



Rapid environmental change during dynastic transitions in Yunnan Province, China



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ABSTRACT

Pollution and eutrophication of Chinese lakes are widely perceived to be 20th century phenomena. However, China has a long history of deforestation, agriculture, mineral resource extraction, and other anthropogenic activities that impact the environment. Here, we present a sediment record from Xing Yun Lake in the Yunnan Province of China that reveals significant alterations to the lake, its ecosystem, and its watershed beginning as early as 500 AD. A comprehensive suite of biogeochemical and isotopic proxies reveal several rapid transitions related to changes in agriculture and lake-level management that coincides with cultural and dynastic transitions. The deterioration of contemporary environmental conditions at Xing Yun arises from a long history of anthropogenic manipulation, eutrophication, and pollution of the lake and its watershed. This study highlights the importance of using historical records of industrial and agricultural activities, including landscape modification, in conjunction with records of climate change, to place present day environmental concerns into a long-term context.

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1. Introduction

Lakes in Yunnan Province of southwestern China have persistent and severe water quality problems related to rapid population growth and industrialization, which have placed great demands on freshwater and agricultural resources (Whitmore et al., 1997; Wang et al., 2012). Today, many lakes in this region are heavily impacted by deforestation, soil erosion, agriculture, urbanization, and industrialization (Whitmore et al., 1994a). As a result of these activities, many lakes on the Yunnan plateau are now eutrophic (Li et al., 2007; Liu et al., 2012) and have high concentrations of toxic metals in their sediments (Zeng and Wu, 2009). Despite an increasing awareness of the negative landscape-scale impacts arising from these disturbances, problems associated with water pollution, sediment quality, and environmental degradation are likely to persist or worsen as the population expands. Further, it is increasingly apparent that many lakes on the Yunnan plateau have been impacted by anthropogenic activities for centuries (e.g., Shen et al., 2006; Dearing et al., 2008). In Yunnan, which has a long history of human occupation, agriculture, and mining, questions

remain regarding which lakes may have been impacted prior to the 20th Century, the nature of the impact, and how anthropogenic effects can be differentiated from other controls such as climate change. Assessment of the relative influence of anthropogenic and climate forcing on the ecosystems of this region has not been the main focus of environmental investigations. A longer-term perspective is therefore necessary to assess the timing, magnitude, and rate of environmental change.

Lacustrine sediments provide a geological and geochemical archive of changes to the lake, its watershed, and the overlying atmosphere at a variety of spatial and temporal scales. For example, trace element concentrations (e.g., lead, mercury, etc.) released during the extraction and processing of metal-rich ores have been used to reconstruct metal inputs through time (Lee et al., 2008; Bindler et al., 2012), while other elements can be used to infer land-use change (e.g., aluminum) and nutrient loadings (e.g., nitrogen and phosphorus) (O'Hara et al., 1993). In addition, the concentration and stable isotopic composition of lake sediment organic matter (Davidson and Jeppesen, 2013) and authigenic carbonate (Hodell et al., 1999) can offer insight into changes in land use, lake trophic status, carbon cycling, and regional climate change. Combined, these proxies provide insight into past human–environment interactions and limnological responses to landscape alterations.

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Here, we present a 3500-year sediment record from Xing Yun Lake, which is ideally situated to answer questions regarding long-term environmental change and the geochemical variations associated with human disturbance. Xing Yun is a lake with a simple catchment and is located in a region containing several well-studied climate records. To reconstruct changes in land-use, lake-level management, and mineral resource extraction, we rely on measurements of sediment composition (% organic, carbonate, and mineral matter), the concentration and isotopic composition of organic carbon and nitrogen ($\%C$, $\%N$, $\delta^{13}C_{org}$, and $\delta^{15}N_{org}$), the oxygen and carbon isotopic composition of authigenic calcite ($\delta^{18}O_{carb}$ and $\delta^{13}C_{carb}$), and several elemental concentrations (Al, P, Pb, and Hg) in sediment. We use these analyses to investigate the timing and magnitude of environmental changes at Xing Yun Lake in the context of anthropogenic activities that coincide closely with periods of cultural transition.

2. Regional setting

2.1. Study site

Southwestern China is heavily influenced by the Asian Summer Monsoon (ASM) system, with 70% of the average annual precipitation falling in the months of June–September (Table 1). Temperature in this region is generally stable throughout the year and winters are mild. The climate is defined as warm temperate with a dry winter and a warm summer (designated Cwb in the Köppen-Geiger classification system) (Kottke et al., 2006). Land clearance for agriculture is extensive around Kunming, but in less impacted areas, subtropical conifer forests predominate (Li and Walker, 1986).

Xing Yun Lake (24°10'N, 102°46'E) is ~70 km south of Kunming, in Yunnan Province of southwestern China (Fig. 1). It is a shallow (Z_{max} : 11 m) and eutrophic lake (Liu et al., 2012) with a surface area of 34 km² and a watershed area of 383 km². The lake catchment contains large flat sections of land used for rice agriculture. The western side of the catchment is dominated by slate and phyllite and the eastern side is composed of sandstone, shale, and limestone (Bureau of Geology and Mineral Resources of Yunnan Province, 1990). The lake's only surficial outflow is the Ge River, which drains into deep Fuxian Lake; however, today several dams exist on the Ge River, regulating water flow. The annual weighted $\delta^{18}O$ mean of modern rainfall for Kunming is -9.86% VSMOW (Table 1) and the $\delta^{18}O$ value of a water sample taken from the lake in 2009 is -4.3% VSMOW, suggesting that substantial water loss from Xing Yun occurs through evaporation (Fig. 2).

