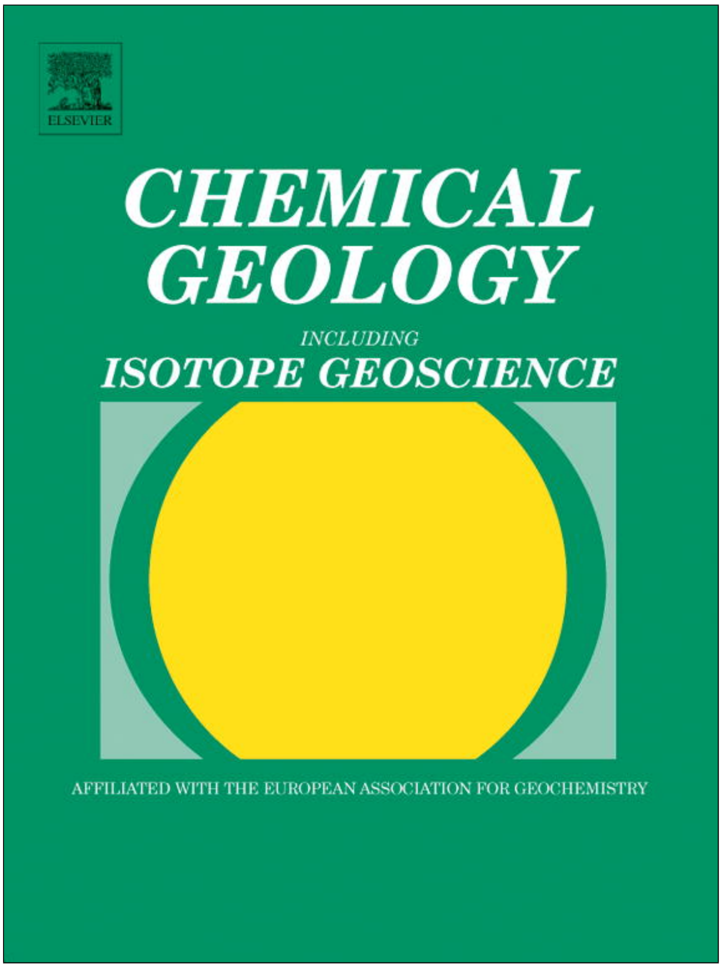
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Stable carbon isotope of black carbon in lake sediments as an indicator of terrestrial environmental changes: An evaluation on paleorecord from Daihai Lake, Inner Mongolia, China

Xu Wang *, Linlin Cui, Jule Xiao, Zhongli Ding

Key Laboratory of Cenozoic Geology and Environment, Institute of Geology and Geophysics, Chinese Academy of Sciences, P.O. Box 9825, Beijing, 100029, China

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ABSTRACT

We measured the carbon isotope ratio of black carbon (BC) from the Daihai Lake sediment core (DH99a) in north-central China with an objective to examine the effectiveness and sensitivity of the $\delta^{13}C$ values of BC ($\delta^{13}C_{BC}$) as a potential indicator of terrestrial environmental changes. We first performed a statistical study on the available data regarding carbon isotope fractionation (CIF) during the conversion of C3 and C4 vegetation to BC and observed that the mean CIF for BC produced from C3 plants is −0.3‰, whereas that for BC from C4 plants is −1.7‰. This result provides a solid reference for reconstructing vegetation and environmental changes using the $\delta^{13}C_{BC}$ values. The $\delta^{13}C_{BC}$ record in the DH99a sediment core spanning the last ca 10,000 years displayed large variations from −23.7‰ to −29.2‰, which suggests that C3 plants dominantly occupied the Daihai Lake region during the Holocene. The most negative $\delta^{13}C_{BC}$ peaks coincided with high values of tree percentages and grain sizes, which occurred under relatively wetter climatic conditions during the middle Holocene (ca 6500–3200 cal. yr BP) and an interval between 1700 and 1350 cal. yr BP. In contrast, the least negative $\delta^{13}C_{BC}$ values corresponded to low values of tree percentages and grain sizes during relatively drier phases of the early and late Holocene. The generally negative correlation of the $\delta^{13}C_{BC}$ values with the tree percentages and grain sizes was thought to reflect a negative correlation of the $\delta^{13}C_{BC}$ values with the monsoon precipitation. This correlation is consistent with the response of carbon isotope in modern C3 plants to precipitation in north China. Therefore, we developed a computational model to reconstruct the changes in annual precipitation over the Daihai Lake region using the $\delta^{13}C_{BC}$ values. The results indicated that the annual precipitation was highly variable, ranging from 170 mm lower to 310 mm higher than that at present during the middle Holocene, whereas the annual precipitation was generally ~70 mm lower than that at present during the early and late Holocene. The general features of the inferred precipitation changes using the $\delta^{13}C_{BC}$ values are generally consistent with those reconstructed using pollen data of the same sediment core. Meanwhile, the $\delta^{13}C_{BC}$ values tend to register some extreme variations of monsoon precipitation, which were not reflected in the pollen assemblages. We conclude that the $\delta^{13}C_{BC}$ values in the Daihai Lake sediments may serve as a sensitive and reliable proxy for monitoring monsoon precipitation.

1. Introduction

Black carbon (BC) is uniquely produced from incomplete combustion processes, such as forest fires and the burning of fossil fuels (Goldberg, 1985). BC contains a variety of materials, ranging from large fragments of slightly charred biomass to microscopic soot and graphitic black carbon (or elemental carbon), which are formed by the condensation of gases produced during combustion (Masiello, 2004). Because BC particles are chemically inert, resistant to oxidation and biodegradation in natural conditions, they can be preserved in soils, lake and marine sediments for as long as thousands to millions of years (Masiello and Druffel, 1998). Therefore, BC signatures in geological deposits can be used as evidence of natural fires that occurred in their surroundings (Wang et al., 2005b). Furthermore, because BC is a product of vegetation fires, the carbon isotope composition of BC may provide an indication of the type of vegetation that was burned and thus, reflect possible environmental changes. If this characteristic was proven valid, the BC carbon isotope would be a very useful and promising proxy indicator, which is especially important for marine and lake sediments that contain organic matter from complicated origins (i.e., including both autochthonous and allochthonous organic matter); therefore, carbon isotopes of bulk organic matter in these settings are not suitable for constraining the nature of terrestrial vegetation.

Despite its potential proxy value, the $\delta^{13}C$ value of BC ($\delta^{13}C_{BC}$) has received only limited use for past vegetation or environmental

* Corresponding author. Tel.: +86 10 8299 8581; fax: +86 10 6201 0846.
E-mail address: xuking@mail.iggcas.ac.cn (X. Wang).

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reconstruction (e.g., Bird and Cali, 1998; Turney et al., 2001; Jia et al., 2003). The reason for this limited use may be partly because studies conducted to determine the effects of combustion on the carbon isotope composition of plant tissues and the products of combustion have produced varied and contradicting results (Hall et al., 2008 and references therein). However, laboratory experiments may not produce results that are completely comparable with the results derived from materials produced in natural fires. The reasons for the difference in the results lie in the following two aspects: first, the types of plant materials burned in experiments cannot easily cover the range of biomass in natural fires; and second, the conditions of combustion, such as oxygen availability, the temperature at which the material is combusted and the length of burning, used in the experiments may be not as the same as those of natural fires. Therefore, carbonization conditions in wildfires are strongly variable and difficult to reproduce experimentally (Resco et al., 2011). In this context, the potential of $\delta^{13}C_{BC}$ values to serve as a vegetation and/or environmental proxy indicator needs to be examined in paleorecords.

Some recent studies have indicated that it was possible to obtain meaningful paleoenvironmental information from the $\delta^{13}C$ values of archeological charcoal and charred cereal grains (e.g., February and van der Merwe, 1992; Ferrio et al., 2006; Aguilera et al., 2008; Hall et al., 2008). Nevertheless, the methods used for sample preparation in these studies are quite different from those employed for BC extraction from soils and sediments. For example, the methods used for extracting BC in this and other studies include acid treatments for removing carbonate/silicates and the oxidative digestion of organic carbon using acid dichromate (e.g., Lim and Cachier, 1996; Bird and Gröcke, 1997). The remaining residue from the aforementioned treatments is called BC, which represents a series of oxidation-resistant carbon components, including charcoal and atmospheric BC particles (or submicron soot spheres). Therefore, the BC extracted from soils and sediments may not be entirely the same component as archeological charcoal. In this case, we should independently assess the potential application of $\delta^{13}C_{BC}$ for paleoenvironmental studies. A previous study regarding the carbon isotope analysis of elemental carbon (namely BC) from plants, modern soils and sediments has shown that the $\delta^{13}C$ value of elemental carbon can provide an indication of the type of vegetation being burned (Bird and Gröcke, 1997). Nevertheless, the usage of BC isotope analysis for paleovegetation and/or paleoenvironmental reconstruction has not yet been evaluated through the paleorecord under changing climatic conditions. In addition, the specific environmental implications that $\delta^{13}C_{BC}$ potentially possessed have not yet been clarified.

