

# Vertical distributions of bound saturated fatty acids and compound-specific stable carbon isotope compositions in sediments of two lakes in China: implication for the influence of eutrophication

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Received: 14 April 2014 / Accepted: 16 June 2014 / Published online: 6 July 2014  
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**Abstract** Lakes Dianchi (DC) and Bosten (BST) were determined to be at different stages of eutrophication, by use of total organic carbon content, bulk carbon isotopic composition, bulk nitrogen isotopic composition, and bound saturated fatty acid (BSFA) concentrations in sediment cores. A rapid increase in the supply of organic matter (OM) to DC began after the 1950s, while the environment and trophic status of BST remained constant as indicated by characteristics of OM input to sediments. The BSFA ratios of nC14+nC16+nC18/nC24+nC26+nC28 increase upward from 7 to 13 in the DC core, which are significantly greater than those from BST (2 to 3). This result is consistent with algae or bacteria being the dominant contribution of the OM increase induced by

eutrophication in DC. The positive shift of nC16 compound-specific  $\delta^{13}\text{C}$  in the upper section might be an indicator of excess algal productivity, which was observed in the two lakes. The positive shifts of compound-specific  $\delta^{13}\text{C}$  of other BSFAs were also observed in the upper section of the core only from DC. The observed trends of compound-specific  $\delta^{13}\text{C}$  of BSFA originated from different sources became more consistent, which reflected the intensified eutrophication had profoundly affected production and preservation of OM in DC. The results observed for BST indicated that accumulation of algae did not affect the entire aquatic ecosystem until now.

**Keywords** Bound saturated fatty acids · Compound-specific  $\delta^{13}\text{C}$  · Core sediments · Eutrophication · Lake Dianchi · Lake Bosten · Asia

Responsible editor: Hongwen Sun

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## Introduction

Most urban lakes in China are facing serious eutrophication. Eutrophication profoundly affects aquatic ecosystems by altering biodiversity, trophic structure, and biogeochemical cycling (Schindler 2006). The effect of eutrophication on production and preservation of organic matter (OM) and alteration of its carbon or nitrogen isotope compositions in situ has been widely studied (Trolle et al. 2010). For example, when the primary productivity of algae increased, it led to greater inputs of the smaller molecular weight PAHs into water, and consequently, increased organic particulates would contribute more of the lesser molecular weight PAHs to sediments. Thus, the molecular ratios of PAHs might not accurately represent the original sources of PAHs (Wu et al. 2012b). Also, biosynthesis and metabolism of enhanced bacterial activities associated with eutrophication are expected to result in an increase

of  $\delta^{13}\text{C}$  of remaining organic matter (Macko and Estep 1984; Macko et al. 1987). This is analogous to the general observation in food webs of enrichment of  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  in higher trophic levels.

Lipids are a large and diverse group of naturally occurring organic compounds that are related by their solubility in non-polar organic solvents. Compound-specific  $\delta^{13}\text{C}$ , combined with distributions of lipids, are routinely used to examine the sources and to provide information for understanding biogeochemical processes and reconstructing past climate and environmental conditions in aquatic environments (Brincat et al. 2000; Chikaraishi and Naraoka 2003; Glaser and Zech 2005; Hu et al. 2008; Lu and Meyers 2009; Machiwa 2010; Tanner et al. 2010; Gómez-Gutiérrez et al. 2011). Among the various lipids that can be analyzed, bound saturated fatty acids (BSFAs) have been more frequently studied, and their identification and quantification has improved (Barakat and Rullkötter 1995). It is well-known that BSFAs occur chemically bound to macromolecular structures of kerogen, asphaltenes, and the mineral matrix in sediments (Barakat and Rullkötter 1995). In a tightly bound state, BSFAs are considered to be less susceptible to early diagenetic process (Lajat et al. 1990; Xiong et al. 2010). However, previous study had shown that bacterial activities associated with eutrophication seemed to have a considerable effect on degradation of BSFAs derived from both algae and terrestrial plants (Wang et al. 2013). Further investigation was deemed necessary to determine whether the eutrophication affected compound-specific  $\delta^{13}\text{C}$  of BSFAs.