The hydrologic and isotopic sensitivity of Xing Yun to changes in precipitation associated with the ASM was previously explored by Hodell et al. (1999) using a 12.5 m sediment core collected near the southern end of the lake at a water depth of 7 m (Fig. 1). Grain size, magnetic susceptibility, and oxygen isotopic values of authigenic carbonate were measured. The Hodell et al. (1999) study suggested that from 20,000 to 12,000 years BP, lake water $\delta^{18}O$ values decreased, indicating an increase in the strength of the ASM, and that from 8000 years BP to present, $\delta^{18}O$ values increased in response to a weakening ASM. These isotopic shifts occurred in

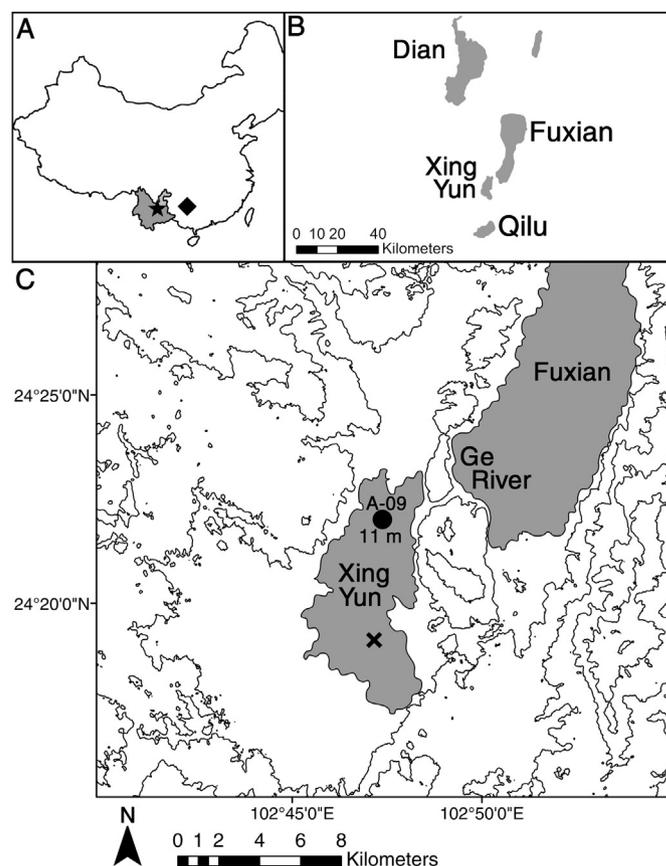


Fig. 1. A – Locations of Xing Yun Lake (star), Dongge Cave (diamond); B – Yunnan lakes near Kunming; C – Xing Yun Lake (1721 m elevation) with 200 m contour intervals. Coring location in this study is marked by the circle. Coring location from Hodell et al., 1999 is marked by the X.

conjunction with changes in Northern Hemisphere summer insolation, similar to conclusions drawn from the Dongge Cave record (Wang et al., 2005). Notably however, the chronology of the Hodell et al. (1999) Xing Yun $\delta^{18}O$ record was constrained by bulk sediment and gastropod shell radiocarbon dates, which are subject to hard-water effects (Deevey et al., 1954). Further, the low sampling resolution (5–6 cm) makes the $\delta^{18}O$ transitions appear abrupt and uneven, a consequence that precludes a direct comparison to the Dongge Cave record (which has a much higher temporal resolution) and makes it difficult to constrain the timing of abrupt shifts in $\delta^{18}O$ values. The Hodell et al. (1999) study acknowledged the possibility of recent anthropogenic disturbance (last 250 years) on Xing Yun, but did not address the possibility of impacts on the lake by earlier human activity.

2.2. Human history

Yunnan has a long history of human occupation, with Neolithic settlement around lake basins occurring between 10,000 and 3500

Table 1

Monthly average temperature, precipitation, and oxygen isotope values from 1986 to 2003 at Kunming GNIP station (25°1'N, 102°40'E, 1892 m) (IAEA/WMO).

	Jan	Feb	Mar	April	May	June	July	Aug	Sept	Oct	Nov	Dec
Average Temperature (°C)	8.9	10.8	14.3	17.7	19.1	20.3	20.0	19.8	18.2	16.2	12.3	9.0
Average Precipitation (mm)	19.4	13.7	16.8	20.9	97.2	191.2	209.2	194.6	107.9	84.2	34.8	11.2
$\delta^{18}O$ (‰VSMOW)	-4.32	-3.15	-3.51	-3.01	-5.42	-8.16	-12.37	-13.1	-10.75	-10.49	-8.58	-5.15

Average weighted $\delta^{18}O$ (‰VSMOW) = -9.86 .