The goal of this study is to assess if it is possible to obtain meaningful vegetation and/or environmental information from the $\delta^{13}C_{BC}$ values in the paleorecord. First, we reviewed recent studies regarding variations in the carbon isotope compositions of the burned and un-burned materials and then presented a common feature of the carbon isotope fractionation during the conversion of C3 and C4 vegetation to BC. On the basis of the above mostly representative fractionation factors, we tried to reconstruct the vegetation and/or environmental changes during the Holocene using the $\delta^{13}C_{BC}$ values in Daihai Lake sediments. The reliability of the reconstructed vegetation and/or environmental changes was then examined by comparison with pollen and other climatic proxies from the same sediment core.

2. Study area

Daihai Lake (40°29′ to 40°37′N; 112°33′ to 112º46′E) is a semi-brackish closed lake that lies in Inner Mongolia in north-central China (Fig. 1). This lake has an area of 133 km^2, a maximum water depth of 14 m and an elevation of 1221 m. The lake basin is bordered by the Manhan Mountains to the north and the Matou Mountains to the south. Hills are distributed on the east of the lake, and plains are present along the western shore. The highest peaks of the Manhan and Matou Mountains reach elevations of 2305 and 2035 m, respectively. The lake has a catchment of 2289 km^2, and five major rivers enter the lake, but no rivers drain the lake (Xiao et al., 2004).

Daihai Lake is located at the transition between semi-humid and semi-arid areas in the middle temperate zone of China (Fig. 1). The mean annual temperature is 5.1 °C with a July average of 20.5 °C and a January average of −13.0 °C (Fig. 2). The mean annual precipitation is 423 mm, and approximately 80% of the annual precipitation falls in June–September, with a peak value of 122 mm in August. The mean annual evaporation reaches 1162 mm and is 2.8 times the annual precipitation (Fig. 2). The lake is covered with ca 50 cm of ice from November to March. In the lake area, the winter climate is controlled by the dry, cold northwesterly winter monsoon that generates frequent dust storms from late autumn to spring, whereas the summer climate is dominated by the warm, moist southeasterly summer monsoon that is responsible for most of the annual precipitation and for rainstorm activities (Gao, 1962; Chinese Academy of Sciences, 1984; Zhang and Lin, 1992).

The modern natural vegetation of the Daihai Lake basin belongs to the southern temperate steppe (Compilatory Commission of Vegetation of China, 1980; Compilatory Commission of Annals of Liangcheng County, 1993). Forests consisting of *Populus davidiana*, *Betula platyphylla*, *Larix*, *Pinus tabulaeformis*, *Pinus sylvestris* var. *mongolica*, *Picea*, *Ulmus* and *Salix* are distributed on the northern slopes of the mountains, and these forests are accompanied by mesophilous scrubs of *Spiraea*, *Hippophae rhamnoides*, *Ostryopsis davidiana*, *Prunus armeniaca* and *Lespedeza bicolor* as well as herbs of *Carex enervis*, *Artemisia argyi*, *Artemisia sacrorum*, *Lilium pumilum*, *Anemarrhena asphodeloides* and *Sanguisorba officinalis* under the trees. Alpine meadows that are mainly composed of *Stipa baicalensis*, *Aneurolepidium chinensis*, *Lilium*, *S. officinalis* and *A. sacrorum* are developed in the mountainous areas above 1900 m a.s.l. The hilly areas are covered by grasses and herbs, such as *Stipa krylovii*, *Stipa bungeana*, *Setaria viridis*, *Artemisia frigida*, *A. sacrorum*, *Medicago falcata* and *Thymus mongolicus*. Halophilic meadows and patches of boggy meadow, which are composed of *Carex stenophylla*, *Achnatherum splendens*, *Elymus dahuricus*, *A. frigida*, *Iris lactea*, *Taraxacum mongolicum*, *C. enervis*, *Suaeda glauca*, *Potentilla chinensis* and *Polygonum aviculare*, dominate over the lakeshore plain and the frontal fringes of diluvial fans. Based on some $\delta^{13}C$ studies on modern plants in the arid areas in northern China (Zhang et al., 2003a; Wang et al., 2005a; Wang, 2007; Li et al., 2009), most of the scrubs, grasses and herbs occupied in the Daihai Lake basin belong to C3 plants, except for the species of *Setaria viridis*.

3. Materials and methods

3.1. Lithology, sampling and chronology of DH99a sediment core

In the summer of 1999, drilling was performed at a water depth of 13.1 m in the central part of Daihai Lake using a piston corer driven by a Japanese-made TOHO drilling rig (Model D1-B). Sediment cores, which were collected in half-cut polyethylene tubes, were extracted to a depth beneath the lake floor of 12.02 m; these cores were designated DH99a. The core sections were split, photographed, and described on location. The cores were continuously cut into 2-cm segments to create samples for laboratory analyses. The core sediments are composed of homogeneous mud and are divided into two parts at a core depth of 6.65 m: a grayish-green to grayish-black upper part with burrows at depths of 2.10–5.70 m and a light to dark gray laminated lower part.

The upper 10.63 m of the DH99a sediment core was used for the present study and was sampled at 4-cm intervals, resulting in a total of 260 samples for analyses of the BC carbon isotope.

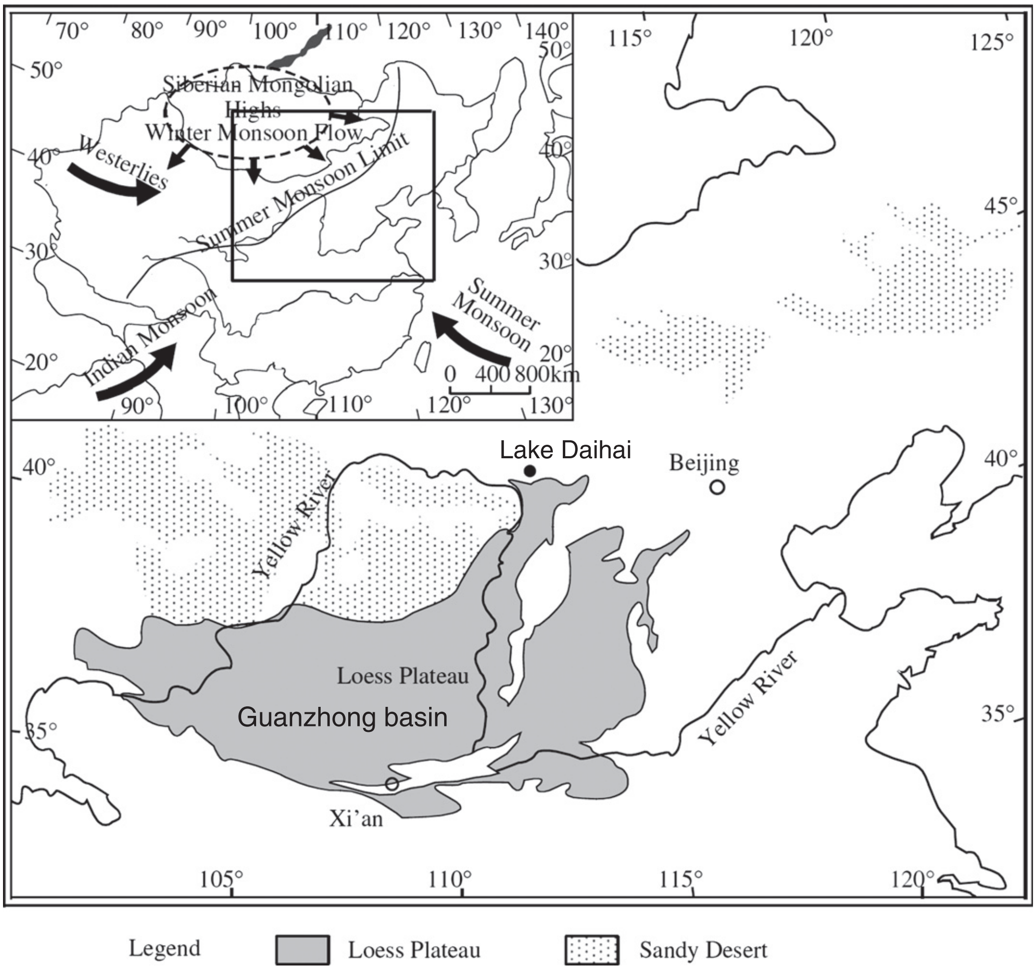

Fig. 1. Map showing the location of Daihai Lake and the climatic system of China, including the East Asian and Indian summer monsoons, the winter monsoon winds associated with the Siberian–Mongolian High, and the Westerly winds. The summer monsoon is a steady flow of warm, moist air from the tropical oceans, and the winter monsoon is a flow of cold, dry air from north-central Asia.

