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Carbon and nitrogen signatures of sedimentary organic matter from Dali Lake in Inner Mongolia: Implications for Holocene hydrological and ecological variations in the East Asian summer monsoon margin

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A B S T R A C T

Environmental changes in the East Asian summer monsoon (EASM) margin have an important impact on the global climate system. This study presents the results of high-resolution analyses of TOC/TN (C/N) ratio, $\delta^{13}C$ and $\delta^{15}N$ of sedimentary organic matter extracted from a sediment core from Dali Lake, Inner Mongolia, in order to monitor Holocene hydrological and ecological variations in the EASM margin. Concurrent increases in the values of these proxies are generally interpreted to reflect intensified surface runoff and vegetation development in the lake catchment, elevated lake levels and enhanced lake productivity; however, changes in $\delta^{13}C$ and $\delta^{15}N$ may also result from within-lake processes due to changes in lake level. These data indicate that Dali Lake experienced gradual rises in water level and primary productivity from 11,500 to 9800 cal yr BP, as documented by increases in TOC and TN concentrations, C/N ratios and $\delta^{13}C$ and $\delta^{15}N$ values. From 9800 to 7700 cal yr BP, high, stable TOC and TN concentrations and C/N ratios together with low $\delta^{13}C$ and $\delta^{15}N$ values suggest a status of high stands and high productivity for the lake and a development of terrestrial vegetation in the catchment. Between 7700 and 5900 cal yr BP, TOC and TN were fluctuating at higher concentrations and C/N maintained high ratios, while $\delta^{13}C$ increased in its value and $\delta^{15}N$ remained at low values, denoting a further rise in lake level and a notable improvement of aquatic and terrestrial ecosystems. Around 5900 cal yr BP, TOC and TN concentrations, C/N ratios and $\delta^{13}C$ values decreased abruptly, while $\delta^{15}N$ value increased rapidly, implying dramatic drops in lake level and water temperature and drastic declines of aquatic and terrestrial ecosystems. Subsequently all geochemical proxies increased in their values until 4850 cal yr BP, indicating a gradual hydrological and ecological recovery. From 4850 to 750 cal yr BP, decreasing trends of TOC and TN concentrations and C/N ratios and increasing trends of $\delta^{13}C$ and $\delta^{15}N$ values denote a general decline in the hydrological and ecological conditions. The last 750 cal yrs witnessed the pattern of hydrological and ecological changes occurring from 5900 to 4850 cal yr BP. We suggest that hydrological and ecological changes in the EASM margin during the Holocene were closely related to the combined effects of regional precipitation and temperature which were ultimately controlled by the Northern Hemisphere summer insolation, the boundary conditions and the physical environment of ocean current.

1. Introduction

The environment of the East Asian summer monsoon (EASM) margin, defined as the semi-arid zone in northern China with mean annual precipitation ranging from 400 to 200 mm, is highly vulnerable to climate change due to the high degree of rainfall

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variability in summer and the frequent occurrence of cold waves in winter (Chinese Academy of Sciences, 1984; Zhang and Lin, 1985). In the geological past, when the climate became dry, the EASM marginal zone acted as a major source of mineral dust production (An, 2000; Zhang et al., 2003) which potentially affected the global climate via regulation of the atmospheric radiation balance (Kinne and Pueschel, 2001; Sokolik et al., 2001) and oceanic plankton productivity (Ridgwell, 2002; Jickells et al., 2005). Recent climate model results suggest that by the end of this century, global warming may have severe environmental consequences in mid-latitude semi-arid regions, including the EASM margin (IPCC, 2013). However, in view of the inherent uncertainties of numerical models, it is important to examine proxy evidence from geological archives in order to characterize the environmental variability of the EASM margin during the current interglacial, the Holocene warm period.

The organic matter accumulated in lake sediments has long been considered to reflect changes in terrestrial vegetation in the lake catchment, aquatic plant productivity within the lake, and hydrological conditions in the lake basin; thus constitutes a source of information about variations in the regional environment through time (Prokopenko et al., 1999; Meyers and Teranes, 2001; Lamb et al., 2004; Lücke and Brauer, 2004; Wischnewski et al., 2011; Aichner et al., 2012). Total organic carbon (TOC) and total nitrogen (TN) concentrations, TOC/TN (C/N) ratio, and the $\delta^{13}C$ and $\delta^{15}N$ values of lacustrine organic matter have been widely used to reconstruct changes in the hydrology and ecology of various lake–catchment systems (Narantsetseg et al., 2013; Wang et al., 2013; Ghosh et al., 2014; Kigoshi et al., 2014; Wang et al., 2014; Sun et al., 2016). For example, in Qinghai Lake, northeastern Tibetan Plateau, C/N ratios of the sedimentary organic matter decreased, while $\delta^{13}C$ values increased, during the past 18,000 yr, indicating an increasing trend of lake primary productivity that was related to climatic warming and increasing humidity in the region (Shen et al., 2005). In Cuoe Lake, central Tibetan Plateau, high TOC concentrations and increased $\delta^{13}C$ values from 8500 to 5700 cal yr BP were inferred to reflect a high level and high primary productivity of the lake under a warm and humid climate (Wu et al., 2006). The organic matter extracted from the varved sediments of Sihailongwan Lake, northeastern China, exhibits increases in TOC and TN concentrations and $\delta^{15}N$ values during the late glacial, suggesting an increase in the productivity of planktonic algae within the lake related to increased inputs of dissolved nutrients under a warm climate (Parplies et al., 2008). Recent studies of sedimentary organic matter from Emerald Peak Lake, Taiwan, located at the core of the EASM region, suggest that high TOC concentrations and C/N ratios, together with low $\delta^{13}C$ values, during the last 2000 cal yr BP denote a high lake stand and dense development of terrestrial C_3 plants resulting from strengthened monsoonal precipitation (Selvaraj et al., 2012).

However, there are discrepancies in interpreting the above proxies with respect to the hydrology and ecology of different lakes, which can vary greatly in terms of sensitivity to factors such as basin structure and climate regime. For example, increased C/N ratios and $\delta^{13}C$ values at Luanhaizi Lake, northeastern Tibetan Plateau, were considered to reflect the presence of submerged macrophytes (Herzschuh et al., 2005). Decreased C/N ratios and $\delta^{13}C$ values at Cuoe Lake were interpreted to reflect the degradation of deposited organic matter due to lowered lake levels (Wu et al., 2006). Increased $\delta^{15}N$ values in Bosumtwi Lake, west Africa, were inferred to result from ammonia volatilization caused by increased lake water alkalinity (Talbot and Johannessen, 1992); while decreased $\delta^{15}N$ values in Tanganyika Lake, east Africa, were interpreted as resulting from nitrogen fixation by the aquatic phytoplankton community (McManus et al., 2015). Therefore, it is important to carefully examine the factors driving variations in organic matter proxies of different lakes in order fully to explore the potential of lakes as reliable sources of information on environmental changes.

Several lakes are located in the EASM margin (see Fig. 1) and are considered as ideal natural archives of past changes in EASM intensity (Li et al., 1992; Xiao et al., 2004, 2006, 2008, 2009; Zhang et al., 2012; Fan et al., 2016). Previous analyses of the TOC concentration and C/N ratio of a sediment core from Dali Lake in the EASM margin revealed the significant impact of monsoon-precipitation-induced soil erosion in the lake catchment on lacustrine processes (Xiao et al., 2008). Nevertheless, hydrological and ecological variations of lake–catchment systems in the EASM margin, and their response to climate change, remain unclear. In the present study, we present high-resolution (~50 yr) records of TN concentration, C/N ratio, $\delta^{13}C$ and $\delta^{15}N$ values of sedimentary organic matter in a Holocene sediment core from the EASM margin. The main aim of the study is to use the dataset to improve our understanding of the environmental variability in mid-latitude semi-arid regions during geological warm periods and the possible response of these fragile environments to future global warming.

2. Regional setting

Dali Lake (43°13'–43°23' N, 116°29'–116°45' E) is located in the northern margin of the E–W-extending Hulandaga Desert Land, 70 km west of Hexigten Banner, Inner Mongolia (Fig. 1), in an inland fault-depression basin that was formed in the Pliocene to Pleistocene (Li, 1993). The lake has an area of 238 km^2, a maximum water depth of 11 m, an elevation of 1226 m above sea level (Fig. 1), and is hydrologically closed. Hills surround the lake to the north and west, and lacustrine plains are present along the eastern shore. Two permanent rivers, the Gongger and Salin Rivers, enter the lake from the northeast; and two intermittent streams, the Holai and Liangzi Rivers, enter from the southwest (Fig. 1); however, there are no outflowing rivers. The Gongger River, the major inflow, rises in the southern terminal part of the Great Hinggan Mountains, where the elevation reaches 2029 m, and has a drainage area of 783 km^2 and a total channel length of 120 km (Li, 1993). Hydrological observations indicate that the discharge of the Gongger River is as large during spring floods in April as during summer floods in July, because of significant melt water runoff from the snow/ice packs covering the mountains (Li, 1993).

Dali Lake is located at the transition from semi-humid to semi-arid areas of the middle temperate zone. The climate of the region is controlled by the East Asian monsoon (Chinese Academy of Sciences, 1984; Zhang and Lin, 1985). In region, mean annual temperature is 3.2 °C with a July average of 20.4 °C and a January average of −16.6 °C (Fig. 2). Mean annual precipitation is 383 mm, and ~70% of the annual precipitation falls from June–August (Fig. 2). Mean annual evaporation reaches 1632 mm, which is more than 4 times the annual precipitation (Fig. 2). Dali Lake has a pH of 9.5, a salinity of 7.4 g/L and an alkalinity of 4.9 $CaCO_3$ g/L (measurements made in June 2010). The $\delta^{13}C$ of the lake's dissolved inorganic carbon ($\delta^{13}C_{DIC}$) averages −0.3‰ (PDB), the $\delta^{13}C_{DIC}$ of Gongger River is −7.8‰ (PDB) and the average $\delta^{13}C_{DIC}$ of the four inflowing rivers is −6.1‰ (PDB) (measurements made in June 2010).

