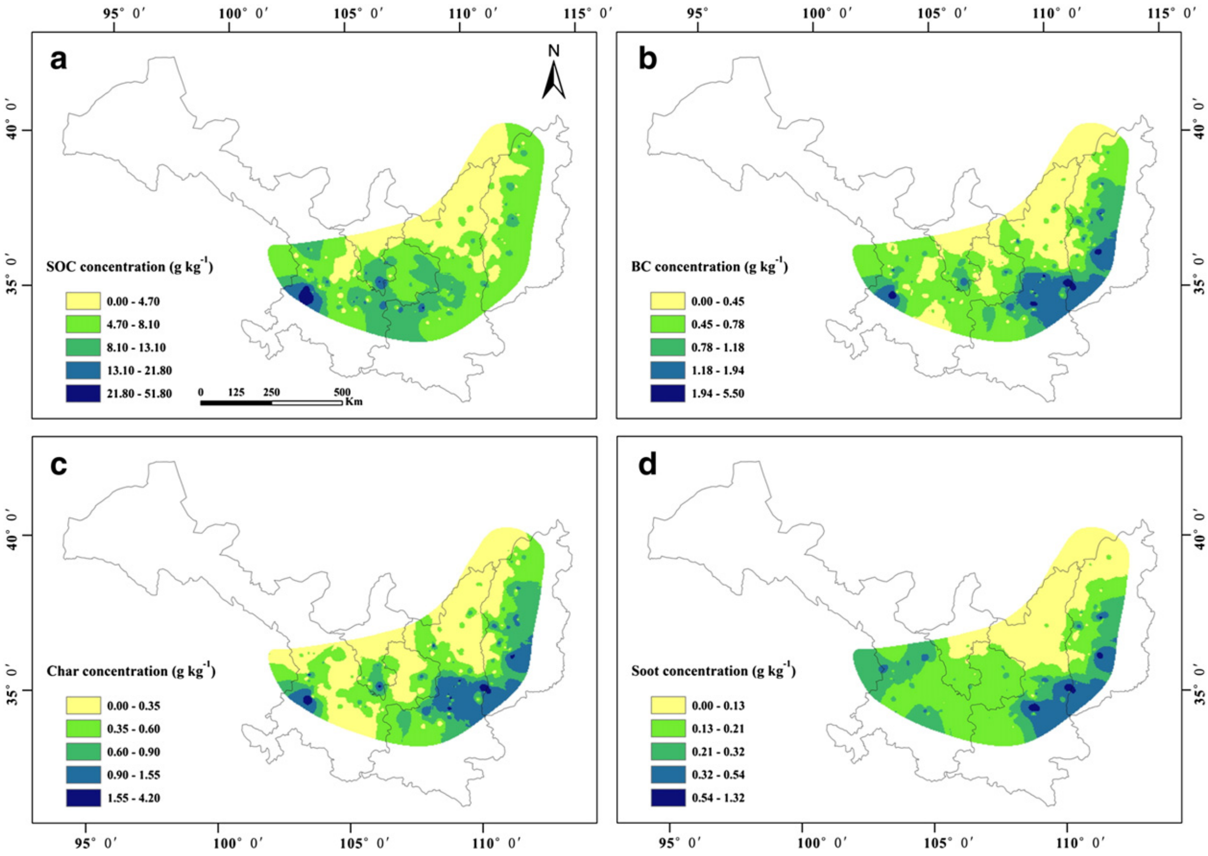

Fig. 2. Spatial distributions of SOC, BC, char and soot concentrations in CLP.

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and therefore, it is the finer ones that are more easily transported long distances. The higher numbers of fine particles in the clayey loess thus indicate that the influences of distant dust sources were relatively stronger when the clayey loess was being deposited compared with when the coarser materials were deposited. This suggests that differences in the amounts of carbon in the dust source regions also may have contributed to the spatial variability in SOC concentrations.

Previous studies have shown correlations between SOC concentrations and clay content (Burke et al., 1989; Glaser and Amelung, 2003; Rumpel et al., 2006), and it has been suggested that interactions of organic carbon with clay minerals can stimulate the formation of organo-mineral complexes, and in this way the SOC can be stabilized (Baldock and Skjemstad, 2000; Czimczik and Masiello, 2007; Schulze and Freibauer, 2005). Indeed, higher clay contents also can inhibit microbial decomposition, and this can lead to the accumulation of SOC. Plant growth also could be favored by clayey soils, and this, too, could increase C contents (Jobbágy and Jackson, 2000). Therefore, it appears that the clay content and interactions with clay and silt are potentially important influences on the concentrations and behavior of OC in soils. Moreover, the large grain-size of sandy loess is consistent with the low SOC concentrations found in that zone. Borchers and Perry (1992), for example, observed that coarse soils had lower total SOC concentrations than silt-loam or sandy-loam soils.

The BC, char and soot contents quantified using the TOR method were estimated to be 0.02 to 5.5 g kg^{-1}, 0.003 to 4.19 g kg^{-1}, and 0.01 to 1.32 g kg^{-1}, respectively (Table 1). The arithmetic mean BC content (0.65 g kg^{-1}) was much lower than that reported for German chernozemic soils (7.6 g kg^{-1}, Schmidt et al., 1999) or soils from the burned forest floor of a Siberian Scots pine forest (13.5 g kg^{-1}, Czimczik et al., 2003) but similar to the levels observed in soils of Tenerife, in the Canary Islands (0.76 g kg^{-1}, Ribes et al., 2003). Overall, BC composed a significant proportion (2.39 to 36.87%) of the SOC, and it was more variable than what has been reported in several previous studies (Table 3). This is possibly related to the different inputs and sources of BC, as well as differences among the physical and chemical properties of the soils. Additionally, the BC quantification methods used for the various studies can measure different fractions of the BC continuum (Hammes et al., 2007), and this complicating issue must be acknowledged. As BC shares some chemical properties with other components of the SOC pool (Preston and Schmidt, 2006), we suggest that BC and SOC may have some similar interactions with the mineral matrix, and in this way BC could be protected from decomposition.

