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A 2100-year trace-element and stable-isotope record at decadal resolution from Rice Lake in the Northern Great Plains, USA

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Abstract: A 2100-year multiple proxy record at decadal resolution from a topographically closed (but hydrologically open) lake basin was used to reconstruct changes in lake salinity and evaporative intensity and to evaluate the reliability of different proxies for inferring climatic change in the glaciated Northern Great Plains (NGP). At Rice Lake, North Dakota, the ostracode-Mg/Ca ratios show good correlation with other ostracode, diatom and aeolian proxy records from other sites in the NGP. All of these records show significant century-scale periodicities, which have been related to solar variability as inferred from atmospheric radiocarbon records. This coherent and consistent pattern suggests that these sites record climatic variability at regional scales. The interval of the Mediaeval Climatic Anomaly from ~900 to 550 cal. yr BP contains two pronounced dry periods. During the following 'Little Ice Age', a single period of frequent drought that peaked at ~300 cal. yr BP was bracketed by wet periods before and after. In contrast, the ostracode-Sr/Ca ratios and ostracode- and mollusc-$\delta^{18}O$ records from Rice Lake have different patterns and do not correlate well with each other or with the same proxies at other sites. This lack of consistent pattern within a site and among several regional sites suggests seasonal and/or site-specific influences on these proxies. Variable contributions of groundwater, with low $\delta^{18}O$ values because of selective recharge from snowmelt, may have played an important role in determining the isotopic composition of the lake. The poor correlation of Sr/Ca with other proxies is caused by mediation of inorganic carbonate mineralogy, owing to active uptake of Sr in the formation of inorganic aragonite. Thus, our sediment data along with present-day lakewater chemistry suggest that carbonate-$\delta^{18}O$ records in the NGP cannot be interpreted simply in terms of climate because of significant local groundwater influences. Ostracode-Mg/Ca ratios tend to be a better indicator of past salinity because of the conservative nature of Mg in oligosaline waters (<10‰ salinity).

Key words: Oxygen isotopes, trace elements, Mg/Ca, Sr/Ca, ostracodes, salinity, palaeoclimate, groundwater, drought, late Holocene, 'Little Ice Age', 'Mediaeval Warm Period', North Dakota.

Introduction

The Holocene has recently received renewed attention in global-change research for exploring natural climatic variability at multiple temporal scales (Forman *et al.*, 2001) and for understanding the forcing mechanisms of the climate system (Duplessy and Overpeck, 1996; Woodhouse and Overpeck, 1998). High-resolution climatic proxy data for the last 2000 years are especially needed to bridge the gap between instrumental climatic records for the last few centuries and conventional century- to millennial-scale palaeoclimatic records. Additional records are always

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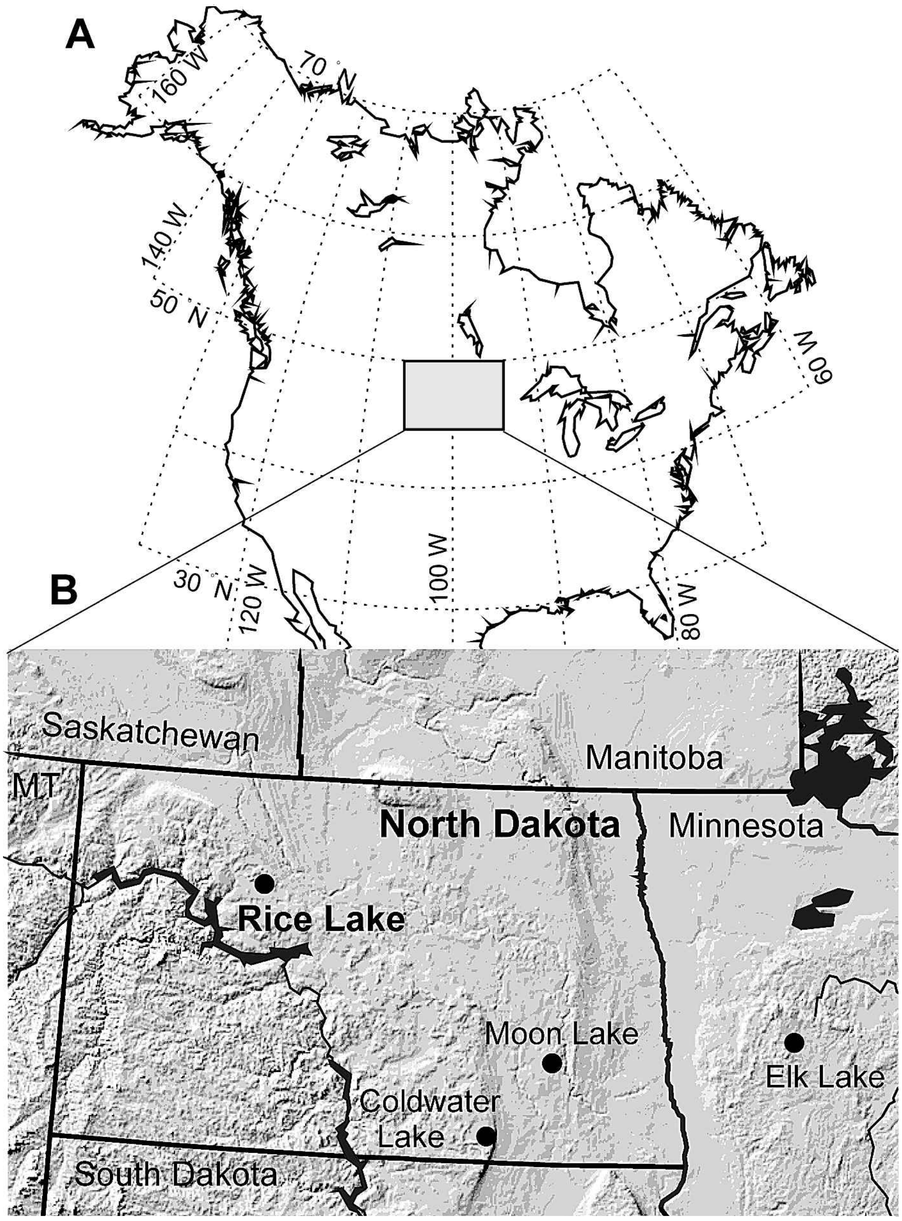

Figure 1 (A) Location map of the study region in the interior of North America. (B) Shaded relief map of the study region and location of the study site (Rice Lake, North Dakota, USA) and several other sites discussed in the text (Moon, Coldwater and Elk Lakes).

needed to define the spatial patterns of climatic variations for a region. These patterns may help reveal climatic forcing as well as response characteristics of different sites. In order to assure robust correlation among different sites, however, a clear understanding of the mechanisms and processes producing individual proxy records is essential for validating and assessing the climatic reconstructions.

In arid and semi-arid regions, oxygen isotopes and Mg/Ca and Sr/Ca ratios of ostracode shells have been used increasingly as indicators of effective moisture through their response to evaporative enrichment of heavy isotopes and solutes in lakewaters (e.g., Chivas *et al.*, 1993; Hodell *et al.*, 1995). In the glaciated North American Great Plains, however, topographic and hydrologic settings determine how local and regional groundwater flow systems interact with climate, controlling the water budget and isotopic and solute-mass balance of lakes (LaBaugh *et al.*, 1987; Winter, 1989). This hydrologic mediation has a significant influence on the reliability of these proxies as palaeoclimatic indicators.

Here we use multiple geochemical proxy data from the sediments of an evaporative groundwater-fed lake in the glaciated Northern Great Plains (NGP) to evaluate the differential responses of stable isotopes and trace elements to climatic and hydrologic variations. Our strategy is to combine palaeorecords together with present-day geochemical data (Yu *et al.*, 2002) to decipher the isotopic and hydrochemical behaviour of lakes in the NGP. We use regional correlation with other sites in the region as a means to demonstrate and assess the validity of these proxies as climatic indicators (Fritz *et al.*, 2000). Our working assumption is that if a proxy or lake records mainly regional climatic signals then palaeolimnological records should be comparable among sites with the same regional climatic history; but if different sites show significantly divergent responses and are not coherent then the record is primarily of local hydrological processes rather than regional climatic variability. Furthermore, by comparing multiple geochemical proxies at a single site, we can evaluate whether the variable patterns of change are attributable to an individual proxy indicator or conversely reflect the individualistic behaviour of the lake itself.