The modern natural vegetation of the Dali Lake basin is categorized as middle temperate steppe and is dominated by grasses (Compilatory Commission of Vegetation of China, 1980; Li, 1993). In the Hulandaga Desert Land, low-growing xerophilous plants including *Polygonum divaricatum*, *Agriophyllum squarrosum* and *Artemisia desterorum* are present, together with the shrubs *Salix*

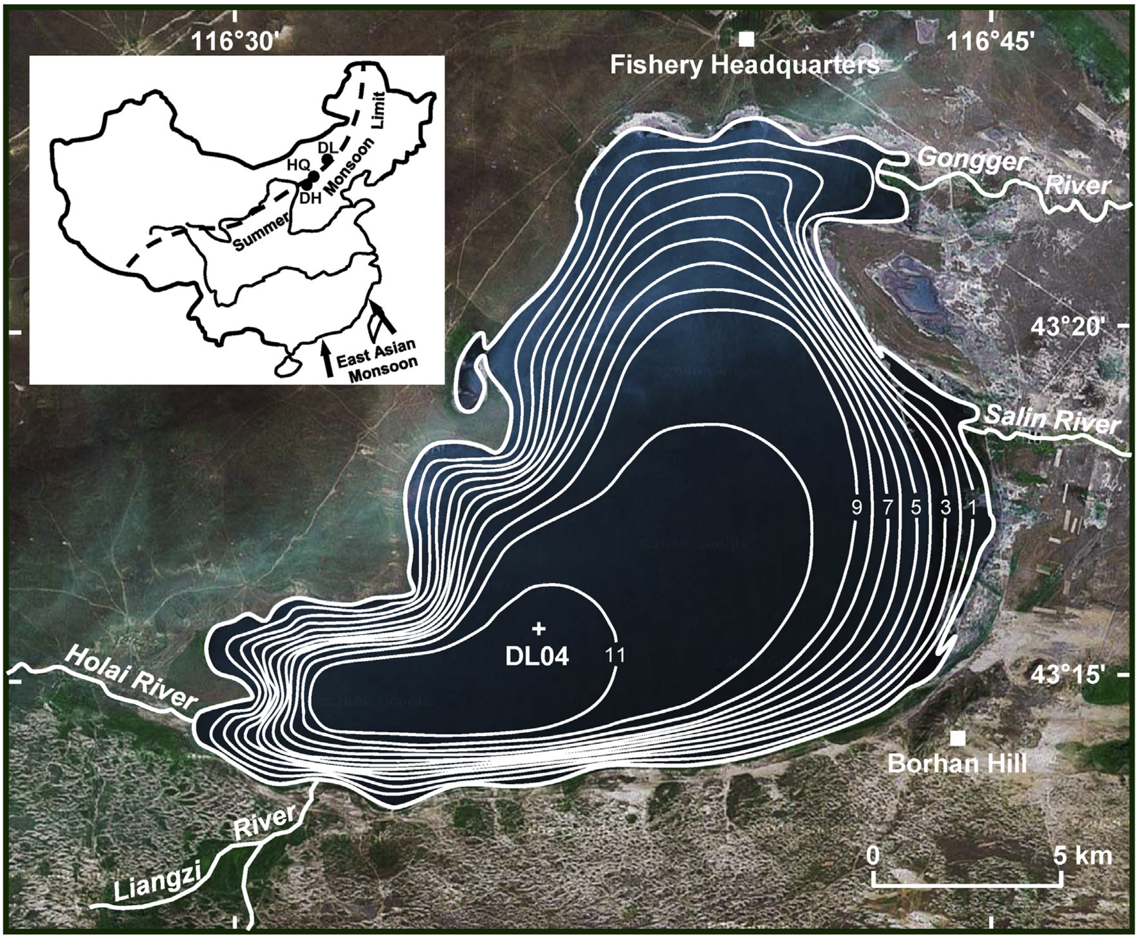

Fig. 1. Map of Dali Lake (from http://maps.google.com) showing the location of the DL04 sediment core (cross). The bathymetric survey of the lake was conducted in June 2002 using a FE-606 Furuno Echo Sounder (contours in m). The inset map shows the location of Dali Lake (DL), Huangqi Lake (HQ) and Daihai Lake (DH) in northern China (solid circles) and the modern northern limit of the East Asian summer monsoon defined by the 400-mm isohyet of mean annual precipitation.

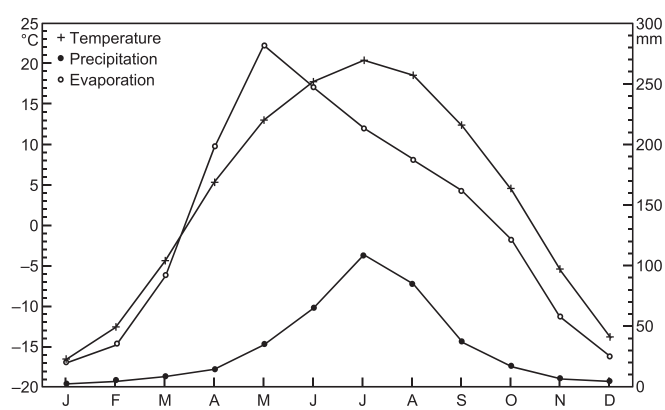

Fig. 2. Mean monthly temperature, mean monthly precipitation and mean monthly evaporation in the Dali Lake region. Data are the average of observations from 1981 to 2010 at Hexigten Banner Meteorological Station, 70 km east of Dali Lake (unpublished data courtesy of Inner Mongolia Meteorological Bureau).

gordeivii, *Ulmus pumila* and *Caragana sinica*. Herbs including *Stipa grandis*, *Leymus chinensis* and *Cleistogenes squarrosa* are developed in the northern and western hilly lands and on the eastern lacustrine plains. Forests consisting of *Larix gmelinii*, *Pinus tabuliformis*, *Betula platyphylla*, *Populus davidiana* and *Quercus mongolica* are distributed on the western slopes of the Great Hinggan Mountains, where the Gongger River rises, together with shrubs and herbs growing beneath the canopy.

Dali Lake has a secchi depth of ca. 1 m in the nearshore zone and 1.5 m in the offshore zone. Aquatic plants living in the lake consist mainly of planktonic algae with scarce vascular plants (Li, 1993; Wang and Dou, 1998). Planktonic algae include *Microcystis salina*, *Lyngbya contorta* and *Anabaenopsis muelleri* of the Cyanophyta, and *Surirella peisohispant*, *Amphiprora paludo* and *Cyclotella meneghiniana* of the Bacillariophyta. Potamogetonaceae, including *Potamogeton crispus* and *Potamogeton pectinatus*, are the dominant aquatic vascular plants; they are generally confined to the areas of the river mouth where the water depth is less than 1.5 m.

3. Material and methods

3.1. Sediment sampling and lithology

Sediment coring was conducted in a water depth of 10.8 m in the depocenter of Dali Lake in February 2004, when the lake surface was frozen (Fig. 1), using a TOHO drilling system (Model D1-B) (Toho Chikakoki Co., Ltd, Japan). Incremental sediment cores were extracted to total a depth of 11.83 m beneath the lake floor and were designated DL04 (43°15.68' N, 116°36.26' E) (Fig. 1). The cores were collected in polyethylene tubes using a piston corer of the drilling system, and sediment recovery approached 100%.

The upper 8.5 m of the DL04 core was used for the present study

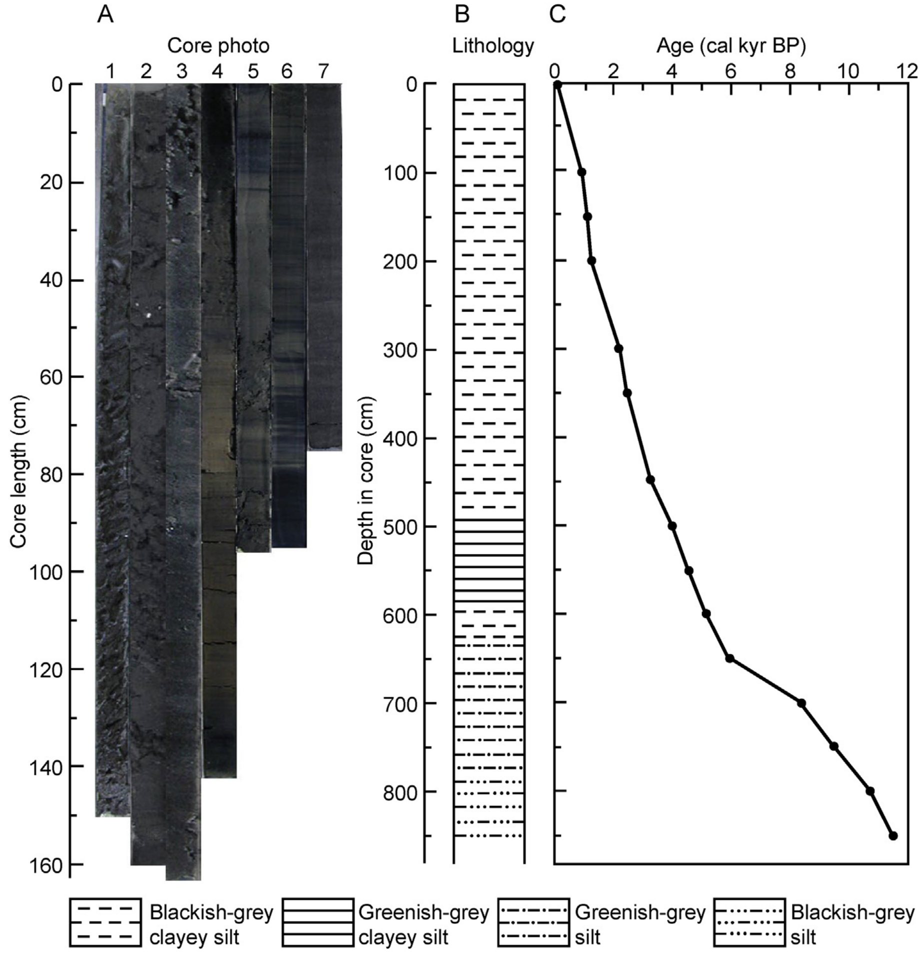

Fig. 3. (A) Photograph of the upper 8.81 m of the DL04 core. The 1st section is the interval 0–150 cm, and the 7th section is the interval 806–881 cm. (B) Lithology of the upper 8.5 m of the DL04 core. (C) Age–depth model for the upper 8.5 m of the DL04 core. Solid circles represent the mean values of 2σ ranges of calibrated ages of reservoir-corrected radiocarbon dates. The dating method and the original radiocarbon dates of the DL04 core are cited from Xiao et al. (2008).