High concentrations of BC were found in samples from the east and south, and much lower levels were seen in the north (Fig. 2b). Consistently high char and soot also were found in the eastern and southeastern CLP (Fig. 2c, d), which is consistent with the population density (Fig. S1), or to say with the patterns of economic development and energy consumption. As BC is produced from the burning of biomass and fossil fuels, we propose that in addition to the influences from natural wildfires, the spatial heterogeneity of BC is mainly linked to anthropogenic activities. Firstly, it is likely that motor vehicle exhaust emissions and coal combustion contribute to the high atmospheric pollution loadings in the east, south and southeast, and these sources may be responsible for the high BC levels in those regions. Secondly, during the time of wheat and corn harvest, straw clearance by in situ burning on the farmland is very prevalent in these regions (Cao et al., 2005), which is also a big source of BC; moreover, land reclamation would lead to grassland and forest fires (Huang et al., 2006). These agricultural activities can significantly increase the production of BC. BC can be transported over long distances to the remote open ocean (Masiello and Druffel, 1998) and polar regions (Hansen et al., 1988). Thus, pollution sources in the eastern and central China also may have elevated the BC in the western and northern parts of CLP. Our findings are in accordance with a study showing that surface soils from industrial sites in Delhi had the highest mean BC concentrations, and the concentrations decreased from a highly trafficked site to a floodplain and decreased further from an agricultural area to increasingly remote sites (Agarwal and Bucheli, 2011). In summary, the spatial patterns in BC evidently reflect the air pollution load in these regions, at least to a degree, and therefore observed variations in soil BC concentrations are likely influenced by anthropogenic activities as well as natural wildfires. The source of char is largely emitted from biomass and coal combustion, because coal burning is very prevalent both in the urban cities and countryside on the CLP in the seasons of winter heating. Furthermore, straw burning for improving soil fertility during the crop harvested seasons, combined with land reclamation, are probably responsible for the char emission.

3.2. Vertical distributions of SOC, BC, char and soot in soil profiles

In general, the SOC concentrations decreased with depth (Table 2). The average SOC abundance in the 0 to 10 cm layer for all profiles (4.79 g kg^{-1}) was approximately three times the average for the 80 to 100 cm layer (1.69 g kg^{-1}). In some profiles, however, most notably QC, TSB, WT, and WS (Table 2), the SOC concentrations were seen to increase again in the deeper layers. Indeed, the highest SOC in the WT profile was found in the 80 to 100 cm layer. The maximum SOC concentration observed in the study was found in 0 to 10 cm layer of the LPS profile (Table 2), which was probably due to the dense vegetative cover in the region. SOCD occasionally decreased with depth in some profiles, which is likely attributed to the distribution of roots in the soil and associated soil processes. The mean SOCD was calculated to be 5.99, 4.31, 6.53, 5.21, 4.55, and 4.19 kg C m^{-2}, respectively, in the 0 to 10 cm, 10 to 20 cm, 20 to 40 cm, 40 to 60 cm, 60 to 80 cm, and 80 to 100 cm layers. It seems that the highest SOCD was always found in the 20 to 40 cm layer (Table 2), and then decreased with depth in the 40 to 100 cm layers for some profiles. Possible reasons for that are the effects of physical and chemical properties of the soils (e.g., pH value, soil bulk density, and mineral elements), or interference of external environment (e.g., vegetation types, microclimate, biological activity).

Several sampling sites (FG, YL, TSB and JB) are located in the sandy loess zone adjacent to the Mu Us Desert, and the mineral particles in these areas are strongly affected by dust storms and wind erosion. As a consequence, conditions there are generally favorable for the decomposition of OC, and this leads to relatively low carbon concentrations (Wang et al., 2006). Furthermore, low soil moisture in the semi-arid lands would support only sparse vegetation cover, and this in turn would cause low inputs of plant matter and low fire-induced BC deposition to the soils. These are other possible reasons for low carbon concentrations throughout the profiles from the four sites near the Mu Us Desert.

In most of the profiles, the BC concentrations determined by TOR decreased with depth, but the concentrations varied greatly in several profiles (Fig. 3). BC measured with benzene polycarboxylic acids molecular markers (BPCAs) method in zonal steppe soil profiles from Russia also showed great variations with soil depth (Rodionov et al., 2006). The amount of BC averaged over all depths for all of the profiles was 1.85 g kg^{-1}, which was about four times that in the 0 to 10 cm layer (0.48 g kg^{-1}). BC concentrations in the 0 to 30 cm layer observed in this study (0.10 to 1.05 g kg^{-1}) were much lower than the values reported previously for Swiss soils at the same depth (10.7 to 91.5 g kg^{-1}, Leifeld et al., 2007), but the range of BC concentrations at 80 to 100 cm (0.05 to 0.69 g kg^{-1}) was significantly higher than Russia steppe soil (0.01 ± 0.0 g kg^{-1}, Hammes et al., 2008). In the soil profiles from WT and DX, the BC concentrations increased approximately 2 to 3 times from the 0 to 10 cm to the 80 to 100 cm layers.

