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# Black carbon records in Chinese Loess Plateau over the last two glacial cycles and implications for paleofires

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## Abstract

To study the temporal and spatial changes in paleofires over the Chinese Loess Plateau, black carbon concentrations were analyzed on the loess–paleosol samples from three sections along a north–south transect. Using the orbitally tuned time scales of the sections, the black carbon mass sedimentation rates (BCMSR) were calculated. Results show that in the last two glacial cycles, the BCMSR values in glacial periods are 2–3 times higher than in interglacial periods, and the BCMSR variability has a relatively strong precession-associated 23 ky period, suggesting that the glacial cold–dry climate conditions were apt to induce natural fires over the Loess Plateau. Comparison of the BCMSR records among the three loess sections demonstrates that natural fire occurrence was much more intensive and frequent in the northern Loess Plateau than in the southern part, coinciding with the previous conclusion that the northern Plateau has experienced a drier climate regime in both glacial and interglacial periods. The substantial increase in BCMSR of the upper S0 relative to the lower S0 at Lingtai and Weinan indicates that human activities have exerted a significant influence on fire regimes in the middle and southern Loess Plateau during late Holocene due to the relatively intensive agricultural usage of land.

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## 1. Introduction

In recent years, natural fire history has attracted an increasing attention of scientists involved in studies of global and regional climate changes, since reconstruction of changes in paleofires would provide an

opportunity to understand the interaction between climate, vegetation and fire events in geological past. Basically, natural fires would leave the combustion products such as charcoal, elemental carbon, polycyclic aromatic hydrocarbons and glucosan in the surroundings, which can be subsequently preserved in various sediments (e.g., Goldberg, 1985; Zepp and Macko, 1997; Elias et al., 2001; Scott, 2003). The concentration and fluxes of the combustion products detected from the sediments can be thus used to

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indicate paleofire occurrence and intensity (e.g., Goldberg, 1985; Zepp and Macko, 1997; Elias et al., 2001; Scott, 2003).

To reconstruct natural fire history, measurements of charcoal abundance, based on point-counting estimation under a microscope, are often used. However, this method proves to be time-consuming and apt to overestimate the concentration by counting some opaque minerals such as pyrite grains (Clark, 1983; Renberg, 1984), or underestimate the concentration by counting merely certain particle-size range of charcoal (Clark and Patterson, 1997). To overcome this shortage, researchers have recently developed thermal (Kuhibusch, 1995) and chemical methods (Winkler, 1985; Wolbach and Anders, 1989; Rose, 1990; Emiliani et al., 1991; Lim and Cachier, 1996; Bird and Grocke, 1997) to document changes in black carbon (BC) concentrations, which are currently employed as a proxy indicator of paleofire occurrence (Lim and Cachier, 1996; Schmidt and Noack, 2000).

The term black carbon is used to describe a relatively inert and ubiquitous form of carbon, comprising a range of materials from char and charcoal to element or graphite carbon produced by the incomplete combustion of fossil fuels and biomass (Goldberg, 1985; Schmidt and Noack, 2000). Due to its inertness, the BC in soils, lacustrine and marine sediments and ice can persist over a long period of time. So BC signatures in geological deposits can be used as evidence of natural fires happened in their surroundings. To date, most of the BC records are derived from marine and lacustrine sediments. However, interpretation of the BC signatures registered in these sediments appears to be often difficult because of their indefinite origins and a relatively long residence time. For instance, studies have shown that the BC particles can remain in the intermediate pools for more than 10,000 years before they are finally trapped in marine deposits (Masiello and Druffel, 1998). Comparatively, BC particles deposited in terrestrial sediments are expected to have a shorter atmospheric lifetime, implying that they may be more accurate to reflect paleofires.

Loess deposits coherently mantle an area of about 440,000 km<sup>2</sup> in the Loess Plateau of north-central China (Liu, 1985). Loess strata are built up by silt and clay particles transported by the winter monsoonal

wind from the deserts north and northwest to the Loess Plateau (An et al., 1991a). Observations have shown that loess particles can remain in air usually no longer than a week, and stratigraphical correlation between far-located loess sections has shown the completeness and continuity of loess deposition and preservation (Ding et al., 2002). These suggest that the Chinese loess deposits may be an ideal reservoir to register natural fire changes over this semi-arid region. In this study, the BC contents in three loess sections were analyzed, with the objective to investigate the spatial-temporal changes in natural fires over the last two glacial cycles.

## 2. Setting and stratigraphy

The selected three loess sections are located, respectively near Lijiayuan (36°7'0"N, 104°51'30"E), Lingtai (35°0'33"N, 107°30'33"E) and Weinan (34°20'14"N, 109°29'45"E) (Fig. 1). At present, both the mean annual temperature and precipitation show a clear northwestward decrease (~13.3 °C to 7 °C in temperature, ~600 to 250 mm in precipitation) along the transect. The precipitation is brought mainly by the southeasterly summer monsoon. The rainfall in summer season (JJAS) accounts for 56%, 66% and 70% of the annual precipitation, respectively at Weinan, Lingtai and Lijiayuan. During winter season, the climate of this region is cold and dry because of the dominance of the northwesterly winter monsoon originated in the Siberian area. Studies of proxy data have shown that this wind system have persisted in the Loess Plateau over the entire Quaternary period (An et al., 1990; Ding et al., 2002).