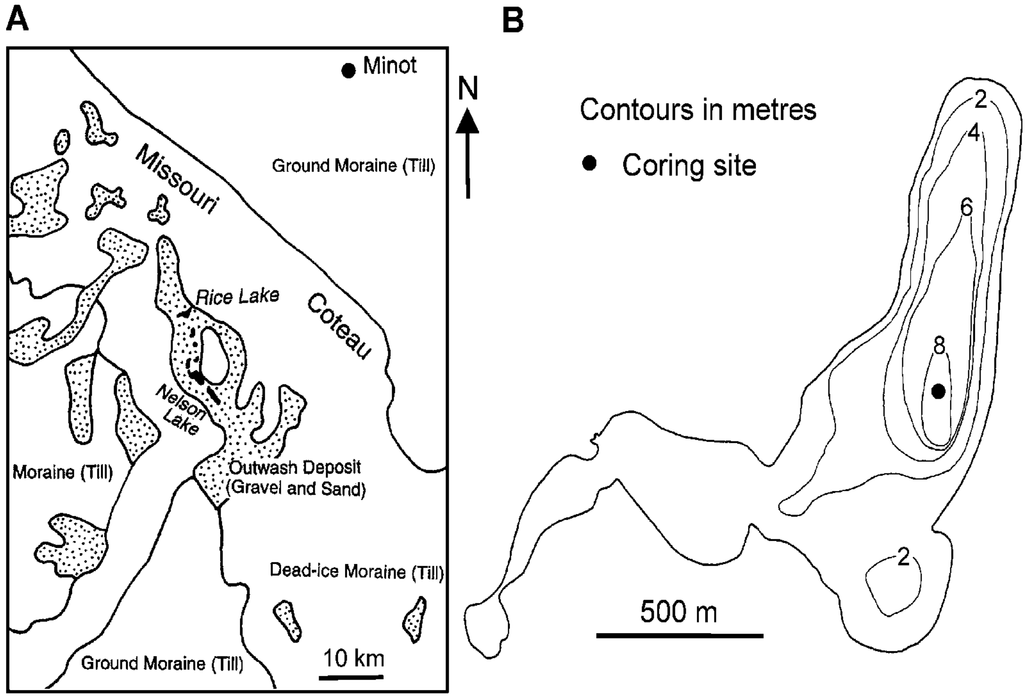

Figure 2 (A) Glacial deposits in the Rice Lake area (adapted from Bluemle, 1991). The dotted areas represent outwash gravel and sand; other areas are dominated by fine-grained till (ground and terminal moraines). (B) Bathymetric map of Rice Lake, North Dakota.

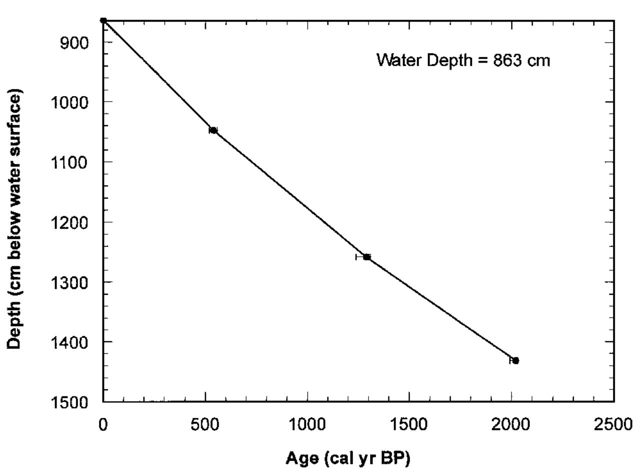

Figure 3 Age-depth plot and age model based on three calibrated AMS ^{14}C dates at Rice Lake, North Dakota, USA (dates from Yu and Ito, 1999; Fritz *et al.*, 2000).

Rice Lake and its hydrologic setting

The Northern Great Plains of central North America (Figure 1) has a subhumid to semi-arid climate, with precipitation significantly less than evapotranspiration. Its highly variable climate reflects its mid-continental location and the variable influences of contrasting regional air masses (Bryson and Hare, 1974). The mean annual precipitation ranges from about 520 mm in the northeast to 350 mm in the southwest, with peak precipitation falling in June (Mock, 1996). The mean temperature ranges from –18 to –10°C for January and from 20 to 24°C for July. The Rice Lake region has an annual precipitation of ~400 mm and evaporation of >600 mm.

Rice Lake (48°00′ N, 101°32′ W; 620 m a.s.l.) is situated on the Missouri Coteau near the edge of the Missouri Escarpment in north-central North Dakota (Figures 1 and 2A). The lake is situated in outwash gravel and sand deposited in a glacial meltwater channel during the last deglaciation (Figure 2A; Sloan, 1972; Bluemle, 1991). Like many other lakes in the glaciated NGP, Rice Lake is a topographically closed but hydrologically open system. The lake has the relatively low salinity of 1.8‰ composed of Mg-Ca-Na-SO_4^{2-}-HCO_3^--Cl ionic composition with a high Mg/Ca molar ratio of 38 (Table 1). It is one of 13 lakes in the Rice–Carlson chain of lakes (Figure 2A; Yu *et al.*, 2002), which owe their presence to the high permeability of underlying outwash gravel and sand deposits. These lakes are probably hydrologically connected by groundwater flow, comprised of a south-flowing N–S limb and a northwest-flowing NW–SE limb (Yu *et al.*, 2002). Rice Lake is located at mid-slope on the N–S limb of the chain. There is no surface-water connection among these lakes, so each lake is topographically closed.

Methods

An 11.5 m sediment core was taken from Rice Lake using a Wright-Livingstone sampler (5 cm diameter) in December 1985 from the deepest part of the basin at water depth of 8.63 m (Figure 2B). A conformable series of 10 AMS ^{14}C dates on grass charcoal or terrestrial macrofossils provides a chronology for the entire Holocene sequence (E.C. Grimm, personal communication). The three most recent dates fit a straight age-depth line and were used to derive a chronology for the last two millennia based on linear interpolation of their calibrated ages (Figure 3; see Yu and Ito, 1999, and Fritz *et al.*, 2000, for ^{14}C dates and their calibrated ages). The lake has a rapid sediment-accumulation rate of 2.5 mm yr^{-1}, and the top 5.5 m of sediment covering the last 2100 years was continuously sampled at decadal time intervals (contiguous *c.* 2.5 cm slices). Due to previous century-scale sampling, there are some gaps in the core, and the final sampling resolution averages about 14 years.

The subsamples (mostly 5–15 g wet weight) were soaked in Calgon and taken through several freeze-thaw cycles before being washed through a set of sieves (150 μm and 250 μm; Forester, 1987). Prior to chemical analysis, the ostracode shells were cleaned with triply distilled de-ionized water and dried in residue-free ethanol. Geochemical analyses were performed at 151 strati-

Table 1 Water isotopic and hydrochemical measurements of Rice Lake (Ward County), North Dakota, USA

Date	pH	Temp. (°C)	$\delta^{18}O$ (‰)	δD (‰)	Na^+ (ppm)	K^+ (ppm)	Ca^{2+} (ppm)	Mg^{2+} (ppm)	Sr^{2+} (ppm)	Cl^- (ppm)	SO_4^{2-} (ppm)	CO_3^{2-} (ppm)	HCO_3^- (ppm)	Mg/Ca (molar)	Sr/Ca (molar)	Salinity (‰)
05/05/91	8.98	5.4			156	42.2	8.9	241	0.021	30.7	600	34.1	701	44.65	0.0011	1.81
16/08/91	9.05	22.6	–3.12	–50.6	166	48.0	6.9	235	0.014	32.5	628	55.9	629	56.16	0.0009	1.79
24/05/98*	8.42	18.0	–6.11	–58.9	123	40.8	23.7	199	0.057	29.0	588	8.7**	709**	13.97	0.0011	1.73
Mean	8.82	15.3	–4.62	–54.8	148	43.7	13.2	225	0.031	30.7	605	32.9	680	38.26	0.0010	1.80

*Average of measurements of two samples.
**Calculated values on the basis of charge balance.

graphic intervals on juvenile shells (instars A-1, A-2) of *Candona rawsoni* – a benthic ostracode species with broad tolerance. Juveniles are more restricted than adults in the season of shell formation (Xia *et al.*, 1997a). Measurements were duplicated on 41 intervals. Bivalve mollusc (*Pisidium* sp.) shells were picked from 52 horizons (Table 2) and cleaned with full-strength Clorox (5.25% NaOCl) for 24 hours. The bleached shells were rinsed thoroughly with triply distilled, de-ionized water and dried in residue-free ethanol.