The BC distributions in the soils may have been influenced by pedogenic processes (Masiello, 2004). Moreover, vertical movements of BC in mineral soils are another potential source for variability in the data (Dai et al., 2005; Leifeld et al., 2007; Major et al., 2010); indeed, these processes may be responsible for the high BC concentrations in

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Table 2
Chemical and physical properties of the soil profiles.

Site	Location	Altitude (m asl)[a]	Depth (cm)	SOC (g kg^{-1})	Bulk density (g cm^{-3})	Density (kg C m^{-2})			
						SOC	BC	Char	Soot
Gaoqiao (GQ)	109.19°N, 36.66°E	1149	0–10	3.25	1.31	4.26	0.28	0.13	0.15
			10–20	2.53	1.30	3.29	0.30	0.08	0.22
			20–40	1.22	1.29	3.16	0.38	0.10	0.29
			40–60	0.75	1.28	1.91	0.45	0.15	0.30
			60–80	0.74	1.27	1.88	0.44	0.17	0.28
			80–100	0.76	1.28	1.94	0.45	0.17	0.28
Jingbian (JB)	108.45°N, 37.51°E	1397	0–10	1.04	1.44	1.49	0.27	0.17	0.10
			10–20	1.00	1.49	1.49	0.22	0.11	0.10
			20–40	0.92	1.42	2.62	0.40	0.20	0.21
			40–60	0.72	1.51	2.18	0.45	0.21	0.24
			60–80	0.62	1.56	1.93	0.53	0.26	0.27
			80–100	0.48	1.49	1.43	0.30	0.08	0.23
Yulin (YL)	108.45°N, 38.13°E	1002	0–10	0.53	1.69	0.89	0.16	0.03	0.14
			10–20	0.54	1.77	0.95	0.18	0.03	0.15
			20–40	0.43	1.71	1.47	0.33	0.06	0.27
			40–60	0.37	1.73	1.33	0.34	0.09	0.25
			60–80	0.39	1.49	1.16	0.31	0.06	0.25
			80–100	0.45	1.63	1.47	0.27	0.03	0.24
Fugu (FG)	111.15°N, 39.11°E	914	0–10	1.48	1.42	2.10	0.23	0.07	0.17
			10–20	0.51	1.52	0.77	0.24	0.09	0.15
			20–40	0.47	1.52	1.43	0.44	0.15	0.28
			40–60	0.45	1.53	1.38	0.44	0.14	0.30
			60–80	0.41	1.52	1.23	0.38	0.10	0.28
			80–100	0.40	1.52	1.23	0.43	0.14	0.29
Muguaping (MGP)	111.09°N, 37.99°E	1119	0–10	3.61	1.23	4.44	0.49	0.28	0.21
			10–20	1.89	1.26	2.38	0.37	0.18	0.19
			20–40	1.41	1.32	3.72	0.66	0.37	0.29
			40–60	1.32	1.30	3.42	0.58	0.35	0.23
			60–80	1.23	1.39	3.42	0.56	0.35	0.21
			80–100	1.22	1.41	3.45	0.58	0.37	0.21
Shikou (SK)	111.16°N, 36.90°E	1422	0–10	9.88	1.37	13.54	0.70	0.47	0.23
			10–20	4.50	1.37	6.17	0.53	0.38	0.15
			20–40	3.19	1.38	8.81	0.88	0.62	0.25
			40–60	3.15	1.32	8.31	0.60	0.30	0.29
			60–80	2.95	1.29	7.60	0.64	0.34	0.29
			80–100	2.62	1.30	6.82	0.82	0.60	0.22
Heyang (HY)	110.12°N, 35.21°E	708	0–10	2.44	1.15	2.81	0.58	0.26	0.32
			10–20	1.72	1.22	2.10	0.39	0.18	0.20
			20–40	0.61	1.47	1.79	0.39	0.13	0.27
			40–60	0.58	1.47	1.71	0.38	0.13	0.24
			60–80	0.55	1.48	1.64	0.33	0.11	0.22
			80–100	0.49	1.46	1.42	0.32	0.10	0.22
Wangtuan (WT)	106.10°N, 36.67°E	1444	0–10	1.74	1.26	2.19	0.30	0.22	0.08
			10–20	2.10	1.29	2.70	0.58	0.49	0.09
			20–40	1.88	1.30	4.87	0.62	0.46	0.16
			40–60	1.46	1.41	4.12	0.81	0.61	0.20
			60–80	1.51	1.33	3.99	0.58	0.44	0.14
			80–100	2.84	1.23	6.97	1.70	1.48	0.22
Qingcheng (QC)	107.93°N, 35.95°E	1091	0–10	6.88	1.31	9.01	0.77	0.63	0.14
			10–20	3.24	1.29	4.19	0.43	0.30	0.12
			20–40	2.39	1.26	6.04	0.87	0.58	0.29
			40–60	1.71	1.31	4.49	1.05	0.69	0.36
			60–80	2.01	1.31	5.25	0.91	0.60	0.30
			80–100	1.47	1.32	3.89	0.51	0.29	0.22
Zhangjiachuan (ZJC)	106.24°N, 34.98°E	1724	0–10	7.43	1.15	8.54	1.17	1.03	0.14
			10–20	7.71	1.11	8.53	0.54	0.39	0.15
			20–40	6.68	1.11	14.83	1.34	1.04	0.29
			40–60	6.06	1.15	13.97	1.34	1.15	0.19
			60–80	3.05	1.11	6.79	0.65	0.46	0.19
			80–100	3.33	1.13	7.51	1.06	0.82	0.24
Tianshuibao (TSB)	106.77°N, 37.19°E	1513	0–10	2.40	1.26	3.03	0.21	0.14	0.07
			10–20	1.99	1.34	2.67	0.20	0.10	0.09
			20–40	1.10	1.42	3.12	0.41	0.29	0.12
			40–60	0.91	1.42	2.60	0.16	0.09	0.07
			60–80	1.60	1.36	4.36	0.30	0.16	0.14
			80–100	0.87	1.42	2.48	0.15	0.07	0.09
Liupanshan (LPS)	106.24°N, 35.68°E	1437	0–10	13.66	1.17	15.98	0.91	0.79	0.12
			10–20	10.93	1.13	12.32	0.49	0.40	0.09
			20–40	9.77	1.10	21.44	0.68	0.48	0.20
			40–60	5.23	1.18	12.35	1.09	0.94	0.15
			60–80	2.98	1.14	6.77	0.59	0.44	0.15
			80–100	2.45	1.14	5.60	0.28	0.19	0.09