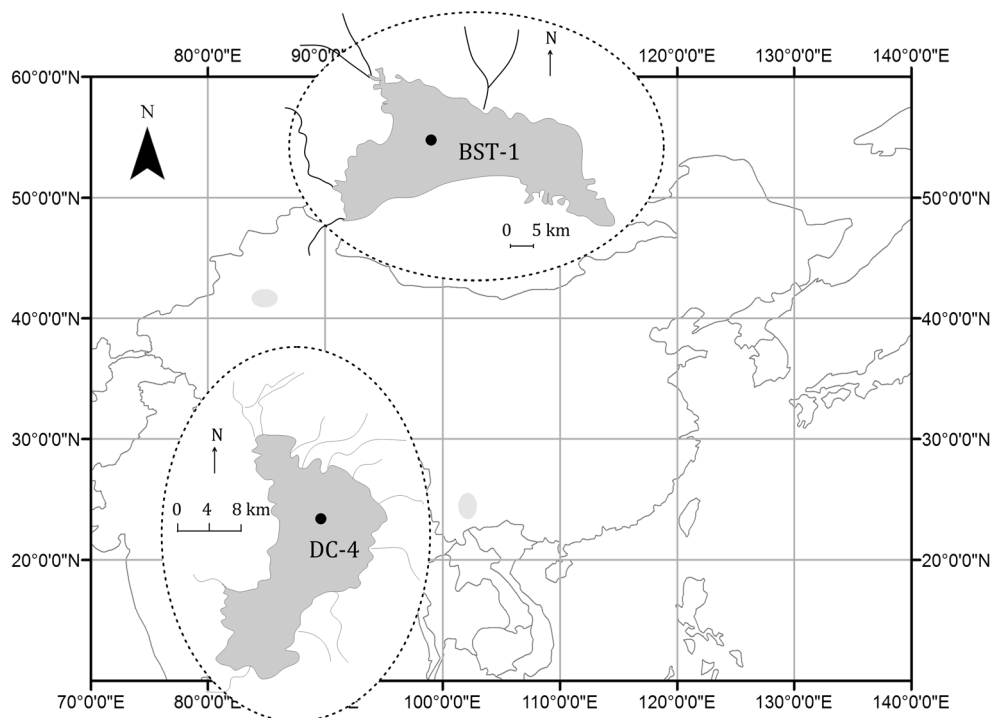
The main objective of this study was to provide an insight into the effect of eutrophication on alteration of compound-specific  $\delta^{13}\text{C}$  of BSFAs. In this study, in order to better evaluate the influence of eutrophication on compound-specific  $\delta^{13}\text{C}$  in a eutrophic lake, two lakes in different stages of cultural eutrophication were compared. Based on previous information on geochemistry of the lakes (Wu et al. 2012a), one polluted lake (Bositeng, BST) and one more polluted lake (Dianchi, DC) were chosen. This key objective was achieved through the following: (1) analyses of total organic carbon (TOC), total nitrogen (TN), and bulk  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values of OM of two sediment cores dated by  $^{210}\text{Pb}$  and  $^{137}\text{Cs}$ ; (2) analyses of distributions and compositions of BSFAs of two dated sediment cores; and (3) measurement of compound-specific  $\delta^{13}\text{C}$  of BSFAs. Here, we present data on BSFA distributions, compositions, and compound-specific  $\delta^{13}\text{C}$  of BSFAs extracted from the sediments collected from DC and BST. These data show vertical variations of compound-specific  $\delta^{13}\text{C}$  in the eutrophic lake and provide understanding of eutrophication on preservation of BSFAs and alteration of its compound-specific  $\delta^{13}\text{C}$ .

### Methods

#### Collection of cores

Two lakes were studied. DC (24° 40' –25° 02' N, 102° 36' –102° 47' E) is a plateau lake located in Kunming City, Yunnan Province, in southwest of China (Fig. 1). It is the

**Fig. 1** Map showing sites of collection of sediment cores



sixth largest freshwater lake in China. Average depth is approximately 5 m and maximum depth is 8 m. The surface area of the lake is approximately 330 km<sup>2</sup>. Historical records show that the watershed of DC has been densely populated since the sixteenth century. Due to growth of the population and associated demands for agricultural land, more and more farmlands have been reclaimed around the lake, and the effects of cultivation on the lake ecosystem have become more and more serious (Huang et al. 2007). Over the past 30 years, DC, which is one of the most eutrophic freshwater lakes in China, has been becoming even more eutrophied. BST is a freshwater lake located 57 km northeast of Korla (41° 30′ –42° 00′N, 86° 30′ –87° 18′ E), in northwest of China (Fig. 1). Covering an area of approximately 1,000 km<sup>2</sup>, it is one of the largest inland freshwater lakes in China. Because the lake is surrounded by the Gobi Desert, the ecological environment is well-preserved due to the relatively small amount of human habitation and associated agricultural and industrial activities. In spite of this, several studies have shown eutrophication has been increasing in during recent years.

Cores of sediment were collected from the center of DC and BST (locations DC-4 and BST-1; Fig. 1) during May and August 2006. Cores were collected by use of a piston percussion corer fitted with a 58-mm internal diameter perspex tube. The cores were sectioned into 1-cm intervals immediately after collection and then freeze-dried. Seven subsamples of the core from DC-4 (upper 20 cm) and ten subsamples from the BST-1 core (upper 20 cm) were selected for organic compound analyses. Samples from other cores collected simultaneously were used for determination of dates of deposition by quantification of <sup>210</sup>Pb and <sup>137</sup>Cs.

#### Determination of <sup>137</sup>Cs and <sup>210</sup>Pb activity

<sup>137</sup>Cs activity was determined by previously described methods (Wan et al. 2005). Briefly, <sup>137</sup>Cs activity was measured by gamma spectrometry with a Canberra S-100 multichannel spectrometer coupled with a GCW3022 H-P Ge coaxial detector (efficiency 50 %). A <sup>137</sup>Cs standard was supplied by the Institute of Atomic Energy, Chinese Academy of Sciences (catalog no. 7137 and source no. 586-26-2). The greatest <sup>137</sup>Cs activity was found at a depth of 8 cm in DC and 7 cm in BST, which was assigned a date of 1963, since that was the year of maximum nuclear explosion tests in the atmosphere.

Activities of <sup>210</sup>Po and <sup>226</sup>Ra were measured according to previously described methods (Wan et al. 1987). Briefly, <sup>210</sup>Po activity in sediment was analyzed by alpha spectrometry with a Canberra S-100 and multichannel spectrometer with a PIPS Si detector, and <sup>226</sup>Ra activity was determined by gamma spectrometry with a Canberra S-100 multichannel spectrometer mated to GCW3022 H-PR Ge well detector. Excess <sup>210</sup>Pb activity was calculated by subtracting <sup>226</sup>Ra activity from the

total <sup>210</sup>Pb activity, which was derived from <sup>210</sup>Po. According to activity profiles of <sup>137</sup>Cs and excess <sup>210</sup>Pb, the collected cores (20 cm in length) were deposited during the periods 1793–2006 in DC and 1883–2006 in BST, respectively.