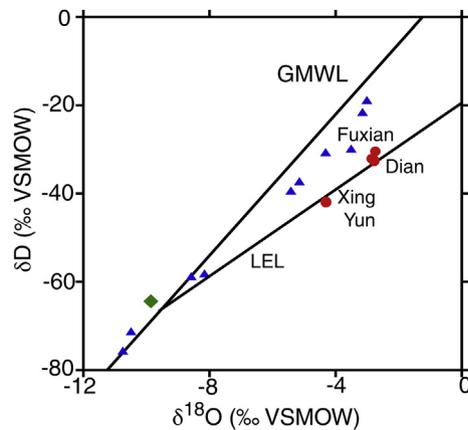


Fig. 2. Water isotope samples from selected lakes in Yunnan (red circles), monthly precipitation isotope values (blue triangles), and the annual weighted mean of modern rainfall for Kunming (green diamond). GMWL = Global Meteoric Water Line. LEL = Local Evaporation Line. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

years BP (Sun et al., 1986). Early habitation sites and associated metal artifacts are understudied relative to later, more abundant sites associated with the Dian culture of 400 BC–100 AD (Chiu-Peng, 2008, 2009). As a result, little is known about agriculture and metalworking activities during the period of earliest habitation, especially around Kunming, the capital of Yunnan. A burial site, Lijiashan, associated with the Dian culture is located within the watershed of Xing Yun (Higham, 1996). After the Dian people, various kingdoms and cultural traditions arose around Kunming, operating independently from the Chinese dynasties of the time. This independence limited the number and availability of historical records (Allard, 1998), though it is clear that Yunnan's rich natural resources were of interest to the Han Dynasty (Yang, 2009). A longstanding industrial site, the Gejiu silver and copper mine, 100 km southeast of Xing Yun, has been active for the past 2000 years (Cheng et al., 2012).

Yunnan became well-known for its silver and copper deposits by the time of the Yuan dynasty (1271–1368 AD) and from then on, was formally part of China (Yang, 2009). Mining and metal resource extraction became an integral part of the Yunnan economy, but the impact of this activity on the landscape was not documented. Numerous studies have focused on recent changes in water and sediment quality (Liu et al., 2010; Zhang et al., 2010) and have attempted to document the changes associated with modern human disturbance through analysis of lacustrine sediment cores (Brenner et al., 1991; Whitmore et al., 1994a). However, bulk sediment radiocarbon measurements and hard-water reservoir effects made it difficult to constrain the timing of observed geochemical changes, thereby limiting this earlier work. Few studies of lake sediment cores from around Kunming have expressly examined the longer term effects of anthropogenic disturbance.

3. Materials and methods

3.1. Core collection

As part of this study, four overlapping cores from Xing Yun were collected in 2009 from the deepest part of the lake (11 m), forming a composite record of 317 cm. Core D-1 was recovered using a 5.5-cm diameter piston core with a removable polycarbonate tube and measures 133.5 cm. A steel barrel Livingston corer (Wright et al., 1984) was used to collect three overlapping cores (D-2, D-3, and D-4) from below this depth. These three Livingston sections were extruded in the field and measure 92 cm, 96 cm, and 63 cm, respectively. An Aquatic Research percussion core measuring 75 cm was taken from the same location, and the upper 56 cm were extruded in the field at 0.5-cm intervals.

3.2. Age control

Radiocarbon ages were measured on seven terrestrial macrofossils and three gastropod shells. Samples were analyzed at Keck Center for Accelerator Mass Spectrometry at the University of California Irvine. Prior to analysis, samples were pretreated using a standard acid, base, acid procedure (Abbott and Stafford, 1996). The resulting ages were calibrated using CALIB 6.0 and the INTCAL09 calibration curve (Reimer et al., 2009). The upper 11 cm of the gravity core were lyophilized and analyzed for ^{210}Pb and ^{214}Pb activities by direct gamma (γ) counting in a broad energy germanium detector (Canberra BE-3825) at the University of Pittsburgh.

3.3. Analytical methods

Water content and bulk density were measured at 2 cm intervals using 1 cm³ samples. Weight percent organic matter and carbonate content within these same samples was determined by loss-on-ignition (LOI) analysis at 550 °C and 1000 °C, respectively (Dean, 1974).

Weight percent nitrogen, weight percent organic carbon, $\delta^{15}\text{N}_{\text{org}}$, $\delta^{13}\text{C}_{\text{org}}$, and atomic C/N ratio were measured at 2-cm intervals. Samples were covered in 1 M HCl for 24 h to dissolve carbonate minerals and rinsed. Samples were then lyophilized and analyzed at Idaho State University using an ECS 4010 (Elemental Combustion System 4010) interfaced to a Delta V mass spectrometer through the ConFlo IV system. Organic carbon isotopes are expressed in conventional delta (δ) notation as the per mil (‰) deviation from the Vienna Pee Dee Belemnite standard (VPDB) whereas nitrogen isotopes are reported relative to atmospheric N₂.

Cores were sampled continuously at 0.5-cm intervals for analysis of the oxygen and carbon isotopic composition of carbonate minerals. Samples were disaggregated with 7% H₂O₂ and sieved through a 63- μm screen to remove biological carbonates derived from ostracod and gastropod shells. Samples were soaked in a 50% bleach and 50% DI water mixture for 6–8 h, rinsed, and lyophilized. Bulk carbonate samples were reacted in ~100% phosphoric acid at 90 °C and measured using a dual-inlet GV Instruments, Ltd. (now Isoprime, Ltd) IsoPrime™ stable isotope ratio mass spectrometer and MultiPrep™ inlet module at the University of Pittsburgh. Oxygen and carbon isotope results are expressed in conventional delta (δ) notation as the per mil (‰) deviation from VPDB. One sigma analytical uncertainties are within $\pm 0.10\%$. Replicate measurements of $\delta^{18}\text{O}_{\text{carb}}$ and $\delta^{13}\text{C}_{\text{carb}}$ of Xing Yun sediment had an average standard deviation of 0.10 and 0.20‰ (VPDB), respectively.

