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# Paleofires and the dynamics of carbon cycling in Chinese Loess Plateau over the last two glacial cycles

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The spatio-temporal changes in trace gas emissions and burnt biomass by paleofires in Chinese Loess Plateau over the last two glacial cycles have been reconstructed using vegetation (C3/C4) specific fire emission factors and black carbon records in three loess-paleosol sections. Results show that the average mass emission rate (AMER) of total trace gases (TTG) and burnt biomass by fires (BBF) in glacial periods are 1~2 times higher than in interglacial periods, and they display a clear southward decrease during both glacial and interglacial periods. This pattern reflects the combined control on paleofire emissions by climate-induced fire regimes and succession of vegetation types. The substantial increases in TTG-AMER and BBF during the late Holocene relative to the middle-to-early Holocene at Lingtai and Weinan support existing conclusion that increased anthropogenic fire activities have occurred in Middle and Southern Plateau during late Holocene. To assess the influence of paleofires on soil carbon stocks, the ratios of BBF to above-ground net primary productivity (ANPP), estimated by magnetic susceptibility-based paleorainfall and paleotemperature reconstruction, were calculated. In the Northern Plateau, the BBF/ANPP ratios during glacial periods are nearly 90%, about 2~3 times higher than during interglacial periods, suggesting paleofires may be the overwhelming force modulating the cycling of terrestrial organic carbon in the region. However, in Middle and Southern Plateau, the large decrease in BBF/ANPP ratios to around 20% and 10% respectively during glacial and interglacial periods suggests that paleofires have had a minor impact on carbon storage in these areas during both glacial and interglacial periods.

#### 1 Introduction

Biomass burning has been recognized as a significant source of a number of important trace gas species and particulate matter to the atmosphere (Seiler and Crutzen, 1980; Lavoue et al., 2000; Andreae and Merlet, 2001). Emissions from biomass burning

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contribute substantially to the global budgets of trace gases and aerosols that affect climate. Recently increased trace gases e.g. carbon dioxide and black carbon particles from both fossil fuel and biomass burning has prompted many researchers to study the changes in carbon budgets associated with increased fire activities (van der Werf et al., 2004, 2010; Mouillot and Field, 2005) and the climatic effects exerted by fires (e.g. Levine, 1991; Menon et al., 2002; Patra et al., 2005; Langmann et al., 2009). In the context, the central issues that emerge are the extent to which terrestrial ecosystems can serve as a sink for carbon and how this could help to mitigate potential human-induced increases in atmospheric CO<sub>2</sub> levels (Scurlock and Hall, 1998; Fang et al., 2001; Keeling et al., 2001; Schimel et al., 2001). As we have known, the main natural disturbance agent in most terrestrial ecosystems is wildfire, which can serve as a major driver of ecosystem carbon cycling and storage (Harden et al., 2000; Houghton et al., 2000; Tilman et al., 2000; Wardle et al., 2003; Bond-Lamberty et al., 2007). To better understand the role of terrestrial ecosystems in global carbon cycle, it is extremely important to know how wildfires influence carbon fluxes among vegetation, soil and atmosphere. To date, many studies regarding the effects of fires on carbon storage of certain ecosystems have been carried out based on modern observation (Page et al., 2002; van der Werf et al., 2010) and/or modeling study (Thornley and Cannell, 2004; Bond-Lamberty et al., 2007; Smithwick et al., 2009; van der Werf et al., 2009). However, these approaches can only provide a limited number of, short-termed (e.g. decades) and discontinuous results. In comparison, paleorecords have an advantage in evaluating the long-term effects of wildfires exerted on the carbon storage of certain ecosystem under the changing climate.

On the other hand, in global carbon budgets, there still exists the so-called "missing carbon" sink, carbon that is evolved as  $CO_2$  but remains unaccounted for (Ojima et al., 1994; Schimel, 1995; Houghton et al., 1999, 2000; Tilman et al., 2000; Hurtt et al., 2002). Generally, the widely accepted assumption is that fires have no net effect on a steady-state world, where losses because of combustion in some places were compensated by gains because of biomass accumulation in others (Crutzen and

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Andreae, 1990). However, if total carbon release from fires is not constant over time, then fire can generate carbon source or sink (e.g. Mouillot and Field, 2005). For example, a trend of increasing combustion through time generates a net source of carbon, while a trend of decreasing combustion causes a sink (Chen et al., 2000; Tilman et al., 2000). Therefore, the changes in carbon fluxes due to the altered fire regimes during different climate periods may account for part of the "missing carbon" sink or source. In this sense, the carbon budgets under different fire regimes should be quantified and compared in order to better understand the influence of wildfires on the carbon storage of terrestrial ecosystems and provide scientific basis for the accurate assessment of global carbon budget in the future.

Since deliberate wildfire suppression and extensive human burning activities have occurred over the past two centuries (e.g. Wardle et al., 2003), observations on modern wildfires are inadequate for understanding the function of natural fires in carbon cycles and calculating wildfire-related carbon budgets during different fire regimes. Instead, paleofire studies may provide an opportunity for studying how long-term fire history can affect the carbon flows and sequestration of ecosystem on various time scales.

Our previous study (Wang et al., 2005) shows that the pattern of paleofire occurrences in Chinese Loess Plateau has undergone a substantial change during the glacial-interglacial periods. Here, we present a quantitative estimate of regional carbon release (including  $\mathrm{CO}_2$ ,  $\mathrm{CO}$  and  $\mathrm{CH}_4$  emission rate) by paleofires over the last two glacial cycles based on the black carbon records in three loess sections along a south-north transect, with objective to gain an insight into the changes in carbon storage associated with the altered fire regimes. Moreover, the ecological effects of different fire regimes during the glacial-interglacial periods on carbon storage in terrestrial ecosystems would also be discussed by comparing the ratio of burned biomass by fires (BBF) to above-ground net primary productivity (ANPP) during the glacial period with that during the interglacial period.