All the three sections consist of the loess–soil units of S0, L1, S1, L2 and S2, and are stratigraphically correlative in the field. The stratigraphic division and correlation of the sections is illustrated in Fig. 2, on the basis of the grain-size records (Ding et al., 1999) and field observations. The Holocene soil of S0, with a thickness of more than 1 m, is well-preserved in the sections, which is characterized by relatively high organic matter content. In the Loess Plateau, the last glacial loess deposit (L1) generally contains two weakly developed soils (L1-2 and L1-4) in the middle portion, which formed during marine isotope stage 3, as indicated by TL dating results (Liu, 1985; Lu et al.,

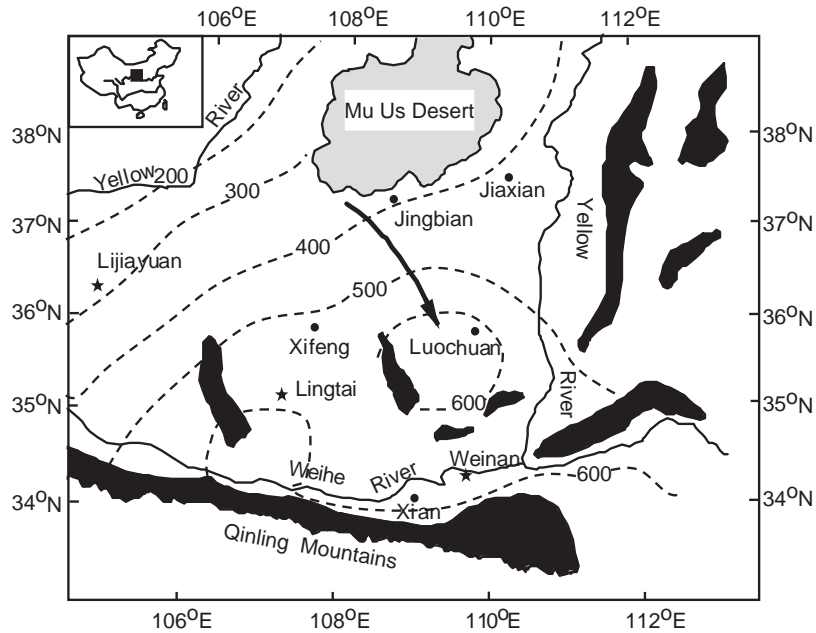


Fig. 1. Schematic map showing the localities of the studied sections which are labeled as five-angle stars. The arrow indicates the dominant subaerial wind direction in winter seasons, coinciding with the observed decrease in grain size and thickness of loess. The desert (dotted) and mountains (black areas) around and within the Loess Plateau are shown. The solid square in the inset map shows the locality of the Loess Plateau in continental China. The isohyets (dashed lines) are averaged values over the 32 years (1970–2001).

1987; Kukla and An, 1989). The two soils are only clearly seen at Weinan and Lingtai. The upper and lower parts of L1 are two typical loess layers (L1-1 and L1-5), which correlate, respectively to isotope stages 2 and 4. The S1 soil formed in the last interglacial period, which is among the best-developed soils of the entire Pleistocene on the Plateau. The S1 soil at Lijiyuan, with a thickness of 6.2 m, can be subdivided into three individual soils and two loess horizons (Fig. 2).

The thickness of the penultimate glacial loess unit (L2) is 8.6 m, 7.0 m and 15.6 m, respectively at Weinan, Lingtai and Lijiyuan. In the Weinan and Lingtai sections, there are also two weakly developed soils (L2-2 and L2-4) in the middle of L2. The S2 soil developed during the oxygen isotope stage of 7 and is usually composed of two separated soils named S2-1 and S2-2. In this study, only the upper soil (S2-1) could be traced at Lijiyuan because of slumping.

The loess–paleosol sequence above S2 at Weinan, Lingtai and Lijiyuan is 25.4 m, 21.1 m and 43.3 m thick, respectively. The similarity of the grain-size

curves (Fig. 2) strongly suggests the completeness of these loess records. We took samples at an interval of 8–10 cm, with a total of 946 samples collected from the three sections. This sample spacing is designed to resolve the paleofire variability on orbital time scales.

