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ORIGINAL ARTICLE

Li/Ca ratios of ostracod shells at Lake Qinghai, NE Tibetan Plateau, China: a potential temperature indicator

Zheng-jie Zhu · Jing-an Chen · Da-hua Li · Shi-cong Ren · Fang Liu

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Abstract Acquiring continuous and high-resolution natural records in recent 2,000 years is the hot issue in the palaeoclimate research. Recent studies revealed that Li/Ca and Mg/Li ratio of carbonate is a potential tracer of past environmental changes in the oceans, and is seldom applied for palaeoclimate reconstruction in lakes. To make full understanding of the potential of Li/Ca ratios of carbonates in lakes, Li/Ca ratios of monospecific ostracod shell *Eucypris inflate* with similar size in the lacustrine sediment core from Lake Qinghai, NE Tibetan Plateau have been analysed for the first time. Single species can effectively avoid the interspecies effects. In combination with the sedimentation rate that existed, data derived from ^{210}Pb and ^{137}Cs, downcore variations of Li/Ca ratios of ostracod shells during the past 800 years in Lake Qinghai have been reconstructed successfully. By comparing Li/Ca ratios and temperature inferred from meteorological records and tree ring widths in Dulan and Qilianshan in adjacent regions, Li/Ca ratios of ostracod shells negatively correlate with temperature. Higher temperature corresponds with lower Li/Ca ratios, and vice versa, indicating that Li/Ca ratio of ostracod shells is an effective indicator for temperature variations. Therefore, ongoing in-depth investigation using Li/Ca ratios of carbonates in more lakes to further reveal its palaeotemperature implications would be deserved.

Keywords Lake Qinghai · *Eucypris inflate* · Li/Ca ratios · Temperature proxy

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Introduction

Over the past three decades, stable carbon and oxygen isotopes as well as trace elemental ratios of ostracod shells from lake sediments have been extensively developed as proxies for the reconstruction of past climate and environmental conditions (Fritz et al. 1975; Chivas et al. 1985, 1986; Lister et al. 1991; Holmes 1996; Holmes et al. 2005; Von Grafenstein et al. 1999; Henderson et al. 2003; Leng and Marshall 2004; Zhang et al. 2004; Li et al. 2007; Liu et al. 2007, 2008a; Hu et al. 2008). There are two advantages to take the ostracod shells as analytical material in paleoenvironmental reconstruction. Firstly, ostracod shells can be selected and identified as pure authigenic carbonate, thus effectively avoiding contamination by detrital carbonates compared with bulk carbonates. Secondly, the composition of ostracod shells is very sensitive to environmental changes. Moreover, the composition of ostracod shells is difficult to change once they are formed (Von Grafenstein et al. 1999; Leng and Marshall 2004; Liu et al. 2008a, b). Most previous studies about trace elemental chemistry have exclusively centred on the magnesium

(Mg) and strontium (Sr). For example, the study by Chivas et al. (1985) suggested that Sr uptake by ostracod shells is completely determined by the salinity and is independent of temperature. Otherwise, Mg uptake is controlled both by salinity and temperature, and it is impossible to separate their relative contributions (e.g. Chivas et al. 1986; Hu et al. 2008). Recently, Li/Ca ratio of carbonate is a relatively new branch of environmental change research in ocean, and such studies have confirmed that Li/Ca ratio of carbonate is a potential tracer of past environmental changes about temperature (e.g. Hall and Chan 2004; Marriott et al. 2004a, b; Montagna et al. 2006; Bryan and Marchitto 2008). According to the analysis of a series of laboratory-grown carbonates and natural coral samples, for example, Marriott et al. (2004a) found that the Li/Ca ratios showed a strong negative correlation with temperature in both inorganic calcite and coral aragonite. Due to its inverse relationship, the temperature effect is of particular importance because it may be a potential palaeotemperature proxy. However, relative to marine system, until now, investigation on the Li/Ca ratios of carbonates in lacustrine sediments is still limited.