(Fig. 3). The sediments consist of greenish-grey to blackish-grey, homogeneous silt and clayey silt, and can be divided into five main sedimentary units (Fig. 3), as follows: 850–789 cm blackish-grey massive silt; 789–634 cm greenish-grey massive silt with occasional and frequent blackish-grey bands at depths of 789–718 and 718–634 cm, respectively; 634–598 cm blackish-grey banded clayey silt; 598–494 cm greenish-grey laminated clayey silt; 494–0 cm blackish-grey massive clayey silt. There was no indication of any sedimentary hiatuses.

3.2. Chronology

The dating method and the original radiocarbon dates of the DL04 core are cited from Xiao et al. (2008). Fifteen bulk samples from the upper 8.5 m of the DL04 core were radiocarbon dated using an Accelerator Mass Spectrometry (AMS) system (Compact-AMS, NEC Pelletron) at the Paleo Labo Co., Ltd (Japan). Organic carbon was extracted from each sample and dated following the method described by Nakamura et al. (2000). The ^{14}C dates of all the samples from the upper 8.5 m of the DL04 core were determined using a half-life of 5568 yr.

As shown in Table 2, the uppermost 1 cm of the DL04 core yields a ^{14}C age of 472 yr. This anomalously old age can be considered to result from a 'hard-water' and other carbon reservoir effects. In order to produce an age–depth model, we first subtracted the reservoir age of 472 yr from all of the original ^{14}C ages, assuming constancy throughout the core. The conventional ages were then converted to calibrated ages using the OxCal4.2 radiocarbon age calibration program (Bronk Ramsey and Lee, 2013) with IntCal13 calibration data (Reimer et al., 2013). The age–depth model indicates that the upper 8.5 m of the DL04 core span the last 11,500 yr (Fig. 3; Table 2). The ages of sampled horizons were estimated by linear interpolation between radiocarbon-dated horizons using the mean values of 2σ ranges of the calibrated ages. The sedimentation rates of ca. 20–280 cm/kyr and sampling intervals of 1–6 cm provide potential temporal resolutions of ca. 40–60 yr for the sedimentary organic carbon and nitrogen data from the upper 8.5 m of the core.

It is worth mentioning that the age–depth model in this study is different from that in Xiao et al. (2008). In Xiao et al. (2008), original ^{14}C ages were directly converted to the calibrated ages without consideration of the reservoir effect. The calibrated ages were then used to produce an age–depth model with a third order polynomial fit, and the surface-intercept age of 611 yr yielded by the age–depth model was considered to be the reservoir age (Table 2).

The reservoir effects of the DL04 core may change due to different lake levels and sedimentation rates or various proportions of autochthonous and allochthonous organic carbon (Kao et al., 2008; Hou et al., 2012) during the Holocene, but accurate ^{14}C

Table 1
Statistics of organic geochemical characteristics of representative samples in the Dali Lake catchment and within the lake. C/N atomic ratios and $\delta^{13}C$ values of terrestrial C_3 and C_4 plants are cited from Li and Chen (1998) and Wang et al. (2003), respectively.

Materials	No. of samples		C/N ratio	$\delta^{13}C$ (‰)	$\delta^{15}N$ (‰)
C_3 plants	3 for C/N	Min.	17.60[a]	−28.66[b]	
	8 for $\delta^{13}C$	Max.	25.30[a]	−24.80[b]	
		Mean	21.73[a]	−27.10[b]	
C_4 plants	1	Min.	33.01[a]	−14.03[b]	
		Max.	33.01[a]	−14.03[b]	
		Mean	33.01[a]	−14.03[b]	
Nearshore surface sediments	15	Min.	12.97	−26.44	7.01
		Max.	13.84	−23.82	7.61
		Mean	13.41	−24.48	7.31
Offshore surface sediments	12	Min.	9.88	−26.25	8.04
		Max.	11.34	−25.68	10.36
		Mean	10.59	−25.97	9.20
DL04 core sediments	225 for C/N	Min.	5.89	−28.01	−1.74
	221 for $\delta^{13}C$, $\delta^{15}N$	Max.	23.07	−23.32	9.19
		Mean	14.91	−25.40	2.95

[a] Data of C/N ratios are cited from Li and Chen (1998).
[b] Data of $\delta^{13}C$ values are cited from Wang et al. (2003).

Table 2
AMS radiocarbon dates of samples from the upper 8.5 m of the DL04 core. The dating method and the original radiocarbon dates of the DL04 core are cited from Xiao et al. (2008).

Laboratory number[a]	Depth interval (cm)	Dating material	$\delta^{13}C$ (‰)	^{14}C age (^{14}C yr BP)	Corrected ^{14}C age[b] (^{14}C yr BP)	Calibrated ^{14}C age (2σ) (cal yr BP)	Calibrated ^{14}C age[c] (cal yr BP)
PLD−4564	1−0	Organic matter	−28.19	472 ± 23	0 ± 33	74−32	611
PLD−4565	100−99	Organic matter	−25.47	1463 ± 24	991 ± 33	965−892	1404
PLD−6250	150−149	Organic matter	−28.05	1629 ± 22	1157 ± 32	1176−982	1763
PLD−4567	200−199	Organic matter	−24.65	1948 ± 25	1476 ± 34	1415−1300	2117
PLD−12450	299−298	Organic matter	−23.24	2652 ± 22	2180 ± 32	2313−2114	2862
PLD−12453	350−349	Organic matter	−24.60	2879 ± 23	2407 ± 33	2503−2347	3299
PLD−12456	449−448	Organic matter	−25.36	3561 ± 24	3089 ± 33	3376−3217	4325
PLD−12459	500−499	Organic matter	−27.80	4113 ± 25	3641 ± 34	4013−3865	4976
PLD−6706	550−549	Organic matter	−23.66	4562 ± 21	4090 ± 31	4654−4516	5715
PLD−4572	600−599	Organic matter	−27.24	4947 ± 28	4475 ± 36	5292−5031	6569
PLD−6255	650−649	Organic matter	−25.58	5709 ± 26	5237 ± 35	6029−5917	7553
PLD−12466	699−698	Organic matter	−30.69	8014 ± 29	7542 ± 37	8415−8309	8658
PLD−12469	750−749	Organic matter	−27.98	8881 ± 30	8409 ± 38	9522−9397	9971
PLD−12470	799−798	Organic matter	−31.56	9969 ± 32	9497 ± 39	10,869−10654	11,404
PLD−12472	849−848	Organic matter	−30.84	10,464 ± 37	9992 ± 44	11,640−11267	13,054

[a] PLD: Paleo Labo Dating, laboratory code of Paleo Labo Co., Ltd., Japan.
[b] The calibrated ages of reservoir-corrected radiocarbon dates. The reservoir correction factor is 472 yr, ^{14}C age of the uppermost 1 cm of the DL04 core.
[c] The ^{14}C age data from the age−depth model with a third order polynomial fit from Xiao et al. (2008).

ages may be difficult to obtain due to the scarcity of terrestrial macrofossils or plant debris in the core sediments.

3.3. *Analyses of total nitrogen concentration*

The upper 8.5 m of the core was sampled at 10-cm intervals in Xiao et al. (2008) ($n = 85$) but at 1–5-cm intervals in this study ($n = 225$) for measurements of TN concentration. Data of TN concentration in this study are totally different from Xiao et al. (2008), although the variation trends of TN concentration in this study and Xiao et al. (2008) are generally same. TN concentrations were determined with a Yanaco CHN Corder MT-5 analyzer. Samples were ground to a powder finer than 61 μm and dried at 40 °C for 24 h. Each sample of 30 mg of dried sediment was fully combusted at 450 °C for 5 min to completely convert nitrogen to NO_x, and the resulting NO_x was then reduced to N_2 by copper. A reference Antipyrine sample (SMA-SP-9), was separated into three weighed portions and routinely analyzed after every thirty sample measurements. All of the TN values were normalized to SMA-SP-9, the certified TN value of which is 14.88%. The relative analytical error of the TN concentration of a sample is less than 0.3%.

Data of TOC/TN (C/N) ratio are calculated based on the TN data in this study and TOC data from the same samples in Xiao et al. (2008). C/N ratio was expressed as mass ratio in Xiao et al. (2008) but atomic ratio calculated using a ratio of 1.17 between the atomic and mass ratios (Meyers, 1994; Lamb et al., 2006) in this study.