### 3. Methods

In this study, we use the chemical method developed by Lim and Cachier (1996) to extract the black carbon in the loess–soil samples. In brief, the carbonates and part of silicates in the samples were removed by the acid treatment with HCl (3 mol/L) and HF (10 mol/L)/HCl (1 mol/L) in sequence. The treated samples were then oxidized by a solution of 0.1 mol/L  $K_2Cr_2O_7$ /2 mol/L  $H_2SO_4$  at 55 °C for 60 h to remove soluble organic matter and kerogen. The remaining refractory carbon in the residues is called black carbon, and includes charcoal and atmospheric BC particles (Lim and Cachier, 1996).

To assess the possible contribution of naturally burnt oil and coal deposits to the remaining

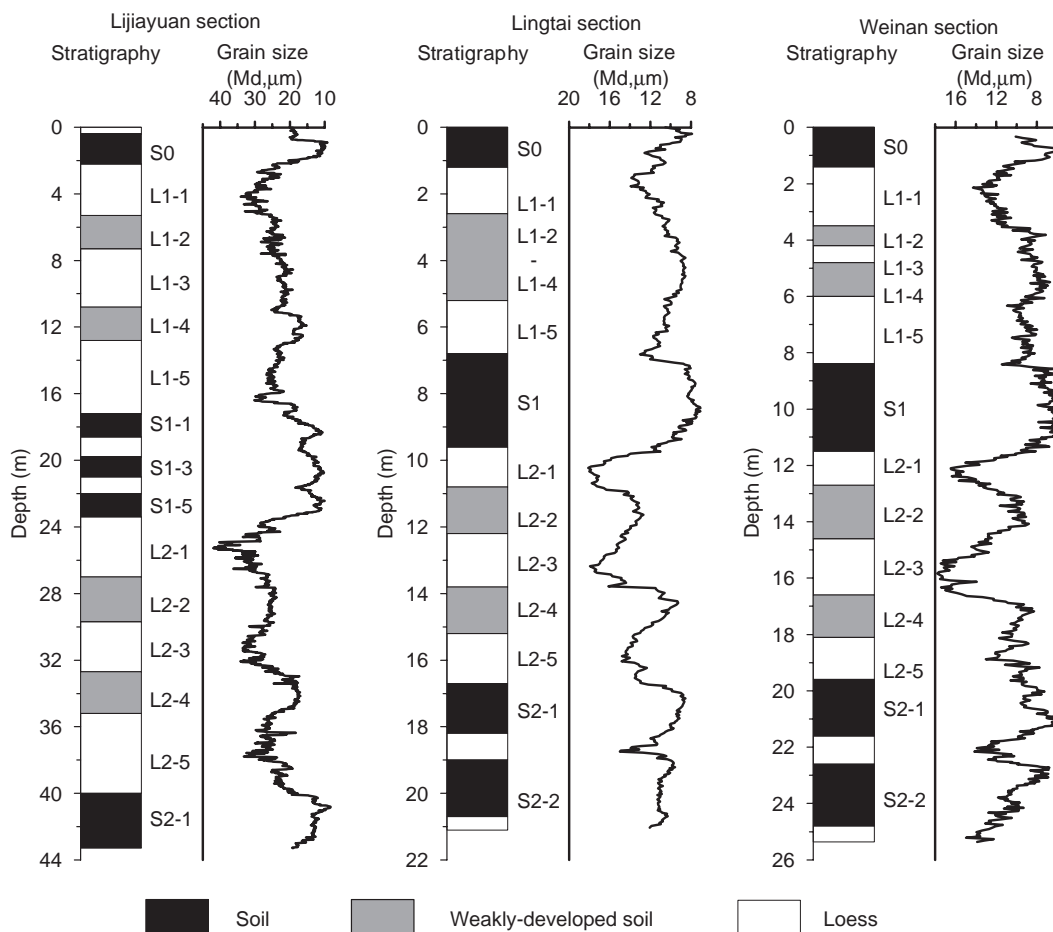


Fig. 2. Stratigraphic subdivision and median grain-size records of Lijiayuan, Lingtai and Weinan sections. The grain-size data are cited from Ding et al. (2001).

refractory carbon, scanning electron microscope (SEM) observations were made on 36 of the 946 samples. The selected SEM photographs are shown in Fig. 3. Previous studies (Griffin and Goldberg, 1979, 1981) have demonstrated that the charcoal or element carbon derived from oil burning is spherical (cenospheres) with a delicate, convoluted, layered structure, while coal combustion carbon particles are spheroidal with a robust network structure. However, the carbon particles produced by biomass burning are commonly elongate with a length to width ratio of over 3.0 and these particles often display the fine details of plant cellular structure. As seen in Fig. 3, the BC particles extracted from the loess samples are predominantly elongate-prismatic

or irregular in shape with various plant cellular structures. Few particles with spherical shapes and porous structures are found. Accordingly, the BC particles in the loess deposits have been produced dominantly by biomass burning, with an insignificant contribution of naturally burnt oil and coal deposits.