Between one and 13 *Candona* or *Pisidium* shells from each sample were reacted with 104% ultra-pure orthophosphoric acid (H_3PO_4) in individual reaction vessels at constant temperature of 70°C in a Kiel II carbonate preparation device. The evolved CO_2 gas was cryogenically purified to remove water and non-condensible gases, and the purified CO_2 was introduced to a Finnigan MAT 252 mass spectrometer. The CO_2 samples were measured for their $^{18}O/^{16}O$ and $^{13}C/^{12}C$ ratios in reference to a standard of known isotopic composition. The isotope results from carbonate shell samples are presented in conventional delta (δ) notation, which is defined as:

$$\delta = [(R_{sample} - R_{standard})/R_{standard}] \times 1000$$

where R is the absolute ratio of $^{18}O/^{16}O$ or $^{13}C/^{12}C$, and the standard is the Vienna Pee Dee belemnite (VPDB). The analytical precision is 0.2‰ for both $\delta^{18}O$ and $\delta^{13}C$. The elemental analysis of ostracode shells was performed on the acid residue remaining after the stable-isotope analysis (Chivas *et al.*, 1993). The residue was diluted about 200 times with 0.5 M high-purity distilled HCl for optimal measurement precision. The analysis of Ca, Mg, Sr and Ba concentrations in diluted acid residues were carried out on a Perkin Elmer/Sciex Elan 5000 inductively coupled plasma mass spectrometer (ICP-MS). The analytical precision is within 5% of concentration measurements of individual elements.

Results

Most sediment from Rice Lake is massive clayey marl, with some bandings (though a laminated section occurs during the early Holocene). The sediments contain ~10–20% organic matter and mostly 30–50% (but up to 78%) calcium carbonates during the last 2100 years, as estimated from loss-on-ignition analysis (E.C. Grimm, personal communication). Based on x-ray diffraction analysis performed on 27 samples from the same interval (J.J. Donovan, personal communication), the mineral fraction of the sediments is dominated by aragonite (mean 57%, with a range of 28–74%). The rest of the mineral components are composed of calcite (mean 4.5%, with a range of 2–9%), dolomite (mean 3.6%, range 2–8%), and non-carbonates (silicates including clays; mean 33%, range 20–57%). Detailed information on carbonate mineralogy is not available at decadal resolution. Throughout the 5.5 m of the core, little variation exists in total ostracode abundance, and the ostracode shells are well preserved. The abundance of mollusc shells is more variable. Several spikes occur in the abundance of macroscopic (>250 μm) charcoal fragments, and charophyte oospore (gyrogonite) concentrations are higher in the lower half of the analysed core section.

The oxygen and carbon isotopic and trace-elemental records show significant and divergent variations over the last 2100 years (Figure 4). The molar ratios of trace elements range from 10 to 32 mmol/mol for Mg/Ca, from 0.35 to 1 mmol/mol for Sr/Ca, and from 0.01 to 0.11 mmol/mol for Ba/Ca (Figures 4, A–C). The $\delta^{18}O$ values range from about –4 to –1.5‰ on ostracode shells (Figure 4D) and from –8 to –3‰ on mollusc shells (Figure 4F). The $\delta^{13}C$ values range from –1 to +1.5‰ on ostracodes (Figure 4E) and from –8 to –4‰ on molluscs (Figure 4G). Sr/Ca and Ba/Ca ratio patterns are generally coherent, although there is little correspondence between those two ratios and Mg/Ca. In general, there is little coherence between patterns of change in trace-element ratios and stable isotopes and between $\delta^{18}O$ and $\delta^{13}C$. Some correlation appears to exist between $\delta^{18}O$ and Mg/Ca ratios during the period of 1000–500 cal. BP, but a phasing shift between these two records is apparent. The two $\delta^{18}O$ peaks during this period tend to lead the Mg/Ca peaks.

Discussion

Interpretation of isotopic and hydrochemical records

The $\delta^{18}O$ values of biogenic carbonates from arid and semi-arid regions are usually interpreted in terms of changes in the $\delta^{18}O$ of lakewater over time, which in turn reflects changes in the intensity of evaporative enrichment. The apparent incoherence between $\delta^{18}O$ profiles of ostracode and mollusc shells from Rice Lake (Figure 5A) suggests a more complicated picture – one that could be caused by (1) the different temporal resolution of ostracode and mollusc samples, (2) ostracodes and molluscs recording different seasonal aspects of the $\delta^{18}O$ composition of lakewater, or (3) spatial variation of respective micro-environments. Ostracodes (low-Mg calcite) moult in a matter of weeks and form their shells (calcification) within 24 hours, so juvenile shells used in the analysis represent only a sampling of summer conditions (when most *C. rawsoni* juveniles moult). In contrast, bivalve molluscs (aragonite) build their shells continuously over their lifespan of many years, and their shell $\delta^{18}O$ probably represents some weighted average of the $\delta^{18}O$ of lakewater over this longer period. Variations in shell chemistry of molluscs are about twice that of ostracodes for both $\delta^{18}O$ and $\delta^{13}C$, suggesting larger variability of annual isotopic composition than that during summer only. The divergent variations in the two proxies suggest a poor correlation between summer and annual average $\delta^{18}O$ composition of lakewater, perhaps owing to seasonal change in the hydrologic budget of the lake.

It has long been recognized that many complicated factors

Table 2 Stable isotopes and trace elements of ostracode (*Candona rawsoni*) and bivalve mollusc (*Pisidium* sp.) shells from sediments of Rice Lake, North Dakota, USA

Age (cal. BP)	Ostracode $\delta^{18}O$ (‰)	Ostracode $\delta^{13}C$ (‰)	Mg/Ca (mmol/mol)	Sr/Ca (mmol/mol)	Ba/Ca (mmol/mol)	Mollusc $\delta^{18}O$ (‰)	Mollusc $\delta^{13}C$ (‰)
0	−2.25	0.78	23.48	0.34	0.04		
14	−4.27	0.48	38.47	0.43	0.06		
23	−3.49	−0.49	26.51	0.39	0.04		
32	−3.54	−1.18	30.64	0.36	0.03		
41	−3.12	−0.63	15.36	0.37	0.02		
50	−1.84	−0.32	19.26	0.68	0.06		
57	−3.76	−0.77	31.04	0.41	0.04		
106	−3.08	1.35	29.78	0.40	0.04		
115	−3.09	0.69	22.22	0.48	0.06		
124	−1.60	0.60	19.92	0.38	0.04		
133	−1.24	0.15	13.32	0.59	0.08		
143	−0.87	−0.19	14.41	0.44	0.08		
151	−3.97	0.02	13.58	0.52	0.07		
162	−2.54	−0.84	16.68	0.41	0.04	−4.12	−6.02
172	−1.64	−1.27	14.50	0.37	0.02	−4.16	−5.25
206	−3.50	0.57	27.35	0.43	0.04		
215	−2.90	0.66	28.10	0.45	0.04	−3.96	−5.37
224	−1.05	1.04	21.90	0.40	0.03		
234	−3.21	0.48	16.07	0.52	0.05	−5.15	−5.79
244	−4.82	0.09	19.37	1.14	0.16	−6.03	−6.49
254	−2.87	0.19	16.00	0.86	0.10		
272	−2.09	1.01	17.31	0.45	0.04		
310	−2.69	0.18	21.65	0.42	0.04	−6.81	−7.79
321	−3.55	0.56	28.64	0.52	0.07	−5.10	−7.50
331	−3.75	0.25	36.50	0.69	0.10	−5.08	−5.94
339	−1.88	−0.40	21.24	0.63	0.07		
349	−3.97	−1.22	32.76	0.72	0.07		
358	−2.50	−0.83	32.72	0.51	0.09		
367	−3.19	−0.32	22.16	0.95	0.05		
375	−3.20	−0.28	31.38	1.08	0.08		
409	−3.54	0.81	20.55	0.46	0.02		
417	−2.00	−0.09	19.48	0.38	0.01		
425	−3.39	−0.05	32.85	0.51	0.03		
434	−2.59	0.05	22.81	0.61	0.03		
443	−3.54	0.72	22.19	0.40	0.03		
451	−2.95	0.15	27.69	0.32	0.03		
460	−2.78	0.35	22.75	0.71	0.08	−7.64	−7.07
469	−3.71	0.99	29.53	0.47	0.04		
476	−3.57	1.13	26.29	0.37	0.03		
509	−2.73	−0.14	19.17	0.38	0.04		
517	−3.54	−0.35	15.54	0.40	0.04		
526	−4.39	−0.45	13.44	0.42	0.04	−5.14	−5.39
534	−3.67	0.39	15.81	0.58	0.07		
543	−3.59	0.28	17.83	0.41	0.05	−4.50	−5.53
552	−4.76	0.12	11.23	0.58	0.06	−5.54	−7.47
561	−3.48	−0.48	14.51	0.48	0.04	−5.02	−7.26
592	−3.16	0.53	22.44	0.94	0.06		
601	−2.50	−0.94	22.59	0.33	0.02		
610	−4.07	−0.27	37.67	0.37	0.03		
621	−4.17	−0.40	29.11	0.75	0.06		
630	−4.07	0.26	27.63	0.61	0.03		
639	−1.29	0.01	16.15	0.36	0.02		
649	−2.29	0.23	24.25	0.60	0.04		
657	−2.51	−0.89	22.88	0.43	0.03		
691	−2.41	0.16	20.13	0.35	0.02	−4.36	−4.81
703	−3.24	−0.16	20.82	0.44	0.03	−4.93	−7.31
712	−3.71	−0.49	22.37	0.48	0.06	−4.46	−5.60
722	−3.33	−0.36	21.60	0.40	0.04	−4.75	−6.92
733	−3.52	−0.33	26.15	0.37	0.03		
744	−3.77	−1.10	23.93	0.46	0.04		
753	−4.23	0.02	14.25	0.43	0.05		
786	−2.96	−0.65	17.22	0.39	0.02	−5.58	−7.96
798	−4.03	0.58	25.67	0.69	0.05		
809	−3.88	0.05	31.33	0.34	0.02		
820	−2.69	0.26	28.31	0.41	0.04		