The time scale for DH99a sediment core was provided by Xiao et al. (2004) using AMS ^{14}C dating on 8 bulk samples collected from the organic-rich sediments. The radiocarbon samples were dated using a HVEE Tandetron AMS-II system at the Center for Chronological Research, Nagoya University. The AMS ^{14}C data indicate that the Daihai Lake sediments reach a thickness of ca 11 m for the Holocene Epoch. An average sedimentation rate of ca 100 cm kyr^{-1} and a sampling interval of 4 cm for the DH99a core provide a potential temporal resolution of ca 40 yr.

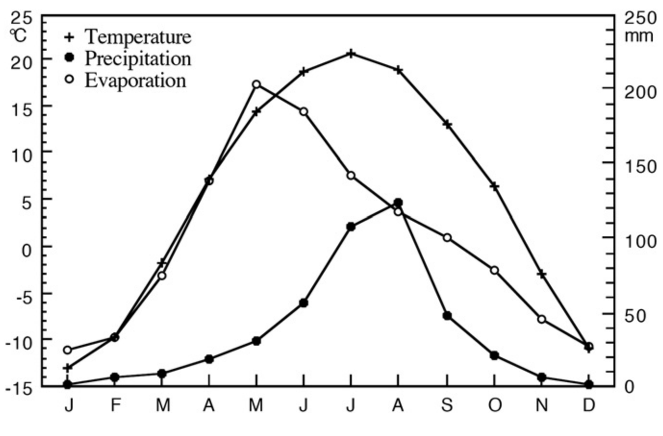

Fig. 2. Monthly changes in the mean annual temperature, mean annual precipitation, and mean annual evaporation over the Daihai Lake region. The data are an average of the observations from 1959 to 1988.

3.2. Extraction of black carbon and stable carbon isotope measurement

In this study, we used the chemical method developed by Lim and Cachier (1996) to extract BC from the lake sediment samples. In brief, the carbonates and part of the silicates in the samples were removed using an acid treatment with HCl (3 mol/L) and HF (10 mol/L)/HCl (1 mol/L) in sequence. The treated samples were then oxidized with 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 the soluble organic matter and kerogen (e.g., Wang et al., 2001). The remaining refractory carbon in the residues is called BC and includes charcoal and atmospheric BC particles (Lim and Cachier, 1996).

To examine the reproducibility of the above chemical treatment method, we extracted BC in six aliquots from one sample of the Daihai Lake surface sediment (DH98 core). The DH98 sediment core was taken from the position with 300 m distance to DH99a in Northwest. This sediment core was not dated and not performed for further study due to a low sediment recovery. The isolated BC containing residues were combusted in a Thermo elemental analyzer (Flash EA 1112) integrated with a ConFlo III system with a Thermo MAT253 isotope ratio mass spectrometer for $\delta^{13}C$ analyses. The analyses were calibrated using glycine ($\delta^{13}C_{VPDB} = -33.3‰$) as an external working standard. The carbon isotope results are expressed in the conventional delta (δ) notation as the per mil (‰) deviation from the standard

Peedee belemnite (PDB). The results indicate that the standard deviation for the measured $\delta^{13}C$ values from those six BC samples is better than ±0.1‰ (see Table 1).

For the sequential measurement on BC samples from the DH99a core, duplicates of each sample were analyzed, and the external working standard materials were inserted between every 6 samples to monitor the working conditions of the analyzer. The precision for analysis of the external standard was better than ±0.1‰, and the error for the BC samples was better than ±0.2‰.

4. Results

4.1. Carbon isotope signatures of BC produced from C3 and C4 plant burning

To obtain a general characteristic of carbon isotope fractionation during the conversion of C3 and C4 vegetation to BC, we compiled all of the currently available data published in literature (see Table 2). The data were adopted for this study based on the following criteria: (1) all of the data from natural burns and controlled field burns were subject to preferential consideration. For the burning experiments performed in laboratories, we chose the data from burns >300 °C because most natural surface fires usually occur at temperatures between 300 and 600 °C (e.g., Swift et al., 1993); (2) The data from charring treatments under limited O_2 conditions (e.g., alu-foil wrap, sand-buried and in Argon atmosphere) were also counted because the smoldering combustion that usually occurs under limited O_2 conditions may be a part of source for BC production (e.g., Czimczik et al., 2002); (3) The data from combustions, even for the same plant species at different temperatures, such as 300, 400, 500, 600 and 700 °C, were considered separately as discrete burnings to encompass the maximum extent of the conditions that might occur in natural fires; (4) The data for each C3 or C4 plant species that were burned under the same conditions were taken as a mean value using an arithmetic average calculation and were counted only once. The dataset used in this study encompasses 40 C3 plants and 58 C4 plants from a total of 134 individual combustion experiments, controlled field burns and natural burns (see references in Table 2).

The results for the carbon isotope fractionation (CIF) during the conversion of C3 and C4 vegetation to BC are shown in Fig. 3. The CIF for BC from the C3 plant combustion ranges from −3‰ to +3‰, with a mean value of −0.3‰. In contrast, the CIF for BC from the C4 plant combustion varies from −10‰ to +3‰, with a mean value of −1.7‰. The relative decrease in the $\delta^{13}C$ values of BC with respect to its C3 wood precursors is presumably due to the lower thermal stability of the isotopically enriched cellulose when compared with aromatic lignin groups (Czimczik et al., 2002). In contrast, the more negative fractionation for BC from the C4 plants is attributed to the existing protected organic matter in phytoliths (Krull et al., 2003).

Table 1
List of the measured $\delta^{13}C_{BC}$ values for the six aliquots of one surface sediment sample in DH98 core.

Sample weight (g)	Residue weight after treatment (mg)	BC content in residue (%)	$\delta^{13}C$ of BC (‰)
1.0035	7.6	21.720	−25.48
1.0080	7.7	21.156	−25.51
1.0009	8.0	21.893	−25.54
0.6078	4.9	20.670	−25.56
0.6015	4.9	18.919	−25.42
0.6055	5.4	17.719	−25.38
S.D. (1σ)			0.07

4.2. Black carbon isotope record of DH99a sediment core

The $\delta^{13}C_{BC}$ values for DH99a core are shown in Fig. 4, and these values are compared with BC concentration, tree percentages in pollen assemblages and grain size records. The $\delta^{13}C_{BC}$ values for the DH99a core range from −23.7‰ to −29.2‰, falling into the isotopic range of C3 plants, with an average of −25.4‰. The $\delta^{13}C_{BC}$ values between ca 10,353 and 7500 cal. yr BP show less negative values with relatively small variations (−25.0 ± 0.5‰). At the same time, both the BC concentration and the percentages of tree pollens are uniformly low (with an average of approximately 0.23% and 20% for BC concentration and tree pollens, respectively) in the lake sediments, and the median grain sizes (Md) of the lake sediments are relatively low and remain constant during this period (Fig. 4). In addition, the $\delta^{13}C_{BC}$ values show a slowly decreasing trend (e.g., from −24.4‰ to a low of −26.0‰) between ca 10,353 and 8000 cal. yr BP, which is coincident with a gradual increase in BC concentration and the percentages of trees during the same period. During the middle Holocene period (ca 7500 to 3000 cal. yr BP), the $\delta^{13}C_{BC}$ values are more negative and display a large variation (mean value: −25.5 ± 1.1‰) with four more negative peaks (<−27‰) centered at ca 6440, 5650, 5150 and 3700 cal. yr BP, respectively. At the same time, the BC concentration increases notably and demonstrates relatively high values during the above four periods. Meanwhile, the percentages of trees exhibit an overall increase with a portion of more than 60% occurring at ca 7380 and 5550 cal. yr BP, and the Md values of the lake sediments also show a large increase with highly variable values during this period (Fig. 4). During the late Holocene (ca 3000 cal. yr BP to present), the $\delta^{13}C_{BC}$ values are less negative with an intermediate size of variation (−25.2 ± 0.9‰), in contrast with the low percentages of trees in the lake basin and the low BC concentration and Md values of the lake sediments. Within this period, a prominent, more negative peak (<−27‰) for $\delta^{13}C_{BC}$ occurs at approximately ca 1500 cal. yr BP, which corresponds with the increased BC concentration, tree percentages and Md values in a short interval between ca 1700 and 1350 cal. yr BP (Fig. 4).

