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Different responses of sedimentary $\delta^{15}N$ to climatic changes and anthropogenic impacts in lakes across the Eastern margin of the Tibetan Plateau

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ABSTRACT

Knowledge of historical variations in sedimentary $\delta^{15}N$ values and their relation to modern ones in lakes is critical in understanding N cycling in inland aquatic systems. Here we present late Holocene sedimentary $\delta^{15}N$ variations in four lakes across the Eastern margin of the Tibetan Plateau (ETP), namely Lake Qinghai and Lake Chaonaqiu over north ETP (N-ETP) area, and Lake Yihai and Lake Lugu over south ETP (S-ETP) area. The results show that: (1) during historical periods when anthropogenic forcing was weak, the centennial/multi-decadal changes in $\delta^{15}N$ values were dominated by climatic changes. Higher precipitation brought greater nutrients supply and led to higher Lacustrine Primary Productivity (LPP) and higher $\delta^{15}N$ values in lake sediments, and vice versa. (2) During the recent/modern epoch with enhanced anthropogenic impacts, sedimentary $\delta^{15}N$ values in Lake Qinghai and Lake Chaonaqiu (mid-high latitude lakes with low LPP) showed decreasing trends, possibly related to the increased global atmospheric reactive nitrogen (Nr) deposition. On the contrary, sedimentary $\delta^{15}N$ values in Lake Lugu and Lake Yihai (subtropical lakes with high LPP) showed increasing trends, most likely because more dissolved inorganic nitrogen in lake water was used during LPP processes. This study highlights that LPP plays an important role in modifying sedimentary $\delta^{15}N$ values both for historical and modern times, whereas atmospheric Nr deposition seems to influence the modern/recent sedimentary $\delta^{15}N$ trends in lakes with low LPP.

1. Introduction

The sedimentary $\delta^{15}N$ value serves as an excellent tool to investigate historical and modern N cycling in inland aquatic systems (e.g., Talbot, 2001). During the past several decades, increasing numbers of studies have focused on sedimentary $\delta^{15}N$ changes and their linkages to climatic changes, global atmospheric reactive nitrogen (Nr) deposition, and various human activities (e.g., Talbot, 2001; Vreca and Muri, 2010; Holtgrieve et al., 2011; Woodward et al., 2012; McLauchlan et al., 2013; Wu et al., 2013). For example, Wolfe et al. (2001) studied N cycling in alpine lakes along the Colorado Front Range, USA, and pointed out that both the rates and magnitudes of recent ecological changes far exceeded the context of natural variability, due to excess N derived from agricultural and industrial sources. Holtgrieve et al. (2011) studied $\delta^{15}N$ in sediments from 25 remote Northern Hemisphere lakes, and attributed the recent decreasing $\delta^{15}N$ trends to anthropogenic N deposition. Hu et al. (2014) studied $\delta^{15}N$ in sediments from two alpine lakes in the western Sichuan plateau, southwestern China, and the results suggest that modern atmospheric Nr deposition may be responsible for the $\delta^{15}N$ variations in those high elevated alpine lakes. Sedimentary $\delta^{15}N$ changes were documented at Lake Qinghai (Xu et al., 2006), and at Lake Lugu (Xu et al., 2014), China, and linked to decadal/multi-decadal variations in regional temperature during the past several hundred years for both lakes. Changes in sedimentary $\delta^{15}N$ values in Lake Balikun, Xinjiang province,

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northwestern China, have been suggested to be closely related to changes in precipitation (Zhong et al., 2013). Changes in sedimentary $\delta^{15}N$ values in Lake Bosten, Xinjiang province, northwestern China, were linked to the proportion of algal to total organic matter, providing insights on both precipitation dominated lake level changes and anthropogenic impacts (Zheng et al., 2012). Both the sedimentary total nitrogen contents and $\delta^{15}N$ values at Lake Ontario (USA, Hodell and Schelske, 1998) and Lake Greifensee (Switzerland) showed increasing trends during the past ~200 years, which reflects the global pattern of increasing nutrient loading due to urbanization and modern land-use practices (see Talbot, 2001 and references therein). The $\delta^{15}N$ values and C/N ratios in lake sediments at Lake Hollingsworth, Florida broadly decreased during the past 100–200 years (Brenner et al., 1999), and $\delta^{13}C$ values increased, suggesting a rise in productivity due to eutrophication and an increasing contribution of N-fixing cyanobacteria to the sediment (Talbot, 2001). These, among many other studies have largely improved our understanding of the causes of $\delta^{15}N$ variations in lake sediments.

However, it remains not fully clear to what extent anthropogenic impacts (such as Nr deposition) influence the N cycling in lake sediments, and how strong it is as compared with the influence from climatic changes. Moreover, because hydrology, climatic setting, water chemistry, and trophic status are diverse between different lakes, N-cycles are also variable between different lakes. It is necessary to study both historical and modern $\delta^{15}N$ changes and the corresponding relationship with other indices ($\delta^{13}C$, C/N ratios, etc.), and comprehensively understand N-cycling. In particular, the relationships between $\delta^{15}N$ and other indices during some key time periods, including the medieval period, the little ice age (LIA), and the last 100–200 years, are critical to understanding changes in lacustrine N-cycling under predominantly natural forcing versus predominantly anthropogenic impacts.