Table 2 Continued

Age (cal. BP)	Ostracode $\delta^{18}O$ (‰)	Ostracode $\delta^{13}C$ (‰)	Mg/Ca (mmol/mol)	Sr/Ca (mmol/mol)	Ba/Ca (mmol/mol)	Mollusc $\delta^{18}O$ (‰)	Mollusc $\delta^{13}C$ (‰)
832	–2.98	0.99	28.52	0.32	0.02		
843	–0.80	1.00	22.41	0.30	0.02		
876	–3.90	0.12	14.48	0.45	0.04		
886	–1.95	1.07	25.34	0.33	0.03		
897	–1.56	1.28	19.87	0.39	0.05	–4.40	–5.15
907	–1.61	1.84	9.82	0.56	0.05		
920	–4.74	–0.48	11.44	0.70	0.09		
932	–2.35	–0.30	13.02	0.40	0.05	–3.58	–6.12
967	–4.31	–1.05	15.45	0.43	0.04	–5.66	–6.14
978	–2.82	–0.49	14.94	0.46	0.04	–5.49	–7.21
987	–4.14	0.49	26.34	0.63	0.04		
998	–3.27	–0.03	20.76	0.42	0.02		
1010	–3.01	0.18	23.78	0.73	0.08	–5.27	–6.09
1020	–3.49	–1.19	20.29	0.48	0.02	–6.58	–7.60
1054	–3.79	0.64	27.53	0.36	0.12		
1065	–4.15	0.22	30.75	0.56	0.04		
1076	–3.14	0.02	21.39	0.39	0.02		
1088	–3.07	0.00	23.30	0.77	0.06		
1102	–3.56	–0.09	24.89	0.41	0.03		
1137	–3.71	–1.00	17.09	0.65	0.03	–4.83	–5.15
1149	–3.46	–0.13	33.26	1.01	0.04		
1162	–3.85	–0.61	28.69	0.79	0.04	–4.39	–6.76
1174	–2.21	–0.75	21.79	1.16	0.04		
1187	–3.12	–1.08	25.45	0.76	0.04		
1220	–4.43	–0.56	20.65	0.64	0.04		
1230	–4.30	0.79	28.57	0.72	0.05		
1240	–3.48	0.81	24.58	0.73	0.06		
1251	–3.29	–0.44	18.54	0.37	0.03		
1258			15.27	0.37	0.03		
1267	–2.59	–0.30	15.08	0.41	0.04	–3.65	–5.23
1273	–3.26	0.13	12.76	0.53	0.06		
1322	–4.71	–0.43	21.91	0.55	0.02		
1352	–4.01	–0.58	20.85	0.66	0.04		
1397	–3.52	–0.28	20.56	0.99	0.11	–5.70	–4.96
1411	–2.21	0.31	15.12	0.47	0.01	–3.49	–6.02
1425	–3.02	–0.98	20.75	0.67	0.06	–4.68	–6.78
1438	–3.69	–0.77	21.46	0.59	0.03	–4.50	–7.12
1452	–3.01	–0.33	24.33	0.46	0.03	–4.64	–5.97
1496	–3.09	–0.57	17.73	0.40	0.02	–2.72	–6.62
1507			26.62	0.26	0.00	–4.14	–8.02
1517	–3.10	0.72	22.51	0.46	0.02		
1527	–2.59	0.35	19.02	0.48	0.05		
1537	–1.39	1.47	17.16	0.61	0.03	–5.69	–4.64
1545	–4.53	–1.59	46.85	0.46	0.08		
1574	–2.56	–1.09	15.88	0.38	0.03		
1584	–1.78	–0.07	21.44	0.29	0.02		
1593	–3.70	0.57	39.41	0.43	0.05	–3.43	–6.06
1602	–3.03	0.40	18.37	0.42	0.04		
1612	–3.45	–1.10	23.79	0.41	0.02	–3.86	–6.80
1630			40.14	0.86	0.17		
1661	–4.03	–0.14	18.62	0.68	0.07		
1670	–1.48	0.17	19.43	0.32	0.02		
1679	–4.37	–0.01	14.14	1.10	0.19	–5.88	–7.70
1688	–3.90	–0.13	18.86	0.47	0.03		
1698	–3.46	–0.71	18.33	0.51	0.02		
1707	–4.02	–0.70	20.31	0.73	0.04		
1715	–3.64	0.52	22.27	0.68	0.02		
1745	–2.24	–1.59	30.21	0.58	0.02	–4.59	–7.87
1755	–3.15	–1.03	24.06	0.38	0.02	–5.42	–8.18
1766	–1.40	0.47	16.23	0.47	0.01		
1776	–3.55	0.65	28.76	0.45	0.01		
1786	–1.96	1.26	20.78	0.69	0.04		
1797	–2.07	1.53	20.52	0.41	0.04		

Table 2 Continued

Age (cal. BP)	Ostracode $\delta^{18}O$ (‰)	Ostracode $\delta^{13}C$ (‰)	Mg/Ca (mmol/mol)	Sr/Ca (mmol/mol)	Ba/Ca (mmol/mol)	Mollusc $\delta^{18}O$ (‰)	Mollusc $\delta^{13}C$ (‰)
1827	–3.99	–0.82	23.46	0.44	0.02		
1837	–2.04	–0.88	22.80	0.31	0.01		
1847	–2.33	–1.19	17.78	0.39	0.01		
1857	–3.45	–0.68	18.16	0.42	0.03	–3.52	–6.49
1867	–2.97	–0.60	18.27	0.45	0.04	–3.54	–4.09
1877	–3.22	–1.10	11.90	0.43	0.03	–4.63	–5.02
1904	–2.92	–2.01	14.98	0.42	0.02		
1912	–2.55	1.37	34.35	0.31	0.04		
1923	–2.43	0.27	23.39	0.51	0.05		
1934	–3.73	0.11	28.21	0.36	0.02	–3.79	–5.92
1944	–3.02	–0.83	19.32	0.43	0.02	–5.60	–6.35
1955	–2.48	–0.40	23.89	0.35	0.02	–5.73	–7.59
1985	–1.81	0.08	25.72	0.36	0.04		
1997	–6.11	–2.15	16.76	0.51	0.03		
2008	–2.23	–0.59	22.60	0.46	0.05		
2019	–3.20	–0.28	23.34	0.54	0.07		
2031	–2.66	–0.36	24.52	0.50	0.04	–4.78	–6.42
2060	–3.01	0.08	19.11	0.55	0.03	–6.61	–7.45
2069	–3.00	0.23	20.18	0.35	0.02	–4.18	–7.04
2078	–3.67	–0.11	24.00	0.38	0.02		
2087	–2.42	–1.40	20.55	0.53	0.04		
2096	–3.21	–0.26	20.42	0.56	0.05	–4.23	–6.15
2103			20.60	0.35	0.05	–3.88	–6.01