5. Discussion

5.1. BC carbon isotope record in the Daihai Lake region as a monsoon precipitation proxy

In previous studies, BC or charcoal carbon isotope records have been used to reflect changes in C3/C4 abundances (Winkler, 1994; Bird and Gröcke, 1997; Bird and Cali, 1998; Clark et al., 2001; Jia et al., 2003), fluctuations of trees versus grasses in vegetation (Cachier et al., 1989; Wang et al., 2012) and variation in aridity or precipitation (Ferrio et al., 2006; Aguilera et al., 2008; Hall et al., 2008). In this study, the $\delta^{13}C_{BC}$ values for the DH99a core are generally correlated with the BC concentration, the tree percentages in the pollen assemblages and the grain size record in the same sediment core (Fig. 4), which suggests that the BC carbon isotope could have recorded terrestrial environmental changes over the Daihai Lake region. Our recent study has found a generally close correlation of the BC mass sedimentation rates (BCMSR) with the tree percentages in the pollen assemblages over Daihai Lake region and interpreted this as a possible indication of the relationship between the vegetation dynamics and the intensity of the fires (Wang et al., 2013). For example, more wood fuels would increase BC production, resulting in high BCMSR. Since the BCMSR is mainly controlled by the BC concentration at our study site, changes in BC concentration may also be a reflection of vegetation dynamics. Meanwhile, the pollen record of DH99a core sediments has been employed to reveal a history of climate changes over Daihai Lake region, i.e., an increasing woody plants suggest a rise in temperature and precipitation (Xiao et al., 2004). In this case, the changes in both the BC concentration and the tree percentages

Table 2
Summary of carbon isotope fractionation for BC from C3 and C4 plants during combustion.

Type of combustion	Carbon isotope fractionation (CIF) for BC from C3 plants				Carbon isotope fractionation (CIF) for BC from C4 plants				References
	$\delta^{13}C$ (‰) plants	$\delta^{13}C$ (‰) BC	Distribution (CIF, n)	Mean	$\delta^{13}C$ (‰) plants	$\delta^{13}C$ (‰) BC	Distribution (CIF, n)	Mean	
Controlled experiment, fire place	−22.8	−23.1	(−0.5, 1)	−0.3					Leavitt et al. (1982)
Natural burns, char residue					−10.5 to −12.5 (n = 2)	−14.5 to −19.5 (n = 2)	(−9.5, 1), (−1.5, 1)	−5.5	Cachier et al. (1985)
Natural burns, char particle					−12.5	−19.5 to −23.5 (n = 2)	(−10.5, 1), (−6.5, 1)	−8.5	Cachier et al. (1985)
Lab charring, in vacuo, 500 °C, 120 min	−27.8 to −29.6 (n = 4)	−28.0 to −29.7 (n = 4)	(−0.5, 4)	−0.2	−12.1 to −12.6 (n = 3)	−12.4 to −14.2 (n = 3)	(−0.5, 3)	−0.6	Bird and Gröcke (1997)
Flame combustion, ash	−24.4 to −29.2 (n = 3)	−23.8 to −27.9 (n = 3)	(+0.5, 2), (+1.5, 1)	+0.8	−12.9 to −13.3 (n = 3)	−12.5 to −14.7 (n = 3)	(−1.5, 1), (−0.5, 2)	−0.8	Turekian et al. (1998)
Flame combustion, char particle	−24.4 to −29.2 (n = 3)	−23.8 to −27.4 (n = 3)	(+0.5, 1), (+1.5, 2)	+1.2	−12.9 to −13.3 (n = 3)	−15.0 to −19.9 (n = 3)	(−7.5, 1), (−3.5, 1), (−1.5, 1)	−4.2	Turekian et al. (1998)
Smolder combustion, ash	−24.4 to −27.7 (n = 3)	−24.0 to −27.0 (n = 3)	(−0.5, 1), (+0.5, 2)	+0.4	−12.9 to −13.3 (n = 3)	−12.6 to −13.5 (n = 3)	(−0.5, 1), (+0.5, 2)	+0.1	Turekian et al. (1998)
Smolder combustion, char particle	−24.4 to −27.7 (n = 3)	−24.6 to −27.3 (n = 3)	(−0.5, 2), (+0.5, 1)	−0.1	−12.9 to −13.3 (n = 3)	−13.8 to −17.5 (n = 3)	(−4.5, 1), (−3.5, 1), (-0.5, 1)	−3.0	Turekian et al. (1998)
Flame combustion, char residue	−26.9 to −31.1 (n = 5)	−27.0 to −30.3 (n = 5)	(−0.5, 3), (+0.5, 2)	−0.1	−5.5 to −14.9 (n = 16)	−10.4 to −14.2 (n = 16)	(−5.5, 1), (−1.5, 1), (−0.5, 5), (+0.5, 4), (+1.5, 3), (+2.5, 2)	+0.1	Beuning and Scott (2002)
Lab charring, Argon, 340–480 °C	−28.2 to −29.4 (n = 4)	−28.7 to −30.5 (n = 4)	(−0.5, 3), (−1.5, 1)	−0.8					Czimczik et al. (2002)
Natural burns	−25.7 to −31.6 (n = 6)	−26.0 to −32.4 (n = 6)	(−1.5, 1), (−0.5, 2), (+0.5, 1), (+1.5, 1), (+2.5, 1)	+1.1	−12.0 to −15.0 (n = 19)	−15.1 to −17.8 (n = 19)	(−5.5, 1), (−4.5, 7), (−3.5, 3), (−2.5, 5), (−1.5, 3)	−3.4	Krull et al. (2003)
Controlled field burns	−27.3 to −30.6 (n = 4)	−27.5 to −30.2 (n = 4)	(−1.5, 1), (−0.5, 1), (+0.5, 2)	−0.3	−12.7 to −14.3 (n = 4)	−11.7 to −15.5 (n = 4)	(−1.5, 2), (−0.5, 1), (+0.5, 1)	−0.8	Krull et al. (2003)
Laboratory experiment, open flame					−11.7	−13.6	(−1.5, 1)	−1.5	Krull et al., 2003
Natural burns	−24.4 to −31.3 (n = 10)	−23.9 to −30.6 (n = 10)	(−1.5, 1), (+0.5, 7), (+1.5, 2)	+0.5	−11.6 to −14.3 (n = 10)	−11.1 to −14.7 (n = 10)	(−1.5, 1), (−0.5, 8), (+2.5, 1)	−0.3	Ning et al. (2004)
Laboratory experiment, sand-buried, 300–500 °C	−22.63 to −26.14	−23.34 to −27.76 (n = 3)	(−2.0, 1), (−1.5, 1), (−0.5, 1)	−1.3					Ferrio et al. (2006)
Burns, >400 °C, char residue			(−2.5, 1), (−1.5, 8), (−0.5, 3)	−1.3					Turney et al. (2006)
Controlled burns, with O_2, 300–600 °C, 120 min	−26.7 to −28.0 (n = 2)	−27.3 to −28.3 (n = 8)	(−1.5, 1), (−0.5, 6), (+0.5, 1)	−0.5					Ascough et al. (2008)
Laboratory experiment, 400–700 °C, char ash	−24.6 to −26.1 (n = 3)	−24.7 to −26.3 (n = 5)	(−0.5, 3), (+0.5, 2)	−0.1	−12.3 to −13.8 (n = 5)	−11.9 to −16.3 (n = 11)	(−3.5, 2), (−2.5, 1), (−1.5, 2), (−0.5, 4), (+0.5, 2)	−1.2	Das et al. (2010)
Laboratory experiment, 400–700 °C, char particle	−26.1 (n = 2)	−25.9 to −26.7 (n = 3)	(−0.5, 2), (+0.5, 1)	−0.2	−12.3 to −13.0 (n = 4)	−12.8 to −18.7 (n = 8)	(−5.5, 1), (−4.5, 1), (−3.5, 1), (−1.5, 4), (−0.5, 1)	−2.5	Das et al. (2010)

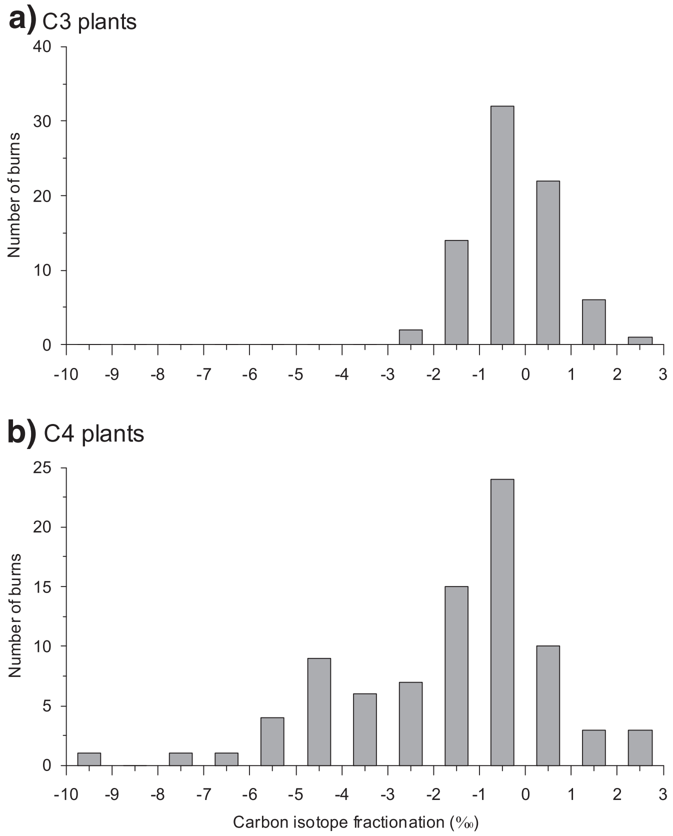

Fig. 3. Diagrams showing the distribution of carbon isotope fractionation during the conversion of C3 and C4 vegetation to black carbon: (a) BC from C3 plants; (b) BC from C4 plants (Data are sourced from the references listed in Table 2).