3.4. Analyses of organic carbon and nitrogen isotopic composition

The upper 8.5 m of the core was sampled at 1–6-cm intervals for measurements of organic carbon and nitrogen isotopic composition ($n = 221$). Carbon and nitrogen isotopic composition were determined with a Delta V Plus mass spectrometer equipped with a Thermo Flash EA 1112 element analyzer and a ConFlo III system. Samples were ground to a powder finer than 61 μm and pretreated with 1 M HCl for 24 h to remove carbonates, and then rinsed and dried at 40 °C for 24 h. Each sample of 0.4–10 mg of dried sediment was fully combusted in the Thermo Flash EA 1112 element analyzer at 960 °C for 1–2 min to generate CO_2 for measurements of $^{13}C/^{12}C$ ratios. For measurements of $^{15}N/^{14}N$ ratios, each sample of 10–20 mg of dried sediment was fully combusted at 960 °C for 1–2 min to generate NO_x, and the resulting NO_x was then reduced to N_2 by copper at 680 °C. A blank sample, a reference sample (Glycine) and a duplicate sample were routinely analyzed after every five sample measurements. $\delta^{13}C$ and $\delta^{15}N$ are expressed in parts per thousand (‰), relative to VPDB and atmospheric nitrogen, respectively. All of the $\delta^{13}C$ and $\delta^{15}N$ values were normalized according to the blank sample and Glycine; the certified $\delta^{13}C$ and $\delta^{15}N$ values of Glycine are −33.3‰ and 10.0‰, respectively. The precision is better than 0.1‰ for $\delta^{13}C$ and 0.2‰ for $\delta^{15}N$.

4. Results

TOC and TN concentrations, C/N ratio, and $\delta^{13}C$ and $\delta^{15}N$ values are plotted against calibrated age in Fig. 4. TOC and TN concentrations vary from 1.0% to 7.3% and 0.13% to 0.56%, respectively, yielding a range of 5.9–23.1 for C/N ratio. $\delta^{13}C$ values range from −28.01‰ to −23.32‰, and $\delta^{15}N$ values from −1.74‰ to 9.19‰. The time series of TOC and TN concentrations, C/N ratio, and $\delta^{13}C$ and $\delta^{15}N$ values spanning the last 11,500 cal yr can be divided into three stages based on the general pattern of temporal changes described below.

4.1. Stage 3 (853–763 cm, 11,500–9800 cal yr BP)

At the beginning of this stage TOC and TN concentrations, C/N ratio, and $\delta^{13}C$ and $\delta^{15}N$ values are at their lowest levels throughout the studied stages; however, they all exhibit an increasing trend towards the end of the stage. TOC and TN concentrations increase from 1.0% to 5.3% and from 0.13% to 0.48%, respectively. C/N ratios rise from 5.9 to 12.8 and exhibit higher values from 10,400 to 10,000 cal yr BP, with a peak value of 18.6. $\delta^{13}C$ values increase from −27.49‰ to −25.13‰, with fluctuations superimposed, but remain relatively lower from 10,400 to 10,000 cal yr BP with a minimum −27.05‰. $\delta^{15}N$ values gradually increase from −0.72‰ to 1.83‰.

4.2. Stage 2 (763–645 cm, 9800–5900 cal yr BP)

This stage can be subdivided into two sub-stages: sub-stage 2b (763–685 cm, 9800–7700 cal yr BP) and 2a (685–645 cm, 7700–5900 cal yr BP). In sub-stage 2b, TOC and TN concentrations and C/N ratios exceed their high values at the end of the preceding stage 3, and remain almost unchanged around average values of 5.6%, 0.45% and 14.6, respectively. $\delta^{13}C$ values decrease from −25.13‰ to −26.43‰ in the early part of this sub-stage, maintain lower values in the middle part, except for a single point with a $\delta^{13}C$ value of −25.32‰, and increase to −25.41‰ towards the end. $\delta^{15}N$ values decrease from 1.83‰ to 0.43‰ at the beginning of this sub-stage, and then remain relatively stable around an average value of 0.43‰ towards the end.

Sub-stage 2a is characterized by high but variable concentrations of TOC and TN and by a general trend of increasing $\delta^{13}C$ values. TOC and TN concentrations are at their highest levels over the last 11,500 yr, and fluctuate between 5.4% and 6.9% and between 0.41% and 0.56%, respectively. C/N ratios resemble the pattern of changes in sub-stage 2b, and exhibit an average of 14.7. $\delta^{13}C$ values increase from −26.11‰ to −24.34‰, while $\delta^{15}N$ values remain almost unchanged with an average of 0.60‰, slightly higher than in sub-stage 2b.

4.3. Stage 1 (645–0 cm, 5900–0 cal yr BP)

This stage can be subdivided into three sub-stages: sub-stage 1c (645–573 cm, 5900–4850 cal yr BP), 1b (573–80 cm, 4850–750 cal yr BP) and 1a (80–0 cm, 750–0 cal yr BP). During the first 250 yr of sub-stage 1c TOC and TN concentrations and C/N ratios decrease to 2.9%, 0.29% and 11.6, respectively, and then increase gradually to 6.3%, 0.48% and 16.4, respectively, by the end. $\delta^{13}C$ values decrease by ~3‰ (from −24.34‰ to −27.50‰) during the first half of sub-stage 1c and then increase by ~2‰ (from −27.50‰ to −25.42‰) during the second half. $\delta^{15}N$ values exhibit a trend of increasing values from −0.68‰ to 5.70‰, with a range of over 6‰.

Sub-stage 1b is characterized by high-frequency, high-amplitude fluctuations of all of organic carbon and nitrogen proxies. TOC and TN concentrations and C/N ratios exhibit trends of decreasing values from 6.3% to 4.6%, 0.43% to 0.33% and 16.4 to 15.0, respectively. $\delta^{13}C$ values increase gradually from −25.84‰ to −23.45‰, and $\delta^{15}N$ values increase gradually from 5.16‰ to 6.88‰.

During the first 350 yr of sub-stage 1a, TOC and TN concentrations and $\delta^{13}C$ values decrease to 2.7%, 0.22% and −27.56‰, respectively, and then increase gradually to their present-day values of 5.5%, 0.50% and −26.70‰, respectively. C/N ratios decrease to 13.9 during the first 350 yr, but reach a peak value of 22.3 at 350 cal yr BP, and then decrease to the present-day value of 12.9. $\delta^{15}N$ values continue the increasing trend in the preceding sub-stage 1b, reaching a present-day value of 8.33‰.

5. Discussion

5.1. Paleoenvironmental implications of the organic carbon and nitrogen proxies

5.1.1. C/N ratios

Organic matter deposited in lakes has two principal sources: aquatic phytoplankton living in the lake (autochthonous) and terrestrial plants growing in the catchment (allochthonous) (Meyers, 1990). Aquatic phytoplankton contain relatively abundant proteins that are rich in organic nitrogen, and therefore autochthonous organic matter is characterized by low C/N ratios, between 4 and 10 (Meyers, 1990). Terrestrial vascular plants are dominated by lignin and cellulose that are poor in nitrogen, and thus allochthonous organic matter has high C/N ratios of 20 and greater (Meyers, 1990). It has been suggested that the degradation of unstable nitrogen compounds in aquatic organic matter (Russell et al., 2009) and the loss of carbon-rich sugars and lipids in terrestrial organic matter (Lamb et al., 2004) may occur during sedimentation and early diagenesis, thus leading to alteration of the C/N ratios of the original organic matter. However, a number of studies have