Lake Qinghai, located on the high-altitude northeastern Tibetan Plateau, and on the junctional zone of the East Asian summer monsoon (EASM), Indian summer monsoon (ISM), East Asian winter monsoon and the westerly jet stream (Fig. 1a), is the largest inland saline lake in China. Because of its unique geographical location, Lake Qinghai is very sensitive to global climate changes, and has attracted international attention during the past two decades, particularly on monsoonal variations (e.g. Lister et al. 1991; Zhang et al. 1994; Yu and Kelts 2002; Henderson et al. 2003, 2009, 2010; Zhang et al. 2003, 2004; Ji et al. 2005; Shen et al. 2005; Xu et al. 2006a, b, 2007; Liu et al. 2007, 2008b, 2009). Stable isotopes of carbonates (including bulk carbonate and ostracod shells) and organic matter, elemental ratios of ostracod shells as well as biomarkers have been investigated in order to reconstruct palaeoclimate in Lake Qinghai. For instance, Lister et al. (1991) have discussed effective humidity (evaporation/precipitation ratio) at Lake Qinghai in terms of $\delta^{18}O$ values of biogenic and bulk carbonates. Ji et al. (2005) observed the monsoon variations during the past 18,000 years on the basis of the varying redness of lake sediments. Xu et al. (2006a, b) revaluated in detail the climatic significance of multi-proxy indices, and observed that the oxygen isotopic composition of bulk carbonate can be used as a proxy for temperature. However, recent studies by Henderson et al. (2010), who believed that both effective moisture and temperature should be taken into account to explain oxygen isotopic composition of bulk carbonate, inconsistent with the commonly accepted viewpoint that the $\delta^{18}O$ of bulk or biogenic carbonates in Lake Qinghai was mainly controlled by effective moisture (Lister et al. 1991; Liu et al. 2007, 2009; Yu and Kelts 2002; Henderson et al. 2003; Shen et al. 2005). Moreover, distribution of C_{37} was normally used as temperature indicator in some lakes, but Liu et al. (2008b) suggested that it may be a potential salinity indicator in Lake Qinghai. It is well known that temperature reconstruction is generally based on the oxygen isotopic composition of carbonate, and the so-called palaeotemperature equations have been established by some researchers (Urey 1947; Epstein et al. 1953; Craig 1965; Leng and Marshall 2004). However, it is impossible to separate the effects of oxygen isotopic composition of lake water and temperature, especially in the monsoon dominated regions, where there is an inverse relationship between the amount and $\delta^{18}O$ value of precipitation (Dansgaard 1964; Zheng et al. 1983; Wei and Gasse 1999). As a consequence, to the best of our knowledge, until now, rare studies have been conducted to discuss the independent temperature indicator at Lake Qinghai. Additionally, abundant ostracod shells were well preserved in the sediments of Lake Qinghai, which also makes the Lake Qinghai as an ideal setting to use geochemistry of ostracod shells to deduce environmental changes.

In this study, with the aim to explore the potential of Li/Ca ratio of carbonate as an environmental indicator in lacustrine sediments, downcore variations of Li/Ca ratios in ostracod shells in the monospecies *Eucypris inflata* with similar size at Lake Qinghai were investigated for the first time, and Mg/Li ratios were also measured for testification. The results revealed that Li/Ca ratios of carbonate may be a promising temperature proxy in lacustrine sediments.

Study area, sampling and methods

Lake Qinghai (36°19′–37°15′N, 99°36′–100°47′E, 3,193 m above sea level), located in the northeastern Tibetan Plateau, China (Fig. 1a), is situated in a semi-arid temperate zone, and now is hydrologically closed, and also is one of the most sensitive regions to global climate changes. Lake Qinghai covers an area of 4,340 km^2, and the catchment area is about 29,660 km^2. The volume of water in the lake is about 778×10^8 m^3 (Wang and Dou 1998). The maximum depth of water is 27 m, with an average depth of 21 m. The mean temperature variations in the Lake Qinghai region range between 10.4 and 15.2°C in July while the lake water is relatively cold, with an average temperature of about 10°C at the surface and 4°C at the bottom. The lake is frozen between December and March. The mean annual precipitation at Lake Qinghai is about 300–400 mm (Wang and Dou 1998), and more than 80% of the total annual precipitation occurs between May and September.

The sediment core, QH2, was retrieved from Lake Qinghai in July 2008 near the centre of the lake with a water depth of approximately 20 m using a self-designed gravitational sediment sampler (Fig. 1b) and a polymethyl tube with a diameter of 59 mm. The sediment core was well preserved as indicated by the clear sediment–water interface. The core with a length of 43 cm was sectioned at 0.5 cm interval for above 40 and 1 cm interval for the remaining bottom sediments in situ, representing the resolution about 10 years.