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Table 2 (*continued*)

Site	Location	Altitude (m asl)[a]	Depth (cm)	SOC $(\mathrm{g\ kg^{-1}})$	Bulk density $(\mathrm{g\ cm^{-3}})$	Density $(\mathrm{kg\ C\ m^{-2}})$			
						SOC	BC	Char	Soot
Jingyuan (JY)	104.68°N, 36.56°E	1431	0–10	3.60	1.11	4.00	0.57	0.44	0.13
			10–20	3.29	1.26	4.15	0.59	0.42	0.17
			20–40	2.66	1.26	6.68	0.57	0.35	0.22
			40–60	1.81	1.28	4.63	0.52	0.26	0.26
			60–80	1.77	1.32	4.65	0.51	0.32	0.19
			80–100	1.85	1.20	4.44	0.47	0.29	0.18
Dingxi (DX)	104.69°N, 35.48°E	1869	0–10	8.14	1.16	9.44	0.48	0.34	0.13
			10–20	8.07	1.04	8.42	0.28	0.15	0.12
			20–40	6.46	1.10	14.21	0.85	0.57	0.28
			40–60	6.13	1.12	13.72	1.31	1.01	0.30
			60–80	6.17	1.10	13.64	0.87	0.55	0.32
			80–100	4.83	1.18	11.37	1.42	1.12	0.30
Wugong (WG)	107.93°N, 34.34°E	537	0–10	6.46	1.21	7.81	0.98	0.81	0.17
			10–20	6.01	1.20	7.22	0.62	0.46	0.16
			20–40	4.50	1.20	10.84	1.20	0.93	0.28
			40–60	3.03	1.20	7.24	1.21	0.95	0.27
			60–80	3.72	1.24	9.20	1.30	1.03	0.27
			80–100	2.37	1.26	5.98	0.98	0.71	0.28
Qianjincun (QJC)	103.43°N, 35.71°E	2440	0–10	4.14	1.36	5.63	0.79	0.58	0.21
			10–20	3.48	1.23	4.26	0.80	0.64	0.15
			20–40	2.59	1.15	5.92	1.22	0.93	0.30
			40–60	2.06	1.19	4.90	0.71	0.49	0.23
			60–80	1.56	1.18	3.67	0.49	0.24	0.24
			80–100	1.30	1.20	3.12	0.51	0.25	0.27
Xinzhuang (XZ)	103.05°N, 36.24°E	1709	0–10	3.21	1.29	4.14	1.36	0.92	0.44
			10–20	1.85	1.15	2.14	0.33	0.15	0.18
			20–40	1.25	1.24	3.10	0.84	0.45	0.38
			40–60	1.18	1.20	2.83	0.59	0.31	0.28
			60–80	1.04	1.22	2.54	0.54	0.28	0.26
			80–100	1.22	1.22	2.97	0.70	0.35	0.35
Wushan (WS)	105.00°N, 34.74°E	1459	0–10	6.26	1.37	8.57	0.59	0.39	0.19
			10–20	3.16	1.25	3.94	0.40	0.23	0.16
			20–40	1.41	1.25	3.55	0.69	0.38	0.31
			40–60	1.10	1.18	2.61	0.32	0.07	0.25
			60–80	0.93	1.20	2.24	0.36	0.08	0.27
			80–100	1.42	1.18	3.36	0.47	0.26	0.20

[a] Altitudes are given in meters above sea level.

the 10 to 20 cm layer of some profiles (Fig. 3a, j). For example, Dai et al. (2005) reported that BC in a temperate mixed-grass savanna ecosystem could be displaced to a depth of 10 to 20 cm within a few years. Leifeld et al. (2007) also detected vertical transport of BC at a peatland site, and they proposed that the mobility of BC was dependent on the soil pore volume and water saturation. A final possible explanation for the increased BC with depth in some profiles (Fig. 3j and o) is that the layer sampled was where paleosol transitioned to loess, that is, when the climate changed from interglacial to glacial conditions. Reconstructions of BC abundances in the Lingtai section of the CLP have shown higher BC in paleosol (interglacial stages) compared with loess (glacial stages), and increases in BC were found when the climate shifted sharply from interglacial to glacial conditions (Zhou et al., 2007).