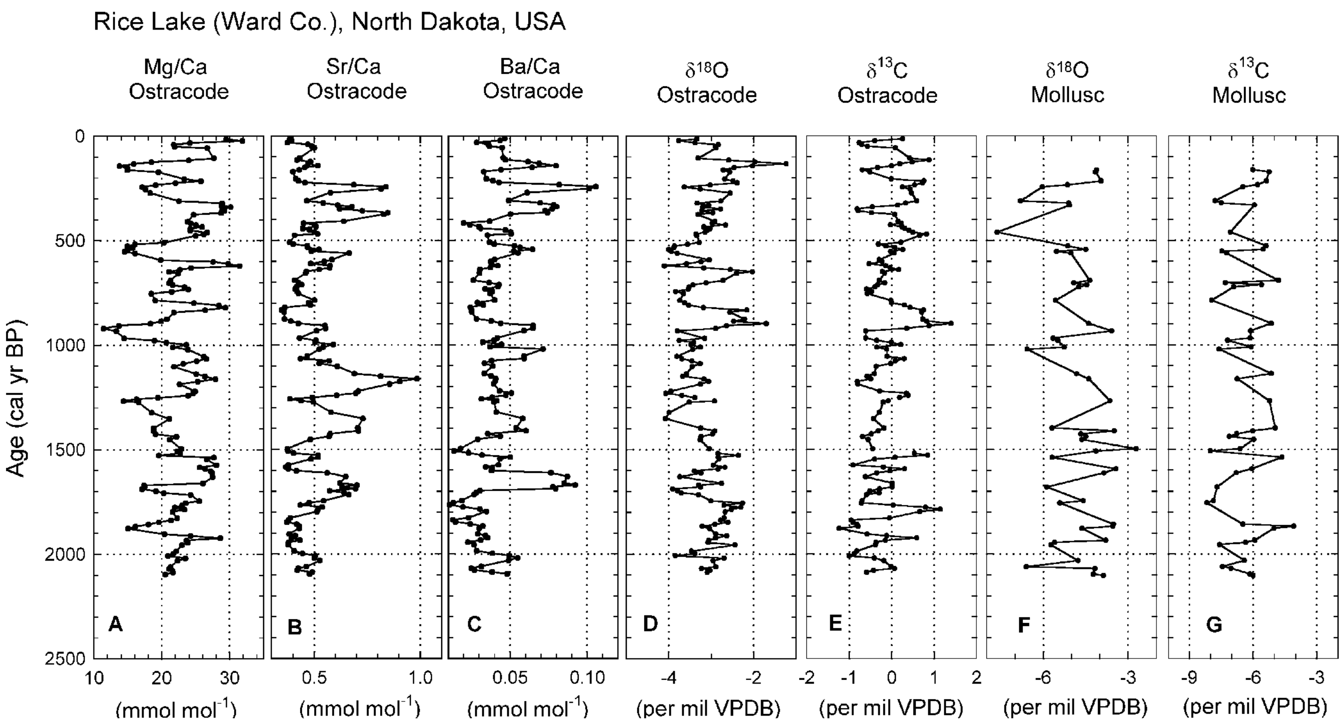

Figure 4 Stable-isotope and trace-element profiles for the last 2100 cal. years from Rice Lake, North Dakota, USA. (A) Ostracode Mg/Ca ratios. (B) Ostracode Sr/Ca ratios. (C) Ostracode Ba/Ca ratios. (D) Ostracode $\delta^{18}O$. (E) Ostracode $\delta^{13}C$. (F) Mollusc $\delta^{18}O$. (G) Mollusc $\delta^{13}C$. All data shown are three-point moving averages, except for mollusc isotopes (raw data). Ostracodes are juvenile (A-1 and A-2 instars) shells of a single species, *Candona rawsoni*, and bivalve mollusc shells are from *Pisidium* sp.

affect the isotopic composition of lakewater and consequently the isotopic composition of endogenically precipitated and biogenic carbonates from the water (Stuiver, 1970; Fritz and Poplawski, 1974; Pearson and Coplen, 1978). Atmospheric precipitation in the study area has an annually averaged $\delta^{18}O$ value of about –11‰, but the groundwater values are –15 to –25‰ (Yu *et al.*, 2002). This implies that groundwaters are recharged by isotopically light late-winter snowmelt and that changes in the seasonality of precipitation – as well as the mean annual moisture budget – strongly influence the isotopic composition of the lake.

The conclusion that local hydrology plays a significant role in the oxygen-isotope composition of lakewater is also illustrated by the lack of correlation between $\delta^{18}O$ records at Rice Lake and Coldwater Lake (Figure 5B). Isotopic data for waters collected from 40 lakes in the NGP suggest no straightforward relation between $\delta^{18}O$ and salinity (Yu *et al.*, 2002), providing additional

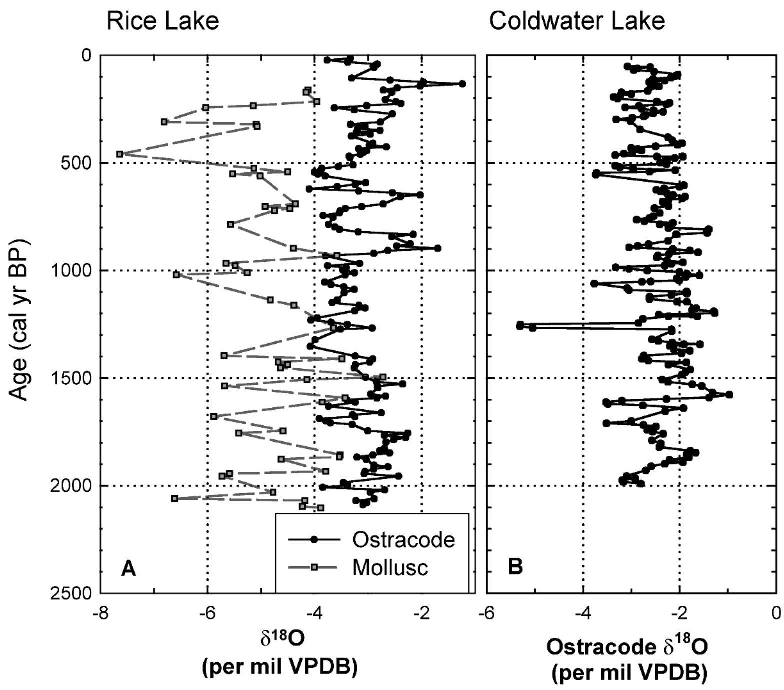

Figure 5 Comparison of oxygen isotopes ($\delta^{18}O$) from (A) ostracode (*Candona rawsoni*) and bivalve mollusc (*Pisidium* sp.) shells at Rice Lake and (B) ostracode (*C. rawsoni*) shells at Coldwater Lake, North Dakota.

evidence that the $\delta^{18}O$ of lakewater and of carbonates is not a simple indicator of evaporative enrichment. A clear example is that from the Rice–Carlson chain of lakes, where 10 of 13 lakes within 12 km distance show uniform lakewater $\delta^{18}O$ values of –6 ± 1‰, despite a large range of total dissolved solids (TDS) of 1 to 65 mg/L (Yu *et al.*, 2002). Smith *et al.* (1997) found that during the dry mid-Holocene increased ostracode-inferred salinity and Mg/Ca ratios corresponded with a 2–3‰ decrease in $\delta^{18}O$ at Elk Lake (Grant Co., Minnesota, USA). They attributed this isotopic depletion to the change in groundwater catchment area and reduction in evapotranspiration during desiccation of peripheral upland lake basins. A decreasing trend in $\delta^{18}O$ during the mid-Holocene dry period at Lake Manitoba, Canada, was also attributed to the increasing influence of isotopically light groundwater inflow (Last *et al.*, 1994).