#### 2.1 Rationale and assumptions

Black carbon (BC) is produced by the incomplete combustion of fossil fuels and biomass, used to describe a relatively inert and ubiquitous form of carbon, comprising a range of materials from char and charcoal to elemental or graphite carbon (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 have been employed as evidence of natural fires happened in their surroundings (e.g. Jia et al., 2000; Wang et al., 2005).

Black carbon particles produced by biomass burning can be initially divided into two groups: one is atmospheric BC emitted with smoke and being transported far in air; the other is BC in fire residues. It has been shown that generally more than 80% of the BC produced by vegetation fires resides in the residues and less than 20% is emitted with the smoke (Kuhlbusch and Crutzen, 1995). Besides, the result from some experimental fire indicates that most charcoal particles accumulated inside the fire, and accumulation declined sharply outside the fire, with only 1% of measured particles transported beyond 20 m from the burn edge (Lynch et al., 2004). Nevertheless, the BC remaining on site after fire in the fine residue, especially in open grassland, tends to be highly accessible to aeolian transport and may significantly contribute to the background concentrations of BC in the atmosphere. However, the aeolian BC has proven to have relatively short atmospheric lifetime, e.g. 40 h to 1 month (Ogren and Charlson, 1983). Therefore, we assume that most the BC particles produced by paleo-biomass burning are ultimately incorporated into contemporary loess and paleosol deposits. In this case, the sedimentary BC record should be well correlated with the carbon released by fires.

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#### 2.2.1 The criteria for data adoption

The prerequisite for quantitative reconstruction of carbon emission from paleo-biomass burning lies in that there should be proper unit e.g. g cm<sup>-2</sup> yr<sup>-1</sup> or g cm<sup>-2</sup> kyr<sup>-1</sup>, for the proxy indicators of past fires from sedimentary records. Therefore, except for the paleofire records using optical charcoal quantification methods, such as pollen-slide and sieved charcoal, which usually provide data expressed in CHAR e.g. mm<sup>2</sup> cm<sup>-2</sup> yr<sup>-1</sup>, mm<sup>2</sup> g<sup>-1</sup> cm<sup>-2</sup> yr<sup>-1</sup> or fragments cm<sup>-2</sup> yr<sup>-1</sup>, other records which adopted chemical methods (e.g. Winkler, 1985; Wolbach and Anders, 1989; Rose, 1990; Lim and Cachier, 1996; Bird and Gröcke, 1997) to quantify charcoal or black carbon in sediments can meet such demand. Fortunately, our previous study (Wang et al., 2005) has provided black carbon records with the unit of g cm<sup>-2</sup> kyr<sup>-1</sup> in Chinese Loess Plateau. This would offer an opportunity to perform quantitative reconstruction of carbon emission from paleo-biomass burning using the BC records.

However, an empiristic relationship between the carbon emission and the black carbon records must be established before we carry out this reconstruction. So far, much work involving both laboratory and field burning experiments has been done to assess the gaseous and particulate emission from biomass burning of different kinds of vegetation (e.g. Hao et al., 1990; Menaut et al., 1991; Turn et al., 1993; Cachier et al., 1995; Kuhlbusch and Crutzen, 1995; Andreae and Merlet, 2001). The provided emission ratios or emission factors enable us estimate quantitatively the carbonaceous gases and particles emission from the paleofires as documented in black carbon records.

#### 2.2.2 Black carbon records and biomass burning in Chinese Loess Plateau

Loess deposits coherently mantle an area of about  $44 \times 10^4 \,\mathrm{km}^2$  in the Loess Plateau of North-Central China (Liu, 1985). Today's Loess Plateau straddles the semi-humid, semi-arid and arid climatic zones, mainly covering natural vegetation zones of the

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warm-temperate forest, warm-temperate forest-steppe and warm-temperate steppe (Sun et al., 1997). However, the carbon isotope analyses of soil organic matter and pollen studies show that the Loess Plateau was covered by grassland vegetation both during glacial and interglacial periods (Sun et al., 1997; Gu et al., 2003; Jiang and Ding, 5 2005), so our BC records may be regarded as an indication of changes in grassland burning on the Plateau.

The temporal and spatial patterns of paleofires in Chinese Loess Plateau over the last two glacial cycles have been studied on the basis of black carbon mass sedimentation rates (BCMSR) documented in three loess-paleosol sections, respectively, at Lijiayuan, Lingtai and Weinan along a north-south transect (Fig. 1) (Wang et al., 2005). Briefly, the glacial cold-dry climate conditions were apt to induce natural fires over the Loess Plateau, and natural fire occurrence was much more intensive and frequent in the Northern Loess Plateau than in the southern part.

This study will focus on quantitative reconstruction of trace gases emission from the paleofires using the BCMSR records presented in our previous study and discuss spatial and temporal pattern of variation in trace gases emission from paleofires in Chinese Loess Plateau over the last two glacial cycles and the linkages to carbon cycle.

#### 2.2.3 Correlation of BC production with gaseous emissions and biomass burnt by fires

Biomass burning is a source of aerosols composed of organic hygroscopic particles, graphitic carbon, and variable amounts of trace gases such as CO<sub>2</sub>, carbon monoxide (CO), and methane (CH<sub>4</sub>) (Andreae, 1991; Kaufman et al., 1992; Ferek et al., 1998; Yamasoe et al., 2000). Both the radiation and greenhouse effects exerted by the released carbonaceous particles and gases have prompted a number of research groups to characterize fire emissions. Till now, a large body of information on emissions from the various types of biomass burning has been accumulated over the past decade. In relevant studies, emission ratios or emission factors have been introduced to describe

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the emission characteristics of biomass burning. These parameters relate the emission of a particular species of interest to that of a reference species, such as CO<sub>2</sub> or CO (emission ratio) or to the amount of fuel burnt (emission factor). Currently, Andreae and Merlet (2001) have critically evaluated the presently available data and integrated them into a consistent format for convenient usage of these data to global estimates of carbon emissions from biomass burning.