BC accounted for 3.18 to 35.3% of the SOC, averaged over all of the soil profiles (Table 2), and while the BC/SOC ratios tended to increase with depth (Fig. 3), the increases were typically discontinuous. When BC was analyzed with BPCA method, studies on Russian steppe soil and Russia Chernozems found that the maximum BC/OC values were in the 30 to 50 cm layer (Hammes et al., 2008; Rodionov et al., 2006), which are consistent with our findings in some profiles (GQ, MGP, QC, TSB, WS, LPS, QJC, and WG, Fig. 3). Considering that BC is biologically and chemically recalcitrant, it seems that a significant proportion of SOC pool in the deeper layer was more inert than those on the upper layers. Char densities ranged from 0.003 to 0.74 kg $\mathrm{m^{-2}}$, while the corresponding range in soot densities was 0.01 to 0.18 kg $\mathrm{m^{-2}}$. Variations in the char abundances were similar to those for BC, but in contrast, there were only minor variations with depth for soot (Fig. 3). The latter result is in agreement with the results of study of Han et al. (2009a), who found that soot concentration from non-disturbed soil profiles from Xi'an showed little variation with depth. This indicates that soot, which is the most highly-condensed and refractory form of BC, may have survived degradation for hundreds to thousands of years according to the average loess sedimentation rate (7.5 cm $\mathrm{kyr^{-1}}$, Heller and Liu, 1984).

Table 3
Comparison of BC/SOC ratio in different regions of the world.

Area	BC/SOC (%)	Method[a]	References
Brazilian, terra preta soils	Up to 35	BPCA	Glaser et al. (2000)
Switzerland, monitoring soils	1–6	CTO-375	Bucheli et al. (2004)
United States, agricultural soils	10–35	UV + NMR	Skjemstad et al. (2002)
Siberian, boreal forest soil	1.6–4.5	BPCA	Czimczik et al. (2003)
German, Chernozems	Up to 45	UV + NMR	Schmidt et al. (1999)
Northern Laos, sloping soils	3–7	$K_2Cr_2O_7/H_2SO_4$	Rumpel et al. (2006)
Northern Michigan, forest soil	4.3–7.6	$NaClO_2$ + NMR	Hockaday (2006)
France, pine forest soil	0.016	CTO-375	Quénéa et al. (2006)
China, Loess Plateau	2.39–36.87	TOR	This study

[a] Abbreviations for the BC isolation methods: BPCA = benzene carboxylic acid molecular markers, CTO-375 = chemical oxidation followed by thermal oxidation at 375 °C, UV = oxidation by ultraviolet radiation, NMR = ^{13}C nuclear magnetic resonance spectroscopy, $NaClO_2$ = oxidation sodium chlorite and acetic acid, $K_2Cr_2O_7/H_2SO_4$ = chemical oxidation after acid treatment with 0.1 M $K_2Cr_2O_7$/2 M H_2SO_4, and TOR = thermal/optical reflectance.

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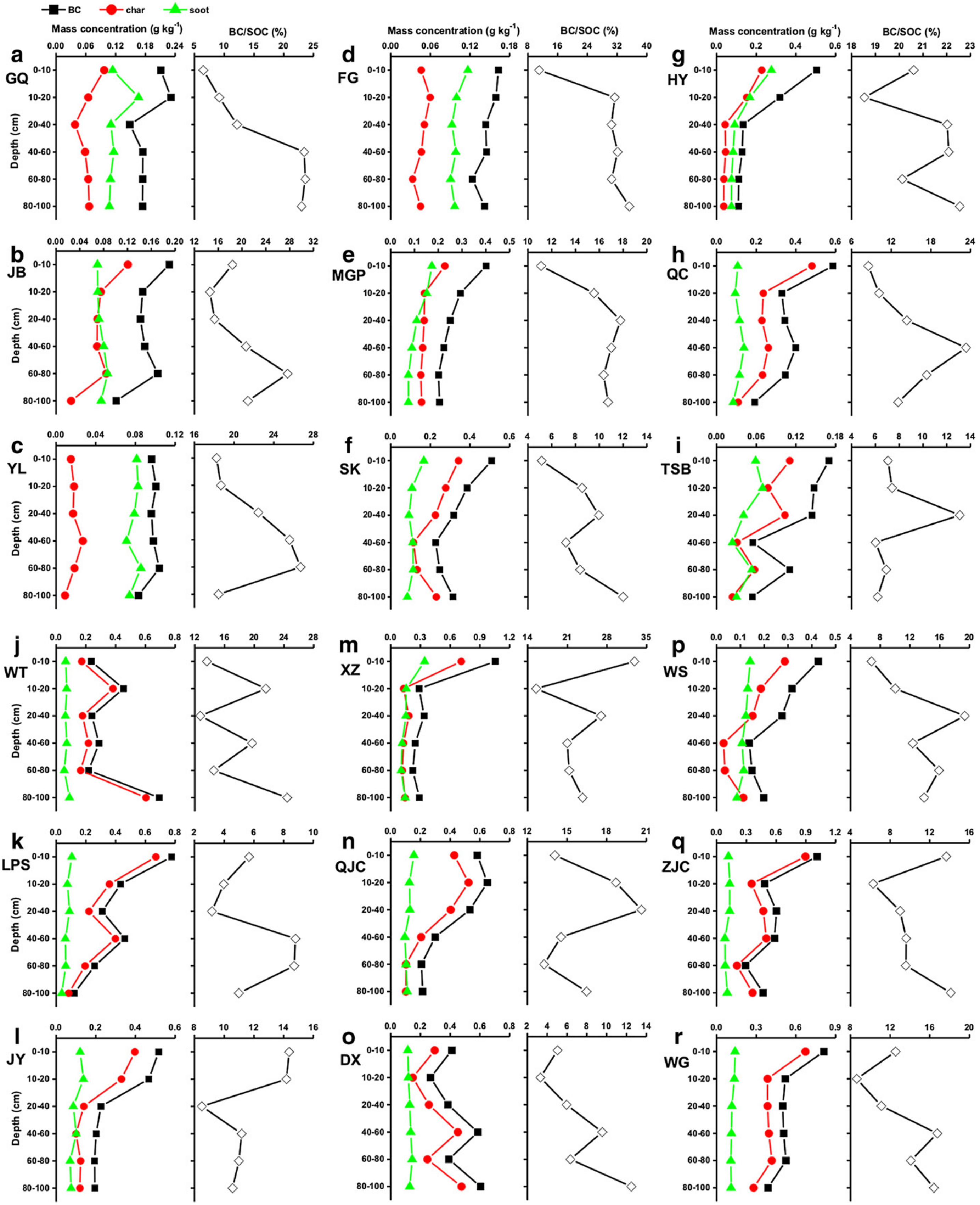