#### Bulk carbon and nitrogen isotope analysis

Concentrations of total organic carbon (TOC) and total nitrogen (TN) were determined on a CHNS Vario E1 III elemental analyzer. Replicate analysis of one sample ( $n=3$ ) gave a precision of  $\pm 0.02$  % for weight. Carbonates were removed by acidification with dilute HCl before determination of TOC.

Values of stable isotopes of carbon (C) and nitrogen (N) were determined by use of a Thermo Finnigan Delta Plus XL mass spectrometer connected to a Flash EA 1112 elemental analyzer via a Finnigan MAT ConFlo III interface.  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  are reported as per mil relative to VPDB and air, respectively. The instrument analytical precision for  $\delta^{13}\text{C}$  is  $\pm 0.1$  ‰ and  $\delta^{15}\text{N}$  is  $\pm 0.2$  ‰.

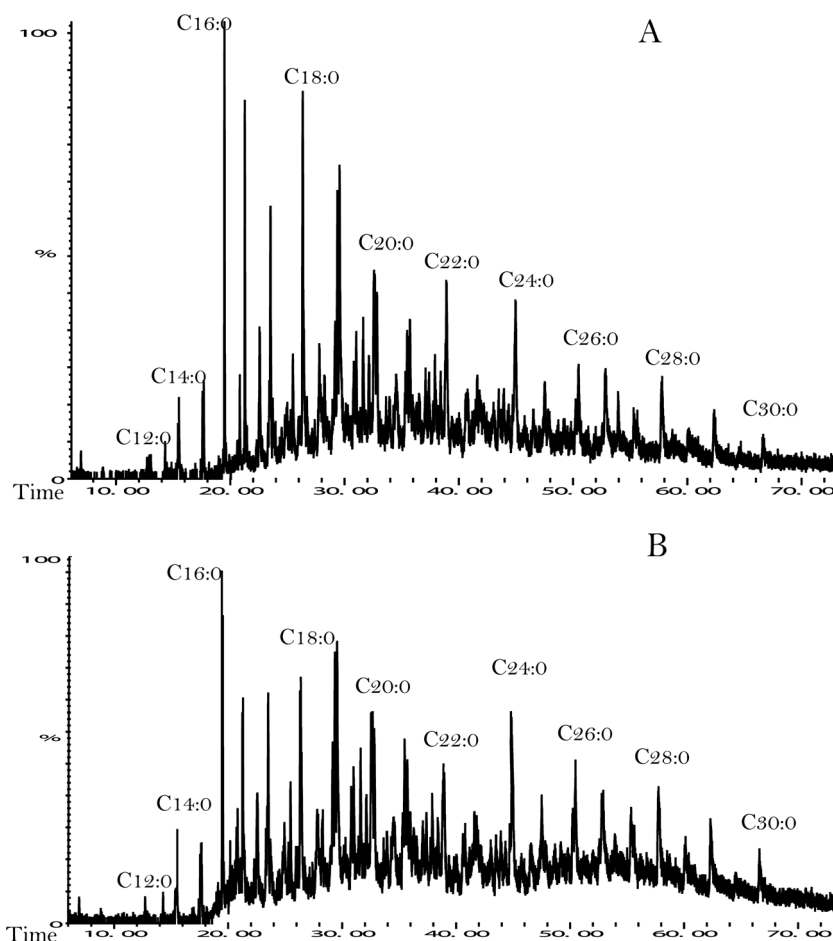
#### Extraction and fractionation of BSFAs

BSFAs were quantified by use of a previously described method (Xiong et al. 2010). For determination of the composition of OM, sediments were first Soxhlet extracted for 72 h with dichloromethane/methanol (9:1 v/v) to obtain the soluble fraction (free lipids). Residual sediments were saponified with 0.5 M KOH in methanol under reflux for 12 h to release bound lipids, centrifuged, and the supernatant decanted. The neutral fraction was extracted with *n*-hexane/diethyl ether (9:1 v/v), then acidified to pH=1 by addition of HCl. The acidic fraction, which contained BSFAs, was extracted with dichloromethane. The acidic fraction was analyzed by gas chromatography-mass spectrometry (GC-MS). Prior to GC-MS analyses, fatty acids were converted to fatty acid methyl esters (FAMES) by methylation with saturated HCl-methanol by heating in an oven at 100 °C for 1 h.

#### GC-MS and GC-IRMS

GC-MS analyses were carried out using a Finnigan Platform II mass spectrometer coupled to a HP 6890 GC and a HP-5 fused silica capillary column (50 m×0.32 mm×0.25 μm). Temperature was programmed at 70 °C for 5 min, then at a rate of 3 °C/min to 290 °C, and held for 30 min at 290 °C. Quantification of BSFAs was achieved by integration of peak areas in total ion current plots (Fig. 2). Deuterated eicosane was used as an internal standard for quantification, and the response factor of individual bound fatty acids relative to the standard was assumed to be 1.0 (Xiong et al. 2010). Compound-specific isotopic analyses were made by use of an Agilent 6890 Series gas chromatograph (GC) interfaced to a Finnigan Delta Plus XL isotope ratio mass spectrometer

**Fig. 2** GC-MS chromatograms on the distribution of bound saturated fatty acids (**a** sample DC-4-1 and **b** sample BST-1-1)