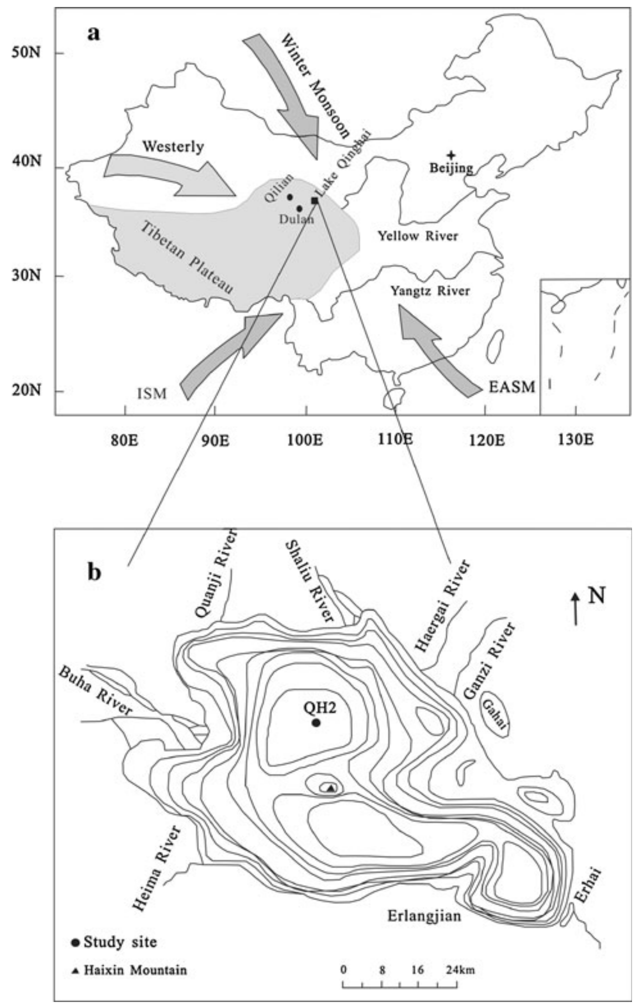


Fig. 1 Location of Lake Qinghai (**a**) and the sampling site (**b**). ISM and EASM denote the Indian summer monsoon and the East Asian summer monsoon, respectively (Xu et al. 2006a, b)

There are only two species of ostracod identified in the sediments of Lake Qinghai including *Limnocythere inopinata* and *Eucypris inflata*, based on this investigation and other studies (Zhang et al. 1994, 2004, 2006; Henderson et al. 2003; Li et al. 2007). Due to the ubiquity of *Eucypris inflata* in the sediment core QH2 and to avoid interspecies effects, this monospecies with similar size has been chosen for the measurement of elemental ratios and the microscopic pictures of *Eucypris inflata* are shown in Fig. 2. In this study, 55 samples have the abundant valves to satisfy the analysis Li/Ca and Mg/Ca ratios. Each sediment sample was wet-sieved by a 63 μm sieve. Clean ostracod shells were selected under a binocular microscope using a fine-hair brush. Before analysis, 20 pieces of the selected ostracod samples were ultrasonically cleaned with distilled water for four times and soaked with ethanol for 12 h to remove detrital grains and clay particles, leached with 0.001 M HNO_3, and heated in hydrazine/ammonium citrate and $NaOH/H_2O_2$ mixed solution (Hall and Chan 2004; Marriott et al. 2004a). After dissolution in 2 mL of 0.1 M HNO_3 acid, the solution was analysed for Li/Ca and Mg/Li ratios by an Inductively Coupled Plasma Mass Spectrometer (ICP-MS) with a standard of known concentration at the State Key Laboratory of Ore Deposit Geochemistry in Guiyang, the capital city of Guizhou Province, southwest China. The analytical precision is better than 2%.