The Eastern margin of the Tibetan Plateau (ETP) is influenced by the Indian summer monsoon (ISM), East Asian summer monsoon (EASM), and the Westerlies during the summer season (Fig. 1). Climates from south to north across ETP areas vary from tropical humid to cool and dry, and are very sensitive to global climatic changes. Therefore, lakes located in ETP areas are ideal sites to study sedimentary $\delta^{15}N$ changes and the causes (e.g., Xu et al., 2006, 2014; Hu et al., 2014). Here we report late Holocene sedimentary $\delta^{15}N$ changes in four lakes across the ETP areas (Fig. 1; Table 1), to understand the key factors that dominate sedimentary $\delta^{15}N$ changes under different climatic conditions and different anthropogenic impacts.

2. Materials and methods

2.1. Lake Qinghai

Lake Qinghai is located in the N-ETP area (Fig. 1; Table 1). Mean temperature varies from −10.4 to −14.7 °C in January and from 10.4 to 15.2 °C in July. Mean annual precipitation is approximately 300–400 mm, and more than 80% of which falls between July and September (Xu et al., 2010). Sediment core (QH10A; 85 cm long) was collected from the southeastern basin of Lake Qinghai (Table 1), in 2010, and was sectioned at 1 cm intervals. An age model of core QH10A during the past ~1300 years was developed based on ^{137}Cs radioactivity (Xu et al., 2015a). Timeseries of proxy indices of core QH10A based on this age model correlated well with dendrochronology nearby, supporting the reliability of the age model (Xu et al., 2015a). Grain size, $\delta^{13}C$, total organic carbon content (TOC), and C/N ratios were previously measured, and their variations were ascribed to changes in precipitation (Xu et al., 2015a). Sedimentary $\delta^{15}N$ values of organic matter of core LQ0407-C from the same lake was studied previously, and its decadal variations (after removal of the stratigraphic trend) were linked to regional temperature variations (Xu et al., 2006). In this study, sediments from QH10A were pretreated with HCl (1 mol/L) and then rinsed repeatedly with distilled water to remove carbonates and instable nitrogen fraction (Xu et al., 2006, 2014). Sedimentary $\delta^{15}N$ values of stable organic matters were then determined (against air; Finnigan Delta Plus XP; error <0.1‰), and expressed in delta (δ) notation ($\delta^{15}N_{org}$).

2.2. Lake Lugu

Lake Lugu is located in the northwestern Yunnan-southwestern Sichuan provinces, S-ETP areas (Fig. 1; Table 1). Mean annual temperature is 12.4 °C, with mean temperatures of 5.2 °C and 17.5 °C for January and July respectively. Mean annual precipitation is 805 mm, with up to 80% falling between June and September (Sheng et al., 2015). Sediment cores were collected with a gravity corer, and were sectioned at 1 cm intervals (Table 1; Sheng et al., 2015). An age model was developed by combination of the ^{137}Cs

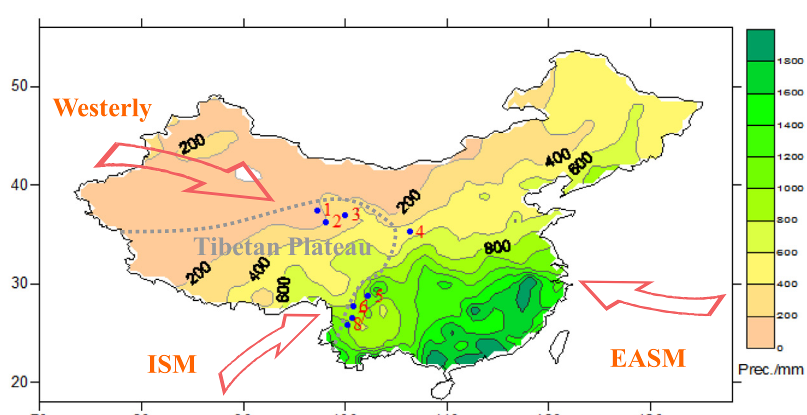

Fig. 1. Locations of the lakes across the ETP, and other comparison sites. 1 Delingha, 2 Dulan, 3 Lake Qinghai, 4 Lake Chaonaqiu, 5 Lake Yihai, 6 Lake Lugu, 7 Lake Chenghai, and 8 Lake Erhai. Color shades show the mean annual precipitation contours (developed from precipitation data during 1951–2007, China). Dotted gray line delineates the approximate boundary of the Tibetan Plateau. The Orange arrows show the broad direction of ISM, EASM, and the Westerlies. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 1
Parameters of the four lakes in this study.