(IRMS) through a combustion furnace. The oven-heating program was the same as for GC analyses. CO<sub>2</sub> gas with pre-calibrated isotopic composition was used as standard. Isotopic compositions of individual compounds were calculated by integration of the mass 44, 45, and 46 ion currents (Merrit et al. 1994). Analyses were done in duplicate with standard deviations better than 0.5 ‰. The measured isotope ratios of the FAMEs were corrected for the isotope ratio of the methyl moiety to obtain the isotope ratios of BSFAs. This was done by using the following formula, and δ<sup>13</sup>C<sub>MeOH</sub> is the δ<sup>13</sup>C of the methanol used for the methylating reaction (−33.78 ‰) in this study.

$$\delta^{13}C_{FA} = [(C_n + 1) * \delta^{13}C_{FAME} - \delta^{13}C_{MeOH}] / C_n$$

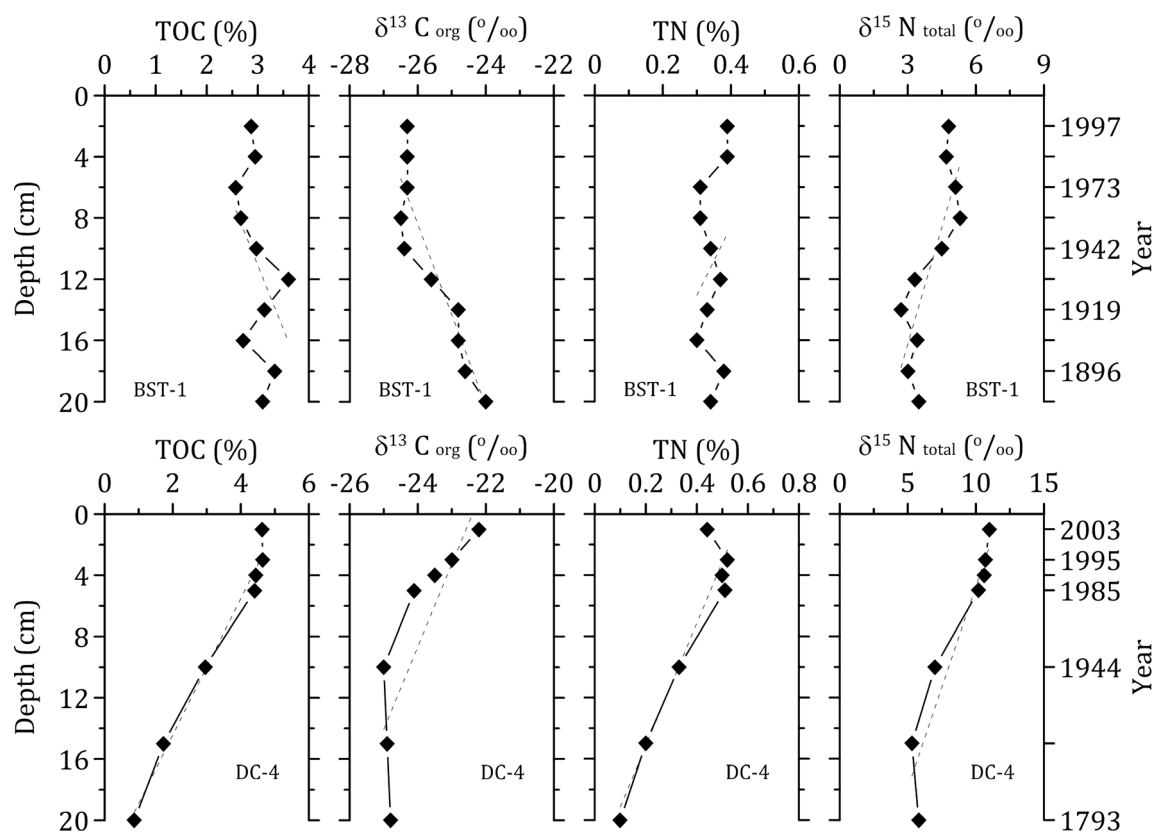
**Results and discussion**

TOC, TN, and bulk δ<sup>13</sup>C, δ<sup>15</sup>N values of OM

Concentrations of TOC and TN and bulk δ<sup>13</sup>C and δ<sup>15</sup>N values are commonly used for the reconstruction of events recorded by constituents in sediment cores (Meyers and Ishiwatari 1993). Vertical profiles of TOC, TN, bulk δ<sup>13</sup>C,

and δ<sup>15</sup>N values are shown in Fig. 3. TOC and TN concentrations varied within a narrow range from 2.7 to 3.3 and 0.31 to 0.39 % in the BST-1 core, while these values ranged from 0.9 to 4.6 and 0.1 to 0.52 % and rapidly increased fivefold upward in the DC-4 core after 1950s. This result indicated a rapid increase in the supply of OM to DC during this period. In comparison, the supply of OM to BST was stable during this period, especially after 1950.