Metal concentrations were measured at 6-cm intervals between 317 cm and 53 cm; above 53 cm depth, samples were measured at 1–3 cm intervals. All samples were lyophilized and homogenized prior to analysis. Elements were extracted by reacting samples with 10 mL of 1 M HNO₃ for ~12 h (Graney et al., 1995). The supernatant was extracted and diluted before measurement on an inductively coupled plasma mass spectrometer (ICP-MS); samples below 260 cm depth were measured on a Perkin–Elmer NeXION 300x ICP-MS at the University of Pittsburgh and samples above 260 cm were measured on a Perkin–Elmer Sciex Elan 6000 ICP-MS at the University of Alberta. Duplicates were run on every 10th sample and were within 5% of each other. Blanks were run every 10 samples and were consistently below our measurement range of interest. Measurement of total Hg was undertaken using a DMA80 direct mercury analyzer at Yale University. Standard reference materials (Mess-3) included in each run were within 10% of certified values.

3.4. Interpretive model

Weight percent carbon, when paired with $\delta^{13}\text{C}_{\text{org}}$, provides information on primary productivity and the organic carbon cycle of the lake and when interpreted in the context of carbon/nitrogen (C/N) ratios, can indicate the source of the organic matter (terrestrial or aquatic) (Meyers, 1994). Terrestrial organic matter typically has higher values of C/N (>14) while aquatic organic matter has lower values of C/N (<10) (Meyers and Lallier-vergues, 1999). Weight percent nitrogen, $\delta^{15}\text{N}_{\text{org}}$, and the ratio of nitrogen/phosphorus (N/P) are used to infer trophic status since eutrophic lakes and lakes with a high percentage of agricultural and sewage runoff have low N/P ratios (<6) (Downing and McCauley, 1992).

We interpret $\delta^{18}\text{O}_{\text{carb}}$ to primarily reflect changes in the lake water balance. Xing Yun water samples suggest that a large portion of lake water is lost through evaporation (Fig. 2), so lake water isotopes should reflect the balance between precipitation and evaporation (P/E). When lake levels rise or fall, the bathymetric characteristics of the lake (volume, surface area, etc.) change and alter the proportion of water lost through evaporation, which in turn alters the isotopic composition of the remaining lake water (Leng and Marshall, 2004; Steinman et al., 2010b; Steinman and Abbott, 2013). Authigenic calcite minerals that form in the Xing Yun water column (and settle to the lake bottom) archive the lake water isotopic composition. By isolating the authigenic calcite and analyzing its isotopic composition, we can estimate how lake levels have changed through time.

Stable isotopes in speleothem carbonate at Dongge Cave in the Guizhou province, 530 km northeast of Xing Yun (Wang et al., 2005) (Fig. 1), were used to reconstruct variations in the strength of the ASM over the Holocene. We use the Dongge Cave record as our primary comparative dataset because of the cave's proximity to Xing Yun, the similarity in climate (Tables 1 and 2), and the well-dated, high-resolution nature of the record.

Table 2

Monthly average temperature, precipitation, and oxygen isotope values at Dongge Cave (25°17'N, 108°50'E, 680 m) (Dykoski et al., 2005).

	Jan	Feb	Mar	April	May	June	July	Aug	Sept	Oct	Nov	Dec
Average Temperature (°C)	5.2	9.9	11.3	16.3	19.7	22.1	23.5	22.8	20.8	16.4	11.4	7.2
Average Precipitation (mm)	29.8	36.7	55.7	135.7	237.8	305.4	324.2	292.3	124.8	116.4	61.8	32.5
$\delta^{18}\text{O}$ (‰VSMOW)	-4.72	-3.45	-4.47	-2.82	-5.16	-7.99	-12.42	-9.67	-12.48	-10.06	-6.99	-5.43

Average weighted $\delta^{18}\text{O}$ (‰VSMOW) = -8.33.

We interpret P concentration to represent nutrient loading to the lake and concentrations of Al, Pb, and Hg to represent land-use change associated with erosion. The size of the lake and catchment area make it unlikely that Pb and Hg could accumulate in the lake solely through atmospheric deposition. However, mining and metalworking activities likely contributed to metal loadings on the landscape through atmospheric transport and they may have entered the lake system via erosion resulting from agricultural activities.

4. Results and discussion

4.1. Core chronology

Radiocarbon ages were measured on seven terrestrial macrofossils and three gastropod shells, the latter of which provide an estimate of the hard-water effect. Dates from the gastropods are approximately 1100 years older than the terrestrial macrofossil ages (Table 3). This offset is consistent with the radiocarbon reservoir age previously noted in Xing Yun and other Yunnan lakes (Whitmore et al., 1994a; Hodell et al., 1999; Xu and Zheng, 2003). The upper 11 cm of the percussion core were dated using the constant rate of supply (CRS) ^{210}Pb age model method (Table 4) (Appleby and Oldfield, 1983). An age-depth model for the Xing Yun sediment record was developed using the ^{210}Pb ages and calibrated macrofossil radiocarbon dates (Fig. 3). A third order polynomial was used to produce an age model using the *clam* 2.1 code (Blaauw, 2010) in the statistical software package “R” (R Development Core Team, 2008). Uncertainty (2σ) associated with the age-depth model is generally limited to ± 60 years. The composite record of 317 cm spans 3500 years.