Parameters		Lake Qinghai	Lake Chaonaqiu	Lake Yihai	Lake Lugu
Locations	Lat.: N Long.: E	36°38′1.545″ 100°36′23.843″	35°15′53.08″ 106°18′35.99″	28°43′49.03″ 102°14′01.59″	27°42′32″ 100°47′24″
Elevation (m)		3200	2430	2283	2694
Lake area (km^2)		4200	0.02	0.15	48.45
Catchment area (km^2)		29,660	0.2	1	171
Water depth of the sampling sites (m)		23	9[b]	12[b]	60[c]
pH		9.15–9.30[a]	7.83[b]	8.22[b]	7.7–8.6[d,e]
Salinity (g/L)		14.53[a]	0.17[b]	0.034[b]	~1.54[e]
TN in lake water (mg/L)		0.08[a]	\	\	~0.11[a]
TP in lake water (mg/L)		0.02[a]	\	\	0.04[a]

[a] Wang and Dou (1998).
[b] This study.
[c] Sheng et al. (2015).
[d] Chen et al. (2014).
[e] Wu et al. (2008).

ages, ^{210}Pb ages, and ^{14}C ages of plant debris (see Sheng et al. (2015) for details). Sedimentary grain size, TOC, and C/N ratios were determined and their variations were ascribed to changes in monsoon precipitation intensity at this lake (see Sheng et al. (2015) for details). Xu et al. (2014) documented variations in sedimentary δ^{15}N of core Lugu07-C (from the same lake) and linked the decadal variations (after removal of the stratigraphic trend) during the past several hundred years to temperature variations. In this study, sedimentary $\delta^{15}N_{org}$ of core LG12-1-3 were determined spanning the past ~3000 years.

2.3. Lake Yihai and Lake Chaonaqiu

Lake Yihai is located at western Sichuan, ~185 km northeastwards from Lake Lugu (Table 1; Fig. 1). Mean annual temperature is 17.1 °C, and the mean temperatures are 22.6 °C and 9.6 °C for July and January, respectively. Mean annual precipitation is ~1014 mm, with ~76% of which falling during June–September. Sediment cores were collected with a gravity corer and sedimentary $\delta^{15}N_{org}$, TOC, and C/N ratios (of core YH12-2-1) were determined at 2 cm interval.

Lake Chaonaqiu (~530 km from Lake Qinghai) is located at Liupan Mt. (Fig. 1; Table 1). Mean annual temperature is 3.4 °C; and mean annual precipitation is ~ 615 mm (Zhou et al., 2010). Sediment cores were collected with a gravity corer, and were sectioned at 1 cm interval. Sedimentary $\delta^{15}N_{org}$, TOC, and C/N ratios of core (CNQ-12-1) were determined.

3. Results

3.1. Sedimentary δ^{15}N, TOC, δ^{13}C, C/N, and grain size in Lake Qinghai

As shown in Fig. 2a, sedimentary δ^{15}N, TOC, C/N, and grain size in Lake Qinghai (core QH10A) showed similar long term trends. Sedimentary δ^{15}N values were relatively higher during ~750–1200 AD (the medieval period); and they were lower during ~1200–1800 AD (broadly corresponding to LIA). During the past 100–200 years, sedimentary δ^{15}N values show an accelerated decreasing trend (Fig. 2a and b). A noteworthy feature is that the relationship between TOC and δ^{15}N is different before and after ~1800 AD. TOC and δ^{15}N in core LQ0407-C were positively correlated before 1800 AD (Fig. 3a; r^2 = 0.40), and were negatively correlated after 1800 AD (Fig. 3b; r^2 = 0.60). Such a shift in this relationship can also be observed in core QH10A (Fig. 2a).

3.2. Sedimentary δ^{15}N, TOC, δ^{13}C, C/N, and grain size at Lake Lugu

The centennial/multi-decadal variations in δ^{15}N values are broadly synchronous with those in TOC, C/N ratios, and grain size at Lake Lugu during the past ~3000 years (Fig. 2c). During ~750–1350 AD, TOC, C/N ratios, mean grain size, and the average δ^{15}N value were all much lower as compared with those before and after. During 1350–1800 AD, sedimentary TOC, C/N ratio, and grain size all increased (Fig. 2c), and the average δ^{15}N value was higher during this period (Fig. 2c). Sedimentary δ^{15}N values show an increasing trend during the past ~ 200 years (Fig. 2c and d). A shift in the relationship between δ^{15}N and other indices can also be clearly seen at Lake Lugu, similar to those observed at Lake Qinghai. For example, δ^{15}N and C/N ratio values in core LG12-1-3 were positively correlated before 1500 AD (r^2 = 0.498; Fig. 3c); but the correlation after 1500 AD is negative as recorded in core Lugu07-C (r^2 = 0.653; Fig. 3d).

3.3. Sedimentary δ^{15}N and C/N in Lake Chaonaqiu and Lake Yihai

The δ^{15}N values in surface sediments of Lake Chaonaqiu showed a decreasing trend for the uppermost section; but total nitrogen contents increased (Fig. 4a). C/N ratios in surface sediments showed a long term decreasing trend at Lake Chaonaqiu (Fig. 4b). δ^{15}N values in sediments of Lake Yihai showed a clear increasing trend towards the present, but total nitrogen contents decreased (Fig. 4c). C/N ratios also decreased at Lake Yihai (Fig. 4d), and the δ^{15}N-C/N relationship in sediments at Lake Yihai is broadly similar to that in Lake Lugu.