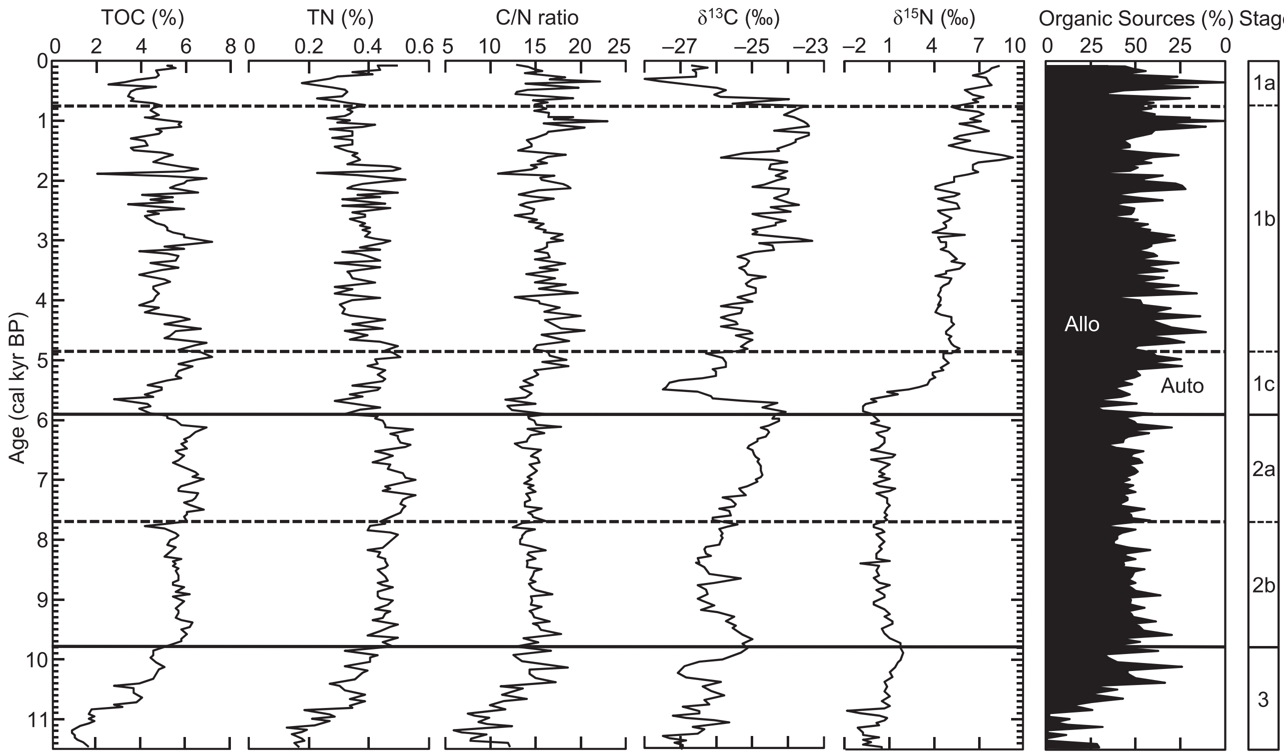

Fig. 4. Time series of total organic carbon (TOC) and total nitrogen (TN) concentrations, atomic TOC/TN (C/N) ratios and organic carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) isotopic composition for the DL04 core during the last 11,500 cal yr. The relative proportions of allochthonous (Allo) and autochthonous (Auto) organic matter in the Dali Lake sediments are estimated on the assumption of an average C/N value of 22 for allochthonous organic matter (Table 1) and 8 for autochthonous organic matter in this study, based on the two-end members model (Ishiwatari et al., 2005, 2009). Horizontal solid and dashed lines indicate the stages and sub-stages of changes in TOC and TN concentrations, C/N ratio, and $\delta^{13}C$ and $\delta^{15}N$ values. TOC data are cited from Xiao et al. (2008). The chronology is derived from the carbon reservoir-corrected age–depth model. The dating method and the original radiocarbon dates of the DL04 core are cited from Xiao et al. (2008).

indicated that the pattern of changes in C/N ratio could be used for distinguishing the relative contributions of autochthonous and allochthonous organic matter in lake sediments (Meyers, 1990, 1994; Talbot and Johannessen, 1992; Lamb et al., 2006).

As shown in Fig. 4 and Table 1, C/N ratios of the organic matter from the DL04 core generally vary within the range from 10 to 20, suggesting a mixture of aquatic and terrestrial components in the Dali Lake sediments. We estimated the relative proportions of allochthonous and autochthonous organic matter in the lake sediments, based on the two-end members model (Ishiwatari et al., 2005, 2009), as follows:

$$TOC/TN_{mea} = TOC/TN_{allo} \times P + TOC/TN_{auto} \times (1-P)$$

where TOC/TN_{mea} represents the measured atomic C/N values of organic matter from Dali Lake sediments; P and (1 − P) represent the proportions of allochthonous and autochthonous organic matter in the lake sediments, respectively.

The relative proportions of allochthonous (Allo) and autochthonous (Auto) organic matter in the Dali Lake sediments (Fig. 4) were estimated on the assumption of an average C/N value of 22 for allochthonous organic matter (TOC/TN_{allo}) (Table 1) and 8 for autochthonous organic matter (TOC/TN_{auto}) in this study. It is tacit that different values of end members of TOC/TN_{allo} and TOC/TN_{auto} adopted in the model would result in different proportions of allochthonous and autochthonous organic matter in the sediments, but the variation trends of these two members through the Holocene were generally parallel.

In addition, soils in the lake catchment commonly have low C/N ratios (with an average of 10.83) (Li and Chen, 1998) falling in the range of aquatic phytoplankton to terrestrial plants, but they have TOC concentrations (less than 1.8%) much lower than those of terrestrial plants (more than 43.2%) (Li and Chen, 1998) and lake sediments (Fig. 4), therefore soil organic matter should have little influence on the geochemical characteristics of organic matter from the Dali Lake sediments during the Holocene. However, cautious attention should be paid to the submerged macrophytes which usually grow in shallow waters, because submerged macrophytes commonly have C/N ratios similar to those of terrestrial plants (Herzschuh et al., 2005).

5.1.2. $\delta^{13}C$

The stable carbon isotopic composition ($\delta^{13}C$) of sedimentary organic matter in Dali Lake potentially indicates changes in the relative proportion of terrestrial and aquatic components, as well as in the primary isotopic signature of one and/or both components because the lake sediments contain a mixture of organic matter from terrestrial and aquatic sources. In the Dali Lake region, the modern terrestrial vegetation is dominated by C_3 plants with $\delta^{13}C$ values ranging from −28.66‰ to −24.80‰ (Table 1). There are no C_4 plants other than *Cleistogenes squarrosa*, sparsely distributed in the lacustrine plains, which has a $\delta^{13}C$ value of −14.03‰ (Table 1). Therefore, the terrestrial component of the sedimentary organic matter in the lake would have primary $\delta^{13}C$ values close to those of C_3 plants, with a mean value of −27.10‰ (Table 1). Moreover, increases in terrestrial inputs into the lake under conditions of intensified surface runoff from the catchment would increase the relative proportion of terrestrial organic matter in the lake sediments, leading to a tendency of changes in $\delta^{13}C$ values of the sedimentary organic matter to trend towards to that of terrestrial

C_3 plants.

The relative proportion of aquatic organic matter in lakes and its primary isotopic signature are closely related to the primary productivity of aquatic phytoplankton. Theoretically, when phytoplankton productivity is enhanced, the relative proportion of aquatic organic matter increases due to increases in the sedimentary flux; while the primary $\delta^{13}C$ values would gradually increase due to progressive enrichment in the ^{13}C of the dissolved inorganic carbon (DIC) pool (Talbot and Johannessen, 1992; Talbot and Lærdal, 2000). With respect to carbon isotopic composition, however, a number of factors may affect the degree of isotopic fractionation during the photosynthesis of aquatic plants, thereby giving rise to changes in the primary $\delta^{13}C$ values of aquatic organic matter (Hinga et al., 1994). Located in a semi-arid area, Dali Lake is a hydrologically-closed, alkaline lake with a pH value of 9.5. The lake experienced significant fluctuations in the water level during the historical past in response to changes in regional rainfall and in the discharge of the inflowing rivers (Xiao et al., 2008). The carbon isotopic fractionation of aquatic phytoplankton in Dali Lake could therefore be controlled principally by two factors that are related to the regional hydrology and the lake-level status: 1) the isotopic composition of the DIC pool; and 2) the isotopic effect caused by significant decreases in the concentration of dissolved carbon dioxide ($[CO_2]_{aq}$). The average $\delta^{13}C_{DIC}$ value of the inflowing waters is −6.1‰, which is much lower than that of the lake water −0.3‰. Therefore the lake DIC would be enriched in ^{12}C, resulting in decreases in $\delta^{13}C$ values of aquatic plants when the riverine DIC inputs increase under conditions of intensified surface runoff in the catchment. When large falls in the lake level occur due to significant decreases in regional precipitation, the $[CO_2]_{aq}$ concentration in the lake water would consequently decrease, which would force aquatic plants to shift from a CO_2− to a HCO_3^--based metabolism (Calder and Parker, 1973; Pardue et al., 1976; Smith and Walker, 1980; Lucas, 1983). Assimilation of HCO_3^- by aquatic phytoplankton would produce organic matter significantly enriched in ^{13}C because HCO_3^- has $\delta^{13}C$ values 7–11‰ higher than $[CO_2]_{aq}$ (Mook et al., 1974). In the case of an alkaline lake such as Dali Lake (pH = 9.5), such a shift would be of particular significance for the growth of aquatic plants. Nevertheless, it is noteworthy that relatively rapid mixing of lake waters during the period of low lake level would promote the injection of dissolved oxygen into the bottom water. This process could facilitate the degradation of deposited organic matter, leading to releases of isotopically-lighter carbon into the lake water and thus to decreases in the $\delta^{13}C$ values of aquatic plants and the resulting organic matter (Leng and Marshall, 2004). In addition, submerged macrophytes in the alkaline lakes commonly have $\delta^{13}C$ values close to those of terrestrial C_4 plants because of bicarbonate metabolism (Aichner et al., 2010, 2012).

5.1.3. δ^5N

The stable nitrogen isotopic composition ($\delta^{15}N$) of sedimentary organic matter in lakes indicates changes in the primary isotopic signature of the aquatic organic matter because organic nitrogen in lake sediments is mainly derived from organic matter of aquatic origin (Meyers, 1990; Talbot and Johannessen, 1992). In theory, aquatic phytoplankton preferentially assimilates ^{14}N from the dissolved inorganic nitrogen (DIN) pool of the lake during the photosynthesis, thus resulting in a progressive enrichment in ^{15}N of the residual DIN pool. Consequently primary $\delta^{15}N$ values of aquatic organic matter tend to increase gradually as the phytoplankton productivity increases (Wada and Hattori, 1978; François et al., 1996). The organic matter in the Dali Lake sediments exhibits a large range of $\delta^{15}N$ values (Fig. 4), indicating significant shifts in the isotopic composition of the DIN pool and/or in the nitrogen metabolism of the dominant phytoplankton in the lake. Nitrate (NO_3^-) and ammonium (NH_4^+) comprise the principal nitrogen source of the lake DIN pool, and are mainly derived from the recycling of organically-fixed N (Talbot and Johannessen, 1992; Talbot and Lærdal, 2000). Nitrification occurred in aerobic environments and denitrification occurred under anaerobic conditions, both of which were closely related to the fluctuations of lake levels, and would result in a ^{15}N-enriched residual DIN pool and thus increase the $\delta^{15}N$ values of aquatic phytoplankton and organic matter. In addition, ammonia volatilization probably occurs in alkaline Dali Lake and the increased pH values would have an important influence on the isotopic composition of the DIN pool because ammonia volatilization from the lake bottom water strongly discriminates in favor of ^{14}N with a fractionation of as great as ca. 34‰ (Collister and Hayes, 1991; Talbot and Johannessen, 1992). Therefore, when the pH value of lake waters increases under conditions of a dry climate and strong evaporation, ammonia volatilization would occur and result in a dramatic enrichment in ^{15}N of the residual DIN pool and the aquatic phytoplankton. In contrast, the input of soil nitrogen into the lake from the catchment would lead to a ^{14}N-enriched DIN pool because soil nitrogen comes initially from atmospheric nitrogen through the fixation of nitrogen-fixing bacteria, atmospherically deposited N and terrestrial plant residues, and has a $\delta^{15}N$ value much lower than that of the lake DIN pool (Peters et al., 1978; Peterson and Howarth, 1987; Meyers, 1997, 2003; Talbot, 2001). Increases in the input of soil nitrogen to the lake would thus give rise to decreases in the $\delta^{15}N$ value of the DIN pool and the aquatic phytoplankton under conditions of enhanced soil erosion in the lake catchment. In the Dali Lake, surface sediments from offshore zone have much lower C/N ratios and higher $\delta^{15}N$ values than those from nearshore zone (Table 1), which may indicate that TN concentrations in the lake sediments are dominated by the primary productivity of aquatic phytoplankton in the lake waters and the lake waters would have potentially higher $\delta^{15}N$ values of DIN pool without ^{14}N-enriched sources input to the offshore zone of the lake.