Fig. 3. Variation of BC, char and soot concentrations, and BC/SOC ratio (%) with depth in different soil profiles. Abbreviations for the sites are as follows: (a) Gaoqiao (GQ); (b) Jingbian (JB); (c) Yulin (YL); (d) Fugu (FG); (e) Muguaping (MGP); (f) Shikou (SK); (g) Heyang (HY); (h) Qingcheng (QC); (i) Tianshuibao (TSB); (j) Wangtuan (WT); (k) Liupanshan (LPS); (l) Jingyuan (JY); (m) Xinzhuang (XZ); (n) Qianjincun (QJC); (o) Dingxi (DX); (p) Wushan (WS); (q) Zhangjiachuan (ZJC); (r) Wugong (WG).

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3.3. Correlations between SOC, BC, char and soot

BC and char were well correlated with SOC while the correlation between soot and SOC was somewhat weaker (Fig. 4a–c). The close correlation between BC and SOC suggests that the two substances may originate from a common source, implying that combustion products are important for SOC because the sources for BC are well established, that is, the burning of biomass and fossil fuels. The close correlation between BC and char ($R^2 = 0.96$, $P < 0.0001$) is consistent with the observation that char is a main component of BC in topsoils from the CLP (Fig. 4g). The BC and soot concentrations had a moderate correlation ($R^2 = 0.53$, $P < 0.0001$), whereas char and soot were weakly correlated ($R^2 = 0.35$, $P < 0.0001$). The strengths of these relationships can be attributed to the different relative contributions of BC, char, and soot to the SOC pools (Fig. 4h, i).

BC accounted for 2.39 to 36.9% of the SOC in the topsoils, and the mean percent contribution of BC to SOC was 10.9% (Fig. 4d). Char, as a proportion of SOC, was relatively similar to BC, ranging from 0.55 to 33.9% of the SOC. Soot on the other hand accounted for a smaller percentage (0.43 to 12.8%) of the SOC (Fig. 4e, f). Fig. 4d–f also showed that the percent contributions of BC, char, and especially soot to SOC decreased when the SOC concentrations are greater than ~10 g kg^{-1}. This implies that non-BC carbon is responsible for the highest observed SOC concentrations. The soot/OC ratios in the soils varied from 0.7 to 24.03%, with a mean value of 7.1% (Fig. 4f), which was much smaller than the range of 16 to 61% reported for North Sea sediments or the range of 25 to 39% found in Eastern Mediterranean sediments (Middelburg et al., 1999). These discrepancies in soot/OC ratios may be a consequence of the different analytical methods used in the studies, and especially the operational definitions for soot carbon inherent in each technique. However the differences in the soot/OC ratios also could be affected by the quantities and types of organic matter produced by terrestrial vegetation versus marine biota. In addition, differences in the mechanisms and processes were responsible for the burial and preservation of the endogenous, and exogenous carbonaceous materials, which would likely influence the concentrations of the various OC compounds in the soils and deep sea sediments. All of these factors also would influence the relative proportions of the various carbon fractions in different environments. Thus, it is of great importance to discriminate between char and soot in the environment, from which we can better understand their environmental and climatic effects.

3.4. Estimation of carbon storage

The quantities of SOC in the 0 to 20 and 0 to 100 cm layers of the CLP were estimated to be 0.741 and 3.63 Pg, respectively (Table 4). Those reservoirs amounted to approximately 1.1% and 5.2% of the total SOC pool in China as estimated by Wu et al. (2003a). On average, 66.4% of the SOC in the upper 100 cm was contained in the 40 to 100 cm layer. When normalized to the total land area of China, the SOC pool in the 0 to 100 cm layer of CLP approaches that average, that is, the CLP makes up about 5% of China's terrestrial area and the CLP holds about the same percentage of the country's SOC. The BC stored in the 0 to 100 cm of CLP (0.456 Pg) was almost six times that in 0 to 20 cm