Results and discussions

Li/Ca and Mg/Ca ratios of ostracod shells in Lake Qinghai

Li/Ca and Mg/Li ratios of ostracod shells *Eucypris inflata* of core QH2 in Lake Qinghai are plotted in Fig. 3 and

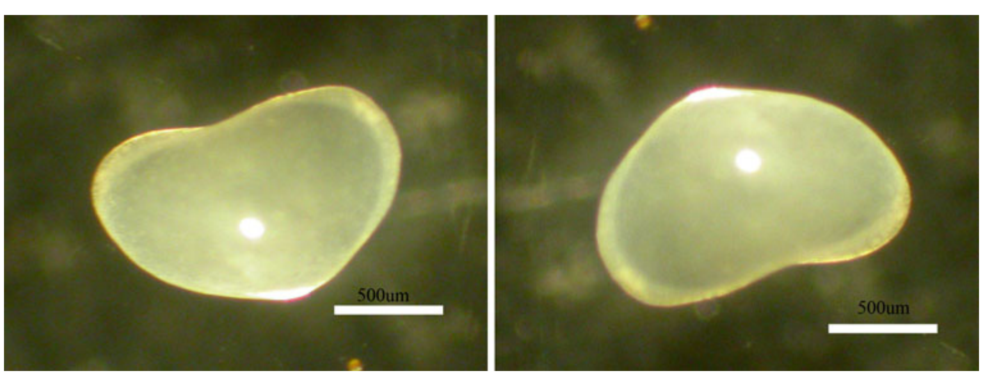


Fig. 2 Microscopic map of two *Eucypris inflate* shells in Lake Qinghai sediments

Fig. 3 Li/Ca and Mg/Li ratios of *Eucypris inflate* of core QH2 in Lake Qinghai

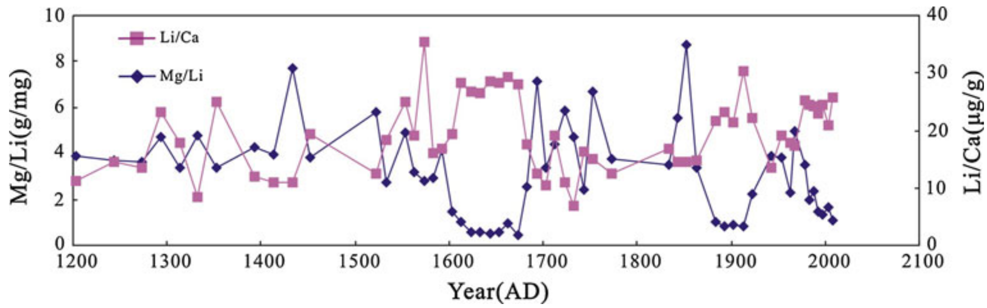


Table 1 Li/Ca and Mg/Li ratios of ostracod shells from core QH2 in Lake Qinghai

Sample no.	Year	Li/Ca (μg/g)	Mg/Li (g/mg)	Sample no.	Year	Li/Ca (μg/g)	Mg/Li (g/mg)
QH2-1	2008	25.84	1.07	QH2-36	1683	17.61	2.57
QH2-2	2003	20.99	1.64	QH2-37	1673	27.99	0.45
QH2-3	1998	24.49	1.32	QH2-38	1663	29.26	0.97
QH2-4	1993	22.89	1.45	QH2-39	1653	28.30	0.56
QH2-5	1988	24.25	2.37	QH2-40	1643	28.59	0.51
QH2-6	1983	24.51	1.99	QH2-41	1633	26.61	0.58
QH2-7	1978	25.34	3.48	QH2-42	1623	26.80	0.59
QH2-9	1968	17.25	4.96	QH2-43	1613	28.35	1.00
QH2-10	1963	17.78	2.30	QH2-44	1603	19.36	1.46
QH2-11	1953	19.01	3.81	QH2-45	1593	16.86	4.12
QH2-12	1943	13.49	3.90	QH2-46	1583	16.13	2.96
QH2-14	1923	22.29	2.24	QH2-47	1573	35.49	2.78
QH2-15	1913	30.34	0.86	QH2-48	1563	19.02	3.21
QH2-16	1903	21.43	0.86	QH2-49	1553	24.99	4.93
QH2-17	1893	23.29	0.82	QH2-51	1533	18.39	2.73
QH2-18	1883	21.61	0.99	QH2-52	1523	12.41	5.81
QH2-20	1863	14.70	3.40	QH2-57	1453	19.40	3.83
QH2-21	1853	14.45	8.71	QH2-59	1433	10.89	7.73
QH2-22	1843	14.43	5.56	QH2-61	1413	10.89	3.93
QH2-23	1833	16.92	3.49	QH2-63	1393	12.06	4.27
QH2-27	1773	12.43	3.76	QH2-67	1353	24.86	3.35
QH2-29	1753	14.99	6.67	QH2-69	1333	8.33	4.80
QH2-30	1743	16.42	2.43	QH2-71	1313	17.88	3.37
QH2-31	1733	6.89	4.68	QH2-73	1293	23.16	4.70
QH2–32	1723	10.85	5.87	QH2-75	1273	13.50	3.62
QH2-33	1713	19.03	4.40	QH2–77	1243	14.46	3.66
QH2-34	1703	10.41	3.36	QH2-81	1203	11.19	3.90
QH2-35	1693	12.52	7.16				