The black carbon content in the residues was determined using a Heraeus CHN-O Rapid elemental analyzer. The combustion temperature was set at 960 °C. Sextuplicate determinations on one sample by the above chemical treatment procedure showed that the relative error was within  $\pm 3.3\%$  for black carbon content. For all the samples, duplicates were analyzed, and each of the data used here is the

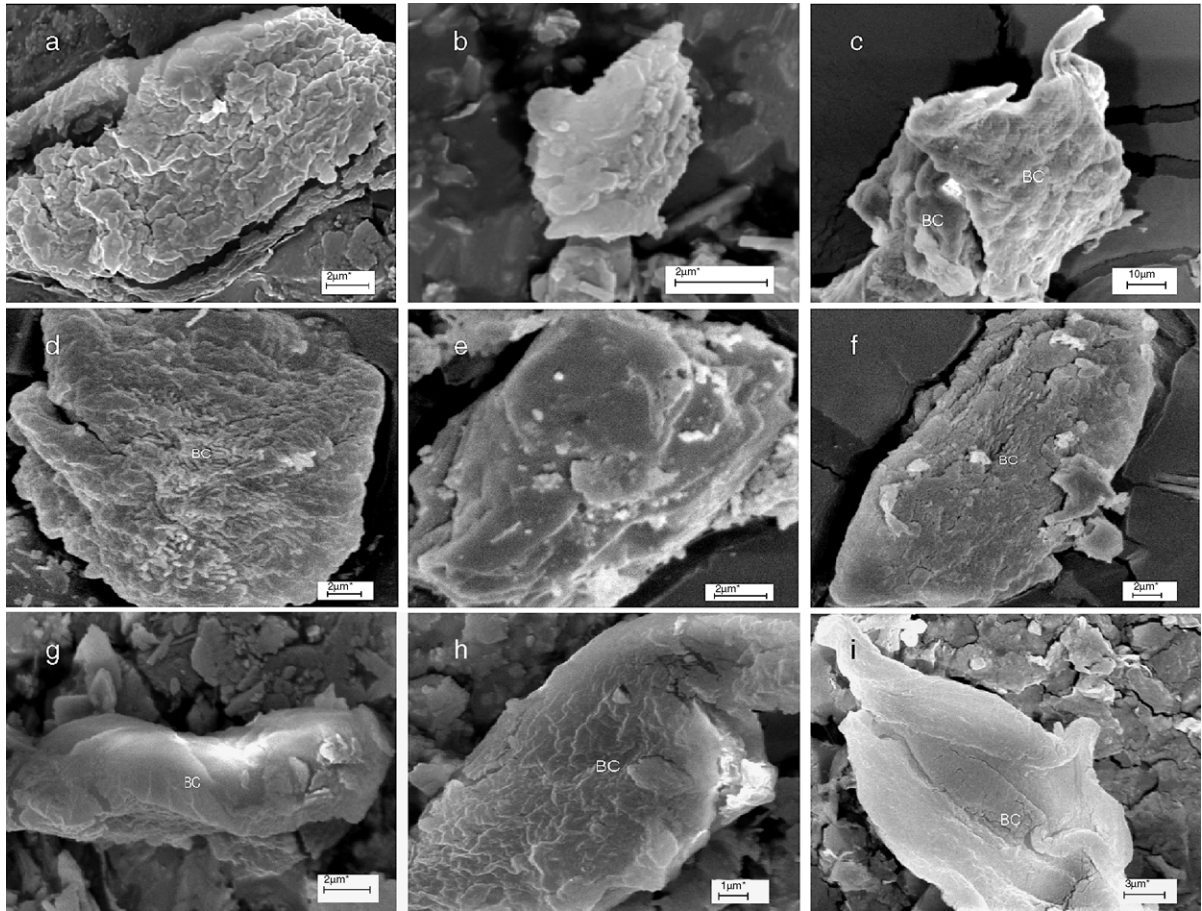


Fig. 3. Selected scanning electron microscopic photographs of the isolated BC particles from loess and paleosol samples of Chinese Loess Plateau. (a) BC particle with elongate/plant structure (Sample No. WN002); (b) small BC particle with irregular shape/layered structure and flat surface (Sample No. WN002); (c) BC particles with irregular shapes and pitted surfaces (Sample No. WN016); (d) a spherical BC particle with pleated surface and layered structure (Sample No. LT49); (e) BC particle with elongate/layered structure and flat surface (Sample No. LT137); (f) BC particle with irregular shape/layered structure (Sample No. LT137); (g) BC particle with prismatic shape/coiled layered and smooth surface (Sample No. LJY1); (h) BC particle with irregular shape/plant structure (Sample No. LJY1); (i) BC particle with irregularly shaped flake and uneven surface (Sample No. LJY14.6).

average of the two measurements with a deviation  $< \pm 3\%$ .

#### 4. Results

Changes in BC concentrations at the three sites are shown in Fig. 4, and compared with the grain-size records. In the sections, BC concentrations range from 0.041% to 0.572%. A most striking feature is that BC concentrations vary frequently on relatively short time scales, in contrast to the obvious glacial–

interglacial variability of the grain-size records. The averaged BC concentration at Lijiayuan, Lingtai and Weinan is 0.184%, 0.178%, and 0.124%, respectively, and shows a decrease from north to south (Fig. 4).