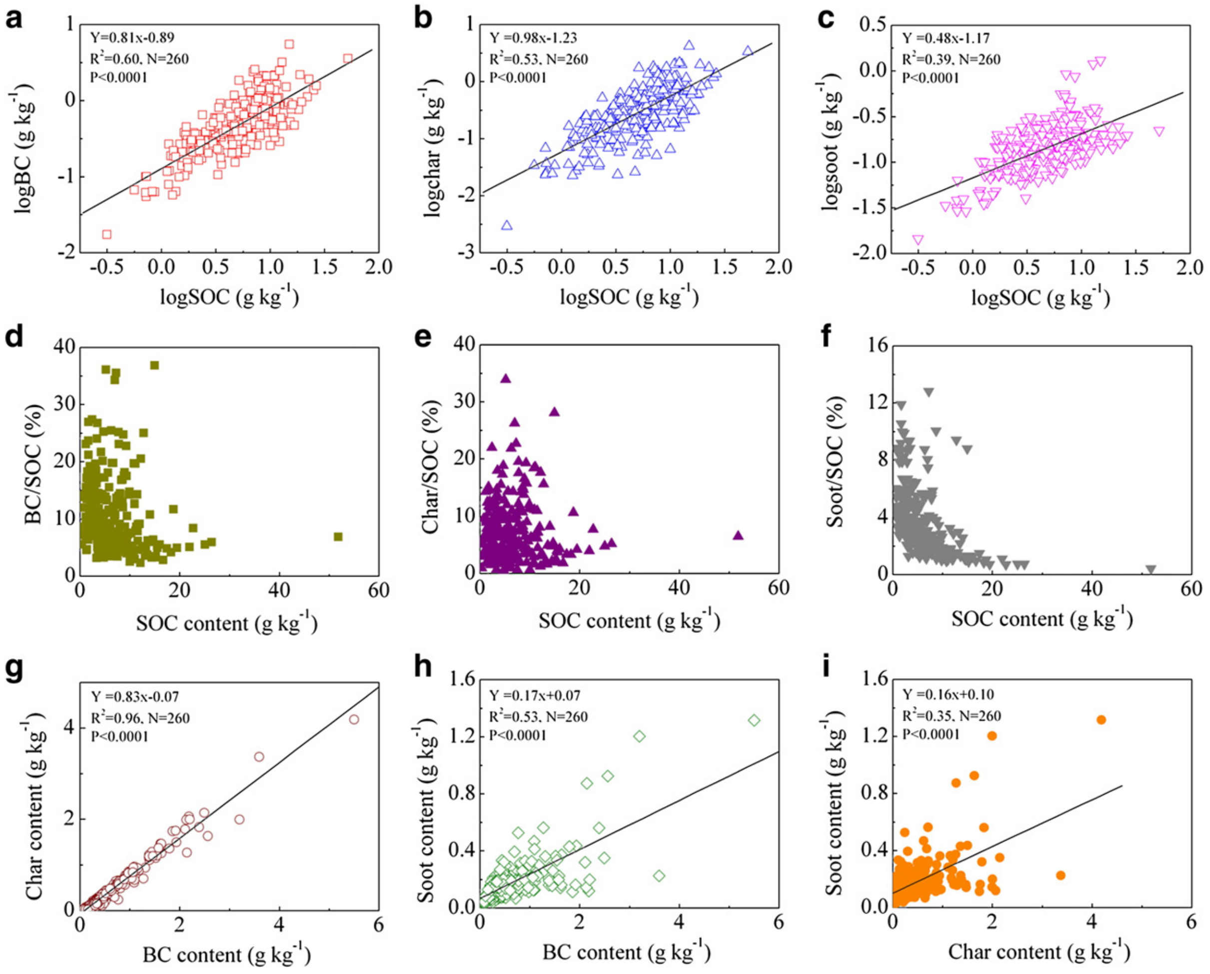

Fig. 4. Relationships between SOC, BC, and different BC fractions in topsoils.

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layer (0.073 Pg) (Table 4). As shown in Table 4, one can see that on a global basis, the char pool is much larger than that of soot. In our study, the char reservoir in the upper 20 cm soil in CLP was 0.053 Pg, which is approximately three times the amount of soot, and char composed 7.16% of the SOC and 72.5% of the BC pools, respectively. Similar to the results obtained for SOC and BC, the quantities of char and soot in the 20 to 100 cm layer accounted for more than 75% of the entire 0 to 100 cm layer. Therefore, one can speculate that a large amount of C may be stored in the deeper layers of CLP.

Our estimation of the SOC pool was ~30% lower than the previous estimate made by Xu et al. (2003) (Table 4), and this can be attributed to several factors. First, variations in climate and complex topography of the CLP play an important role for distribution of SOC, and this heterogeneity leads to uncertainties to the estimation of the SOC stock as discussed below. Furthermore, land-use changes, such as conservation tillage, can cause the soils to lose carbon to the atmosphere. During the past twelve years, the implementation of the Grain-for-Green project (Uchida et al., 2005) has led to increased human impacts on the environment of the CLP. Agricultural activities and associated changes in land use have altered the soil structure and the carbon cycle itself, including SOC sources, composition, storage, and dynamics. Wu et al. (2003b) suggested that land use had led to a reduction of the overall SOC pool by ~7.1 Pg in China, which represents ~9.5% of the world's decrease. Along these lines, based on the data from 18 previous studies on ten Mollisols of North American grasslands, three tropical forest sites and five forests in temperate or boreal regions, a paired comparative study conformed that 20 to 40% of the soil C inventory could be lost following cultivation (Davidson and Ackerman, 1993).

For centuries, deforestation, overgrazing and the collection of firewood have led to the destruction of vegetation on the CLP, and this in turn increased the vulnerability of soils to erosion. It has been estimated that 4.56×10^7 km^2 of the CLP are affected by severe erosion; this amounts to 70% of the total area of the CLP (Lal, 2002), and the average annual soil loss has been estimated to be 2000 to 2500 t km^{-2} (Shi and Shao, 2000). Erosion preferentially removes surface sediments and this results in a disproportionately large fractional loss of the total soil C (Davidson and Ackerman, 1993). This not only applies to SOC, but as our data suggests, also BC, char and soot. Unfortunately, we are not aware of any other attempts to quantify the BC sequestered in terrestrial soils, even much less the terrestrial pools of char and soot, which making the comparisons with other reservoirs difficult.