presented in Table 1. Results show that Li/Ca ratios vary between 6.89 and 29.26 μg/g, with an average value of 18.34 μg/g (Fig. 3; Table 1). Mg/Li ratios vary between 0.45 and 8.71 g/mg, with an average value of 3.23 g/mg. Li/Ca ratios in *Eucypris inflate* is much higher than previous reported values in Brachiopods (Delaney et al. 1989; Bryan and Marchitto 2008), experimental inorganic calcite (Marriott et al. 2004a), coral (Marriott et al. 2004a), and benthic forams (Marriott et al. 2004b). Solution or lake water Li/Ca ratios and interspecies effects may be responsible for these differences. Li/Ca ratio of Lake Qinghai water is 31,786 μg/g and the distribution coefficient ($Li/Ca_{carb}/Li/Ca_{water}$) is 0.0006, much lower than the published values shown in Fig. 4.

Possible implications of Li/Ca ratios of ostracod shell in lake sediments

Lithium (Li) is a common minor element in lake and ocean, and is incorporated into calcite crystals with carbonate

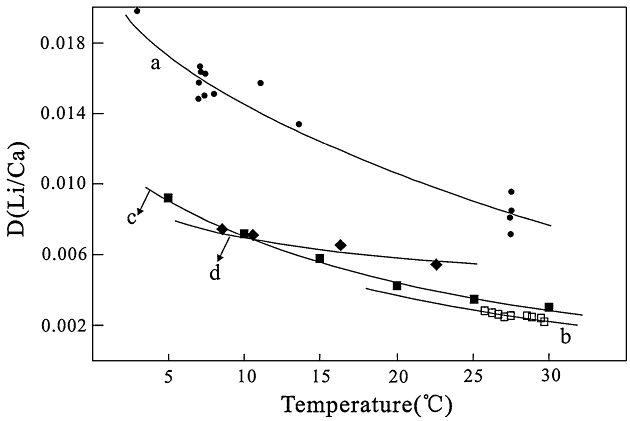


Fig. 4 The temperature dependence of Li/Ca incorporation into various species carbonates (redrawn from Marriott et al. 2004a). D(Li/Ca) represents Li/Ca distribution coefficients of carbonate precipitated in solution or water. *a* Brachiopods (Delaney et al. 1989), *b* Inorganic calcite (Marriott et al. 2004a), *c* Coral (Marriott et al. 2004a), *d* Benthic forams (Marriott et al. 2004b)

precipitation. Previous investigation has demonstrated that Li is preferentially incorporated into the 0001 face in calcite (Titiloye et al. 1993). Compared with other faces, the 0001 face in calcite is exothermic, and is likely to be favoured at lower temperatures (Parker et al. 1993; Marriott et al. 2004a). Therefore, during the processes of carbonate precipitation, Li is easily incorporated into calcite crystals at lower temperature, and Li/Ca ratios of carbonate are potentially capable of providing reliable palaeotemperature records.

Delaney et al. (1985) first cultured foraminifera to study their Li/Ca ratios, and the relationship between Li/Ca ratios of foraminifera and the seawater was noticed. However, in their subsequent study, they suggested that the lithium input to the ocean has been unchanged during the past 116 Ma (Delaney and Boyle 1986). In order to develop these pioneering works, several studies have investigated Li/Ca ratios in a line of experimental and natural carbonates. Possible factors can influence the Li/Ca ratios of carbonates to certain extent other than temperature, such as growth rate, solubility, salinity, interspecies effect.