Despite the emission factors of the various smoke constituents are quite variable due to the different type of fuel, the fuel moisture and density, and the wind direction during a burn, only the average vales of emission factors from Andreae and Merlet (2001) are considered in this study. Since the paleofires in Chinese Loess Plateau have been recognized to be grassland burning (Wang et al., 2005), the emission factors for some pyrogenic species emitted from savanna and grassland burning were adopted for the quantitative reconstruction (see Table 1). At the same time, the emission factor of black carbon in the fire residues (BC<sub>residue</sub>) were calculated based on the BC/CE (carbon exposed to fire) ratios of savanna fires provided by Kuhlbusch and Crutzen (1995) and an average carbon content of 40% of dry matter for the grass species. For convenient purpose of computation, the ratios of three traces gases (CO<sub>2</sub>, CO, CH<sub>4</sub>) to BC in fire residues were also presented in Table 1.

#### 2.3 Estimate of trace gases emission and biomass burnt by paleofires from sedimentary BC records

The mass emission rates of trace gases released by paleo-biomass burning and the biomass burnt by paleofires (BBF) in Chinese Loess Plateau are respectively calculated using the following equation

$$TGMER(gC - TG m^{-2} yr^{-1}) = BCMSR(g cm^{-2} kyr^{-1}) \times (f \cdot gC - TG/gC - BC_{residue,C3} + (1 - f) \cdot gC - TG/gC - BC_{residue,C4}) \times (kyr 1000 yr^{-1}) \times (10 000 cm^{2} m^{-2})$$
(1)

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 $BBF(g dm m^{-2} yr^{-1}) = BCMSR(g cm^{-2} kyr^{-1}) \times (f/EF_{BCresidue,C3} + (1-f)/EF_{BCresidue,C4})$  $\times$  (kyr 1000 yr<sup>-1</sup>)  $\times$  (10 000 cm<sup>2</sup> m<sup>-2</sup>)  $\times$  (1000 g kg<sup>-1</sup>)

Where TGMER represents trace gases mass emission rates, BCMSR is black carbon mass sedimentation rates, and TG/BC<sub>residue.C3</sub> or TG/BC<sub>residue.C4</sub> denotes the ratios of trace gases, such as CO<sub>2</sub>, CO or CH<sub>4</sub> to BC in fire residue from C3 or C4 vegetation combustion, EF<sub>BCresidue C3</sub> or EF<sub>BCresidue C4</sub> is the emission factor of BC in fire residue from C3 or C4 vegetation combustion. The letter f is the relative abundance of C3 plants in the burned vegetation, whereas (1-f) means the relative abundance of C4 plants in the vegetation. These two parameters can be estimated by the following equation

$$\delta^{13}C_{\text{soil}} = \delta^{13}C_{C3}f + \delta^{13}C_{C4}(1-f)$$
(3)

Where  $\delta^{13}C_{soil}$  is the  $\delta^{13}C$  value of soil organic matter in loess or paleosol sample,  $\delta^{13}C_{C3}$  and  $\delta^{13}C_{C4}$  are the average  $\delta^{13}C$  value of C3 and C4 plants in the area, -27.3% and -12.6%, respectively (see Liu et al., 2003).

The  $\delta^{13}$ C values of soil organic matter (SOM) since last glacial maximum (LGM) in Lingtai and Weinan section are cited from Gu et al. (2003), whereas  $\delta^{13}$ C values of soil organic matter since LGM in Lijiayuan section are measured by Flash 1112 NC elemental analyzer coupled with MAT253 isotope ratio mass spectrometer (EA-IRMS) in this study. Due to the shortness of carbon isotope data downcore, we have obtained the  $\delta^{13}$ C values of SOM early than LGM using the strong correlation of the SOM  $\delta^{13}$ C with growing season precipitation (GSP) in Chinese Loess Plateau since LGM (Fig. 2). This correlation is also demonstrated by observation on the correlation between C4 biomass (or soil organic matter  $\delta^{13}$ C) in modern ecosystem and annual precipitation (An et al., 2005). The GSP used here were calculated by the modern seasonal distribution of precipitation in the study area (corresponding to 85%, 83%, 75% of MAP respectively in Lijiayuan, Lingtai and Weinan) and the reconstructed mean annual precipitation (MAP) in geological past as shown in the following section.

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Above-ground net primary productivity (ANPP) has been shown to be mainly controlled by rainfall e.g. mean annual precipitation (MAP) and temperature e.g. mean annual temperature (MAT) in the grasslands of Northern China and somewhere else in the world (Austin and Sala, 2002; Ni, 2004). Therefore, we applied Thornthwaite Memorial Model (Lieth and Box, 1972), which is based on mean annual precipitation and temperature, to estimate net primary productivity in Lijiayuan, Lingtai and Weinan areas. Moreover, Thornthwaite Memorial Model has been proven very useful and practical for net primary productivity (NPP) estimate in Chinese Loess Plateau (Wang and Zhang, 2005; Wang et al., 2006). The mean annual precipitation (MAP) and temperature (MAT) in Lijiayuan, Lingtai and Weinan were quantitatively reconstructed from the magnetic susceptibility (MS) data in these areas over last two glacial cycles (data cited from Ding et al., 1996, 2001) using the following equations from Porter et al. (2001)

$$MS(10^{-8} \text{ m}^3 \text{ kg}^{-1}) = 8.8078e^{0.0042 \text{ MAP(mm)}}, R^2 = 0.60 (n = 66)$$
 (4)

$$MS(10^{-8} \text{ m}^3 \text{ kg}^{-1}) = 11.18e^{0.1908 \text{ MAT}(^{\circ}\text{C})}, R^2 = 0.58 (n = 56)$$
 (5)

In order to obtain above-ground net primary productivity (ANPP), an R/S (root/shoot) ratio of 3.7 for grassland (Jackson et al., 1996) is used to apportion the above-ground biomass and below-ground biomass. Thus, the changes in paleo-biomass (e.g. ANPP) in Chinese Loess Plateau over the last two glacial cycles could be reconstructed.