4. Discussion

4.1. Climatic significance of the proxy indices

Generally, the content of organic matter indicates the biomass in both the lake and the catchment. Terrestrial plants and/or emerged plants are rich in fiber but poor in proteins and the atomic C/N ratios of organic matter are therefore high (generally greater than 20; Meyers, 2003). In contrast, lake algae and/or plankton contain less fiber but more proteins and hence have low atomic C/N ratios (generally less than 10; Meyers, 2003). So the atomic C/N ratios of organic matter have been widely used to indicate the relative contribution of authigenic and terrigenous organic matter. Higher C/N ratios correlate to larger proportions of

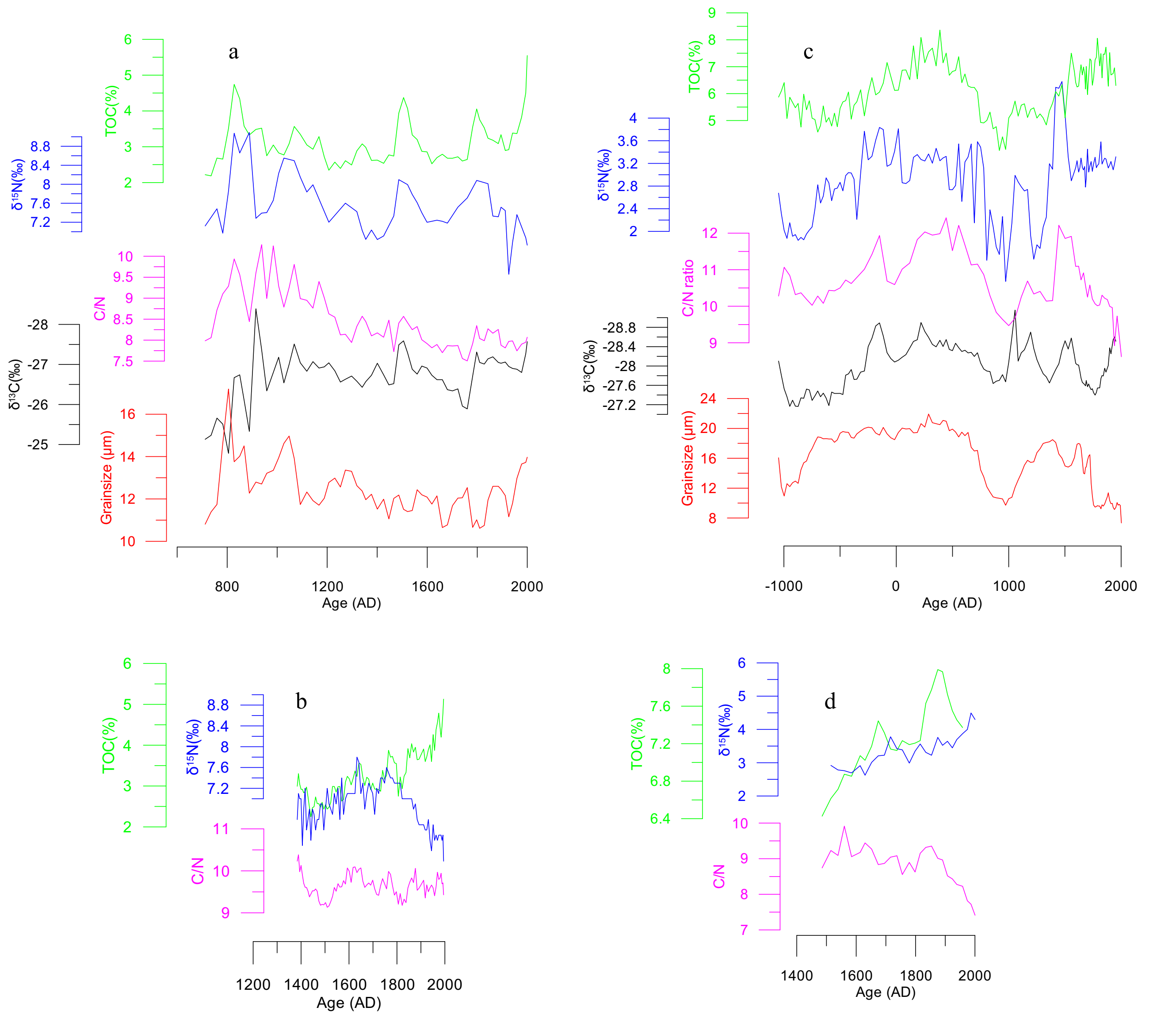

Fig. 2. Comparisons between sedimentary $\delta^{15}N$ and other indices from Lake Qinghai and Lake Lugu. a. $\delta^{15}N$ from core QH10A from Lake Qinghai over the past ~1300 years (this study). b. $\delta^{15}N$ from core QH0407-C Lake Qinghai during the past ~600 years (Xu et al., 2006). c. $\delta^{15}N$ from core LG12-1-3 from Lake Lugu during the past 3000 years (this study). d. $\delta^{15}N$ from core Lugu07-C from Lake Lugu during the past ~500 years (Xu et al., 2014).