Monospecies *Eucypris inflate* ostracod shells with similar size selected for the measurement of Li/Ca and Mg/Li ratios can availably avoid the effects of growth rate and interspecies. Additionally, on the basis of the previous research by Marriott et al. (2004a), it is obvious that solubility is not the main control on Li/Ca ratios of carbonate, since the solubility of Li_2CO_3 decreases with higher temperature (Dean 1992). Consequently, salinity and temperature may affect Li/Ca ratios of ostracod shells at Lake Qinghai to certain degree. On the one hand, lithium and calcium are both conservative elements, and previous studies proposed that they have long residence time in the ocean and large lakes (Broecker and Peng 1982; Huh et al. 1998). The long residence time results in homogeneous distribution of Li and Ca in the oceans or lakes, which would keep the Li/Ca ratios of water constant on centennial to millennial scales. Numerous works have revealed that the residence time of Li and Ca are 2.5 Ma and 1 Ma in the ocean, respectively (Broecker and Peng 1982; Huh et al. 1998; Hall and Chan 2004). Relative to studies of oceans, Tomascak et al. (2003) have calculated the residence time of Li in Lake Mono, which has a surface area of 200 km^2 and a volume of 27 × 10^8 m^3, and the result is 28 ka. Compared with Lake Mono, Lake Qinghai has a larger surface area (4,340 km^2) and volume (778 × 10^8 m^3) (Wang and Dou 1998), potentially indicating that Li and Ca in Lake Qinghai have longer residence times than in Lake Mono. Consequently, due to the long residence time of lithium in Lake Qinghai, although this was not understood directly, Li/Ca ratios would be unchanged during the past 800 years, which further implied that Li/Ca ratios of ostracod shells are not controlled by Li/Ca ratios of lake water, thereby salinity.

On the other hand, studies on a wealth of species of carbonates have been performed to study the sensitivity between Li/Ca ratios and temperature (Delaney et al. 1989; Marriott et al. 2004a, b). As seen in Fig. 4, despite the variations of Li/Ca ratio of carbonate with temperature is not statistically significant in various species carbonate (including brachiopods, inorganic calcite, coral and benthic forams), it is striking that Li/Ca ratios of all species of carbonates are inversely correlated with temperature, with higher Li/Ca ratios at lower temperature. In order to further assess the controls on Li/Ca ratios of carbonate, Marriott et al. (2004a) have investigated a series of laboratory-grown carbonates. The results indicated that Li/Ca ratios of carbonate are inversely correlated with temperature, and the correlation coefficient is −0.98 (Marriott et al. 2004a). Meanwhile, Marriott et al. (2004b) have studied the single species foraminifera in the Arabian Sea, and they also revealed that Li/Ca ratios of foraminifera are inversely correlated with water temperature, and the correlation coefficient is −0.99 (Marriott et al. 2004a). Montagna et al. (2006) have confirmed the temperature dependence of Li/Ca ratios of carbonate on the basis of high-resolution Li/Ca ratios of *C. caespitosa* in the Mediterranean Sea. Bryan and Marchitto (2008) also found that Li/Ca ratio for all species shells in Florida Straits is negatively correlated with temperature. Moreover, the carbonates precipitated in ocean and Lake have the similar formation processes, and thus Li/Ca ratios of ostracod shells at Lake Qinghai would be a reliable temperature indicator.

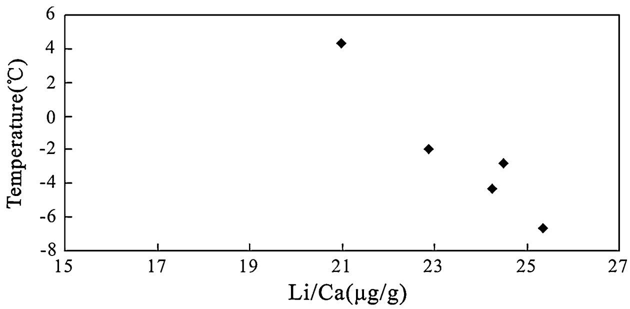


Fig. 5 The dependence between mean summer lake water temperature and Li/Ca ratios of ostracod shells in Lake Qinghai, showing strong correlation ($r = -0.97$, $p < 0.01$)