Generally, the δ<sup>13</sup>C of terrestrial plants ranges from −30 to −25 ‰ while that of aquatic plants and alga is in the range of −25 to −18 ‰ (Meyers and Ishiwatari 1993). Bulk δ<sup>13</sup>C values enriched in the heavier isotope (from −25 to −22 ‰) at more shallow depths of the DC-4 core, suggesting a gradual increase in OM from aquatic plants to DC in more recent decades. In contrast, bulk δ<sup>13</sup>C values (−26.6 ‰) in the core from BST-1 exhibited contribution from both aquatic and terrestrial plants. Stable nitrogen isotope composition is also a good indicator of OM sources. For example, the δ<sup>15</sup>N value of dissolved NO<sub>3</sub><sup>−</sup> used by algae is typically 7–10 ‰, typical δ<sup>15</sup>N values range from 3 to 8 ‰ for soil organic nitrogen nitrate and 10 to 20 ‰ for human waste nitrate (Aravena et al. 1993). Bulk δ<sup>15</sup>N values increased upward in both lakes, with ranges of 3.0 to 5.3 ‰ in the BST-1 core and 9.7 to 12.3 ‰ in the DC-4 core, respectively. This further supported the



**Fig. 3** Vertical profiles of TOC, TN, Bulk  $\delta^{13}\text{C}$ , and Bulk  $\delta^{15}\text{N}$  in cores from BST-1 and DC-4

conclusion that the contribution of anthropogenic nitrogen rapidly increased in DC during this period.

Vertical variations of TOC, TN, bulk  $\delta^{13}\text{C}$ , and bulk  $\delta^{15}\text{N}$  in the DC-4 core are consistent with results from field investigation and records of total phosphorus contents from the center of DC (Gao et al. 2005). Over the past 30 years, with rapid economic development, excessive nitrogen and phosphorus loadings in the water further accelerated eutrophication in DC (Gao et al. 2005). In contrast, the trophic status of BST, as indicated by characteristics of OM in the sediments, has been stable.

#### Compositions and vertical variations of BSFAs

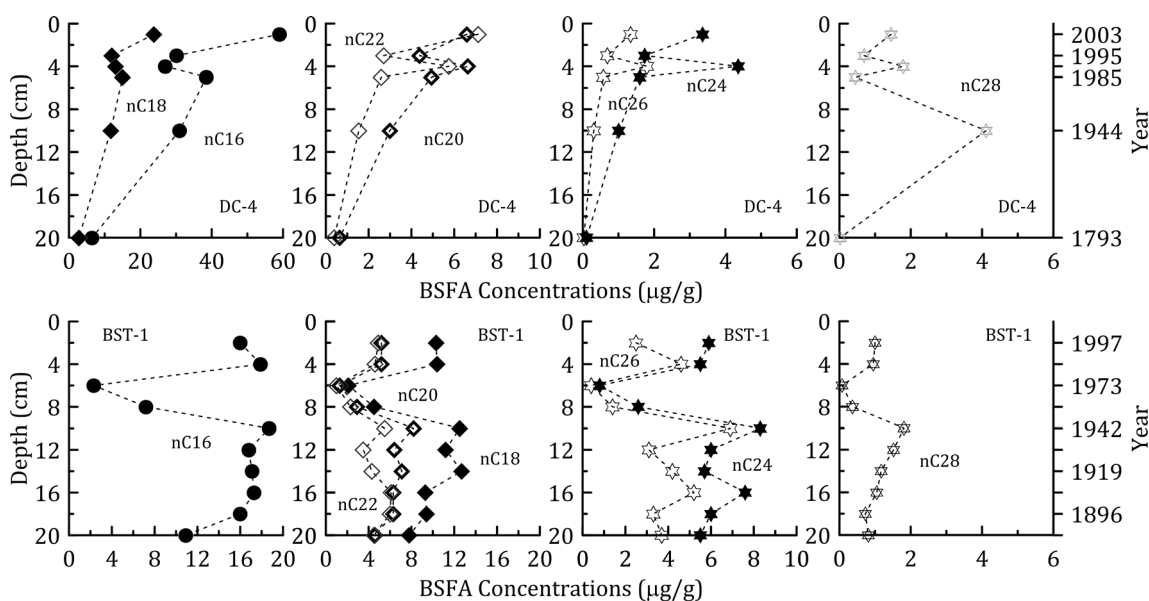
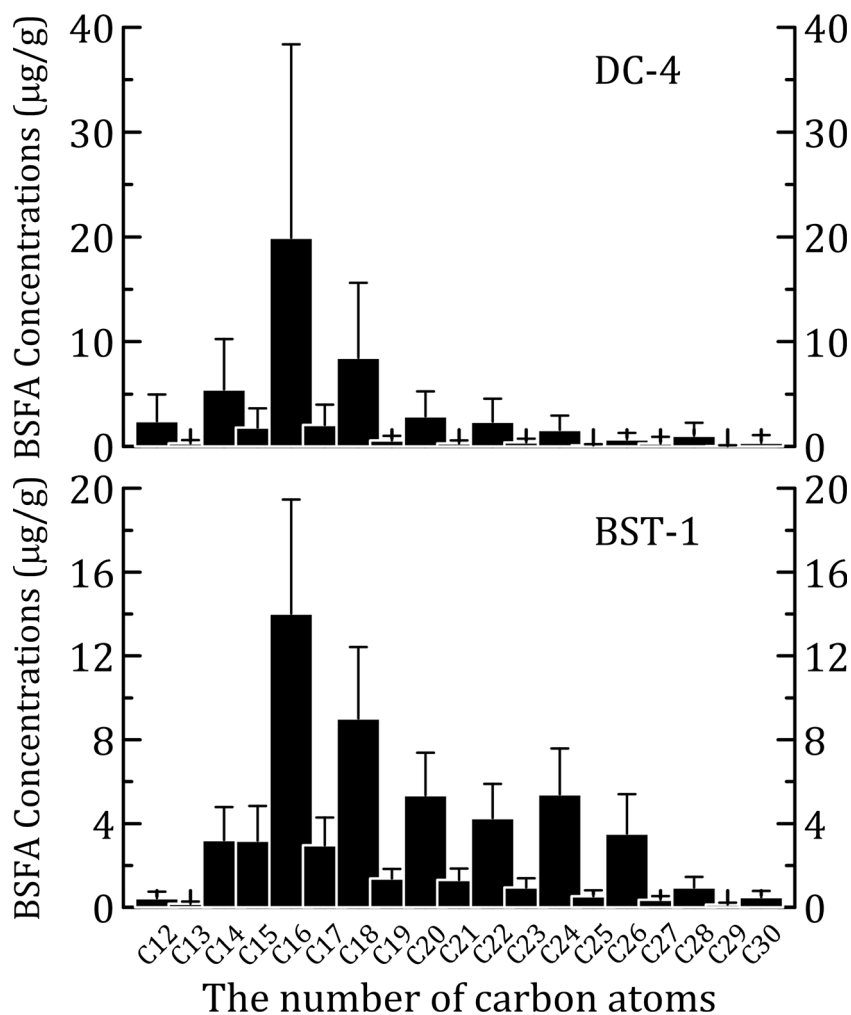
BSFA compounds in the sediments ranged from nC12 to nC30 in seven samples and had a maximum concentration on nC16 in the DC-4 core (Fig. 4), while BSFAs exhibited a bimodal distribution with nC16 being the maximum and nC24 the secondary maximum in the BST-4 core (Fig. 4). In the DC-4 core, the predominant BSFAs were nC16 and nC18, and the two compounds comprised  $41\pm 6$  and  $17\pm 3$  % of the total BSFA concentrations (sum of nC12 to nC30), respectively. A bimodal distribution of BSFAs was observed in the sediments at different depths in the BST-1 core. The predominant BSFAs were C16 ( $24\pm 2$  %) $>$ C18 ( $16\pm 2$  %) $>$ C24 ( $9\pm 1$  %) $>$ C26 (6