may reflect the monsoon precipitation variability at our study site. In general, high precipitation rates would enhance the soil erosion over the lake region and increase the transport capacity of streams and rivers, leading to more, coarser clastic materials available for river transport and subsequent deposition in the central part of the lake (Håkanson and Jansson, 1983). Therefore, Peng et al. (2005) inferred that the grain size distribution of Daihai Lake sediments can be linked predominantly with the hydrological cycles over the lake region, i.e., an increase in the median grain size of clastic particles, resulting from a increase in the content of the silt-size fraction, is generally related to an increase in the precipitation intensity in the lake region. Because the BC concentration, the tree percentages in the pollen assemblages and the grain sizes of the lake sediments are ultimately governed by monsoon precipitation in the Daihai Lake region, $\delta^{13}C_{BC}$ variation may readily reflect changes in monsoon precipitation at our study location. To examine the use of the $\delta^{13}C_{BC}$ value as a potential precipitation proxy, the contribution to the $\delta^{13}C_{BC}$ signals from different sources deserves further discussion.

5.1.1. Influence of C3/C4 abundances on BC carbon isotope in the Daihai Lake region

Because C3 and C4 plants have a distinct carbon isotope composition, e.g., with mean $\delta^{13}C$ values of −27‰ for C3 plants and −13‰ for C4 plants (Farquhar et al., 1989), changes in the C3/C4 abundances in a specific vegetation ecosystem should affect the variation of $\delta^{13}C_{BC}$ values. The modern vegetation in the Daihai Lake region is dominated by C3 plants (e.g., some deciduous trees, C3 grasses and shrubs), which may be attributed to its relatively cold and dry climate. Some recent studies have shown that the C4 abundance in the Inner Mongolia grassland is mainly controlled by the temperature of the warmest month, i.e., the percentage of C4 plant plants was significantly lower below the 22 degree isotherm of the warmest month

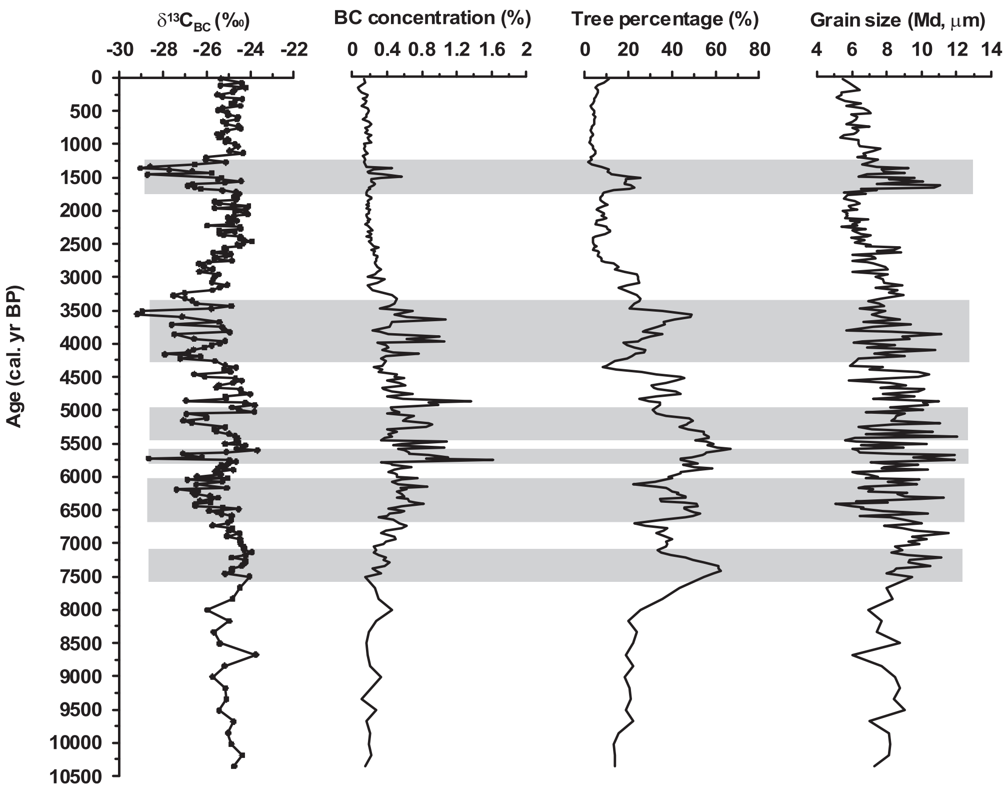

Fig. 4. Variation in the $\delta^{13}C$ values of black carbon for the DH99a sediment core in comparison with BC concentration, tree percentages in the pollen assemblages and median grain sizes.

(Auersward et al., 2009; Wittmer et al., 2010). The average temperature of ~20.5 °C for July, the warmest month in Daihai, is apparently lower than the above-threshold temperature, which may account for the extremely low abundance of C4 plants in the modern vegetation of the region.

To reconstruct the changes in the C3/C4 abundances over the Daihai Lake region during the Holocene, we should first choose proper $\delta^{13}C$ end-member values for the C3 or C4 plants. In a previous study, Liu et al. (2003) have used a $\delta^{13}C$ value of −27.3‰ and −12.6‰ as the end-member values for the C3 and C4 plants, respectively, on the Chinese Loess Plateau. Because there is a 1.3‰ decrease in the $\delta^{13}C$ of atmospheric CO_2 since the industrial revolution due to the combustion of ^{13}C-depleted fossil fuels (Marino et al., 1992), we adopted −26.0‰ and −11.3‰ as the $\delta^{13}C$ end-member values for C3 and C4 plants, respectively, during the Holocene in this study. Considering that the average CIF was −0.3‰ and −1.7‰ during the conversion of C3 and C4 vegetation to BC (See Fig. 3), we finally decided to choose −26.3‰ and −13.0‰ as the end-member $\delta^{13}C$ values for BC produced from the C3 and C4 plants, respectively, during the Holocene. Therefore, the average $\delta^{13}C$ value of −25.6‰ for BC throughout the DH99a record indicates that the vegetation in the Daihai Lake region was primarily composed of C3 plants during the Holocene. Even if the most positive value of −23.9‰ is considered to be a pure contribution from the increase in C4 abundance in the vegetation, the highest possible C4 abundance is approximately 18%. This result further confirms the dominance of C3 plants in the Daihai Lake region throughout the Holocene.

The most positive shifts in $\delta^{13}C_{BC}$ mainly occurred at ca 7000, 5000, 2500 and 1200 cal. yr BP, which correspond to certain key periods of temperature declines, which were inferred from the total inorganic carbon (TIC) record from the DH99a core (Xiao et al., 2006). If the variation in the $\delta^{13}C_{BC}$ value accurately reflects the changes in the C3/C4 abundances, then these more positive isotope shifts would indicate increases in the C4 abundance in the Daihai Lake region during the above-mentioned cold periods. However, this assumption is unreasonable because many previous studies have observed warm temperatures and the enhanced seasonality of monsoon precipitation that favor the growth of C4 plants in northern China (Gu et al., 2003; Zhang et al., 2003b; Liu et al., 2005a; Yang et al., 2012). In contrast, the most likely occurrence of C4 plants may exist in the warm and/or enhanced summer precipitation periods with high percentages of trees appearing in the region, which corresponds to the most negative $\delta^{13}C_{BC}$ values in our record (indicated by the gray bars in Fig. 4). Therefore, the possible influence of C4 occurrences on $\delta^{13}C_{BC}$ values during those periods is that it makes the $\delta^{13}C_{BC}$ values less negative, which is similar to the case as if there was no existence of C4 plants. In this case, although there may be some BC produced from a small amount of C4 plants, the net effect of $\delta^{13}C_{BC}$ variations in the total BC for the DH99a core reveals the change of monsoon precipitation over the Daihai Lake region.