#### 3 Results and interpretation

## 3.1 The changes in trace gases emission from paleo-biomass burning over the last two glacial cycles

In Fig. 3 we present the mass emission rates (MER) of trace gases released by paleobiomass burning in Lijiayuan, Lingtai and Weinan over the last two glacial cycles. BGD

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Data are expressed as the average MER value calculated for each marine isotopic stage, with the chronological constraints provided by Imbrie et al. (1984). During the glacial periods (including last glacial period: 12~71 kyr and penultimate glacial period: 125~190 kyr), the averaged MER for CO<sub>2</sub>, CO and CH<sub>4</sub> is respectively about  $39.9 \sim 43.5$ ,  $4.2 \sim 4.6$  and  $0.33 \sim 0.37$  g C m<sup>-2</sup>yr<sup>-1</sup> (totally  $44.4 \sim 48.5$  g C m<sup>-2</sup>yr<sup>-1</sup>) at Lijiayuan, and about  $19.5 \sim 23.6$ ,  $1.7 \sim 2.1$  and  $0.12 \sim 0.15 \,\mathrm{g\, C\, m^{-2}\, yr^{-1}}$  (totally 21.3 ~ 25.8 g C m<sup>-2</sup> yr<sup>-1</sup>) at Lingtai and Weinan, showing a clear southward decrease (Fig. 3). The averaged MER in the interglacial periods (including last interglacial period: 71~125 kyr and penultimate interglacial period: 190~216 kyr) is generally about  $2 \sim 3$  times lower than in the glacial periods; this value is respectively about 15.0  $\sim 16.5$ ,  $1.5 \sim 1.7$  and  $0.12 \sim 0.13$  g C m<sup>-2</sup> yr<sup>-1</sup> (totally  $16.6 \sim 18.3$  g C m<sup>-2</sup> yr<sup>-1</sup>) at Lijiayuan and about  $8.2 \sim 14.5$ ,  $0.6 \sim 1.1$ ,  $0.04 \sim 0.08$  g C m<sup>-2</sup> yr<sup>-1</sup> (totally  $8.8 \sim 15.7$  g C m<sup>-2</sup> yr<sup>-1</sup>) at Lingtai and Weinan. In addition to the glacial-interglacial variability, the MER values also vary notably within the two glacial periods. For instance, the MER in the stadial periods of L1-1, L1-5, L2-1, L2-3 and L2-5 (at 12~25, 60~71, 125~140, 155~165 and 175~190 kyr, respectively) is much higher than in the interstadial periods of L1-2, L1-4, L2-2 and L2-4 (at 25~35, 45~60, 140~155 and 165~175 kyr, respectively) (Fig. 3, see Fig. 7 for stratigraphic indication). For the Holocene period (0~12 kyr), the averaged MER for CO<sub>2</sub>, CO and CH<sub>4</sub> is about 47.0, 4.1 and 0.31 g C m<sup>-2</sup> yr<sup>-1</sup> at Lijiayuan and about  $7.6 \sim 33.3$ ,  $0.61 \sim 2.6$  and  $0.04 \sim 0.18$  g C m<sup>-2</sup> yr<sup>-1</sup> at Lingtai and Weinan. It should be emphasized that at Lingtai and Weinan the average MERs of total trace gases (TTG) in late Holocene is respectively 41.6 and 10.2 g C m<sup>-2</sup> yr<sup>-1</sup>, apparently higher than those in early Holocene (e.g. 30.5 and 6.4 g C m<sup>-2</sup> yr<sup>-1</sup>, see Fig. 3).

To examine regional TGMER dynamics during the glacial-interglacial periods, the trace gas mass emission rate over Chinese Loess Plateau (CLP) is approximately estimated provided the 44 × 10<sup>4</sup> km<sup>2</sup> of CLP could be north-southwards divided into three equal-sized zones, each of which has an average TGMER as the same as that in Lijiayuan, Lingtai and Weinan, respectively. The estimated TGMERs at different time intervals over CLP are shown in Fig. 4. It shows that the CO<sub>2</sub>, CO and CH<sub>4</sub> MERs

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during the glacial periods (e.g. MIS  $2 \sim 4$  and 6) range respectively from 6.5 to 19.7, 0.65 to 1.84 and 0.050 to 0.140 Tg C yr<sup>-1</sup>, with an average value of 12.6, 1.20 and 0.09 Tg C yr<sup>-1</sup>. Comparatively, the CO<sub>2</sub>, CO and CH<sub>4</sub> MERs during interglacial periods (e.g. MIS 5a–5e, 7a and 7b) are much lower, ranging respectively from 4.3–7.5, 0.37–0.67 and 0.028–0.050 Tg C yr<sup>-1</sup>, with an average value of 6.1, 0.51, 0.038 Tg C yr<sup>-1</sup>. However, a striking feature is that the CO<sub>2</sub>, CO and CH<sub>4</sub> MERs in Holocene (e.g. 12.6, 1.05 and 0.076 Tg C yr<sup>-1</sup>, respectively) are not as low as those during interglacial periods, but showing nearly the same level as those in glacial periods (Fig. 4).