It is noteworthy that the flourishing of aquatic phytoplankton in Dali Lake, located in a semi-arid area, under favorable conditions would result in the possible limitation of nitrogen in the lake DIN pool. In this case, other populations of phytoplankton taxa would be limited, while blue-green algae tend to dominate the phytoplankton owing to their capacity of fixing atmospheric nitrogen (Hecky and Kling, 1981, 1987). Shifts in the nitrogen metabolism of the dominant phytoplankton in the lake would cause a significant decrease in the $\delta^{15}N$ value of the aquatic organic matter due to little or no isotopic fractionation during the fixation of atmospheric nitrogen (Arthur et al., 1983).

5.2. Holocene hydrological and ecological variations in the Dali Lake basin

The TOC and TN concentrations, C/N ratio, and $\delta^{13}C$ and $\delta^{15}N$ values of the DL04 core exhibit distinct and regular temporal patterns of changes during the last 11,500 yr that can be readily correlated with each other (Fig. 4). Based on the above interpretations, past changes in these organic carbon and nitrogen proxies can potentially be used to reconstruct the history of variations in the hydrology (surface runoff and the lake level) and ecology (terrestrial vegetation and aquatic phytoplankton productivity) of the lake basin during the Holocene (Fig. 4; Table 4).

From 11,500 to 9800 cal yr BP, C/N ratios generally increased from 6 to 15, accompanied by increases in TOC concentration, suggesting a trend of gradually increasing input of organic matter from terrestrial plants to the lake related to intensified fluvial erosion in the lake catchment (Håkanson and Jansson, 1983; Cohen,

Table 3
Coefficients of determination for the linear correlations between the organic carbon and nitrogen proxies for each stage/sub-stage of the DL04 core spanning the last 11,500 cal yr. See Fig. 5 for plots of the organic carbon and nitrogen proxies.

Stage	No. of samples	TN vs. TOC	$\delta^{13}C$ vs. TOC	$\delta^{15}N$ vs. TN
1a	16	0.67, positive	0.15, positive	0.09, positive
1b	83	0.62, positive	0.03, negative	0.06, negative
1c	19	0.65, positive	0.04, negative	0.45, positive
2a	37	0.50, positive	0.07, negative	0.05, positive
2b	40	0.12, positive	0	0.03, positive
3	30	0.78, positive	0.15, positive	0.64, positive

progressive enrichment in both ^{13}C and ^{15}N of the DIC and DIN pools (Talbot and Johannessen, 1992; Talbot and Lærdal, 2000). During this interval, there are significant positive correlations between $\delta^{13}C$ values and TOC concentrations, as well as between $\delta^{15}N$ values and TN concentrations (Figs. 4 and 5; Table 3), providing further support for the inference of increased phytoplankton productivity. In addition, there should be little influence of submerged macrophytes on the geochemical characteristics of organic matter from the DL04 core sediments during this period, because C/N ratios and $\delta^{13}C$ values of the sediments were much lower than the

Table 4
An overview of variations in the hydrology and ecology of the Dali Lake basin during the Holocene.

Stage	Age interval (cal yr BP)	Organic matter input	Hydrology	Ecology
1a	750–0	Both terrestrial and aquatic inputs decreased during the first half of the sub-stage and increased during the second half.	Surface runoff in the lake catchment weakened in the first half of the sub-stage and then gradually intensified in the second half. The lake contracted in the first half and then gradually expanded in second.	Both terrestrial vegetation and aquatic phytoplankton declined in the first half of the sub-stage and then gradually recovered.
1b	4850 –750	Both terrestrial and aquatic inputs decreased gradually, and the terrestrial input generally predominated.	Surface runoff in the lake catchment gradually decreased and the lake gradually shrank.	Both terrestrial vegetation and aquatic phytoplankton declined gradually.
1c	5900 –4850	Both terrestrial and aquatic inputs drastically decreased at the beginning of the sub-stage and then gradually increased through the sub-stage.	Surface runoff in the lake catchment dramatically weakened at the beginning of the sub-stage and then gradually intensified. The lake largely contracted at the beginning of the sub-stage and then gradually expanded.	The status of both terrestrial vegetation and aquatic phytoplankton abruptly deteriorated at the beginning of the sub-stage and then recovered.
2a	7700 –5900	Both terrestrial and aquatic inputs increased further, and the terrestrial input began to increase more rapidly.	Surface runoff in the lake catchment intensified further. The lake reached its highest stand for the entire Holocene.	The status of terrestrial vegetation and aquatic phytoplankton increased further.
2b	9800 –7700	Both terrestrial and aquatic inputs maintained high levels, and the terrestrial input began a slightly decreasing trend.	Surface runoff in the lake catchment remained at a high level. The lake reached a high stand, which facilitated the deposition and burial of organic matter.	Both terrestrial vegetation and aquatic phytoplankton attained a highly-developed state.
3	11,500 –9800	Both terrestrial and aquatic inputs continued to increase, and the terrestrial inputs gradually became dominant.	Surface runoff in the lake catchment began to intensify and the lake level began to rise.	Both terrestrial vegetation and aquatic phytoplankton gradually developed.

2003). At the same time, TN concentrations increased continuously, indicating increases in the phytoplankton productivity, despite that the proportion of aquatic organic matter decreased due to increased terrestrial organic matter. $\delta^{13}C$ values increased from −27.5‰ (close to the value of C_3-dominated plants in the catchment) to −25.1‰, while $\delta^{15}N$ values increased from −0.7‰ to 1.8‰. This indicates that increases in lake productivity caused common values of submerged macrophytes (Aichner et al., 2010; Herzschuh et al., 2005) (Fig. 4).

During the period from 9800 to 7700 cal yr BP, C/N ratios and TOC and TN concentrations remained at high values, indicating that the hydrology and ecology of the lake basin maintained a similar condition as occurred at the end of the preceding stage. The lake reached a higher stand and the aquatic productivity attained a

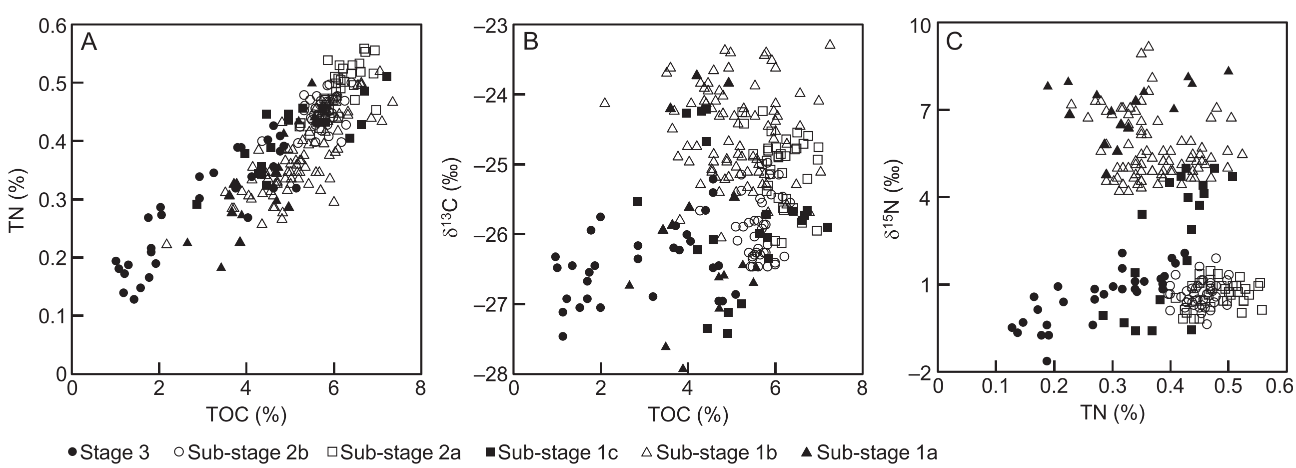

Fig. 5. Biplots of organic carbon and nitrogen proxies for the DL04 core spanning the last 11,500 cal yr (A) TN vs. TOC. (B) $\delta^{13}C$ vs. TOC. (C) $\delta^{15}N$ vs. TN. See Table 3 for coefficients of determination for the linear correlations for each stage/sub-stage. TOC data are cited from Xiao et al. (2008).

higher level under conditions of increased fluvial inputs, which promoted the steady accumulation of aquatic and terrestrial organic matter on the lake floor. $\delta^{13}C$ values decreased by ca. 1.5‰ during the first half of this period, presumably reflecting the enrichment of the lake DIC pool in ^{12}C and thus the decrease in $\delta^{13}C$ of aquatic organic matter as a result of large inputs of isotopically-lighter riverine DIC to the lake (Talbot and Johannessen, 1992; Talbot and Lærdal, 2000). The decrease in $\delta^{13}C$ may also have been the result of an increase in the relative contribution of isotopically-lighter, terrestrial organic matter (Talbot and Johannessen, 1992; Talbot and Lærdal, 2000). During the second half of this period, the $\delta^{13}C$ values increased by ca. 1‰, which may have been a result of the decreases in the relative contribution of terrestrial organic matter. During the entire period, both C/N ratios and TOC concentrations exhibit an overall trend of decreasing values, implying gradually decreasing inputs of terrestrial organic matter to the lake. $\delta^{15}N$ maintained low values of around ca. 0.4‰ through the entire period, following a rapid decrease by ca. 1.4‰ at the beginning. This may have been related to the effect of inputs of isotopically-lighter soil nitrogen on the lake DIN pool (Peters et al., 1978; Peterson and Howarth, 1987) and/or to the effect of atmospheric nitrogen fixation by Cyanophyta due to the nitrogen limitation of the lake DIN pool caused by the high productivity of aquatic phytoplankton (Hecky and Kling, 1981, 1987).