5.1.2. Influence of trees versus grasses on BC carbon isotope in the Daihai Lake region

Although the substantial correlation between the $\delta^{13}C_{BC}$ values and the tree percentages in the pollen was thought to reflect a strong correlation between the $\delta^{13}C_{BC}$ values with the monsoon precipitation, changes in the trees/grasses ratios may also affect $\delta^{13}C_{BC}$ variation if there are apparent isotope differences between them. Because the $\delta^{13}C_{BC}$ values in our record mostly fall in the range of carbon isotope for C3 plants, we only consider the influence of trees versus C3 grasses on the changes in $\delta^{13}C_{BC}$ values. Since the $\delta^{13}C$ difference between trees and C3 grasses has not been well constrained so far, the best way to achieve the net isotope differences between these plants should be limiting the comparisons to plants at the same geographic site. However, no such data can be currently obtained at our study site. Since our study site lies in semi-arid/semi-humid transition zone, we could make a comparison for $\delta^{13}C$ values between trees and grasses within the contiguous areas of these zones such as the Chinese Loess Plateau (CLP) instead. Liu et al. (2005b) have reported a mean $\delta^{13}C$ value of −27.2‰ for trees (range: −26.3 to −28.2‰; n = 5) and a mean $\delta^{13}C$ value of −28.0‰ for C3 grasses (range: −25.0 to −29.6‰; n = 22) on the CLP. Recently, Zheng and Shangguan (2007) have investigated foliar $\delta^{13}C$ values of nine dominant species on the CLP, and obtained a similar result that trees had higher $\delta^{13}C$ values than herbs with the mean $\delta^{13}C$ value of −26.7‰ for trees (n = 39) and −27.9‰ for herbs (n = 49). In this study, we adopted the weighted mean $\delta^{13}C$ values for these two results by the sampling numbers, which are −26.7‰ and −27.9‰ for trees and C3 grasses, respectively. The higher $\delta^{13}C$ values for trees than those for C3 grasses have been also observed for plants on Changbai Mountains, Northeast China (Tan et al., 2009). Besides, the influence of changes in gymnosperm vs. angiosperm wood on carbon isotope has been found substantial in some previous studies (Leavitt and Newberry, 1992; Diefendorf et al., 2010). For example, deciduous angiosperms and evergreen angiosperms have higher carbon isotope fractionation factors (Δ_{leaf}) than evergreen gymnosperms, by 2.7‰ and 2.2‰ respectively, at the same geographic site (Diefendorf et al., 2010). To constrain this influence in our study, we assumed that a mean value of 2.5‰ could represent the carbon isotopic difference between angiosperm and gymnosperm trees over Daihai Lake region. Since the trees on the CLP used for the above $\delta^{13}C$ statistical calculation contained nearly identical numbers of angiosperms and gymnosperms (Zheng and Shangguan, 2007), we consider the mean $\delta^{13}C$ value of −26.7‰ as an approximate median value between the $\delta^{13}C$ of these two groups. Therefore, the mean $\delta^{13}C$ values of angiosperm and gymnosperm trees should be −25.5‰ and −28.0‰, respectively.

To assess the influence of changes in trees versus C3 grasses on $\delta^{13}C_{BC}$, we should first determine the end members of carbon isotope composition for BC produced from trees (angiosperm vs. gymnosperm) and C3 grasses. In this study, we use −25.5‰, −28.0‰ and −27.9‰ as the end member $\delta^{13}C$ values for modern angiosperm, gymnosperm trees and C3 grasses, respectively. Because the $\delta^{13}C$ value of atmospheric CO_2 ($\delta^{13}C_{air}$) during the pre-industrial part of Holocene was prominently different from its value today (Leuenberger et al., 1992; Marino et al., 1992) and there were also some variations in the $\delta^{13}C$ value of atmospheric CO_2 ($\delta^{13}C_{air}$) within the pre-industrial part of Holocene, we used a time-dependent $\delta^{13}C_{air}$ value to calculate the end member $\delta^{13}C$ values for angiosperm, gymnosperm trees and C3 grasses at a specific time in the Holocene based on the above three end member $\delta^{13}C$ values for modern angiosperm, gymnosperm trees and C3 grasses, respectively, using the following three equations:

$$\delta^{13}C_{AG}(T) = \delta^{13}C_{AG}(T_0) + \delta^{13}C_{air}(T) - \delta^{13}C_{air}(T_0) \tag{1}$$

$$\delta^{13}C_{GM}(T) = \delta^{13}C_{GM}(T_0) + \delta^{13}C_{air}(T) - \delta^{13}C_{air}(T_0) \tag{2}$$

$$\delta^{13}C_{GR}(T) = \delta^{13}C_{GR}(T_0) + \delta^{13}C_{air}(T) - \delta^{13}C_{air}(T_0) \tag{3}$$

where $\delta^{13}C_{AG}(T)$, $\delta^{13}C_{GM}(T)$ and $\delta^{13}C_{GR}(T)$ represent the end member $\delta^{13}C$ values for angiosperm, gymnosperm trees and C3 grasses, respectively, at a specific time in the Holocene, $\delta^{13}C_{AG}(T_0)$, $\delta^{13}C_{GM}(T_0)$ and $\delta^{13}C_{GR}(T_0)$ are the end member $\delta^{13}C$ values for modern angiosperm, gymnosperm trees and C3 grasses, respectively (i.e., $\delta^{13}C_{AG}(T_0) = -25.5‰$; $\delta^{13}C_{GM}(T_0) = -28.0‰$; $\delta^{13}C_{GR}(T_0) = -27.9‰$), $\delta^{13}C_{air}(T)$ denotes the $\delta^{13}C$ value of atmospheric CO_2 at a specific time in the Holocene and $\delta^{13}C_{air}(T_0)$ is the $\delta^{13}C$ value of atmospheric CO_2 at present. Here, we use the $\delta^{13}C_{air}$ value of −8.1‰ in 2003 as the value of $\delta^{13}C_{air}(T_0)$. The $\delta^{13}C_{air}(T)$ value was inferred by interpolating values from Antarctic ice-core records, together with modern data from two Antarctic stations (Halley Bay and Palmer Station) of the

CU-INSTAAR/NOAA-CMDL network for atmospheric CO_2 measurements, as described in Ferrio et al. (2005). In brief, we simply input our interested date ranges into the files on the internet (http://web.udl.es/usuaris/x3845331/AIRCO2_LOESS.xls) to obtain the mean $\delta^{13}C_{air}$ at a specific time period. The resulting smoothed $\delta^{13}C$ curve of atmospheric CO_2 from ca 10,353 cal. yr BP to present is shown in Fig. 5b. According to this record, the $\delta^{13}C_{air}$ value used to calculate $\delta^{13}C_{AG}(T)$, $\delta^{13}C_{GM}(T)$ and $\delta^{13}C_{GR}(T)$ varies between −7.51‰ and −6.24‰. Considering a mean CIF of −0.3‰ for BC from C3 plant combustion (Fig. 3), we finally use the values of $\delta^{13}C_{AG}(T)-0.3$‰, $\delta^{13}C_{GM}(T)-0.3$‰ and $\delta^{13}C_{GR}(T)-0.3$‰ as the end member $\delta^{13}C$ values for BC produced from angiosperm, gymnosperm trees and C3 grasses, respectively, at a specific time in the Holocene.

Based on the end member $\delta^{13}C$ values for BC produced from angiosperm, gymnosperm trees and C3 grasses at a specific time in the Holocene and the portion of angiosperm, gymnosperm trees and grasses in the pollen record at the same time, we construct a predicted-$\delta^{13}C_{BC}$ time series that reflects the changes in trees (angiosperm vs. gymnosperm)/grasses in the Daihai Lake region using the following equation:

$$\delta^{13}C_{BC}(\text{trees/grasses}) = \delta^{13}C_{BC}(AG) * f(AG) + \delta^{13}C_{BC}(GM) * f(GM) + \delta^{13}C_{BC}(GR) * f(GR) \quad (4)$$

where $\delta^{13}C_{BC}$(trees/grasses) represents the $\delta^{13}C_{BC}$ value caused by the changing trees (angiosperm vs. gymnosperm)/grasses proportion at a specific time in the Holocene, $\delta^{13}C_{BC}(AG)$, $\delta^{13}C_{BC}(GM)$ and $\delta^{13}C_{BC}(GR)$ are the end member $\delta^{13}C$ values for BC produced from angiosperm, gymnosperm trees and C3 grasses, respectively, at a specific time in the Holocene, and f(AG), f(GM) and f(GR) denote the percentages of angiosperm, gymnosperm trees and grasses in the pollen record at a specific time in the Holocene. In this study, f(AG), f(GM) and f(GR) are obtained from the pollen data provided by Xiao et al. (2004). The results indicate that the $\delta^{13}C_{BC}$(trees/grasses) varies between −25.7‰ and −27.5‰ (Fig. 5c). Furthermore, for the most time during the Holocene (i.e., before ~85 cal. yr), the $\delta^{13}C_{BC}$(trees/grasses) only changes from −25.7‰ to −26.7‰. The amplitude for this variation (~1.0‰) is substantially smaller than that for our $\delta^{13}C_{BC}$ record (amplitude: 5.5‰), only accounting for ~18% of the full amplitude. The remaining part of the variation in the $\delta^{13}C_{BC}$ values may result from changes in the monsoon precipitation.