## 3.2 The changes in biomass matter burnt by paleofires over the last two glacial cycles

Based on the emission factor of BC in fire residue, we work out the weights of dry biomass matter burnt by the paleofires per unit area per year in Lijiayuan, Lingtai and Weinan (Fig. 5). During the glacial periods (including last glacial period:  $12\sim71\,\mathrm{kyr}$  and penultimate glacial period:  $125\sim190\,\mathrm{kyr}$ ), the biomass burnt by paleofires is respectively  $52.3\sim164.5$  (mean value:  $\sim97$ ),  $22.8\sim78.8$  (mean value:  $\sim44$ ) and  $13.8\sim93.0$  (mean value: 48) g dm m<sup>-2</sup> yr<sup>-1</sup> in Lijiayuan, Lingtai and Weinan. However, the biomass burnt by paleofires during the interglacial periods have been largely reduced, with the burned biomass of  $25.5\sim46.1$  (mean value:  $\sim36$ ),  $14.2\sim35.7$  (mean value:  $\sim24$ ) and  $16.5\sim43.7$  (mean value:  $\sim30$ ) g dm m<sup>-2</sup> yr<sup>-1</sup> in Lijiayuan, Lingtai and Weinan. Moreover, the burned biomass also shows a southward decline from Lijiayuan to Weinan.

## 3.3 The changes in estimated aboveground net primary productivity (ANPP) over the last two glacial cycles

The reconstructed ANPP over the last two glacial cycles in Lijiayuan, Lingtai and Weinan are shown in Fig. 6. In general, the ANPP during glacial periods are relatively lower than those during interglacial periods. During the glacial periods, the ANPP is

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respectively around 127  $\sim$  139, 197  $\sim$  206 and 216  $\sim$  257 g dm m<sup>-2</sup> yr<sup>-1</sup> in Lijiayuan, Lingtai and Weinan. Comparatively, the ANPP during interglacial periods is about 147  $\sim$  163, 248  $\sim$  260 and 263  $\sim$  278 g dm m<sup>-2</sup> yr<sup>-1</sup> in Lijiayuan, Lingtai and Weinan, respectively. However, the changes in the Holocene ANPP at the three study sites are more complicated. For examples, the ANPP in the Holocene in Lijiayuan is around 196 g dm m<sup>-2</sup> yr<sup>-1</sup>, much higher than those during both glacial and interglacial periods whereas the ANPP in the Holocene in Lingtai and Weinan are between those during glacial and interglacial periods with the value of 239 and 246 g dm m<sup>-2</sup> yr<sup>-1</sup>, respectively.

# 3.4 Comparison of the biomass burnt by paleofires with the reconstructed ANPP over the last two glacial cycles

To examine the impact of paleofires on soil organic carbon storage, we use the ratio of the biomass burned by paleofires to the reconstructed ANPP (BF/ANPP) as a proxy indicator. The changes in the BF/ANPP ratios in Lijiayuan, Lingtai and Weinan over last two glacial cycles are illustrated in Fig. 7. The results show that the average BBF/ANPP ratio during glacial periods in Lijiayuan is nearly 75%, three times higher than that during interglacial periods. However, the average BBF/ANPP ratio during glacial periods in Lingtai and Weinan is respectively 22% and 20%, two times higher than those during interglacial periods in the areas.

#### 4 Discussion and conclusions

## 4.1 Trace gases emissions from paleo-biomass burning in Chinese Loess Plateau and potential implications

Our results show that the trace gas mass emission rates (TGMER) from paleofires in Lijiayuan, Lingtai and Weinan clearly display both glacial-interglacial and spatial

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variability (Fig. 3). Within the temporal series, the average TGMER of CO<sub>2</sub>, CO and  $CH_4$  during glacial periods are  $2 \sim 3$  times higher than those during interglacial periods. Geographically, the average TGMER of CO₂, CO and CH₄ in Lijiayuan are about 2 times higher than those in Lingtai and Weinan during both glacial and interglacial periods. This changing pattern is clearly different from the modeling study which shows a reduction in global fire carbon emission during LGM relative to interglacial period (Thonicke et al., 2005). It may be attributed to different methods used for estimating fire carbon emission between this study and the previous modeling study. The estimate in this study is based on sedimentary BC records by emphasizing empirical usage of paleo-record whereas the above modeling study seems to embed general features of biomass burning in the model by emphasizing on the strong control of vegetation cover on biomass burning rather than the regional climate conditions e.g. "Cold/dry regions with sparse or no vegetation show little or no biomass burning." (Thonicke et al., 2005). This difference implies that some region-specific fire regimes as revealed by paleo-record should be considered in modeling global fire carbon emissions during glacial-interglacial periods so that a more reasonable and reliable estimate would be obtained.

Comparatively, the  $CO_2$ , CO and  $CH_4$  MER in Holocene show the values close to those in glacial periods, much higher than the  $CO_2$ , CO and  $CH_4$  MER in glacial periods (Fig. 3). The far difference between TGMER in Holocene and those during interglacial periods could not be explained by the pattern of climate-induced natural fires, but possibly by the intensification of human burning activity in Holocene (Wang et al., 2005). To examine the contribution of human burning activity to TGMER in Holocene, we use the average TGMER during interglacial periods as an analog of the TGMER from natural fires in Holocene period provided the climates and vegetation during these two periods are almost under the same condition. Therefore, over Chinese Loess Plateau, the anthropogenic fire emission in Holocene can be obtained by subtracting the average of regional TGMER during interglacial periods (including last interglacial period: 71 ~ 125 kyr and penultimate interglacial period: 190 ~ 216 kyr) from the

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regional TGMER in Holocene (0  $\sim$  12 kyr) (Fig. 4). Thus, the anthropogenic fire emission in Holocene is respectively 6.53, 0.54 and 0.038 Tg C yr<sup>-1</sup> for CO<sub>2</sub>, CO and CH<sub>4</sub> (totally 7.108 Tg C yr<sup>-1</sup>), comparable with the TGMER difference caused by altered fire regimes between glacial and interglacial periods (e.g., 6.51, 0.68 and 0.054 Tg C yr<sup>-1</sup> for CO<sub>2</sub>, CO and CH<sub>4</sub>, totally 7.244 Tg C yr<sup>-1</sup>). This means the fire emission resulted from Holocene human activities in Chinese Loess Plateau did not exceed the natural range of fire emission under different fire regimes.