terrigenous organic matter; and lower C/N ratios imply higher proportions of algal organic matter. Besides, the aquatic plants will partly use the dissolved inorganic carbon (DIC) during the photosynthesis, this usually leads to higher $\delta^{13}C_{org}$ of algal and/or plankton than that of C_3 plants in catchments (Leng and Marshall, 2004; Xu et al., 2014). Therefore, lower $\delta^{13}C$ of total organic matter indicates higher contribution of terrestrial organic matter; but higher $\delta^{13}C$ of total organic matter indicates higher contribution of algal organic matter (Xu et al., 2006). Variations in sedimentary grainsize on decadal/centennial scales are dominated by runoff intensities, which are closely related changes in precipitation (e.g., Xu et al., 2014, 2015a,b).

4.2. $\delta^{15}N$ in lake sediments

Generally, $\delta^{15}N$ values of atmospheric N_2 are close to 0‰ (Peters et al., 1978; Meyers, 1997, 2003). $\delta^{15}N$ values in precipitation range from −18‰ to 4‰, with average values generally <0‰ (Talbot, 2001). $\delta^{15}N$ values are ~7–10‰ for DIN, ~8‰ for plankton, and ~0.5‰ for terrestrial C_3 plants (Peters et al., 1978; Meyers, 1997, 2003). $\delta^{15}N$ values in man-made N-fertilizer are generally low, e.g., NH_3 and NO_x typically have $\delta^{15}N$ values of ~0 ± 3‰ (Wolfe et al., 2001). As a result, atmospherically deposited N is generally regarded as a $\delta^{15}N$-depleted end-member, and is widely used to interpret low $\delta^{15}N$ values (or decreased $\delta^{15}N$ trends) in lake sediments. For example, decreasing $\delta^{15}N$ trends in 25 remote high latitude lakes have been associated with increased Nr deposition during the modern epoch (Holtgrieve et al., 2011). $\delta^{15}N$ of nitrate from the Greenland ice sheet also showed significant decreases over the past 150 years (~15‰ depletion; Hastings et al., 2009).

The $\delta^{15}N$ fractionation in lakes can be influenced by many factors, like the concentration of dissolved nitrate, N_2-fixing processes, microbial nitrogen fixers, diagenetic processes, bacterial decomposition, and kinetic isotopic effect, etc (Hodell and Schelske, 1998; Xu et al., 2006; Chu et al., 2009). In general, N_2-fixing algae prefer to use ^{14}N to synthesize organic matter. As the $\delta^{15}N$-depleted fraction is selectively removed, the left N is relatively ^{15}N-enriched. When LPP increases greatly, part of the ^{15}N-enriched N in DIN pool may be used to synthesize organic matter, which would lead to an increase in $\delta^{15}N$ value in organic

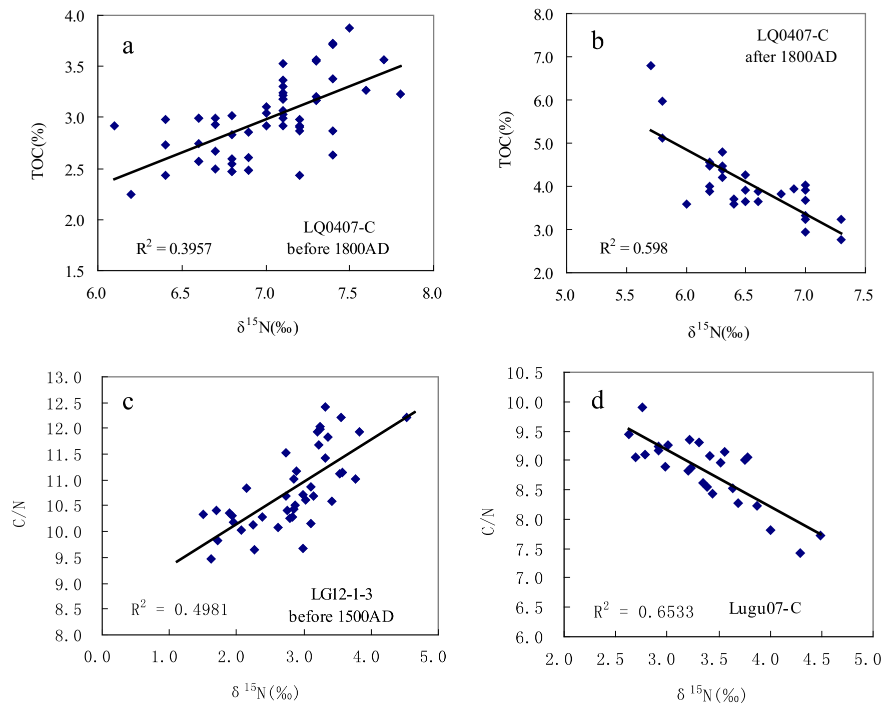

Fig. 3. (a and b) Show relationship between $\delta^{15}N$ and TOC at Lake Qinghai before and after ~1800 AD. (c and d) Show relationship between $\delta^{15}N$ and C/N ratios at Lake Lugu before and after ~1500 AD.