The interval between 7700 and 5900 cal yr BP is characterized by a further increase in the productivity of aquatic phytoplankton, as documented by significant increases in the TN concentration. C/N ratios remained at a value around 15, close to that of the preceding sub-stage, denoting a concomitant increase in both aquatic and terrestrial contributions to the sedimentary organic matter. These data suggest that the vegetation in the catchment was well-developed and surface runoff was intensified, while aquatic phytoplankton flourished in the conditions of a high lake level and warm lake waters. $\delta^{13}C$ values continued to increase through this interval, indicating the progressive enrichment of ^{13}C of the lake DIC pool due to enhanced aquatic productivity (Talbot and Johannessen, 1992; Talbot and Lærdal, 2000). $\delta^{15}N$ values remained relatively stable at a value close to that of atmospheric nitrogen, suggesting the likely effect of nitrogen fixation by blue-green algae on the $\delta^{15}N$ value of aquatic organic matter (Hecky and Kling, 1981, 1987). It is notable that the sedimentation rate during this period was the lowest of the entire Holocene (Fig. 3), implying a possible different reservoir age for the ^{14}C dating of bulk organic carbon of the DL04 core (Kao et al., 2008; Hou et al., 2012). We suggest the reservoir age during this period may be not as large as that of the surface sediment, because of the significantly increased terrestrial organic carbon input to the lake. Therefore several hundred years of ^{14}C dating error may be imposed upon the environmental interpretation of this period.

The several hundred years at the beginning of the interval from 5900 to 4850 cal yr BP were marked by drastic declines both in the input of terrestrial organic matter and in the productivity of aquatic phytoplankton, as demonstrated by large decreases in the TOC and TN concentrations. The relative proportion of autochthonous organic matter increased at the beginning of this interval, which may be related to the largely decreased allochthonous organic matter. These data imply that the vegetation deteriorated and surface runoff weakened in the lake catchment, while the lake underwent a lowering in level and a cooling in lake water temperature at the beginning of the interval. At the same time, $\delta^{13}C$ values decreased by up to 3‰, suggesting the possible degradation of deposited organic matter and the resulting release of CO_2, with low $\delta^{13}C$ values, into the lake water under conditions of increased concentration of dissolved oxygen in the lake bottom water (Talbot and Johannessen, 1992; Talbot and Lærdal, 2000). The relatively low C/N ratios and $\delta^{13}C$ values excluded the influence of submerged macrophytes on the total organic matter (Aichner et al., 2010). Subsequently, TOC and TN concentrations, C/N ratios and $\delta^{13}C$ values exhibit a trend of gradually increasing values, indicating a significant recovery of the hydrological and ecological status of the lake basin that is similar to that which occurred at the beginning of the Holocene. $\delta^{15}N$ values increased continuously through the entire period, which may have been the effect of the enrichment in ^{15}N of the lake DIN pool resulting from nitrification occurring during low lake stands (Talbot and Johannessen, 1992; Talbot and Lærdal, 2000).

During the period from 4850 to 750 cal yr BP, the terrestrial input of organic matter gradually decreased and aquatic productivity gradually declined, as indicated by the overall trend of decreases in TOC and TN concentrations; while the relative proportion of autochthonous organic matter exhibited an increasing trend based on the estimation from C/N ratios. $\delta^{13}C$ values increased by ca. 2.5‰, which would have resulted from the combined effects of decreases in the supply of isotopically-lighter terrestrial organic matter and increases in the $\delta^{13}C$ value of aquatic organic matter. The latter could be related to a shift in the phytoplanktonic metabolism from assimilating $[CO_2]_{aq}$ to HCO_3^- caused by increases in the alkalinity of lake waters during this period of lowered lake level (Smith and Walker, 1980; Lucas, 1983). The $\delta^{15}N$ continued to increase during this period, supporting the inference of falling lake level and rises in water alkalinity, because these processes could promote ammonia volatilization within the lake and thus the enrichment in ^{15}N of the lake DIN pool (Collister and Hayes, 1991; Talbot and Johannessen, 1992).

During the last 750 cal yr, changes in the hydrological and ecological conditions of the lake basin exhibit a pattern similar to that during the period from 5900 to 4850 cal yr BP. During the first 350 cal yr of this sub-stage, decreases in TOC and TN concentrations probably reflect a reduction in the input of terrestrial organic matter and decreased productivity of aquatic phytoplankton in the lake. The rapid decreases in $\delta^{13}C$, by as much as 3‰, probably indicate the degradation of deposited organic matter in the context of a lowered lake level. During the last 400 cal yr of this sub-stage, TOC and TN concentrations and $\delta^{13}C$ values all exhibit a trend of increasing values, while C/N ratios exhibit higher values, indicating a gradual increase in surface runoff and thus the lake level and a recovery of the terrestrial vegetation and aquatic productivity. $\delta^{15}N$ values increased continuously through the last 750 cal yr, suggesting an overall status of low lake level and high water alkalinity with the occasional occurrence of ammonia volatilization within the lake.

5.3. *Possible causes of the Holocene hydrological and ecological variations*

The carbon and nitrogen signatures demonstrate that Dali Lake was characterized by high water level and high aquatic productivity from 9800 to 7700 cal yr BP, at which time, however, low tree percentages and TOC concentrations from Daihai Lake sediments indicate that the precipitation in the region remained at a low level (Xiao et al., 2004) (Figs. 1 and 6). It is noticeable that TN and TOC concentrations are not significantly correlated for the period from 9800 to 7700 cal yr BP, although they are strongly positively correlated during the remainder of the Holocene (Fig. 5; Table 3). These findings suggest that soil erosion in the lake catchment and thus clastic input to the lake was less significant during this period, although the inflowing rivers were able to maintain a high lake stand. Summer insolation in the Northern Hemisphere began to increase 12,000 yr ago and reached a maximum (7% greater than the present value) from 10,000 to 9000 yr ago (Laskar et al., 2004).

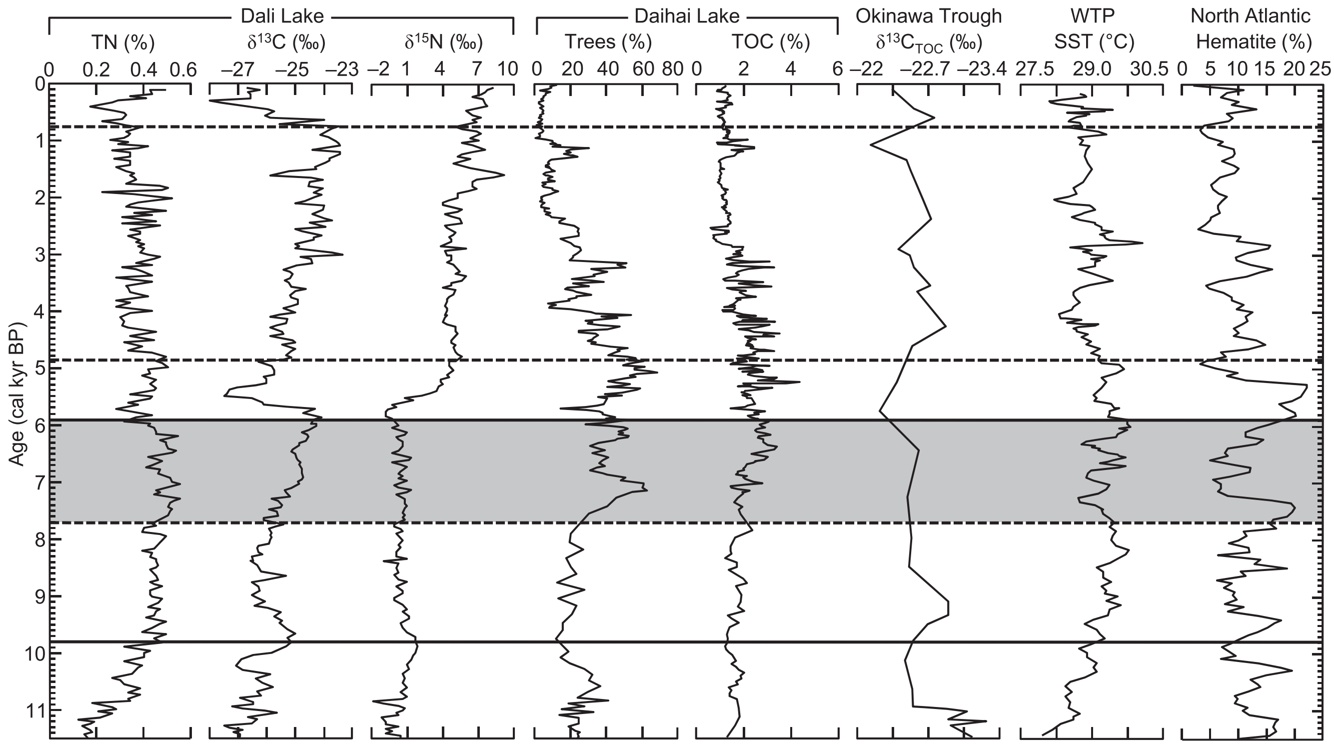

Fig. 6. Correlation of TN concentration, $\delta^{13}C$ and $\delta^{15}N$ of the sedimentary organic matter from the DL04 core spanning the last 11,500 cal yr with tree percentage and TOC concentration from Daihai Lake (Xiao et al., 2004, 2006), $\delta^{13}C$ value of TOC ($\delta^{13}C_{TOC}$) from the southern Okinawa Trough (Kao et al., 2008), sea surface temperature (SST) from the western tropical Pacific (WTP) (Stott et al., 2004) and hematite-stained grain concentration from the North Atlantic (Bond et al., 2001). High hematite-stained grain concentration implies high ice-rafted debris and thus cold temperature in the North Atlantic. Horizontal solid and dashed lines indicate the stages and sub-stages characterizing the pattern of changes in the hydrological condition in the Dali Lake region during the last 11,500 cal yr. Shaded bar indicates the highest water level and primary productivity of Dali Lake through the entire Holocene.