5.1.3. Interpretation of BC carbon isotope using monsoon precipitation in the Daihai Lake region

Previous studies have shown that soil humidity or water availability is an important climatic factor that influences plant carbon isotopic discrimination (Deines, 1980; Francey and Farquhar, 1982). The restricted soil water would cause plants to close their stomata to conserve water for photosynthesis. This process in turn reduces the intercellular partial pressures of CO_2, which increases the plant $\delta^{13}C$ values accordingly. Recently, investigations on modern ecosystems in northern China have also found that the plant $\delta^{13}C$ values shifted toward more negative values with the increase of mean annual precipitation (Wang et al., 2003; Liu et al., 2005c). In the Daihai Lake region, both the tree percentages in pollen assemblages and the grain-size distribution of lake sediments have been inferred to be proxy indicators for past changes in the East Asian monsoon precipitation such that greater tree percentage and higher median grain size reflect increased monsoonal precipitation (Xiao et al., 2004; Peng et al., 2005). Therefore, the overall negative correlation of $\delta^{13}C_{BC}$ in our record with the tree percentage and the median grain size (Fig. 4) reveals a generally negative correlation of $\delta^{13}C_{BC}$ with monsoon precipitation in the Daihai Lake region.

5.2. Reconstruction of precipitation using $\delta^{13}C_{BC}$ in the Daihai Lake region

Because the variation in precipitation results in a large extent of the changes in the $\delta^{13}C_{BC}$ values in the Daihai Lake region, we attempt to reconstruct the precipitation using the $\delta^{13}C_{BC}$ value in this study.

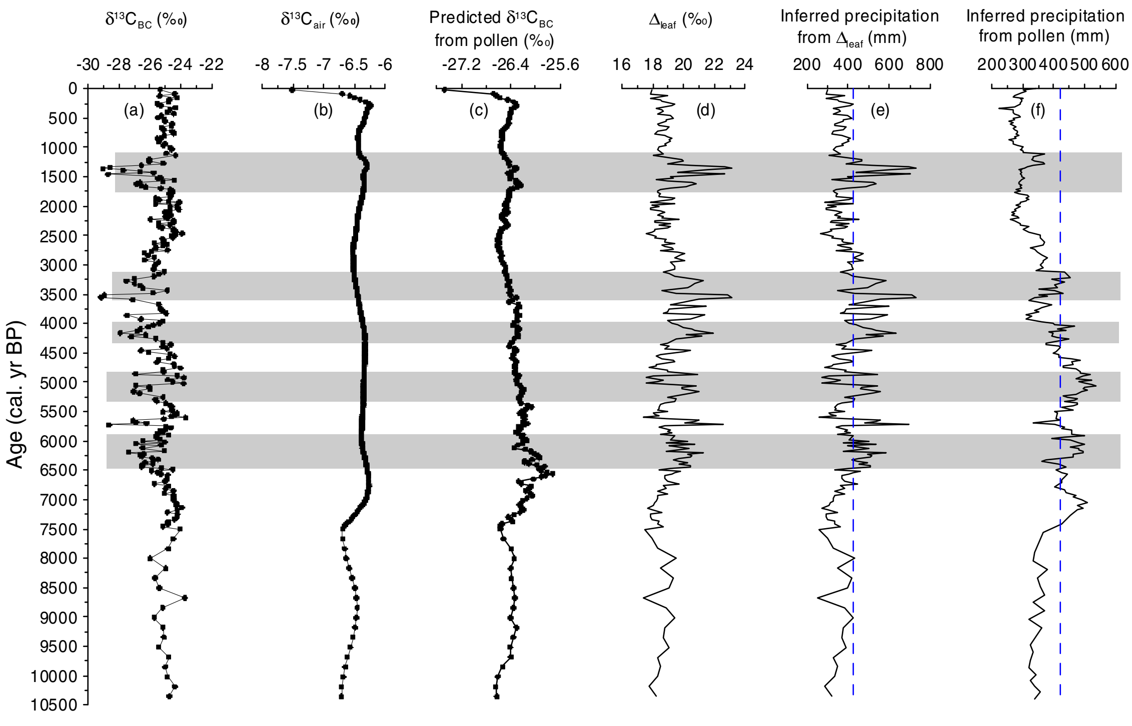

Fig. 5. Variations in the measured $\delta^{13}C_{BC}$ values (a), inferred $\delta^{13}C_{air}$ values (data source from Ferrio et al., 2005) (b), predicted $\delta^{13}C_{BC}$ values using tree/grass percentages (c), Δ_{leaf} calculated using the $\delta^{13}C_{air}$ and $\delta^{13}C_{BC}$ values (d), inferred annual precipitation using Δ_{leaf} values (e), and reconstructed precipitation using pollen data cited from Wen et al., 2012 (f). The blue vertical dish lines represent the annual precipitation at present. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

For the purpose of simplification in the precipitation reconstruction, the influence of changes in trees versus C3 grasses on $\delta^{13}C_{BC}$ values could be neglected since it results in a limited variation in BC carbon isotopes (Fig. 5c). Before the reconstruction, we should eliminate a small portion of changes in $\delta^{13}C_{BC}$ value caused by the changing $\delta^{13}C$ value of atmospheric CO_2 from the original BC carbon isotope record. To eliminate this contribution, we then use the leaf ^{13}C discrimination (Δ_{leaf}), which reflects a balance of photosynthesis and stomatal conductance and their coupled response to the environment. Values of Δ_{leaf} generally decrease with reductions in water availability, reflecting a down-regulation of stomatal conductance and increased water-use efficiency (Farquhar et al., 1989; Ehleringer and Cerling, 1995). Recently, a strong positive correlation between Δ_{leaf} and mean annual precipitation (MAP) has been observed at the global scale (Diefendorf et al., 2010; John, 2010).

The Δ_{leaf} values were calculated using the following equation:

$$\Delta_{leaf} = \left(\delta^{13}C_{air} - \delta^{13}C_{leaf}\right) / \left(1 + \delta^{13}C_{leaf}/10^3\right) \tag{5}$$

where $\delta^{13}C_{air}$ represents the $\delta^{13}C$ value of atmospheric CO_2 and $\delta^{13}C_{leaf}$ is the $\delta^{13}C$ value of leaf or plant. For the certain period of Holocene, $\Delta_{leaf}(T)$ could be obtained using the $\delta^{13}C_{air}(T)$ (shown in Fig. 5b) and $\delta^{13}C_{leaf}(T)$. $\delta^{13}C_{leaf}(T)$ was estimated using $\delta^{13}C_{BC}$ at the same time period plus 0.3‰ since the mean CIF for BC from C3 plant is −0.3‰.

To reconstruct the past precipitation, we must select a proper transfer function to convert the Δ_{leaf} values to precipitation changes at our study site. Wang et al. (2008) have measured the $\delta^{13}C$ value of C3 plants occurring in north China and found that the mean $\delta^{13}C$ values of C3 plants were well correlated with mean annual precipitation (MAP). Using their data set, we established a transfer function between Δ_{leaf} and MAP. To calculate the Δ_{leaf} values for those C3 plants, we adopt −8.1‰ (i.e., $\delta^{13}C_{air}(T_0)$) as modern atmospheric CO_2. The established transfer function is shown in Fig. 6. As can be seen, there is a strong positive correlation ($R^2 = 0.44$) between Δ_{leaf} and MAP in north China, which is similar with that observed at the global scale. Therefore, we transferred the $\Delta_{leaf}(T)$ to precipitation

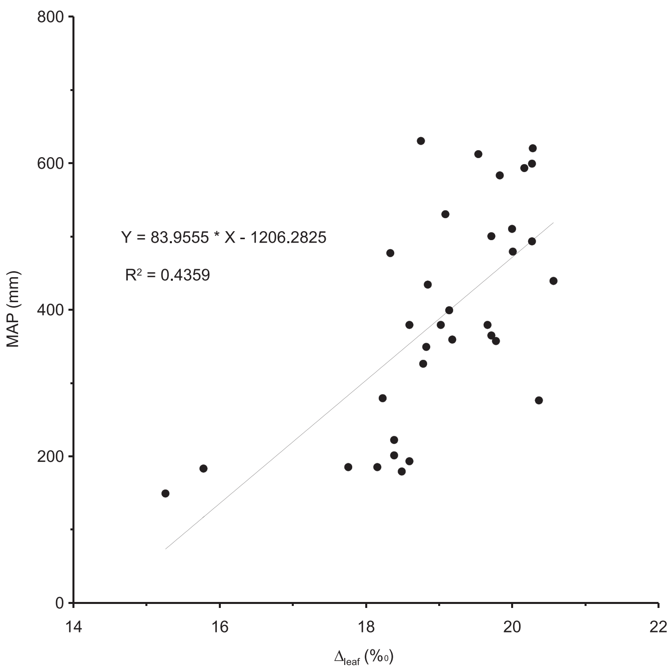

Fig. 6. Plot showing the correlation between MAP (mm) and Δ_{leaf} values in north China (Data are cited from Wang et al., 2008). The transfer function for precipitation is also shown.