4.2. Sedimentology

Two distinct sedimentological units characterize the Xing Yun sediment record. Unit I, (317–200 cm) spans 1500 BC–500 AD and is composed of ~50% carbonate, 40% mineral matter, and 10% organic matter (Fig. 4 and S1). The transition from Unit I into II (200–157 cm) spans 500–1000 AD and is characterized by a

Table 3

AMS radiocarbon dates for samples from Xing Yun.

UCI number	Composite core depth (cm)	Sample type	^{14}C age (BP)	\pm	Median probability calibrated age (yr AD/BC)	2σ calibrated age range (yr AD/BC)
71481	35.5	Wood	110	25	1838	1954–1682
71482	37.5	Charcoal	110	50	1827	1954–1673
84722 ^a	42.5	Gastropod Shell	1245	20	739	863–685
71483	59	Charcoal	130	25	1826	1953–1677
84866	145	Charcoal	1060	25	988	1022–898
84723 ^a	222.5	Gastropod Shell	3000	25	-1252	-1130 to -1372
84867	250	Wood	2105	20	-129	-54 to -191
84724 ^a	259.5	Gastropod Shell	3285	20	-1566	-1508 to -1615
84815	263	Charcoal	2220	20	-277	-204 to -378
122325	310.5	Charcoal	3070	15	-1350	-1299 to -1407

^a Denotes date excluded from age model due to reservoir effect.

marked increase in residual mineral content, decrease in carbonate, and little change in organic matter. Unit II (157–0 cm) spans 1000 AD-present and mainly consists of iron-rich red clays and silts that are low in both calcium carbonate (<10%) and organic matter (10–20%) content; the color and composition of these Unit II sediments match closely with catchment soils. These sedimentological units also broadly correspond to geochemical changes.

Samples taken at even intervals within Units I and II were analyzed using X-ray diffraction (XRD). XRD analysis identified calcite as the primary carbonate mineral in all the samples. We calculated the theoretical $\delta^{18}\text{O}$ value of calcite using the equation: (Kim and O'Neil, 1997)

$$1000 \ln \alpha(\text{Calcite} - \text{H}_2\text{O}) = 18.03 \left(10^3 T^{-1} \right) - 32.42$$

where $T = 20$ °C (the mean June, July, August temperature of Kunming). Given the observed $\delta^{18}\text{O}$ value of -4.3‰ VSMOW from the water sample collected in 2009, the $\delta^{18}\text{O}$ value of calcite precipitated during the summer should be -6.5‰ VPBD. This value is very close to the $\delta^{18}\text{O}$ value of calcite in the surface sediments (-6.2‰), supporting our assertion that the calcite is authigenic and that it precipitates in isotopic equilibrium with the lake water.

4.3. Geochemistry

4.3.1. Unit I- Pre-human disturbance

Within Unit I, some of the proxies (%C, %N, C/N, N/P, $\delta^{13}\text{C}_{\text{org}}$, $\delta^{15}\text{N}_{\text{org}}$, and Al) exhibit slight shifts, particularly from 280 to 240 cm (680 BC–5 AD) (Fig. S1). This time period corresponds with the Dian culture (400 BC–100 AD); these slight changes may indicate early, but minimal, land use change. However, in the context of the full record, these geochemical variations are minimal and are not accompanied by any sedimentological changes (e.g., sediment color or % residual mineral matter). Since many of the other proxies are stable prior to 500 AD, we focus our attention on the last 2000 years (Fig. 4). Trace metal concentrations are steady and low in this interval (P_{average} : 600 ± 30 $\mu\text{g/g}$; Pb_{average} : 10 ± 1 $\mu\text{g/g}$; Hg_{average} : 20 ± 1 ng/g). Notably, the oxygen isotopic composition of the authigenic calcite is highly stable, averaging -8.2 ± 0.2 ‰. This is ~1‰ lower than the average $\delta^{18}\text{O}$ value (-7.4 ± 0.2 ‰ over the past 3500 years) of a speleothem from Dongge Cave (Wang et al., 2005). The difference between the Xing Yun and Dongge Cave $\delta^{18}\text{O}$ values can be explained by disparity in the annual weighted $\delta^{18}\text{O}$ means of modern rainfall in these two regions, which are offset by ~1.5‰ (Tables 1 and 2). This suggests that Xing Yun was an overflowing, hydrological open system prior to 500 AD and that the calcite precipitated during this time primarily records the oxygen isotopic composition of regional rainfall (with secondary influence by lake water temperature) (Kim and O'Neil, 1997; Leng and Marshall, 2004). This portion of the record agrees well with previous analyses of Xing Yun $\delta^{18}\text{O}_{\text{carb}}$ values, when accounting for the aforementioned reservoir effect of 1100 years (Hodell et al., 1999).