3.5. Uncertainty and implications

Major uncertainties still exist in the estimates of SOC and BC storage both for the CLP and on a global scale as well. Uncertainties arise from a variety of factors, including BC quantification methods, limited soil samples and profiles, and missing or incomplete data. Moreover, the heterogeneity of the SOC and BC distributions and the dynamics of the biogeochemical cycles make it infeasible to precisely estimate the sizes of the SOC and BC reservoirs. Nevertheless, this study presents the first attempt to estimate the BC, char and soot reservoirs in soils from the CLP.

In order to quantify the role of BC in the CLP as a potential long-term organic carbon sink, further studies on the stocks and dynamics of BC within soils are needed. Although we still do not know how much of the BC is mineralized or degraded in the loess sediments, the refractory organic carbon fraction is likely to be significant for a variety of geochemical reasons. Studies have showed that the degradation products of BC can be transported in both the dissolved and colloidal states in riverwater (Kim et al., 2004) and seawater (Dittmar, 2008; Mannino and Harvey, 2004). Thus, rain-induced runoff and soil erosion are potentially important pathways for the loss of BC from the CLP and transport to other environments. For instance, the BC mobilized by these processes may be transported long distances as the runoff flows into the Yellow River and beyond; the export of this material also presents opportunities for the BC to become involved in a multitude of biogeochemical processes. Another portion of the less-condensed BC (char or charcoal) probably decomposes gradually and is lost to the atmosphere as CO_2, thereby becoming involved in the global carbon cycle. Moreover, certain soil mineral fractions, when present, could adsorb BC and form a stable carbon pool (Czimczik and Masiello, 2007), and the more refractory BC constituents (e.g., soot), too, are likely left to form C reservoirs in the soils and sediments which may eventually degrade.

In the context of global warming, the more refractory organic carbon compounds are most likely to decompose at relatively high temperatures (Davidson and Janssens, 2006; Knorr et al., 2005), and therefore the balance between the deposition of SOC to the CLP and the SOC emissions from it are likely to be affected by temperature. This sensitivity of SOC sequestration to climate has potentially important implications for the carbon cycle. When we consider soil carbon in relation to global climate change, both the size of BC pools and the distribution of BC should be taken into consideration; that is, the dynamics of BC could influence carbon dioxide emissions from the terrestrial ecosystem, which is likely sensitive to changes in temperature or other environmental conditions (Lehmann et al., 2008).

4. Conclusions

BC is ubiquitous in topsoils and soil from the Chinese Loess Plateau. The spatial distributions of SOC and BC were found to be highly variable, but a large-scale trend emerged; that is, both SOC and BC decreased from the southeastern to northwestern parts of the CLP, and both exhibited low concentrations in the northern part of the plateau. Compared with the sandy loess zone or loess zones, the carbonaceous materials were more likely to be enriched in the clayey loess zone. In most of soil profiles, SOC, BC and char contents decreased with depth, whereas

Table 4
Estimated SOC, BC, char and soot pools in the Chinese Loess Plateau and comparisons with selected geological reservoirs.

Region	Area ($\times 10^5$ km^2)	Soil depth (cm)	Pool (Pg)				References
			SOC	BC	Char	Soot	
Global terrestrial soils	1352.15	0–100	1576	–[a]	–	–	Eswaran et al. (1993)
Terrestrial soils, China	88.18	0–100	70.31	–	–	–	Wu et al. (2003a)
Uncultivated soils, China	73.97	0–30	38.45	–	–	–	Song et al. (2005)
Cultivated soils, China	13.77	0–30	5.09				Song et al. (2005)
Grassland soils of Qinghai-Tibetan Plateau, China	12	0–100	7.4	–	–	–	Yang et al. (2008)
Paddy soils, China	2.98	0–20	1.3	–	–	–	Pan et al. (2004)
Loess Plateau, China	4.298	0–20	1.068	–	–	–	Xu et al. (2003)
Loess Plateau, China	4.4	0–20	0.741	0.073	0.053	0.020	This study
Loess Plateau, China	4.4	0–40	1.218	0.125	0.082	0.043	This study
Loess Plateau, China	4.4	0–100	3.629	0.456	0.288	0.168	This study

[a] Blank indicates unknown.

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the BC/SOC ratio showed the reverse pattern even though the increases were typically discontinuous. Soot showed comparatively little variation with depth in the profiles, implying that it is highly resistant to degradation. BC was tightly correlated with char and moderately correlated with both SOC and soot. On average, char and soot accounted for 57% and 43% of BC, respectively, indicating char and soot were the important components of BC. Our study showed that ~80% of the carbon in a 100 cm section was below 20 cm while the BC in the upper 0 to 20 cm layer accounted for just ~10% of the SOC, suggesting that BC was not the dominant fraction of the sequestered SOC. The quantity of char sequestered in soils amounted to over 63% of the BC pool, and it was approximately two times the stock of soot. Although the information obtained from this study concerning the storage of BC, char and soot in the CLP is preliminary, the results will not only be useful for understanding the relationships and dynamics among different carbonaceous substances, but also as a source for basic data relative to the sequestration of global BC, char, and soot in sedimentary materials.

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2012.10.113.

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