We thus infer that the expansion of Dali Lake during the early Holocene before 7700 cal yr BP may have resulted from the input of snow/ice melt water from the surrounding mountains in response to increases in Northern Hemisphere summer insolation. Modern hydrological observations show that the major inflow to Dali Lake, the Gongger River, generally discharges melt water during spring floods as large as during summer floods in the rainy season (Li, 1993). These observations support the inference that inputs of melting snow and ice to the lake played a key role in driving the lake level rises during the early Holocene.

Modern observations and historical documents indicate that the level of Dali Lake, and the area of pasture around the lake, increases during years of high summer rainfall (Li, 1993). This suggests a close relationship between the hydrological and ecological conditions within the lake basin and the regional monsoonal precipitation during the middle to late Holocene. From 7700 to 5900 cal yr BP, Dali Lake attained its highest stand of the entire Holocene, and aquatic phytoplankton and terrestrial vegetation flourished. We infer that the monsoonal precipitation intensified in the region during this period which promoted high stands of the lake and the flourishing of aquatic and terrestrial ecosystems in the lake catchment. In the Daihai Lake basin (Fig. 1), large-scale covers of mixed coniferous and broadleaved forests were developed (high tree percentages in Fig. 6) and TOC concentrations increased significantly after 7900 cal yr BP (Fig. 6), indicating a prominent enhancement of the temperature and precipitation in the semiarid regions of northern China 7900 cal yr ago (Xiao et al., 2004). In the Huangqihai Lake (Fig. 1), the water level was low before 8500 cal yr BP but increased significantly at 8500 cal yr BP, implying the monsoonal precipitation in the region significantly enhanced at 8500 cal yr BP (Li et al., 1992; Zhang et al., 2012). These data, within dating error, provide substantial support for our inference. On the other hand, given the significant warmth in middle and low latitudes from 8000 to 6000 yr BP (Shi et al., 1993; Kim et al., 2002; Stott et al., 2004), the results of this study present a favorable scenario of hydrological and ecological conditions for the EASM margin if the current global warming trend continues.

The variations of the hydrology and ecology in the EASM margin was closely related to the monsoonal precipitation in the region as discussed above. Our geochemical data imply a development of hydrology and ecology in the EASM margin during the middle Holocene after 7700 cal yr BP, however, in southern China the hydroclimate generally improved since the early Holocene at 11,000–10,000 cal yr BP. For example, high-resolution, absolutely dated oxygen isotope records of stalagmites from Dongge Cave indicate that Asian summer monsoon intensified significantly during the early to middle Holocene (Wang et al., 2005; Dykoski et al., 2005). C/N ratios-based rainfall intensity recovered from the lake sediments from Tung-Yuan Pond in southern Taiwan significantly increased in the several hundred years after 10,600 and 9500 cal yr BP (Yang et al., 2011). The onset of significant improvement of the hydroclimate in northern China lagged behind that in southern China by 4000–3000 yr during the Holocene. We suggest that during the early Holocene, the northward migration of the monsoon rain belt may have been hampered by the Northern Hemisphere (NH) remnant ice sheets (Dyke et al., 2003) as well as the relatively lower global sea level (Peltier and Fairbanks, 2006) despite gradual increases in the NH summer insolation (Laskar et al., 2004) and in sea surface temperature (SST) of the western tropical Pacific (Stott et al., 2004). At 7000–6000 cal yr BP, ice sheets in the NH high-latitudes retreated to the present extent (Dyke et al., 2003; Carlson et al., 2008), while the global sea level rose to its current configuration (Peltier, 2002; Peltier and Fairbanks, 2006). The relatively stable and high $\delta^{13}C_{TOC}$ values from the southern Okinawa Trough indicate that the East China Sea reached a high and stable level after ca. 8000 cal yr BP (Kao et al., 2008) (Fig. 6), accompanied by significantly intensified Kuroshio Current evidenced by decreased total sulfur contents (Kao et al., 2005). Such changes in the boundary conditions and the ocean current could help enhance the thermal contrast between the tropical Pacific and the Asian continent, and shorten the distance from the Asian interior to the source areas of the monsoon moisture, thereby leading to further northerly penetration of the monsoon rain belt beyond its modern northern limit, and significant increases in the precipitation and thus the status of hydrology and ecology in the present monsoon margin.

During the intervals from 5900 to 4850 and 750–0 cal yr BP, Dali Lake experienced intervals of dramatic falls in water level and inferred drastic decreases in the productivity of aquatic and terrestrial ecosystems, followed by gradual recovery of the hydrological and ecological conditions. These changes were consistent well with the variations in the tree percentages and TOC concentrations from Daihai Lake (Xiao et al., 2004) (Fig. 6), both of which indicate decreases in the precipitation in the region at the beginning of these intervals. The significantly weakening of the monsoonal precipitation in the EASM margin at ca. 5900 and 750 cal yr BP corresponds in time to the decrease in SST of the western tropical Pacific (Stott et al., 2004) and the occurrence of ice-rafted debris in the North Atlantic (Bond et al., 2001) (Fig. 6). These data imply a close link between the summer monsoon variability and climatic processes in the high and low latitudes on millennial and centennial scales. The cooling of the North Atlantic could generate the negative state of North Atlantic Oscillation/ Arctic Oscillation (Bond et al., 2001), thus leading to an anomalously weakening in the intensity of the summer monsoon (Sung et al., 2006). The decrease in SST of the western tropical Pacific could reduce the formation of water vapor over the source areas of the summer monsoon, thereby decreasing the moisture available for transport onto the Asian inland via the monsoon circulation.

The interval between 4850 and 750 cal yr BP was characterized by decreasing status of hydrology and ecology in the lake catchment, and the fluctuations during this interval were more frequent and much larger than other intervals of the Holocene (Fig. 6). The regional precipitation gradually decreased as indicated by the decreased tree percentages and TOC concentrations from Daihai Lake (Xiao et al., 2004) (Fig. 6), which may be caused by gradually decreased SST of the western tropical Pacific (Stott et al., 2004) (Fig. 6) and the NH summer insolation (Laskar et al., 2004). These data indicate that the hydrology and ecology in the EASM margin would become much more fragile when regional climate reached a status of gradual deteriorating trend, in other words, small decreases in the precipitation and temperature superimposed on the long-term deteriorated climate may cause large declines in the hydrology and ecology in the semi-arid regions of northern China.

6. Conclusions

Carbon and nitrogen signatures of the sedimentary organic matter extracted from a sediment core from Dali Lake, Inner Mongolia, document significant variations in the hydrology and ecology of the lake basin during the Holocene. From 11,500 to 9800 cal yr BP, fluvial input to the lake gradually increased, leading to gradual rises in lake level and aquatic productivity. From 9800 to 7700 cal yr BP, surface runoff from the catchment intensified, and terrestrial plants flourished, while the lake attained a high stand and high productivity. From 7700 to 5900 cal yr BP, surface runoff intensified further, resulting in the highest lake stand during the entire Holocene, while the status of the terrestrial and aquatic ecosystems significantly improved. Around 5900 cal yr BP, surface runoff dramatically weakened and the lake level fell, which was accompanied by a significant deterioration in the status of the terrestrial vegetation and aquatic productivity. Subsequently, until 4850 cal yr BP, a gradual recovery of the hydrological and ecological conditions occurred. From 4850 to 750 cal yr BP, surface runoff continued to weaken, the lake continued to shrink, and the status of terrestrial and aquatic ecosystems gradually declined. During the last 750 cal yr, the hydrological and ecological conditions of the lake basin varied in a pattern similar to those from 5900 to 4850 cal yr BP. Hydrological and ecological variations in the EASM margin during the Holocene were closely related to the combined effects of regional precipitation and temperature which were ultimately controlled by the NH summer insolation, the boundary conditions and the physical environment of ocean current. Our data suggest that future global warming scenarios would potentially be beneficial for the hydrological and ecological conditions of the EASM margin, while small decreases in the precipitation and temperature superimposed on the long-term deteriorated climate may cause large declines in the hydrology and ecology in the semi-arid regions of northern China.

Carbon and nitrogen isotopes of sedimentary organic matter in lakes are greatly affected by the biogeochemical cycles within lakes such as the degradation and nitrification/denitrification of organic matter and the HCO_3^- assimilation and nitrogen fixation by aquatic phytoplankton. Comprehensive investigations into the internal processes occurring within different types of lakes in different climate regions would help improve our understanding of the environmental implications of carbon and nitrogen isotopic signatures of lacustrine organic matter.

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