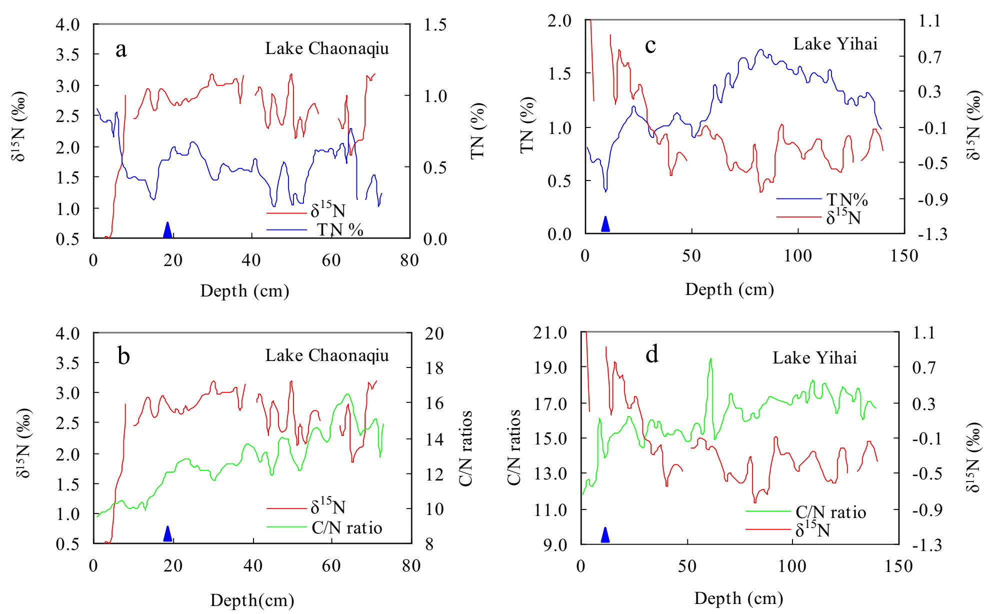

Fig. 4. $\delta^{15}N$, C/N, and total organic nitrogen contents in Lake Chaonaqiu (a and b) and Lake Yihai (c and d). Blue triangles show the depth of the ^{137}Cs peaks (corresponding to 1964 AD) for core CNQ-12-1 (18 cm) and core YH12-2-1 (10 cm). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

matters produced within lakes, and eventually result in higher $\delta^{15}N$ values in lake sediments (e.g., Hodell and Schelske, 1998; Xu et al., 2006, 2014). Such an effect has also been widely used to interpret high organic matter $\delta^{13}C$ values in lake sediments due to partial utilization of the dissolved inorganic carbon (DIC) during LPP processes (Meyers, 1997; Leng and Marshall, 2004; Xu et al., 2006). Therefore, increases in nutrient supply and temperature could lead to increased LPP and higher sedimentary $\delta^{15}N$ values, and vice versa (e.g., Xu et al., 2006, 2014). It is interesting to note that similar effects have also been observed in the $\delta^{15}N$ variations in tropical trees. For example, $\delta^{15}N$ in tree rings from Thailand showed increasing trends during the past ~100 years (Hietz et al., 2011). Even though the N-fertilizer $\delta^{15}N$ values are lower than those of the tree leaves; such an increasing $\delta^{15}N$ trend can still be observed (Hietz et al., 2011). Therefore, the long term $\delta^{15}N$ trends in tropical tree rings seem to be independent of $\delta^{15}N$

values in N-fertilizer, but should be ascribed to the increased biomass due to use of fertilizer (Hietz et al., 2011).

Changes in lake levels influence the lake areas and water depth of the thermocline/chemocline, and could influence the limnological/geochemical processes, such as nutrients supply from the lake peripheral areas, changes in biomass and/or biological community within the lake aquatic system, overturning/mixing of the lake water and the related geochemical processes, such as denitrification (e.g., Talbot, 2001; Chu et al., 2009). These influences on sedimentary $\delta^{15}N$ trends seem to work better on long term timescales, like millennial and/or glacial-interglacial timescales (e.g., Talbot, 2001). For example, at Lake Bosumtwi, Ghana, the sedimentary $\delta^{15}N$ values ranged between ~0‰ and 15‰ during the early-mid Holocene (Talbot and Johannessen, 1992), which could be closely associated to the lake water stratification and the related changes in algal/phytoplankton community in the lake (see details and more examples in Talbot, 2001). However, on short term time scales, although changes in lake levels may influence the $\delta^{15}N$ values in N-species in lake water, the influences on sedimentary $\delta^{15}N$ trends are likely to be limited.

4.3. Historical variations in sedimentary $\delta^{15}N$ and climates: LPP dominating

The similar trends between $\delta^{15}N$, TOC, C/N ratios, and grain size during the historical period (before 1800 AD for Lake Qinghai, and before 1500 AD for Lake Lugu; Fig. 2) suggested that the historical variations in $\delta^{15}N$ values in lake sediments are closely related to climatic changes across ETP areas.