$\pm 1$  %) in decreasing order of concentration. The proxy  $\text{TAR} = (\text{nC14} + \text{nC16} + \text{nC18}) / (\text{nC24} + \text{nC26} + \text{nC28})$  can be used to evaluate compositional differences between the two lakes. Ratios range from 2 to 3 ( $n=10$ ) in samples from the BST-1 core, which are significantly less than those from the DC-4 core (7–13,  $n=7$ ). In general, the presence of the longer-chain fatty acids (LCFAs,  $\geq \text{nC24}$ ) is indicative of contribution from higher plants, especially of terrestrial origin, while the shorter-chain fatty acids (SCFAs, nC14, nC16, and nC18) have been widely found in many aquatic organisms including phytoplankton, bacteria, and algae, and they are generally found in greater concentrations in lower organisms than in higher organisms (Logan and Eglinton 1994; Meyers 1997). Different compositions of BSFAs between the two lakes could represent different source contributions, which was consistent with the result obtained from bulk  $\delta^{13}\text{C}$  values.

Total concentration of BSFAs (sum of nC12 to nC30) was greatest in the top sediment ( $>130$   $\mu\text{g/g}$ ) and decreased rapidly downward to  $\sim 10$   $\mu\text{g/g}$  at a depth of 20 cm in the DC-4 core (Fig. 5). The greater concentrations near the top of the core were coincident with higher TAR values. This result suggests that phytoplankton, algae, or bacteria were the dominant sources for the observed increase in BSFAs over time. In comparison, total BSFA concentrations remained relatively constant except the sharply decreasing trend during the period



**Fig. 4** Different profiles of BSFA composition in the samples from the DC-4 core and BST-1 core



**Fig. 5** Concentration variations of individual BSFA with depth in the DC-4 core and BST-1 core

1940–1985 in the BST-1 core (Fig. 5). The lake was characterized with low water levels in this period due to increased consumption of water, which has led to rapid decreasing of OM input to the lake.

#### Compound-specific $\delta^{13}\text{C}$ values of BSFAs

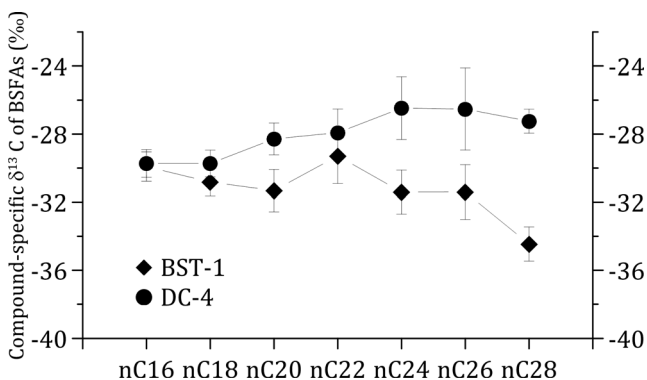
Compound-specific  $\delta^{13}\text{C}$  of BSFAs in the two sediment cores were reported for the nC16, nC18, nC20, nC22, nC24, nC26, and nC28 homologues but could not be determined for compounds with odd numbers of carbon atoms because these occurred at concentration less than the limit of quantification (Figs. 2, 6, 7). The compound-specific  $\delta^{13}\text{C}$  of the individual BSFAs were somewhat less than those of bulk  $\delta^{13}\text{C}$ . This is in agreement with the results of other researchers who have reported that labile OM contained more  $\delta^{13}\text{C}$  (Benner et al. 1987). Overall, the compound-specific  $\delta^{13}\text{C}$  of BSFAs from DC-4 ranged from  $-31.3$  to  $-24.5$  ‰ with a maximum for nC26 and minimum for C16 (Fig. 6), while the compound-specific  $\delta^{13}\text{C}$  of BSFAs in the BST-1 core ranged from  $-36.2$  to  $-27.3$  ‰ and the greater and lesser values were found for nC16 and nC28, respectively (Fig. 6). In general, aquatic organisms such as phytoplankton, algae, and macrophytes usually use dissolved  $\text{CO}_2$  ( $\delta^{13}\text{C} = -7$  ‰) or  $\text{HCO}_3^-$  ( $\delta^{13}\text{C} = 1$  ‰) in water as their carbon source, while land plants are dependent on atmospheric  $\text{CO}_2$  ( $\delta^{13}\text{C} = -7$  ‰) (Lesniak and Sakai 1989). Isotopic ratios for different organisms and its pathway of carbon fixation are roughly preserved in the compound-specific  $\delta^{13}\text{C}$  of the lipids. The compound-specific  $\delta^{13}\text{C}$  of LCFAs deposited in sediments from the BST-1 core were isotopically 4–6 ‰ lighter than those deposited in the DC-4 core (Fig. 6). One primary reason was the isotopic depletion (or enrichment) in different higher plants, with lipids derived from C3 and C4 land plants having  $\delta^{13}\text{C}$  values of  $-35$  and  $-20$  ‰, respectively, in average (Cerling et al. 1993; Rieley et al. 1993). This result is consistent with the fact that C3 vegetation predominates in the wet and cold area surrounding BST,