For a purpose of comparison, we also carried out an estimate of the trace gas emission from modern biomass burning (forest fires and grassland fires) in Chinese Loess Plateau based on the typical annual amounts of biomass burned in Gansu, Ningxia, Qinghai, Nei Mongol, Shaanxi, Shanxi and Henan provinces of this area (data cited from Streets et al., 2003) and the emission factors listed in Table 1. The results show that modern fire carbon emissions in Chinese Loess Plateau (CLP) are respectively 14.52, 0.95 and 0.063 Tg C yr<sup>-1</sup> for CO<sub>2</sub>, CO and CH<sub>4</sub> with a total emission of 15.533 Tg C yr<sup>-1</sup>, clearly higher than the average TGMER in both Holocene and glacial periods (Fig. 3). This indicates that forest and grassland fires are still one of main sources for trace gas emissions although the fire suppression practices have been undertaken for many years. Therefore, more effective measures should be taken to reduce the fire emissions in the region. In addition, if we assumed the Holocene TG-MER over CLP (e.g. 13.726 Tg C yr<sup>-1</sup>) could represent the pre-industrial level of carbon emission from biomass burning in this region, then the pre-industrial emission would be ~88% of present biomass burning. This result, although only from CLP region, is comparable with ~70% of present biomass burning on global scale as provided by Mouillot et al. (2006), but far different from the frequently-used assumption that pre-industrial emissions were only 10% of present biomass burning (Crutzen and Zimmermann, 1991).

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#### Fire regimes and the carbon storage in soils in Chinese Loess Plateau

The carbon isotope analyses of soil organic matter and pollen studies show that the Loess Plateau was covered by grassland vegetation both during glacial and interglacial periods (Sun et al., 1997; Gu et al., 2003; Jiang and Ding, 2005). Currently, Grassland 5 soil carbon stocks amount to at least 10% of the global total (Eswaran et al., 1993). Even modest changes in inputs to grassland carbon storage may there result in significant and long-lived sequestration (Scurlock and Hall, 1998). Here we will assess the effects of fire regime changes during glacial-interglacial periods on the carbon storage in soils in Chinese Loess Plateau. Comparisons of the biomass burnt by paleofires (BBF) with the above-ground net primary productivity (ANPP) show that the average BBF/ANPP ratio during glacial periods in Lijiayuan is about 75%, three times higher than that during interglacial periods (Fig. 7). Moreover, the BBF/ANPP ratios in the stadial periods of L1-1, L1-5, L2-1, L2-3 and L2-5 are higher than 100%, indicating some below-ground biomass were also burnt by paleofires in those periods. This suggests that paleofires may be the overwhelming force modulating the cycling of terrestrial organic carbon in the northern part of Chinese Loess Plateau. However, the average BBF/ANPP ratio during glacial periods in Lingtai and Weinan is respectively 22% and 20%, two times higher than those during interglacial periods in the areas. It means that paleofires have had a minor impact on carbon storage in the southern part of Chinese Loess Plateau during both glacial and interglacial periods. This case study proposes that wildfires is a very important disturbing factor in determining carbon inputs into soils in semi-arid region like northern part of CLP. At the same time, the extent to which the wildfires influence the carbon storage is highly variable even at relatively small regional scale (e.g. within CLP). The data sets in this study may be used as fine constraints in model parameterization where wildfires are considered in C-cycle model.

Since the spatio-temporal patterns of fire occurrences in Chinese Loess Plateau during the glacial-interglacial periods were governed by the wet-dry cycles (Wang et al., 2005), the extent to which wildfire influenced the carbon storage in the region should ultimately determined by the precipitation. In Lijiayuan, northern part of CLP, the mean annual precipitation when 100% ANPP was burnt during the glacial periods is about 280 mm. This may provide a baseline value for environmental conditions in future assessment of carbon storage in semi-arid or arid regions of Northwestern China as well as elsewhere in the world.