Because the dust deposition rate varies largely at both spatial and temporal scales over the Loess Plateau, the BC concentration records cannot be directly used to describe the paleofire history. This means that the BC concentration records should be transferred into BC mass sedimentation records by development of loess chronology. Previous studies



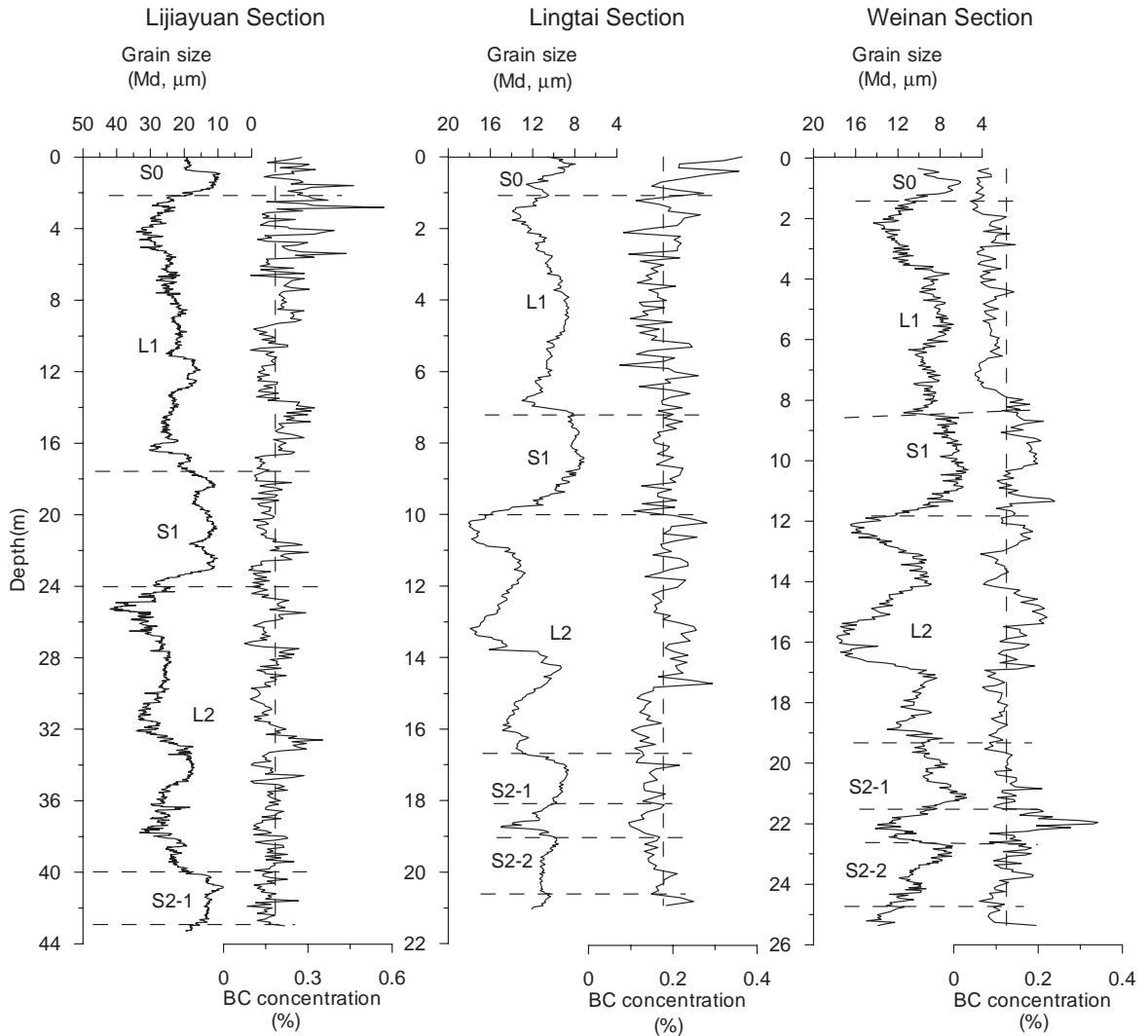


Fig. 4. Black carbon (BC) concentrations and comparison with median grain-size records of the studied sections, plotted on depth scales. The vertical dashed lines indicate the averaged BC concentration for each record.