Our results indicate that anthropogenic impact on the lake was relatively limited prior to ~500 AD. This is despite a well-known burial site associated with the Dian culture within Xing Yun's

Table 4
Down-core ^{210}Pb activities, ^{214}Pb activities, cumulative weight flux, and constant rate of supply (CRS) sediment ages.

Depth (cm)	^{210}Pb activity, $\text{Bq}\cdot\text{g}^{-1}$	1σ error ^{210}Pb activity	^{214}Pb activity, $\text{Bq}\cdot\text{g}^{-1}$	1σ error ^{214}Pb activity	Cumulative weight flux, g cm^{-1}	CRS age (yr AD/BC)	1σ error age
0.5–1.0	0.2500	0.0335	0.1120	0.0097	0.09	2008	2.08
1.0–1.5	0.2270	0.0318	0.0745	0.0087	0.15	2006	2.14
3.0–3.5	0.2180	0.0343	0.0767	0.0088	0.46	1996	2.39
4.0–4.5	0.2020	0.0280	0.0660	0.0072	0.66	1988	2.64
5.0–5.5	0.1770	0.0249	0.0583	0.0064	0.91	1976	2.98
6.0–6.5	0.1140	0.0170	0.0623	0.0057	1.09	1971	3.19
7.0–7.5	0.1310	0.0163	0.0614	0.0048	1.28	1963	3.67
8.0–8.5	0.1100	0.0158	0.0656	0.0053	1.50	1954	4.10
9.0–9.5	0.0998	0.0134	0.0663	0.0048	1.70	1947	4.62
10.0–10.5	0.1030	0.0128	0.0672	0.0047	1.92	1936	5.68
11.0–11.5	0.0876	0.0091	0.0556	0.0048	2.15	1919	8.22

watershed (Higham et al., 2011) and historical indication of rice agriculture in Yunnan since the Han Dynasty (ca 200 BC) (Herman, 2002). This also contrasts with a previous paleolimnological study of Erhai Lake, located ~300 km to the northwest of Xing Yun, which found palynological evidence for landscape modification as early as the middle Holocene (Deearing et al., 2008). Archaeological and historical evidence indicate that the political and cultural traditions around Xing Yun were different from those of Erhai, near which there are no Dian burial sites (Higham, 1996; Elvin et al., 2002).

4.3.2. Transition period

The transition from Units I to Unit II, which spans the period from 500 to 1000 AD, is marked by substantial shifts in most of the proxies (Fig. 4). Percent carbon and nitrogen roughly double before declining, and $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ shift briefly towards more negative values before increasing by 1‰ and 2‰ respectively. Additionally, gradual increases in the $\delta^{18}\text{O}_{\text{carb}}$ and $\delta^{13}\text{C}_{\text{carb}}$ values occur at the beginning of this transition. Around 800 AD, P, Pb, and Hg concentrations roughly double while Al concentrations increased by ~3-fold. These geochemical changes occur in conjunction with increased mineral matter and decreased carbonate content. A plot of $\delta^{18}\text{O}$ values versus concentrations of phosphorus, aluminum, lead, and mercury (Fig. S2) reveal that changes in the concentrations of metals occurred independently from changes in lake level. The change in water level from 500 to 800 AD did not cause a significant change in metal concentrations. In particular, we note the transition across $\delta^{18}\text{O}$ values of -7‰

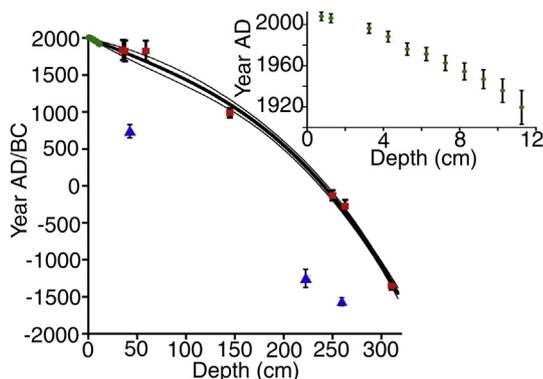


Fig. 3. Age–depth model and 95% confidence intervals. ^{210}Pb (green circles), radiocarbon (red squares), gastropod shell dates (blue triangles), all with 2 sigma error bars. Gastropod shell dates exhibit a relatively consistent 1100 year offset from terrestrial macrofossils although this could change over time. ^{210}Pb age model detail is displayed in the inset. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

where metal concentrations increase while lake level remains relatively constant. This occurs from 800 to 1120 AD and demonstrates that the increase in metals at this time is unrelated to lake level.

The transition between Units I and II exhibits many of the biogeochemical fingerprints commonly ascribed to eutrophication. For example, a two-fold increase in P, which limits phytoplankton growth in many lakes, is contemporaneous with decreases in %C and N/P ratio and increases in $\delta^{13}\text{C}_{\text{org}}$ and $\delta^{15}\text{N}_{\text{org}}$. Lakes heavily impacted by eutrophication typically have high fertilizer and sewage inputs, which are characterized by positive $\delta^{15}\text{N}_{\text{org}}$ values (Brenner et al., 1999), high sediment $\delta^{13}\text{C}_{\text{org}}$ values, and low N/P ratios (Schindler et al., 2008). Our conclusion is further supported by a previous analysis of the diatom community composition that documented an increase in the proportion of hypereutrophic and eutrophic diatom species (from 60 to >80%) during this transition (Whitmore et al., 1994b). However, the timing of this transition in the previous study was thought to be around 2500 years BP. Bulk sediment radiocarbon measurements combined with large hard-water reservoir effects led to limited age control; the results presented here demonstrate that this transition actually occurred >1000 years later.