#### 4.3 Fire regimes and carbon cycling mechanism

Early study has shown that a large carbon sink ("missing sink") in the global carbon budget may be located in the terrestrial regions of the Northern Hemisphere (Tans et al., 1990). Recent study has shown that the aboveground biomass of China's grassland may have functioned as carbon sink in the past two decades (Piao et al., 2007). How has the changes in fire regimes influenced the carbon sequestration in the grassland in Chinese Loess Plateau? For entire Loess Plateau, the overall difference in fire-burned biomass between glacial period (27.73 Tg dm yr<sup>-1</sup>) and interglacial period  $(12.87 \,\mathrm{Tg}\,\mathrm{dm}\,\mathrm{yr}^{-1})$  is  $14.86 \,\mathrm{Tg}\,\mathrm{dm}\,\mathrm{yr}^{-1}$ , corresponding to  $\sim 5.944 \,\mathrm{Tg}\,\mathrm{C}\,\mathrm{yr}^{-1}$ . It means this amount of vegetation carbon must have been burnt during the glacial periods instead of having been incorporated into soil organic carbon cycles during the interglacial periods. Recent observations on wildfires in tropical savannas have shown that a high grass biomass accumulates during the wet seasons and burns occur at 1-3 years intervals and generally during the dry season (e.g., Gillon, 1983; Coutinho, 1990; Mistry, 1998). Due to similar ecological and fire weather features, natural fires in Chinese Loess Plateau may share the same burning pattern as that in savannas. In this case, about 14.86 Tg dm (~5.944 Tg C) of extra biomass may be annually burnt by more frequent or intense wildfires in the dry season during the glacial period and largely contribute to intra- and/or inter-annual variation of atmospheric carbon budgets in Chinese Loess Plateau. To date, several researchers have estimated using a carbon cycle model, CASA (Carnegie-Stanford-Ames Approach) that the size of biomass carbon sink is  $\sim 7.2 \,\mathrm{Tg}\,\mathrm{C}\,\mathrm{yr}^{-1}$  in China's grassland (with an area of  $331 \times 10^4 \,\mathrm{km}^2$ ) from 1982 to 1999 (Fang et al., 2003; Piao et al., 2005). Applying the estimate of biomass

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C sink per area  $(7.2 \, \text{Tg} \, \text{C} \, \text{yr}^{-1} / 331 \times 10^4 \, \text{km}^2)$  to our study, the biomass C sink could be  $\sim 0.96 \,\mathrm{Tg}\,\mathrm{C}\,\mathrm{yr}^{-1}$  in grassland of CLP (with an area of  $44 \times 10^4 \,\mathrm{km}^2$ ). Thus, the net loss of ~ 5.944 Tg C yr<sup>-1</sup> resulted from fire regime changes in CLP over interglacialto-glacial cycle, although having some uncertainties, accounts for more than six times 5 of the estimated carbon sink. It indicates that long-term changes in fire regimes over past two glacial cycles may have substantial impact on the carbon sink in grassland in Northwestern China. Therefore, more frequent wildfires in Chinese Loess Plateau during the glacial periods must have shifted the grassland from being a carbon sink to net carbon source.

Comparatively, the biomass left unburnt due to less frequent wildfires during the interglacial periods would incorporate into soil carbon pool and experience multiple carbon cycling processes with different turnover times (e.g. Parton et al., 1988; Trumbore, 2000). For example, coarse particles that are mainly fresh litter have mean residence times between 0.5 and 20 years, whereas the carbon in the clay fraction has mean residence times of about 60-80 years (Balesdent, 1996); Baties and Sombroek (1997) quote a global mean of 22 years (litter included), and a maximum of up to 5000 years. That means carbon release from soil carbon pool needs much longer time than that from biomass burning. In soil carbon cycling, outputs are dominated by the efflux of CO<sub>2</sub> from the soil and the production of CO<sub>2</sub> in soils is almost entirely from root respiration and microbial decomposition of organic matter. Because the efflux of CO<sub>2</sub> from soils (often lumping microbial and root respiration together as "soil respiration") tends to increase with temperature and often soil water content or precipitation (Davidson and Janssens, 2006), higher level CO<sub>2</sub> efflux from soil in Chinese Loess Plateau must occur at summer time. However, the produced CO<sub>2</sub> can be immediately sequestered by local vegetation in this growing season and thus would not contribute to intra-annual variation of atmospheric carbon budgets. Therefore, it seems that the altered fire regimes from glacial to interglacial period can change carbon flow partly from rapid cycling process (vegetation-atmosphere cycle) to a relatively slow cycling process (vegetation-soil-atmosphere cycle). In this sense, less frequent wildfires in

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Chinese Loess Plateau during the interglacial periods would still make the grassland serving as a transient carbon sink.

For the long-term carbon cycle, the more consumed biomass of  $\sim 14.86\,\mathrm{Tg}\,\mathrm{dm}\,\mathrm{vr}^{-1}$ by frequent wildfires during glacial periods than during interglacial periods could yield  $_{5}$  ~ 0.117 Tg BC yr<sup>-1</sup> in loess surface, which represents a net sink of biospheric carbon and thus of atmospheric CO<sub>2</sub> as well as a source of O<sub>2</sub> (e.g. Seiler and Crutzen, 1980), but can only compensate ~ 1.95% of the annually more released trace gases from wildfires during glacial periods than that during interglacial periods.

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Table 1. Emission factors used in this study for the pyrogenic species of CO₂, CO, CH₄ and black carbon produced by biomass burning.

Species	C3 Emission factors	C4 Emission factors	Species/BC <sub>residue</sub> ratio for C3	Species/BC <sub>residue</sub> ratio for C4
	g species kg <sup>-1</sup> dry matter	g species kg <sup>-1</sup> dry matter	g C-species g <sup>-1</sup> C <sup>-1</sup>	g C-species g <sup>-1</sup> C <sup>-1</sup>
CO <sub>2</sub>	1568 <sup>a</sup>	1664 <sup>a</sup>	54.55	113.45
CO	106 <sup>a</sup>	63 <sup>a</sup>	5.79	6.75
CH <sub>4</sub>	4.8 <sup>a</sup>	2.2 <sup>a</sup>	0.46	0.41
BC <sub>smoke</sub>	0.56 <sup>a</sup>	0.48 <sup>a</sup>	0.07	0.12
BC <sub>residue</sub>	7.84 <sup>b</sup>	4.0 <sup>b</sup>	1	1

<sup>&</sup>lt;sup>a</sup> Data by Andreae and Merlet (2001).

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<sup>&</sup>lt;sup>b</sup> Adapted values from mean BC/CE (carbon exposed to fire) (%) provided by Kuhlbusch and Crutzen (1995) with a conversion factor of 4.0, where C3 emission factor for  $BC_{residue}$  is from extratropical forest (pine needle and deciduous wood) and C4 emission factor for is BC<sub>residue</sub> from savanna grass.