(Liu, 1985; An et al., 1991b; Ding et al., 1992) have shown that the particle sizes in loess horizons are consistently larger than in soils, in response essentially to the orbital-scale variations in the intensity of the winter monsoon wind which is the major agent transporting the dust. A recent study (Ding et al., 2001) has calculated, by tuning the grain-size records to the orbital parameters of the Earth, the changes in sedimentation rates of eolian dust for the three sections. Based on the orbital time scales, we calculated the BC mass

sedimentation rate (BCMSR) using the following equation

$$\text{BCMSR} = C_{\text{BC}} (\%) \times \text{BD}_{\text{loess}} \left( \text{g/cm}^3 \right) \\ \times \text{SR}_{\text{dust}} (\text{cm/ky})$$

where  $C_{\text{BC}}$  represents BC concentration,  $\text{BD}_{\text{loess}}$  is the bulk density of loess or paleosol, and  $\text{SR}_{\text{dust}}$  denotes dust sedimentation rate. Since there are no considerable spatial and temporal differences in bulk

Table 1  
Bulk densities used in this study

Layer	S0	L1	S1	L2	S2
Bulk density (g/cm <sup>3</sup> )	1.40	1.48	1.65	1.49	1.69

densities for both loess and paleosol horizons over the Loess Plateau, we here adopted the average bulk density of each horizon provided by Liu (1985) (Table 1).

The resulting BCMSR records of the three sections are shown in Fig. 5, in relation to the grain-size time

series and the SPECMAP record of the last 220 ky (Imbrie et al., 1984). The three records all exhibit a distinctive orbital time-scale variability in BC mass sedimentation rates. To examine the BCMSR difference over different time intervals, the average BCMSR value is calculated for each marine isotopic stage, with the chronological constraints provided by Imbrie et al. (1984); the results are shown in Fig. 5b, e, h. During the glacial periods, the average BCMSR is about 0.07–0.09 g/cm<sup>2</sup>/ky at Lijiayuan, and about 0.03–0.05 g/cm<sup>2</sup>/ky at Lingtai and Weinan, showing a