The Mg/Ca ratios in ostracode shells are directly related to the Mg/Ca ratios in lakewater and the water temperature at the time of shell formation (Chivas *et al.*, 1986; Forester, 1986; Engstrom and Nelson, 1991; Xia *et al.*, 1997a). The Mg/Ca of lakewater in the NGP is directly proportional to salinity in the range of 0.6 to 12 mg/L of TDS – conditions under which Mg behaves conservatively and Ca is held constant by carbonate precipitation (Yu *et al.*, 2002). The arid climate of the NGP makes ostracode-Mg/Ca a robust proxy for salinity, as has been shown for Devils Lake (Engstrom and Nelson, 1991), Coldwater Lake (Xia *et al.*, 1997b; Fritz *et al.*, 2000) and Rice Lake (Yu and Ito, 1999), similar to the situation observed in Australia (De Deckker *et al.*, 1999). A comparison of Mg/Ca and $\delta^{18}O$ records from Rice Lake shows some similarity in broad-scale trends during certain time intervals (Figure 6A), but in general there is little covariance (Figure 6B). For example, two pronounced peaks are apparent at 1000–600 cal. BP during the Mediaeval Climatic Anomaly (MCA; Stine, 1998), although the Mg/Ca seems to lag behind $\delta^{18}O$ by about 100 years. Also both proxies show similar trends during the intervals of 1500–1350 cal. BP and 600–300 cal. BP (indicated by grey lines in Figure 6A).

In the NGP, Sr/Ca ratios in both ostracodes and lakewater are usually poorly correlated with salinity and thus are difficult to interpret in palaeoclimatic terms. The weak correspondence results from variation in endogenic carbonate mineralogy (calcite versus aragonite), which mediates Sr/Ca ratios because of active uptake of Sr in aragonite precipitation (Haskell *et al.*, 1996; Xia *et al.*, 1997a). At Rice Lake, dry intervals indicated by high Mg/Ca ratios frequently correspond with low Sr/Ca ratios (Figure 7A), when increased salinity induces a change in endogenic carbonate formation, possibly from calcite to even more aragonite. The precipitated inorganic aragonite incorporates and therefore removes Sr from lakewater, decreasing the Sr concentration in remaining water and in moulting ostracode shells. This relation between Mg/Ca and Sr/Ca is illustrated by double-arrowed lines in Figure 7A. However, both ratios do not show an overall correlation (Figure 7B).

Regional climatic signals and teleconnections

To evaluate the validity of our different proxies for climatic reconstruction, we compare our records from Rice Lake with geochemical and biological proxy records from other sites in the NGP (Figure 8). The working assumption is that if a proxy responds to the same climatic forcing in a similar manner at all sites then it probably represents regional climatic patterns. If no similarity in the response pattern exists among sites for any given proxy, it suggests that local factors, particularly hydrologic processes, play a more important role in the response of that proxy. Among the Rice Lake proxy records (Figure 4), Mg/Ca ratios show strong similarity to records from other sites (Figure 8), especially in terms of long-term patterns. This regional correlation suggests that the Mg/Ca ratios faithfully record salinity changes in lakewater and in turn large-scale climatic changes in moisture balance. The minor differences among sites indicate different local influences, but regional climatic signals prevail. Fritz *et al.* (2000) compared diatom-inferred salinity and ostracode-Mg/Ca records from the NGP sites in fine stratigraphic detail and reached similar conclusions regarding the robustness of climatic inferences from proxies that record ionic concentration.

Previously, Yu and Ito (1999) discussed the spectral nature of the Mg/Ca ratios at Rice Lake, which show statistically significant periodicities of about 400, 200, 130 and 100 years. The Mg/Ca record is similar to the atmospheric ^{14}C variations (Figure 9; Stuiver and Braziunas, 1989), a proxy of past solar variability, and its phasing is correlated with that of other NGP sites and with

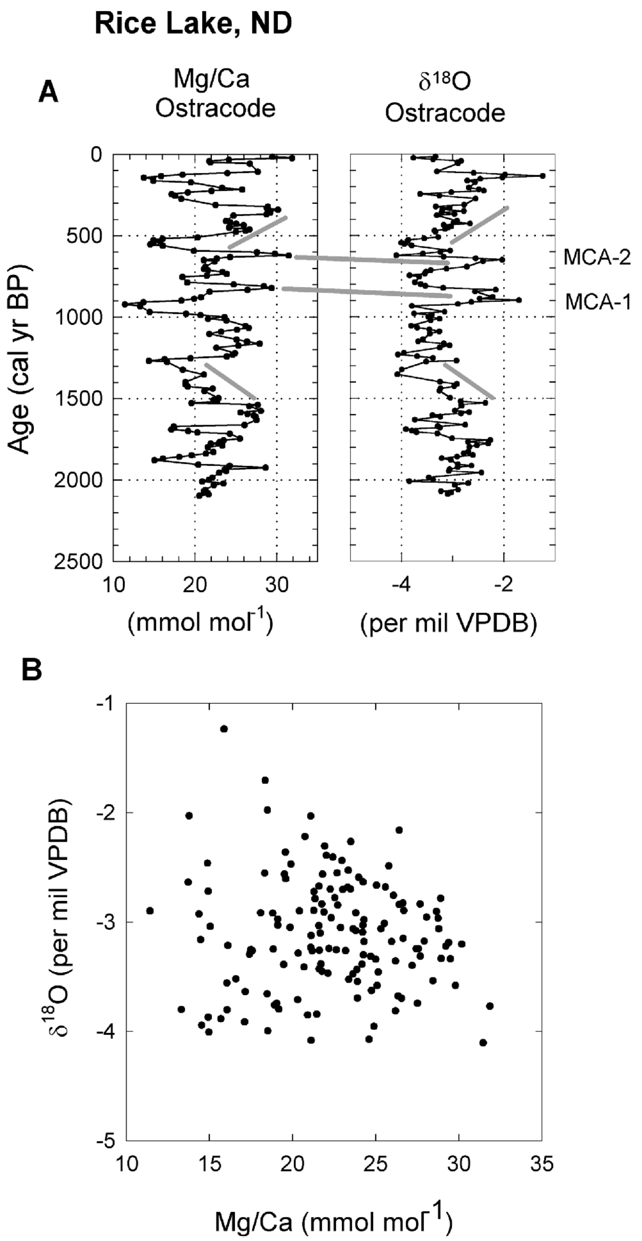

Figure 6 Detailed correlation of ostracode-Mg/Ca ratios and ostracode-$\delta^{18}O$ from Rice Lake. (A) Stratigraphic correlation. (B) Covariance plot. Two distinct peaks (MCA-1 and MCA-2) during the Mediaeval Climatic Anomaly at ~1000–600 cal. BP correlated well, although with a timelag of ~100 years for Mg/Ca. Other common trends were indicated by grey lines.