4.3.1. Lake Qinghai

Our previous work showed that higher precipitation led to higher TOC and C/N ratios in lake sediments both for Lake Qinghai (Xu et al., 2006) and Lake Lugu (Xu et al., 2014). Higher precipitation also brought coarser particles and resulted in larger sedimentary grain size for both lakes (Xu et al., 2014, 2015a). During ~750–1200 AD, higher average values of TOC, C/N ratio, grain size, together with the relatively lower $\delta^{13}C_{org}$ values, indicate a warmer and wetter climatic condition at Lake Qinghai, corresponding to a medieval warm-wet climatic optimum (Xu et al., 2015a). Lines of evidence suggested that the N-ETP areas were warm and wet during this period (Zhang et al., 2003; Liu et al., 2009; Xu et al., 2015a). The increased precipitation led to higher nutrient loads to the lake; and higher temperatures also resulted in higher LPP, which would increase the utility of DIN, and eventually lead to higher $\delta^{15}N$ values in lake sediments.

During 1200–1800 AD, mean sediment grain size at Lake Qinghai was smaller, TOC and C/N ratios were much lower, and $\delta^{13}C_{org}$ was higher, suggesting weakened precipitation and decreased influx of terrestrial materials (Xu et al., 2015a), corresponding to a cold-dry LIA climatic condition. Such a cold-dry climatic pattern is also consistent with data extracted from tree rings nearby, e.g., Dulan (Zhang et al., 2003) and Delingha (Shao et al., 2005; see locations in Fig. 1). Because of the cold LIA climatic conditions and reduced nutrient supply, LPP was weak, which eventually led to lower $\delta^{15}N$ values in lake sediment as compared with those during the medieval period.

4.3.2. Lake Lugu

During ~750–1350 AD, TOC, C/N ratios, and mean grain size in Lake Lugu were all much lower as compared with those before and after, suggesting a dry hydroclimatic condition during the medieval period. Similar hydroclimatic patterns also existed at Lake Chenghai and Lake Erhai (Fig. 1) as inferred from lake level changes, pollen records, as well as other multiple geochemical indices (Xu et al., 2015a,b). Such a medieval dry climatic condition occurred not only over S-ETP areas, but also over most ISM areas (e.g., Xu et al., 2015a,b; Sheng et al., 2015), and it is out-of-phase/anti-phase with those over N-ETP areas (e.g., Xu et al., 2015a,b; Sheng et al., 2015). Decreased precipitation during the medieval period led to decreased nutrient supply, and decreased lacustrine biomass at Lake Lugu, which eventually resulted in low $\delta^{15}N$ values in lake sediment. Sedimentary TOC, C/N ratio, and grain size all increased during 1350–1800 AD (Fig. 2c), indicating increased precipitation during the LIA, which is also contrast with the dry climatic condition at Lake Qinghai. The increased precipitation at Lake Lugu during the LIA led to higher nutrient supply and higher LPP, which eventually led to higher $\delta^{15}N$ values in lake sediment (Fig. 2c).

Both Lake Lugu and Lake Qinghai are thermal stratified during warm seasons (Wang and Dou, 1998). Lake Lugu is a hydrologically semi-closed deep lake, and the lake levels could keep relatively steady during the late Holocene (Sheng et al., 2015). Changes in lake levels of Lake Qinghai during the past millennium were also expected to be relatively small (Liu et al., 2014). This means that changes in lake levels could have minor influence on the sedimentary $\delta^{15}N$ trends both in Lake Qinghai and Lake Lugu on the timescales focused in this study. Taken together, the centennial/multidecadal variation in sedimentary $\delta^{15}N$ at both Lake Qinghai and Lake Lugu were dominated by the contemporaneous hydroclimatic changes during historical periods (when human influences were weak).

4.4. Recent/modern $\delta^{15}N$ variations: natural influence and anthropogenic impacts

Sedimentary TOC, C/N ratios, and grain size data from Lake Qinghai indicate a warmer and wetter climatic pattern during the last ~200 years (as compared with that during the LIA). This climatic pattern is very similar to that during the medieval period, and is supported by lines of evidence from N-ETP areas (e.g., Shao et al., 2005; Liu et al., 2009; Xu et al., 2015a). However, sedimentary $\delta^{15}N$ values show a decreasing trend during the past ~200 years (Fig. 2a and b), which is clearly in contrast with those during the medieval period. If the sedimentary $\delta^{15}N$ response during the past ~200 years was similar to those before (e.g., during the medieval period and LIA), then $\delta^{15}N$ values should be expected to increase rather than decrease during the past ~200 years. Therefore, a shift in the $\delta^{15}N$ response pattern could have occurred during the modern/recent epoch. Because human activities during the past ~200 years around Lake Qinghai have been relatively weak, and the LPP in Lake Qinghai has been low due to the cold water and oligotrophic setting, we argue that the decreasing $\delta^{15}N$ trend in lake sediment here should be ascribed to globally increased Nr deposition (generally with lower $\delta^{15}N$ values). Changes in the relationship between $\delta^{15}N$ and TOC (Fig. 3a and b) support such an inference. This also suggests that the impacts of Nr deposition on sedimentary $\delta^{15}N$ values are considerably strong for lakes with low LPP (because they can trigger a shift of the sedimentary $\delta^{15}N$ response pattern).