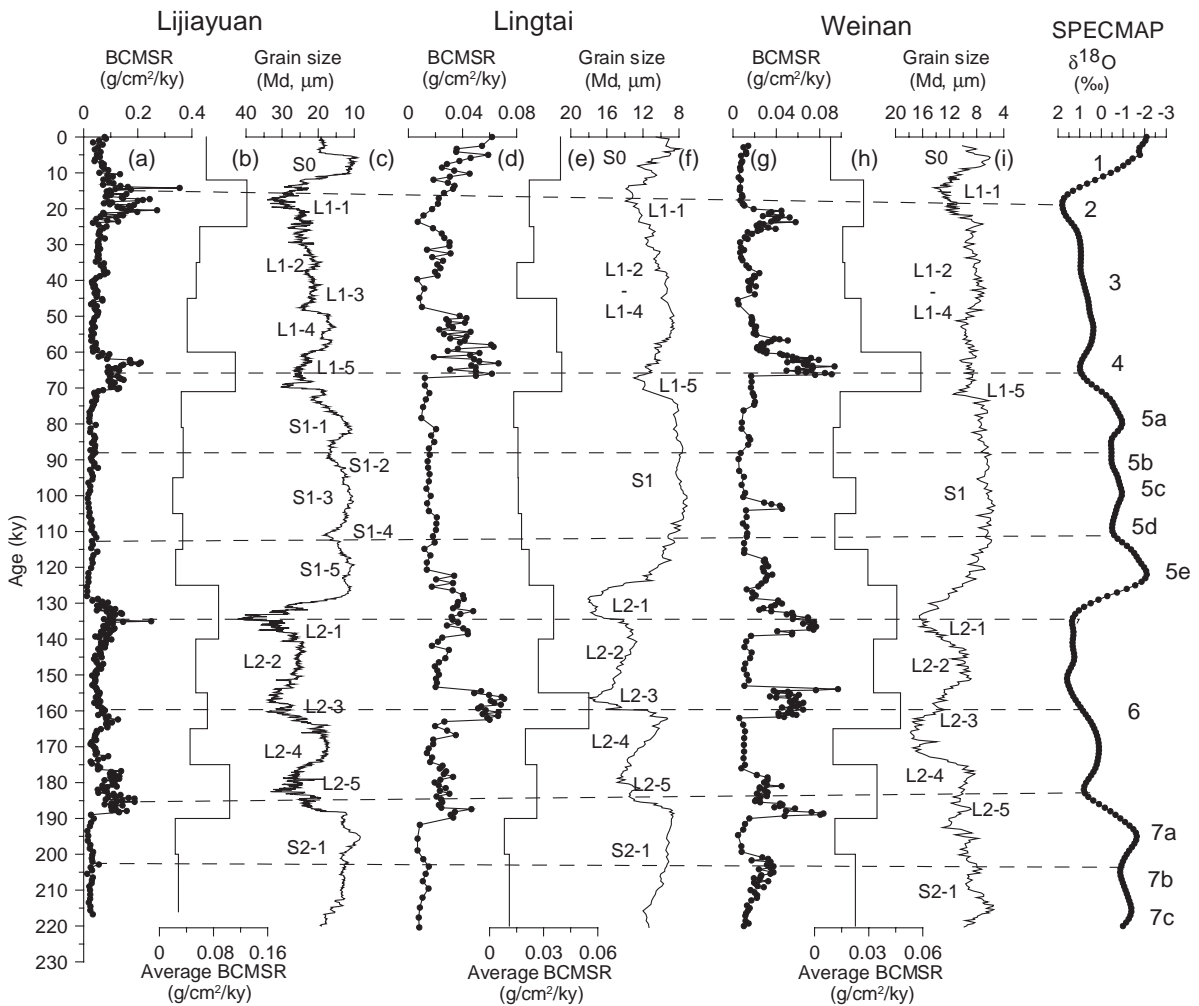


Fig. 5. Black carbon mass sedimentation rate (a, d, g), averaged BCMSR (b, e, h) and median grain-size (c, f, i) records of the studied sections (plotted on orbital time scales), and correlation with the SPECMAP  $\delta^{18}\text{O}$  record (Imbrie et al., 1984). In each section, the BCMSR is averaged for each oxygen isotopic stage (b, e, h), using the chronology of the SPECMAP record.

clear southward decrease. The average BCMSR in the interglacial periods is generally about 2–3 times lower than in the glacial periods; about  $0.03 \text{ g/cm}^2/\text{ky}$  at Lijiayuan and about  $0.015 \text{ g/cm}^2/\text{ky}$  at Lingtai and Weinan. In addition to the glacial–interglacial variability, the BCMSR values also vary notably within the two glacial periods. For instance, the BCMSR in the stadial periods of L1-1, L1-5, L2-1, L2-3 and L2-5 is much higher than in the interstadial periods of L1-2, L1-4, L2-2 and L2-4 (Fig. 5).

To examine the periodicities of BCMSR variations, we conducted maximum entropy spectral analyses on the three BCMSR time series. Results (Fig. 6) show

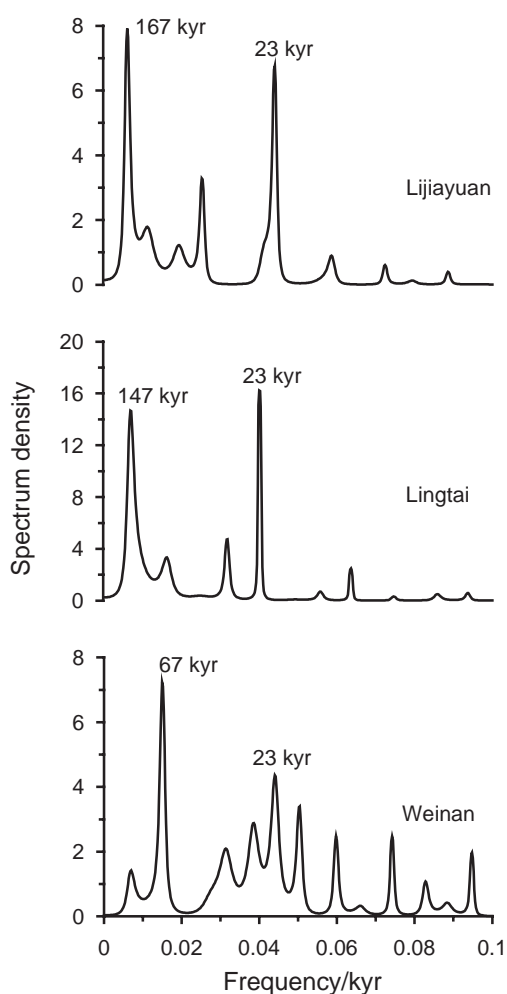


Fig. 6. Periodicities of the BCMSR time series of the studied sections, using the Maximum entropy spectrum method.

that there is a strong variance at about 23 ky in all the three records, which may be associated with the orbital precession changes, as envisaged in the stadial–interstadial variability of the BCMSR records (Fig. 5). There is also a periodicity of about 67 ky in the Weinan record and a periodicity of about 150 ky in the Lijiayuan and Lingtai records. Since our records are relatively short, these low frequency variations may not have meaningful implications.

## 5. Discussion and conclusions

Our results show that black carbon mass sedimentation rates over the Loess Plateau clearly display changes at orbital time scales, and the BCMSR values during glacial periods are 2–3 times higher than during interglacial periods. Within the glacial periods, BCMSR in stadial periods is much higher than in interstadial periods, which is evidently associated with the 23 ky period of changes in the orbital precession. As the Chinese loess is transported by wind from the deserts north and northwest to the Loess Plateau (Liu, 1985), it is likely that part of the BC particles in the records have been transported with the dust. To assess this possible contribution, we employed the sample with the lowest BC concentration among the sections as the upper limit of BC content transported with dust. This yields a datum of 0.041% (at depth of 1.42 m at Weinan section), being 22.28%, 23.03% and 33.06% of the averaged BC content, respectively at Lijiayuan, Lingtai and Weinan, thereby indicating the major contribution of local fires to the loess BC concentrations.