The gradual increase in $\delta^{18}\text{O}_{\text{carb}}$ values likely resulted in part from catchment water diversion for agriculture and a consequent decrease in water throughflow to the Ge River leading to longer residence times and increased enrichment of the heavy isotopes of oxygen by evaporation. While Dongge Cave records an abrupt and brief decrease in ASM strength at 400 AD (Wang et al., 2005), the gradual, prolonged nature of the increase in $\delta^{18}\text{O}_{\text{carb}}$ values at Xing Yun, the difference in timing, and the other geochemical changes that co-occurred with this shift suggests that a decrease in the strength of the ASM likely had little influence on the drop in lake level. Combined, these data indicate that intensive land-use within Xing Yun's watershed, probably dominated by agriculture activities, began between 500 and 1000 AD. The lagged increase in Al, P, Pb, and Hg suggests that initial land-use activities were limited in scope, and were not aimed at exploiting mineral resources within Xing Yun's watershed.

The onset of intensive land-use – as indicated by the rapid deposition of red, iron-rich clays, decrease in carbonate, and increase in P, Pb, and Hg concentrations – occurred soon after the regional establishment of the Nanzhao Kingdom (~700 AD) (Fig. 4). This sedimentological transition was noted in the previous study of Xing Yun, but was dated to 1000 BC (Hodell et al., 1999) because of the large uncorrected hard-water reservoir effects. Studies of other Yunnan lakes, including Qilu (Brenner et al., 1991) and Dian (Sun et al., 1986) (Fig. 1), also noted this transition and attributed it to the onset of 20th Century industrialization, though these studies acknowledged that the timing of this transition was unclear. Our more accurately dated sediment record places this transition into a more reliable temporal context. Additionally, the timing of this matches quite closely with a period of enhanced erosion and landscape disturbance at Erhai (Shen et al., 2005). The characteristics of Xing Yun sediments from this time, namely, a low organic carbon content and high residual mineral matter content composed primarily of catchment soils, are similar to those from Guatemalan lakes known to have been impacted by human deforestation and erosion (Brenner, 1983; Binford et al., 1987) supporting our hypothesis that the transition from Unit I to II was caused by anthropogenic activities.

4.3.3. Unit II- intensive human disturbance

The ~100 years (1270–1380 AD) that include the Yuan Dynasty, better known as the Mongols, are marked by a clear departure from the last ~700 years in many of the proxies (Fig. 4). In general,