Lake Chaonaqiu is an alpine lake with weak human activities during the historical period. The $\delta^{15}N$ values showed a decreasing trend in the upper 40 cm of the core (corresponding to the past about one century; unpublished results), which is also likely related to enhanced global atmospheric Nr deposition, similar to that at Lake Qinghai. However, the $\delta^{15}N$ values of the uppermost 10 cm (corresponding to the past about two decades) decreased much more sharply, which may be related to the increasing recent human activities within the watersheds, e.g., road construction, touring development, and other agriculture activities.

Mean grain size at Lake Lugu varied at the lowest sizes during the past ~200 years, indicating weakened monsoon precipitation, which is similar to the precipitation trends over S-ETP areas (Sheng et al., 2015). The influx of terrestrial organic matter could have decreased under an arid climatic condition. However, TOC in lake sediments of Lake Lugu increased slightly, but C/N ratios decreased clearly during the past ~200 years, suggesting that the lacustrine biomass increased under an arid climatic condition during this time, which is in contrast with the synchronicity between precipitation and lacustrine biomass during the medieval period and LIA (Fig. 2). We attribute this shift to increased nutrient supply due to enhanced human activities during the past several hundred years. C/N ratio values decreased during the past ~150 years, possibly indicating enhanced anthropogenic impacts after the Industrial Revolution. As LPP increased sharply, more DIN was used to synthesize organic matter, resulting in increased sedimentary $\delta^{15}N$ values (Fig. 2c and d). A similar $\delta^{15}N$ trend can also be observed at Lake Yihai nearby (Fig. 4). If Nr deposition determines the sedimentary $\delta^{15}N$ trends in Lake Lugu and Lake Yihai, then the sedimentary $\delta^{15}N$ trends would be decreasing (like Lake Qinghai and Lake Chaonaqiu) rather than increasing, suggesting that the influence of LPP on sedimentary $\delta^{15}N$ clearly exceeds that of contemporaneous atmospheric Nr deposition. The lowest sedimentary $\delta^{15}N_{org}$ values in Lake Lugu over the late Holocene occurred during the medieval period (Fig. 2), but not during the modern epoch, suggesting again that LPP most likely dominated the sedimentary $\delta^{15}N$ changes in lakes with relatively high LPP.

McLauchlan et al. (2013) compiled and analyzed sedimentary $\delta^{15}N$ changes in 86 lakes globally, and the results showed that $\delta^{15}N$ values in tropical/sub-tropical lakes broadly increased during the past 500 years, but those in high-latitude lakes generally decreased. The sedimentary $\delta^{15}N$ response patterns during the past several hundred years across ETP areas are broadly consistent with the results of McLauchlan et al. (2013). However, the results of this study further suggest that although global atmospheric Nr deposition increased during the modern epoch, changes in $\delta^{15}N_{org}$ values in lake sediments across ETP areas cannot be fully ascribed to Nr deposition, but are also associated with changes in LPP. For lakes with low LPP (e.g., high-latitude lakes, remote high-elevated alpine lakes), changes in $\delta^{15}N$ values in lake sediments are closely related to atmospheric Nr deposition. However, for lakes with high LPP (e.g., tropical/sub-tropical lakes, and/or lakes with strong anthropogenic impacts), changes in sedimentary $\delta^{15}N$ are more likely controlled by changes in LPP.

5. Conclusions

Sedimentary $\delta^{15}N$ variations and the related causes were investigated in four lakes across ETP areas. During historical periods with weak anthropogenic forcing, the centennial/multi-decadal changes in sedimentary $\delta^{15}N$ values were dominated by hydroclimatic changes. Higher precipitation brought more nutrients, which led to higher LPP and higher $\delta^{15}N$ values in lake sediment due to the utilization of DIN during LPP processes. On the other hand, lower precipitation led to lower sedimentary $\delta^{15}N$ values. In this sense, the long term sedimentary $\delta^{15}N$ values can also be used as an indicator of long term changes in precipitation. During the recent/modern epoch with enhanced anthropogenic impacts, $\delta^{15}N$ values in lakes with low biomass (e.g., high-latitude/high-elevation lakes) showed decreasing trends due to atmospheric Nr deposition; but $\delta^{15}N$ values in lakes with higher biomass (e.g., tropical/subtropical lakes with high temperatures and great nutrient supply) showed increasing trends due to increasing utilization of DIN. We contend that LPP plays a most important role in controlling sedimentary $\delta^{15}N$ values during historical periods; and that the atmospheric Nr deposition seems to modify the modern/recent sedimentary $\delta^{15}N$ trends in lakes with low LPP.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.jseaes.2016.03.024.

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