while C4 plants are more predominant in the warmer area surrounding DC. However, there was no significant difference in the compound-specific  $\delta^{13}\text{C}$  of SCFAs, reflecting some similar characteristics of algae or bacteria in the freshwater environments.

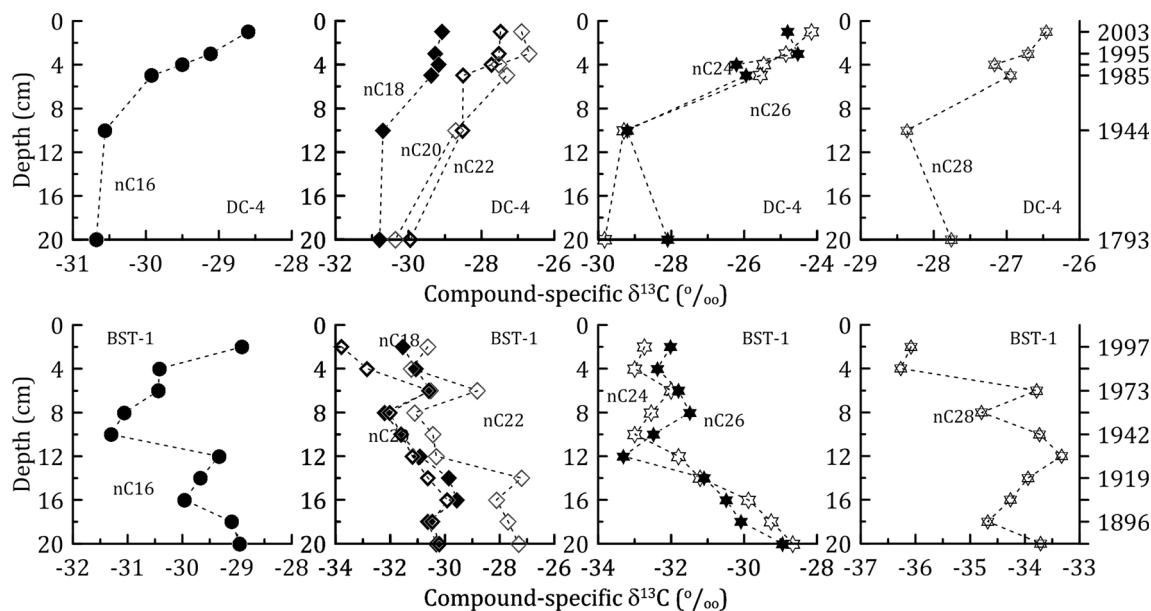
#### Influence of eutrophication on isotopic composition

Different vertical trends in compound-specific  $\delta^{13}\text{C}$  of BSFAs were observed in the two lakes. The trends of compound-specific  $\delta^{13}\text{C}$  of all BSFAs in DC-4 were similar that a sharp increase occurred after the 1940s. Except for nC16 BSFA, the compound-specific  $\delta^{13}\text{C}$  of BSFAs in the BST-1 core decreased from the bottom to the top, but for the nC16 BSFA, there was an obvious increasing trend after 1950s.