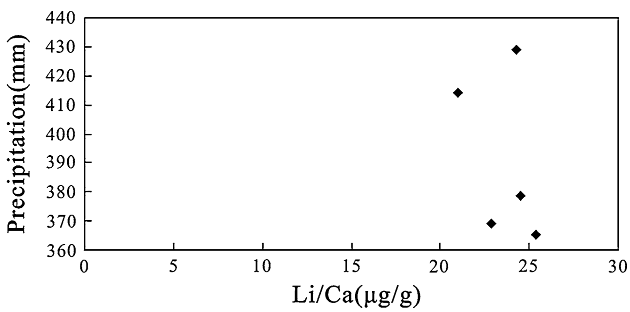


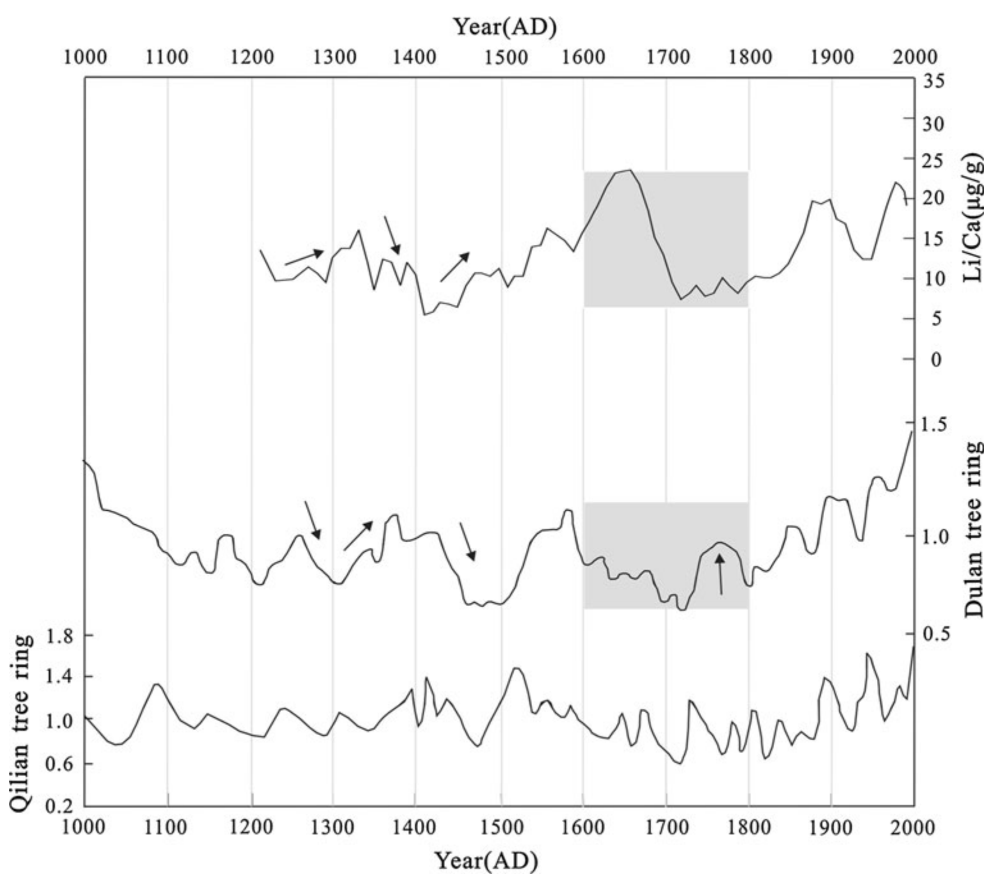
Fig. 6 The correlation between precipitation and Li/Ca ratios of ostracod shell at Lake Qinghai, and the correlation coefficient is low ($r = -0.34$, $p < 0.05$)

Comparisons between Li/Ca ratios and climates inferred from tree rings and meteorological records

As mentioned above, Li/Ca ratios of ostracod shells at Lake Qinghai would be a reliable palaeotemperature indicator. In order to better understand their potential, Li/Ca ratios of ostracod shells at Lake Qinghai have been applied to compare with the meteorological records, as well as the climates inferred from tree rings in the nearby regions, as shown in Figs. 5, 6 and 7. Meteorological records (precipitation and lake water temperature) are collected from the nearest Gangcha station, which is about 10 km northwards to Lake Qinghai (Xu et al. 2006b). Obviously, there is a negative correlation between lake water temperature and Li/Ca ratios of ostracod shells (Fig. 5), and no correlation between precipitation and Li/Ca ratios (Fig. 6), which imply that Li/Ca ratio may be a promising temperature proxy at Lake Qinghai.