based on this regional transfer function. The reconstructed changes in annual precipitation over the Daihai Lake region during the Holocene are shown in Fig. 5e. The inferred annual precipitation varied from about 250 mm to 730 mm. The results suggest that the change in precipitation over the Daihai Lake region during the Holocene is characterized by three stages: a dry episode before ca 6500 cal. yr BP, then a wet period between ca 6500 and 3200 cal. yr BP and a dry episode after ca 3200 cal. yr BP. Before ca 6500 cal. yr BP, the average annual precipitation was ~350 mm, which is about 70 mm lower than at present, but slightly intensified at two intervals of ca 9500–9000 cal. yr BP and ca 8500–8000 cal. yr BP. During the period of ca 6500 and 3200 cal. yr BP, the climate was generally wet with the average annual precipitation of ~440 mm, but the annual precipitation displayed high-frequency and high-amplitude fluctuations from 170 mm lower to 310 mm higher than those at present. For example, precipitation levels higher than those at present (i.e., 50–240 mm by average higher than today) occurred mainly at the intervals of ca 6450–6000, 5750–5650, 5200–5050, 4260–4025, 3930–3850, 3600–3510 and 3400–3230 cal. yr BP. The episode after ca 3200 cal. yr BP was dominated by a climate drier than that at present with the average annual precipitation of ~390 mm, but it was intercalated with a short interval of enhanced precipitation rates (e.g., the average annual precipitation was ~510 mm, being ~80 mm higher than at present) from ca 1680 to 1200 cal. yr BP (Fig. 5e).

In north-central China, the changes in the Holocene climate remain controversial. For instance, multi-proxies in the Daihai Lake sediments have convergently indicated that the climate was warm and dry during the early Holocene (e.g., before ca 8000 cal. yr BP) but became warm and humid during the middle Holocene (e.g., defined as Holocene climate optimum, ca 8000–3300 cal. yr BP) (Xiao et al., 2004; Peng et al., 2005; Xiao et al., 2006). A similar conclusion is also inferred from pollen studies in other lake sediments and loess deposits in north-central China (e.g., Zhang et al., 1981; Sun and Zhao, 1991; Cui and Kong, 1992; Sun et al., 2008). However, some other studies in north-central China suggested that the climate was either persistently humid during the early and middle Holocene (Feng et al., 2004, 2006) or humid during the early Holocene but then dry during the middle Holocene (Chen et al., 2003a; Chen et al., 2003b). Our BC carbon isotope record independently demonstrates that conditions wetter than that at present did not occur in the Daihai Lake region until ca 6500 cal. yr BP. This result is synchronous with the advent of primitive agriculture in the Daihai Lake region (e.g., Lian and Fang, 2001), which suggests the dependence of early agriculture on favorable climate conditions, particularly precipitation. Meanwhile, the amplitude (50–240 mm) for the inferred rainfall increase relative to today is comparable to a 100–200 mm increase for annual precipitation during the Holocene climate optimum with respect to that of today, as inferred from archeological and pedogenetic studies in the Daihai Lake region and other locations in northern China (e.g., Zhu et al., 1998; Fang, 1999; Lian and Fang, 2001). In addition, there was a major lithologic change occurred at 6.65 m (i.e., at ca 5100 cal. yr) with some borrows at depths of 2.10–5.70 m of DH99a, corresponding to a time interval from ca 4350 to 1650 cal. yr. The occurrence of those borrows may indicate more oxygen contained in lake water, probably reflecting an overall drop of lake level during this period. It may be roughly in accordance with the inferred rainfall decreases since ca 3300 cal. yr over Daihai Lake region in this study.

At the same time, most of the intervals with precipitation levels higher than those at present, as inferred using the $\delta^{13}C_{BC}$ values, are in agreement with the high precipitation periods that were reconstructed using pollen data from the DH99a sediment core by Wen et al. (2012) (see gray bars in Fig. 5). However, the annual precipitation reconstructed using the $\delta^{13}C_{BC}$ values at those intervals increased by approximately 110 mm on average with respect to today, and these values are two to three times higher than the amplitude for rainfall increases during the middle Holocene, as inferred from the pollen data. In addition, the inferred precipitation using the $\delta^{13}C_{BC}$ value is highly variable in comparison with the precipitation reconstructed using the pollen assemblages in the DH99a sediment core. This variability may be attributed to the high sensitivity for carbon isotopic variation in plants in response to the changing precipitation. In contrast, the composition in vegetation would change slowly with rainfall fluctuations. In this sense, BC carbon isotope records may intend to register some extreme changes in precipitation and a long-term trend, whereas the precipitation reconstructed using the pollen assemblages may represent a long-term average.

At the interval between ca 1680 and 1200 cal. yr BP, the inferred annual precipitation using the $\delta^{13}C_{BC}$ value was ~80 mm on average higher than the present-day level (Fig. 5e). This stage is in agreement with the so-called 'Sui–Tang warm period' (A.D. 600–1000), which was initially proposed by Zhu (1973) based on the historical documents, mammal fossils and plant remains from archeological relics. During this period, the mean annual temperature was 0.5–1 °C higher than that at present, and the northern boundary of farming moved 1° northward, which resulted in higher crop yields (Wu and Dang, 1998; He et al., 2010). Except for the warm climate, the inferred precipitation level being higher than that at present may also be an important contribution to the higher crop yields during the Sui–Tang Dynasty period. Some recent studies based on historical documents have shown that the precipitation during the Sui–Tang Dynasty was higher than that at present in the Guanzhong region (i.e., north-central China) (Zhu et al., 1998) and that severe persistent flood events frequently occurred in northern China during the Sui–Tang Dynasty (Hao et al., 2010). These studies provide support for our inference of precipitation changes over the Daihai Lake region during the Sui–Tang warm period. In contrast, the precipitation reconstructed using the pollen assemblages during the Sui–Tang warm period was still approximately 50 mm lower than that at present, although it showed an increase with respect to the other periods during the late Holocene. The reason for this difference deserves further study.

The inferred precipitation levels being higher than those at present were reasonable during the middle Holocene and the Sui–Tang warm periods. These inferred values are in accordance with the general features of rainfall occurrences in the monsoon climatic region. The basic physical process is that the temperature being higher than that at present would enhance the atmospheric pressure gradient between the Asian continent and the Indo-Pacific Oceans by increasing the thermal contrast between them and eventually drive an inland expansion of the monsoon rainfall belt (e.g., Xiao et al., 2006; Yang and Ding, 2008). Although warmer conditions can sometimes lead to drier conditions because evaporation increases (effective precipitation decreases) in some places, e.g., northeastern China (Seki et al., 2009), this may be not the case at our study site. As can be seen in Fig. 2, the highest evaporation occurred in May, which is respectively two and three months earlier than the occurrence of highest temperature (e.g., July) and highest precipitation (e.g., August) in Daihai Lake region. In contrast, the temperature and the precipitation tend to show a coeval change at our study site.

In summary, the $\delta^{13}C_{BC}$ values of the Daihai Lake sediments can serve as a reliable proxy for past changes in monsoon precipitation. An increase in the $\delta^{13}C_{BC}$ value implies a decrease in the monsoon precipitation over the lake region. The variation in the $\delta^{13}C_{BC}$ values of the DH99a sediment core recorded generally synchronized, but more sensitive, changes along with the proxy of pollen assemblages for the past monsoon precipitation intensity. Note that the application of the $\delta^{13}C_{BC}$ values of lake sediments as a monsoon precipitation proxy is currently limited to the areas that are not occupied by an abundance of C4 plants. Otherwise, the changes in C3/C4 abundances will also lead to large variations in the $\delta^{13}C_{BC}$ value, which affects the usefulness of the BC carbon isotope as an indicator for monsoon precipitation.

6. Conclusions

In this study, we demonstrated that carbon isotopic composition of black carbon ($\delta^{13}C_{BC}$) in the Daihai lake sediment (DH99a core) can be a useful indicator of monsoon precipitation changes. Our data show that decreased $\delta^{13}C_{BC}$ values generally corresponded to high values of the tree percentages in the pollen assemblages and the grain size records of the same sediment core, suggesting that the $\delta^{13}C_{BC}$ values could have recorded monsoon precipitation changes over the Daihai Lake region. The reconstructed changes in annual precipitation using $\delta^{13}C_{BC}$ values are overall consistent with the variation in inferred annual precipitation using pollen assemblages. Nevertheless, the $\delta^{13}C_{BC}$ values also registered some extreme variations of precipitation, which could not be recorded by pollen assemblages. We conclude that the $\delta^{13}C_{BC}$ values in the Daihai Lake sediments may serve as a sensitive and reliable proxy for monsoon precipitation.

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