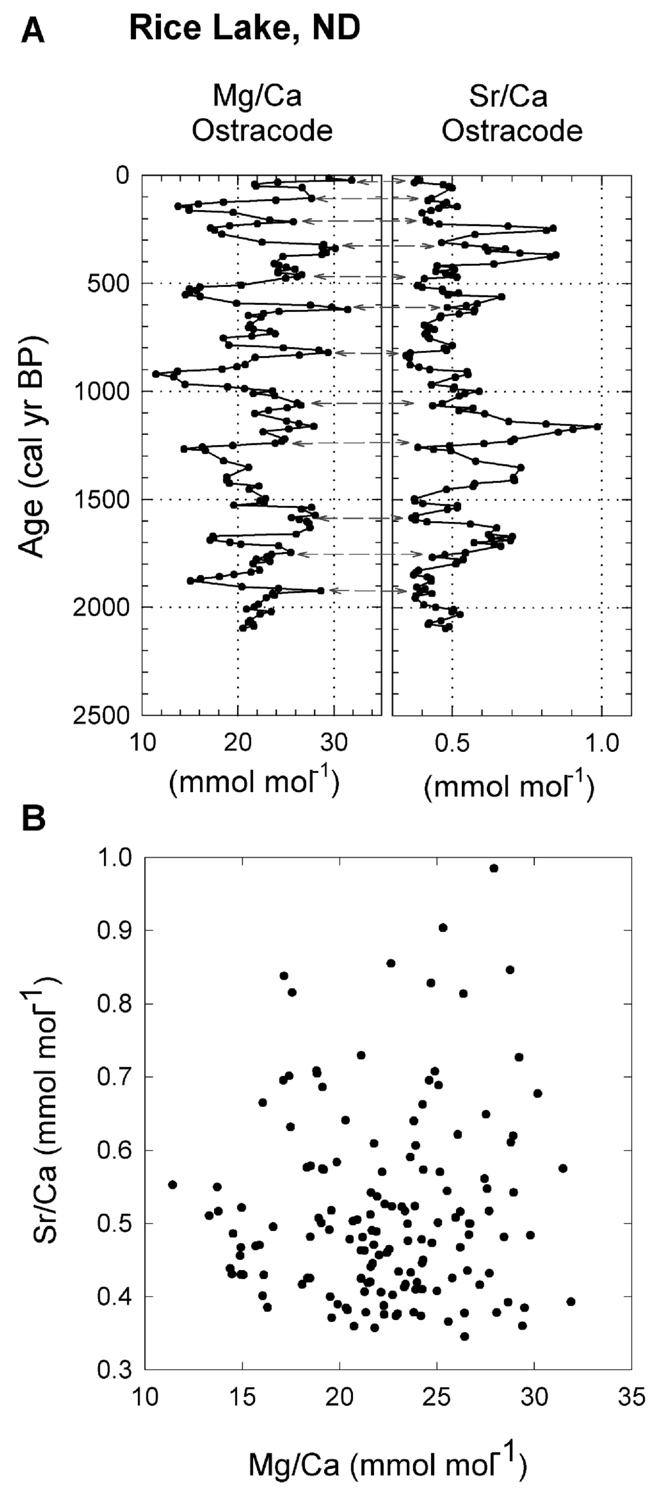

Figure 7 Detailed correlation of Mg/Ca ratios and Sr/Ca ratios from Rice Lake. (A) Stratigraphic correlation. (B) Covariance plot. The arrowed lines indicate an inverse relation between the two ratios, suggesting aragonite mediation of Sr concentration at higher salinity.

the Greenland GISP2 $\delta^{18}O$ record (Grootes *et al.*, 1993; Figure 8A). Yu and Ito (1999) proposed that solar forcing caused the century-scale drought frequency in the NGP. In addition to the generally strong correlation among these records, the NGP sites also show periodicities similar to those of Rice Lake: ~130 years at Coldwater Lake (Fritz *et al.*, 2000), 400 years at Moon Lake (Laird *et al.*, 1996; 1998), 400 and 88 years at Elk Lake (Dean, 1997) and 400 years at Pickerel Lake, South Dakota (Dean and Schwalb, 2000). Given the cumulative chronological uncertainty of at least 100 years, a pronounced feature in most time-series from the NGP is that the relative dry periods were separated by low salinity/wet period at about every 400 years, as indicated by alternate grey and black bars in Figure 8. Although both the Mediaeval period and the 'Little Ice Age' (LIA) were hydrologically complex and neither can be characterized by uniformly wet or dry conditions (Fritz *et al.*, 2000), certain general structures and long-term patterns appear. The second 400-year dry period between ~1000 and 500 cal. BP (MCA: Mediaeval Climatic Anomaly; Stine, 1998) apparently has two peaks of major drought, especially at Rice Lake, Coldwater Lake and Elk Lake. The MCA-1 occurred at ~900–800 cal. BP, whereas the MCA-2 occurred at ~700–600 cal. BP. The increasing trend in salinity in the first half of the LIA, which peaked around 300 cal. BP, was documented at all four NGP lakes (arrows in Figure 8, B–E), and LIA drought has also been noted in the Devils Lake (North Dakota) record (Fritz *et al.*, 1994). The double Mediaeval droughts are probably correlated with the Generation-1 (killed in AD 1110) and Gener-

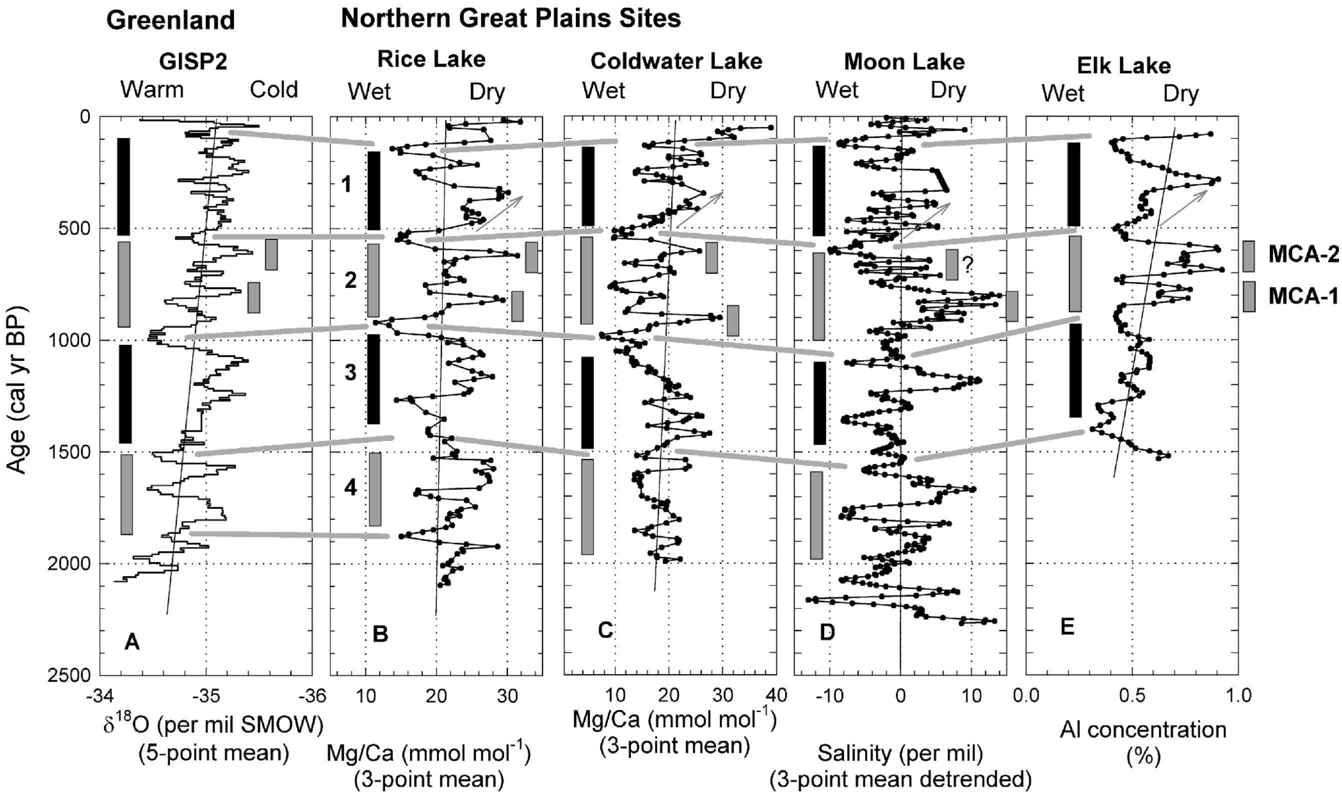

Figure 8 Regional and global correlation of geochemical and biological proxies of palaeoclimates. (A) GISP2 $\delta^{18}O$ profile (20-year average) from Greenland (Grootes *et al.*, 1993). (B) Mg/Ca molar ratios on ostracode shells (three-point moving average) from Rice Lake (Yu and Ito, 1999). (C) Mg/Ca molar ratios on ostracode shells (three-point moving average) from Coldwater Lake, ND (Fritz *et al.*, 2000). (D) Diatom-inferred salinity (detrended three-point moving average) from Moon Lake, ND (Laird *et al.*, 1996; 1998). (E) Al concentration (three-point moving average) from Elk Lake, MN (Dean, 1997). The distinct multicentury-scale cycles are indicated by alternate grey and black bars (numbered 1 to 4), with a spacing of ~400 years. Arrows show the drying trend in the first half of the 'Little Ice Age'. Thin lines represent long-term trends, except for detrended Moon Lake record.

ation-2 relict tree stumps (killed in AD 1350) from California in the Great Basin as presented by Stine (1994; 1998). These century-scale fluctuations in moisture conditions during the Mediaeval period appeared at other sites in the NGP (e.g., Vance *et al.*, 1992) and elsewhere in North America (see review by Stine, 1998).