Previous studies have demonstrated that tree growth is mainly controlled by temperature variations, and thus tree ring widths from Dulan and Qinlian were both interpreted to reflect temperature variations (Yao et al. 2001; Liu et al. 2004). As a whole, there are some broad similarities between Li/Ca ratios of ostracod shells and the trends of temperature inferred from tree ring width in adjacent regions, including Dulan and Qilian (Fig. 7), despite they are obtained at

Fig. 7 Comparisons between Li/Ca variations in *Eucypris inflate* and temperature records (10-year running mean). Temperatures reconstructed from tree ring widths in Dulan and Qilian (see location in Fig. 1a, Yao et al. 2001; Liu et al. 2004)



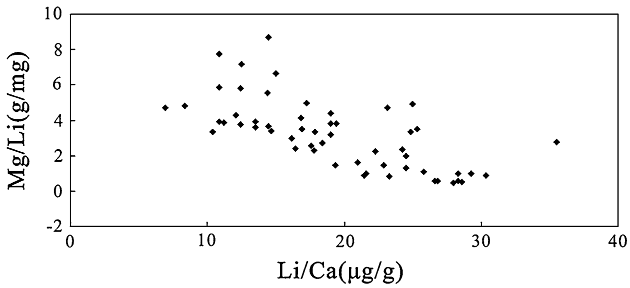


Fig. 8 The relation between Li/Ca and Mg/Li ratios of ostracod shells in Lake Qinghai, showing strong correlation ($r = 0.68$, $n = 55$, $p < 0.01$)

different resolutions and therefore difficult to compare in a statistical sense. As shown in Fig. 7, there are somewhat differences between Li/Ca ratios of ostracod shells and climates derived from tree rings, such as during the period of 1500–1600 AD, Li/Ca ratios and tree ring width showing the synchronous variations rather than the inverse variations. These discrepancies may be attributed to the different dating and climatic signals for different archives, such as lake sediments and tree rings. Therefore, variations of Li/Ca ratios of ostracod shells likely correspond with temperature variations in Lake Qinghai, rather than other factors that could potentially contribute to Li/Ca ratios changes. Li/Ca ratios of ostracod shells at Lake Qinghai are in turn mainly controlled by temperature; higher water temperatures result in lower Li/Ca ratios of ostracod shells, and vice versa.

In this paper, Mg/Li ratios of ostracod shells in Lake Qinghai were also measured to testify Li/Ca ratio as a temperature indicator. In a core study from Florida Straits, Bryan and Marchitto (2008) suggested that Mg/Ca ratios of carbonate are not solely representative of temperature signal, and also may be affected by high carbonate ion concentrations, and this influence can be adjusted by Mg/Li ratios. Mg/Li ratios display strong correlation with temperature, indicating that Mg/Li ratios may be a reliable temperature proxy as well as Li/Ca ratios. As shown in Fig. 8, the good correlation between Li/Ca and Mg/Li ratios in Lake Qinghai ($r = -0.68$, $n = 58$, $p < 0.01$) further and reliably proves that Li/Ca ratios of carbonate may be a potential temperature indicator.

Conclusions

Generally, there are currently rather few suitable indicators for temperature reconstruction. The first measurement of downcore Li/Ca ratios of ostracod shells in Lake Qinghai, NE Tibetan Plateau were carried out to reveal their potential palaeoenvironmental implications. Comparisons between Li/Ca ratios of ostracod shells in Lake Qinghai and temperature inferred from meteorological records and tree ring widths in Dulan and Qilian in adjacent region, the good correlation between them was observed, suggesting that Li/Ca ratio of carbonate may be a promising temperature proxy in lacustrine sediments. The good correlation between Li/Ca and Mg/Li ratios of ostracod shells in Lake Qinghai further supports this conclusion. Otherwise, this paper affords a new direction for temperature reconstruction in some regions where the temperature indicator is limited. Whereas the knowledge about Li/Ca ratios of carbonate is not comprehensive and further research is necessary, requiring detailed investigation on Li/Ca ratios of authigenic carbonates in more lakes and experimental research for adequate understanding.

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