In previous studies, Van der Kaars et al. (2000) found, based on the records derived from core SHI-9014 of Banda sea, that the charcoal and elemental carbon concentrations in glacial periods are higher than in interglacial periods, a pattern being the same as our records. However, Bird and Cali (1998) reported that more frequent natural fires in sub-Saharan Africa occurred during the transition from interglacial to glacial periods, suggesting that the destabilized climate conditions of the periods be apt to induce natural fires. These findings show that there is a regional difference in fire regimes in the world during the late Pleistocene.



Recently, many efforts have been made to reconstruct the paleo-vegetation history of the Loess Plateau (Liu et al., 1996; Sun et al., 1997; Li et al., 2003). A conclusion made on the basis of pollen analyses is that the Loess Plateau was covered by an *Artemisia*-dominated grassland vegetation both during glacial and interglacial periods, although moisture-loving species increase to some degree in interglacial periods. Even at Weinan, one wettest location of the Plateau, pollen results still indicate a succession of steppe and meadow-steppe environment during the last glacial cycle (Sun et al., 1997). This conclusion is also corroborated by carbon isotope analyses of organic matter. Recently, Gu et al. (2003) reported  $\delta^{13}\text{C}$  results of five loess sections including the Weinan and Lingtai section, and concluded that grassland vegetation cover has been dominant on the Loess Plateau during the last glacial cycle. Accordingly, our BC records may be regarded as an indication of changes in grassland burning on the Plateau.

The more frequent occurrence of natural fires in the Loess Plateau during glacial periods may be explained by a synergistic influence of climate conditions and fuel accumulation. It has long been known that the climate of the Plateau was very dry and cold during the glacial periods of the late Pleistocene (Liu, 1985). Under such conditions, the biomass litter tends to dry quickly and decompose slowly, leading to a built-up of a thick fuel layer on the surface, and subsequently to more intensive and frequent natural fires that may be started by lightning strikes (e.g., Scott et al., 2000).

A spatial comparison of the BCMSR records (Fig. 5) shows that natural fires in the northern part of the Loess Plateau have been more intensive than in the southern part during both glacial and interglacial periods. This may be explained by a relatively steep gradient of precipitation over the Loess Plateau under the influence of Asian monsoon climate (see Fig. 1). Various proxy records from loess sediments have demonstrated that the monsoonal rainfall over the Plateau significantly decreases from south to north both in glacial and interglacial periods, coinciding with today's pattern (Liu and Ding, 1998). Obviously, this precipitation pattern has exerted a significant control on paleofire occurrence. The precessional-associated 23 ky period of BCMSR changes may also be related to the monsoon system. The monsoon

originates in the low latitudes where changes in the orbital precession exert a significant control on variations in insolation values (Ruddiman, 2000). Changes in precession-modulated monsoon strength will in turn induce wet–dry oscillation in the Loess Plateau, thereby controlling the intensity and frequency of biomass burning there. A study by Haberle and Ledru (2001) also concluded that fluctuations in tropical biomass burning were partly controlled by orbital precession forcing.

The more intensive natural fires in the northern Plateau may imply that even in the glacial periods, this place was still covered by relatively dense vegetation. This is consistent with the observation that thick, complete dust deposits have continuously accumulated during glacial periods on the northern Loess Plateau (Liu, 1985), as a result of dense vegetation cover trapping falling dust.

Intensive and extensive human activity has had a relatively long history in the Loess Plateau. It is therefore interesting to assess the anthropogenic impact on fire regimes, based on the BC records of the S0 Holocene soil. After cultivation for several thousand years, the S0 soil on the Loess Plateau can be generally divided into two parts: the upper cultivated soil and the lower undisturbed soil. Fire regime difference of human period with pre-human period may be examined by comparing BC mass sedimentation rates between the two soil horizons. The averaged BC mass sedimentation rate of the upper and lower soils is 0.060 and 0.074 g/cm<sup>2</sup>/ky at Lijiyuan, 0.047 and 0.036 g/cm<sup>2</sup>/ky at Lingtai, and 0.012 and 0.008 g/cm<sup>2</sup>/ky at Weinan, respectively. An obvious northward increase in BCMSR during both the human and pre-human periods indicates the first-order control of humidity on the occurrence of natural fires. Lijiyuan is located at the northwestern edge of the Loess Plateau with a mean annual precipitation below 300 mm (Fig. 1) and a low population density. In the Lijiyuan section, no increase of BCMSR in the upper part of S0 is observed. However, we do see the influence of human activity on fire regimes, as suggested by the substantial increase in BCMSR of the upper soil relative to the lower soil at Lingtai and Weinan. This may be explained by the relatively intensive agricultural usage of land in the middle and southern Loess Plateau where climatic conditions are more suitable for agriculture. In addition, BCMSR is

exceptionally high for the entire Holocene soil at Lingtai, also suggesting human-induced increase in fires. This anthropogenic mechanism is also documented in the records derived from both marine and lake sediments (Bird and Cali, 1998; Moreno, 2000).

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