Contributions of algae/bacteria were dominant for OM input in the DC-4 core, and thus, their effects on bulk isotope values were primary. Compound-specific  $\delta^{13}\text{C}$  of the nC16 BSFA was in good agreement with the bulk  $\delta^{13}\text{C}$ , covarying and showing the same excursions down-core in the DC-4 core ( $R^2 = 0.98$ ,  $n = 7$ ) (Fig. 7). The rapid positive shift of both nC16 compound-specific  $\delta^{13}\text{C}$  and bulk  $\delta^{13}\text{C}$  that was observed in the upper section further confirmed the excess phytoplankton productivity in DC. Not only that, the fit of compound-specific  $\delta^{13}\text{C}$  between nC16 homologue and other individual BSFA yielded  $R^2 = 0.73$ – $0.87$  (Fig. 7). However, the difference in isotope between individual BSFAs was more than 3 ‰. In general, fractionation of isotope during extension of the carbon chain in an individual organism commonly does not exceed 1–2 ‰ (Schouten et al. 1998). Thus, they were unlikely to be from the same source. The compound-specific  $\delta^{13}\text{C}$  of BSFAs from different sources showed similar trends in the DC-4 core. This result suggested that excessive algal productivity seemed to have a significant effect on compound-specific  $\delta^{13}\text{C}$  of BSFAs. Actually, algae preferentially removed dissolved  $^{12}\text{CO}_2$  from water and left the remaining dissolved inorganic carbon enriched in  $^{13}\text{C}$  (Hollander and MacKenzie 1991; Hodell and Schelske 1998). When alga productivity continues to increase to a certain extent, a progressively increased fraction of the  $^{13}\text{CO}_2$  would be available as a source of carbon for both algae itself and macrophytes (nC20 and nC22 BSFAs) and incorporated into the lipids, which resulted in similar enrich trends in their isotopic compositions (Raven et al. 2002). For LCFAs, correlations of concentrations of nC16 and LCFAs in  $\delta^{13}\text{C}$  is also noteworthy. This could indicate a causal relationship of input of OM from terrestrial plants that had been transformed by bacteria. Metabolism and biosynthesis of the bacterial decomposers are expected to result in an increase in the  $\delta^{13}\text{C}$  of remaining organic matter (Nakatsuka et al. 1997). Provided an increase in the activities of bacteria, coupled with an increase in primary bacterial productivity in DC (Wang



**Fig. 6** Compound-specific  $\delta^{13}\text{C}$  of individual BSFA in the DC-4 and BST-1 sediment cores



**Fig. 7** Vertical variations of compound-specific  $\delta^{13}\text{C}$  values of individual BSFA in the DC-4 and BST-1 cores

et al. 2013), the magnitude of removal of  $^{12}\text{C}$  by bacteria was greater with enhanced degradation of terrestrial BSFAs, which would have resulted in the positive  $\delta^{13}\text{C}$  excursion observed in residual LCFAs in the sediments. Although the mechanism behind the correlation of nC16 and LCFAs compound-specific  $\delta^{13}\text{C}$  remains elusive, the ultimate result is that the observed trends of BSFA specific-compounds  $\delta^{13}\text{C}$  from different organisms varied towards more consistent with the intensified eutrophication. Strong bacterial/algal activities associated with long-term eutrophication seemed to cause over-enrichment of  $^{13}\text{C}$  during production and preservation of OM.

There was a correlation between compound-specific carbon isotope values of nC28, nC26, or nC24 and bulk isotope values in the BST-1 core, which demonstrates a linear relationship when cross-plotted ( $R^2=0.63$ ,  $n=10$ ) (Fig. 7). Alternation of bulk isotope values is characteristic of higher plant fraction in the core, likely due to the fact that LCFAs derived from terrestrial plants can survive in deeper samples. Selective preservation of terrestrial relative to aquatic organic carbon is a well-documented phenomenon in sedimentary processes (Zonneveld et al. 2010). In addition, there were significant correlations between isotope ratios of nC18, nC20, and nC22. BSFAs can be divided into three categories, nC16, nC18–nC22, and nC24–nC28, based on the different sources and vertical variation features (Fig. 7). The positive shift in  $\delta^{13}\text{C}$  of the nC16 BSFA in the upper section might be a signal related to a shift towards eutrophication in BST. This result also suggests that accumulation of algae has not yet had a large effect on the BST ecosystem.

### Conclusions

Paleo-limnological investigations of the two lakes examined patterns of BSFAs of sediments in the cores and investigated TOC contents, bulk carbon, and nitrogen isotopic compositions. Concentration variations of BSFAs in the sediment cores indicate that there has been a rapid increase in the supply of OM to DC, especially in the uppermost 10 cm, which corresponds to a time period since the 1950s. It is consistent with remarkable excursions to positive of  $\delta^{13}\text{C}_{\text{org}}$  and  $\delta^{15}\text{N}$  and greater contents of TOC. These observations suggest that there was an abrupt change in the lacustrine environment, indicating that the lake had entered a hypereutrophic stage. This organic geochemical evidence was also in good agreement with the results of the BSFA compositions. The sum of nC16 and nC18 was primarily composed up to 90 % of the total BSFAs in the surface sediment, and increased BSFA concentrations upward were simultaneous with the increasing trend of  $\text{nC14}+\text{nC16}+\text{nC18}/\text{nC24}+\text{nC26}+\text{nC28}$  ratios. Due to the long-term and continuous algal blooms in DC, observed trends of BSFA compound-specific  $\delta^{13}\text{C}$  from different organisms varied towards positive shift of  $\delta^{13}\text{C}$ . Strong bacterial/algal activities associated with eutrophication seemed to cause over-enrichment of  $^{13}\text{C}$  during production and preservation of OM. In comparison, there was no significant variation of TOC, TN, and total BSFA concentrations in the BST-1 core, suggesting that BST was in a period of stable lake environment. The positive shift of nC16 compound-specific  $\delta^{13}\text{C}$  in the upper section might be a signal. This signal seemed to be related to a shift towards eutrophication in BST. However, the accumulation of algae would not affect entire aquatic ecosystem in BST so far.



**Acknowledgments** This work was supported by the National Natural Science Foundation of China (41103082) and the Ministry of Science and Technology 973 early special (2012CB426506). Prof. Giesy was supported by the program of 2012 “High Level Foreign Experts” (no. GDW20123200120) funded by the State Administration of Foreign Experts Affairs, the People’s Republic of China to Nanjing University and the Einstein Professor Program of the Chinese Academy of Sciences. He was also supported by the Canada Research Chair program and a Visiting Distinguished Professorship in the Department of Biology and Chemistry and State Key Laboratory in Marine Pollution, City University of Hong Kong.

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