To further investigate the century-scale drought patterns in the Northern Great Plains, we resampled the four time-series (Rice, Coldwater, Moon and Elk) at even 10-year intervals during the last millennium. These time-series were smoothed with a Savitzky-Golay smoothing filter in the software AutoSignal Version 1.0 (by SPSS Inc., Chicago, Illinois). The options used for all data sets were 4 for window width, 3 for polynomial order and 2 for pass. The smoothed time-series were normalized to a range from 0 to 1 in arbitrary units, and the four normalized series were averaged to generate a combined time-series as an aridity index for the Northern Great Plains (Figure 10). We emphasize that the index only reflects multiple century-scale patterns in a relative sense, for the finer details have been smoothed out by the resampling procedure. The MCA-1 and MCA-2 also show up in this combined aridity index, along with a drought peak around 300 cal. BP during the LIA. The double-peak pattern of the MCA has also been recorded in summer sea-surface temperatures in the Norwegian Sea, reconstructed from diatom transfer functions by Jansen and Koç (2000), who found high temperatures at 1100–1000 cal. BP and at 700–600 cal. BP, as well as increasing temperature at the first half of the LIA, which peaked at 300–200 cal BP. This correlation and that with the California tree-stump record (Stine, 1998) suggest possible teleconnection of century-scale climatic variations during the last 1000 years.

Summary and conclusions

(1) The ostracode-Mg/Ca record from Rice Lake shows a consistent and coherent pattern with other sites in the NGP, all of which exhibit several significant century-scale periodicities. The spectral similarity and phasing correlation of Mg/Ca ratios at Rice Lake and the atmospheric ^{14}C record indicate a possible link of lake salinity and drought frequency in the NGP to solar forcing.

(2) The Mg/Ca ratios at Rice Lake together with salinity proxies from other NGP lakes suggest a coherent pattern of century-scale drought variability, although both the Mediaeval Climatic Anomaly (MCA) and the 'Little Ice Age' (LIA) were hydrologically complex (Fritz *et al.*, 2000). Two peak droughts during the MCA occurred at 900–800 cal. BP and 700–600 cal. BP, whereas the LIA contains a single broad drought peak centred at ~300 cal. BP.

(3) Our multiple proxy records indicate that the ostracode-Mg/Ca ratios are a reliable indicator of lake salinity and of evaporative intensity. The present-day lakewater hydrochemical data (Yu *et al.*, 2002) support this conclusion, which shows the conservative nature of Mg at salinities ranging from 0.6 to 12 mg/L of total dissolved solids.

(4) The ostracode-Sr/Ca ratios are strongly influenced by shifts in inorganic carbonate mineralogy, which may distort their response to changes in salinity. In contrast to a conventional interpretative model, the Sr/Ca ratios decrease when lake salinity increases, because of the non-conservative nature of Sr and its incorporation into inorganic aragonite.

(5) Our multiple proxy records along with lakewater data (Yu *et al.*, 2002) suggest that $\delta^{18}O$ records in the NGP cannot be interpreted simply in terms of climate, because of significant local

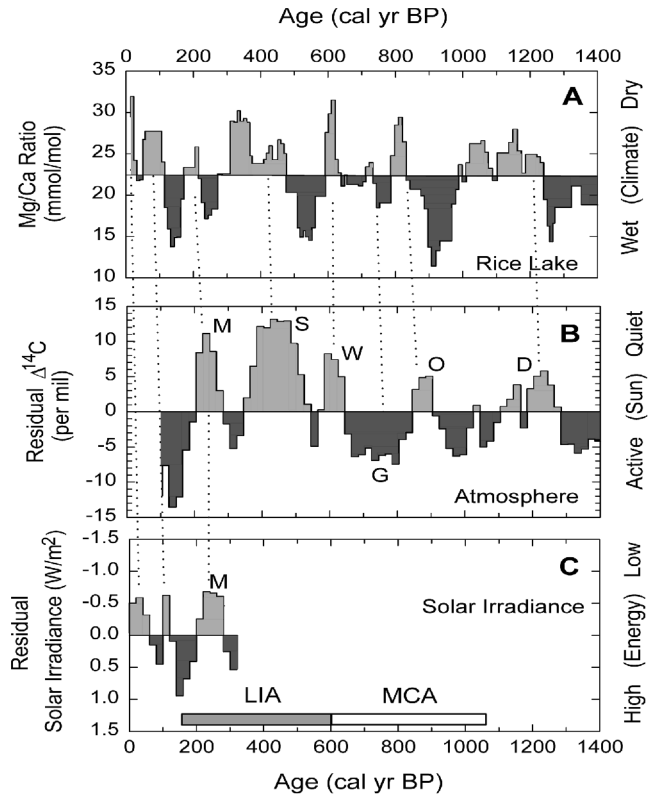

Figure 9 Correlation of historical drought and solar proxies. (A) Mg/Ca ratios (mmol mol^{-1}) of ostracode *Candona rawsoni* shells from Rice Lake (Yu and Ito, 1999), as a proxy of drought frequency. (B) Bidecadal atmospheric $\Delta^{14}C$ (‰ residual) from tree-rings (Stuiver *et al.*, 1998). Classical historical $\Delta^{14}C$ maxima and solar (i.e., sunspot) minima include M – Maunder (AD 1645–1715) S – Spörer (AD 1420–1530), W – Wolf (AD 1280–1340), O – Oort (AD 1010–50) and D – Dark Age (AD 660–740). G – Grand Solar Maximum (AD 1100–1250). Solar minima correspond to dry periods (A). (C) Bidecadal solar irradiance (W m^{-2} residual; Lean *et al.*, 1995). LIA – 'Little Ice Age' (600–150 yr BP). MCA – Mediaeval Climatic Anomaly (1050–600 yr BP). All curves are plotted on a common age scale (calendar years before AD 1950, i.e., cal. yr BP).

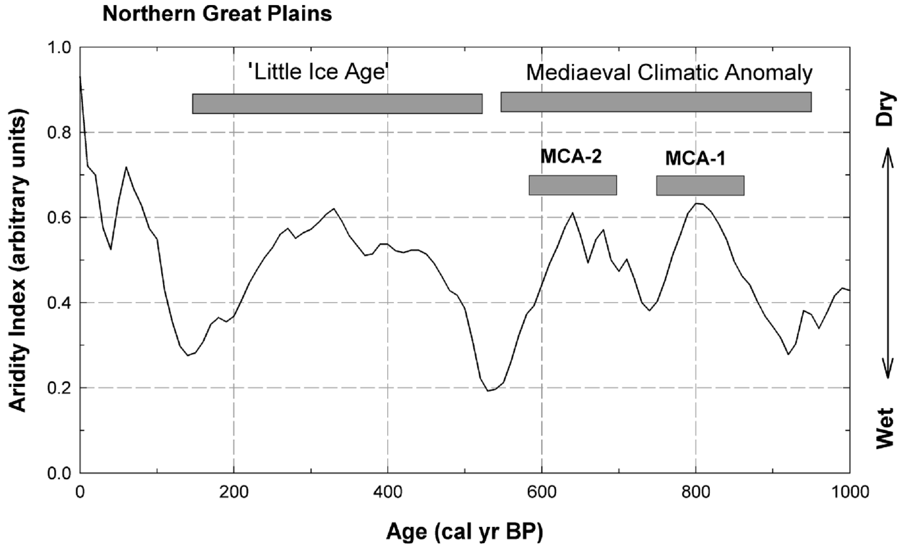

Figure 10 Aridity index for the Northern Great Plains of North America. The curve was derived by averaging resampled, normalized salinity/drought proxies from four lakes (Rice, Moon, Coldwater and Elk Lakes) in the NGP. The index shows a double-peak pattern during the Mediaeval Climatic Anomaly (MCA; Stine, 1998). During the 'Little Ice Age' (LIA), drought peaks around 300 cal. BP, with low aridity at the beginning and end of this episode.

groundwater influences. In some hydrological settings, fluctuations in the isotopic composition of groundwater and the seasonality of recharge may dictate the isotopic mass balance of these NGP lakes.

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