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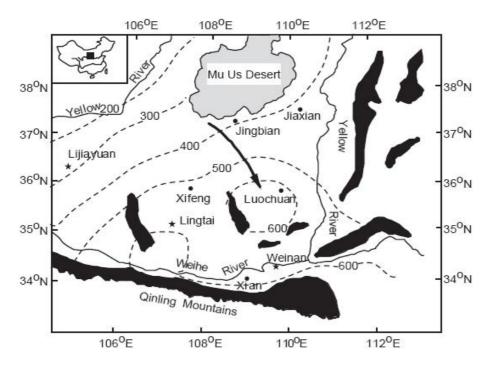


Fig. 1. Schematic map showing the locality of the studied section labeled as the five-angle star. The arrow indicates the dominant subaerial wind direction in winter seasons, coinciding with the observed decrease in grain size and thickness of loess. The desert (dotted) and mountains (black areas) around and within the Loess Plateau are shown. The solid square in the inset map shows the locality of the Loess Plateau in Continental China.



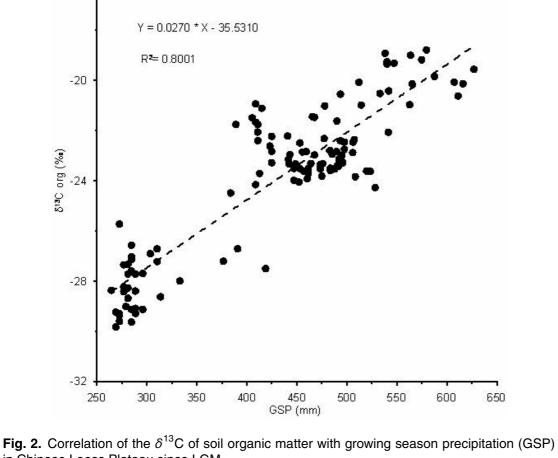
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in Chinese Loess Plateau since LGM.

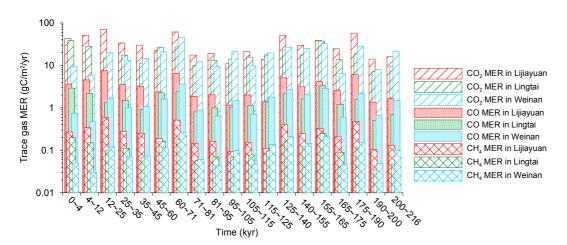


Fig. 3. The mass emission rates (MER) of CO<sub>2</sub>, CO and CH<sub>4</sub> released by biomass burning in Lijiayuan, Lingtai and Weinan over the last two glacial cycles.

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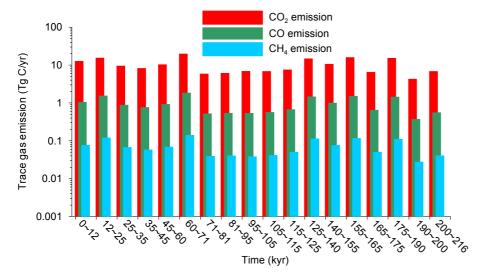


Fig. 4. The estimated TGMERs in the whole Chinese Loess Plateau at different time intervals over the last two glacial cycles.



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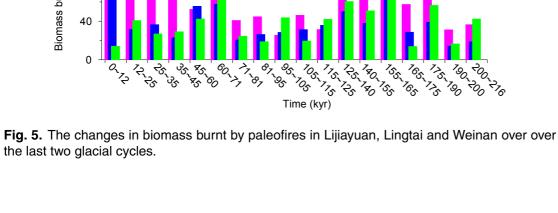
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BBF in Lijiayuan

BBF in Lingtai

BBF in Weinan

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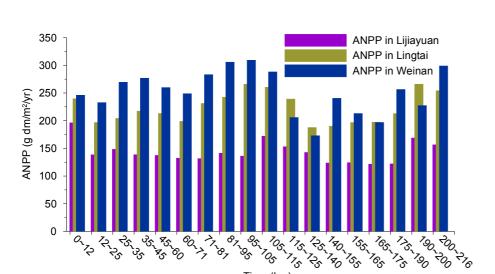
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the last two glacial cycles.

Biomass burned by fires (g dm/m²/yr)



**Fig. 6.** The reconstructed aboveground net primary productivity (ANPP) in Lijiayuan, Lingtai and Weinan over the last two glacial cycles.

Time (kyr)

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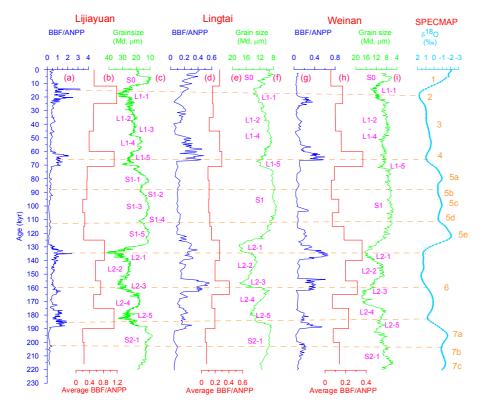


Fig. 7. Comparison of the biomass burnt by paleofires with the reconstructed ANPP in Lijiayuan, Lingtai and Weinan over the last two glacial cycles. All the three loess-soil sections consist of the Holocene soil S0, the last glacial loess deposit L1, the last interglacial soil S1, the penultimate glacial loess unit L2 and the penultimate interglacial soil S2. The 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, and 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 loess unit (L2) also consists of two weakly developed soils (L2-2 and L2-4) in the middle part. 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.