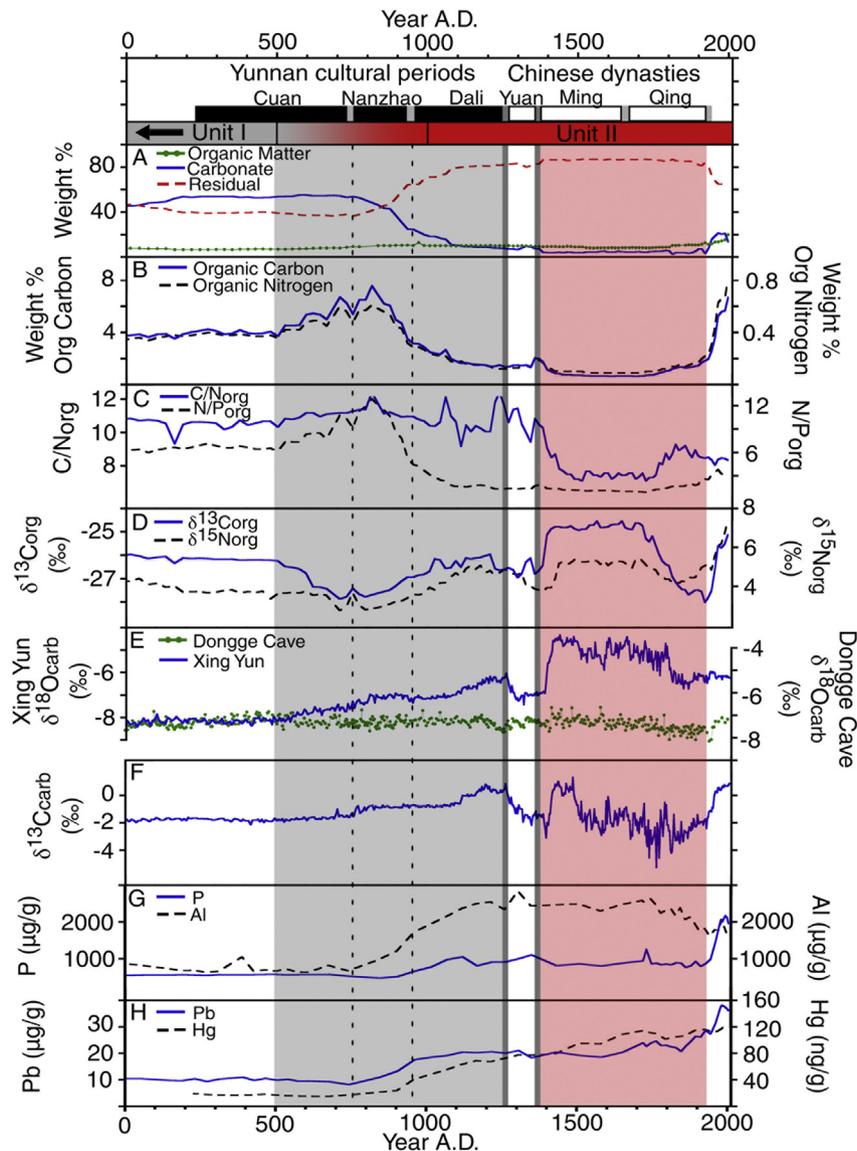


Fig. 4. A – Weight percent organic matter (green circles), calcium carbonate (blue solid line), and residual mineral matter (red dotted line); B – Percent organic carbon (blue solid line), percent organic nitrogen (black dotted line); C – Carbon to nitrogen ratio (blue solid line) of organic matter, nitrogen to phosphorus ratio (black dotted line); D – Carbon isotope values of organic matter (blue solid line), nitrogen isotope values of organic matter (black dotted line); E – Oxygen isotopic composition of Xing Yun calcite (blue solid line), Dongge Cave (green circles) (Wang et al., 2005); F – Carbon isotopic composition of Xing Yun calcite; G – Concentration of phosphorus (blue solid line), aluminum (black dotted line); H – Concentration of lead (blue solid line) and mercury (black dotted line). The gray bar is the initiation of human disturbance on the lake; the dotted lines are the establishment and decline of the Nanzhao Kingdom; the white with gray bars are the establishment and decline of the Yuan Dynasty; the red bar is the establishment and decline of the Ming and Qing Dynasty. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

$\delta^{18}\text{O}_{\text{carb}}$, $\delta^{13}\text{C}_{\text{org}}$, and $\delta^{15}\text{N}_{\text{org}}$ values are lower and %C and C/N values are higher than in sediment from adjacent time periods. These shifts occur over several decades or less and are not matched by changes in either regional land-use or mineral extraction, as indicated by stable inputs of Al, P, Pb, and Hg, or ASM intensity, as recorded by the Dongge Cave $\delta^{18}\text{O}$ record (Wang et al., 2005). We suggest that these changes reflect lake-level management during this period; historical data confirm that the Mongols implemented a series of regional political changes, including the construction of dams and artificial reservoirs near Kunming largely for transportation purposes (Herman, 2002). Lower $\delta^{18}\text{O}_{\text{carb}}$ values therefore likely reflect some combination of higher lake levels and/or greater water throughflow at Xing Yun during this time.

Wet-rice agriculture, which requires the routine flooding and drying of flat fields (Bray, 1986), became widespread in Yunnan

shortly after the Ming defeated the Mongols in 1368 AD (Herman, 2002). Trenching of the Ge River to control Xing Yun water levels took place multiple times during the Ming and early Qing Dynasties, resulting in a water level drop of ~4 m (Wang, 1994). Our sediment core suggests that these changes in regional cultural activity at the Mongol-Ming transition had important limnological consequences. Sediment %C, C/N, N/P, $\delta^{18}\text{O}_{\text{carb}}$, and $\delta^{13}\text{C}_{\text{org}}$ rapidly shift to values (1%, 8, 1, -4‰, and -25‰, respectively) that are unprecedented within the sediment record (Fig. 4). The low %C, C/N, and N/P values indicate eutrophic conditions, and high Al, P, and $\delta^{15}\text{N}_{\text{org}}$ values suggest that soil erosion, manure, and sewage inputs supplied nutrients to the lake. The combination of lower lake levels and eutrophic conditions likely accelerated the remineralization of autochthonous organic matter in this broad shallow lake, limiting organic carbon preservation.

This time period also encompasses the Little Ice Age, which was characterized by a weakened ASM and decreased precipitation across much of southwestern China (Morrill et al., 2003). Changing climate during this period may have reduced catchment water availability (leading to lower lake levels) and exacerbated eutrophication. The Dongge Cave record indicates an abrupt and brief drop in the strength of the ASM at 1500 AD (Wang et al., 2005). However, the drop in ASM strength occurs slightly later and is much shorter in duration than the changes in the Xing Yun record. Since historical records confirm intentional manipulation of lake level (Wang, 1994), and since other geochemical changes (C/N, N/P, $\delta^{13}\text{C}_{\text{org}}$, and $\delta^{15}\text{N}_{\text{org}}$) occur with the shift in $\delta^{18}\text{O}_{\text{carb}}$ values, we attribute the majority of lake-level change to human activity, including catchment water diversion for agriculture. The high variability in $\delta^{18}\text{O}_{\text{carb}}$ and $\delta^{13}\text{C}_{\text{carb}}$ values during this period suggests that the lake was more hydrologically closed, lake levels were likely on average lower than during prior periods (Steinman et al., 2010a), and that lake level and water use were managed. This rapid shift to more positive $\delta^{18}\text{O}_{\text{carb}}$ values is present in the earlier study of Xing Yun, but the low sampling resolution makes the exact timing difficult to discern, and the large hard-water effect makes it appear ~1000 older (Hodell et al., 1999).

The onset of wet-rice agriculture and lake-level management during this time period was likely enhanced by regional population expansion within Yunnan as more Han Chinese immigrants moved into the province to work in the mines (Durand, 1960). The intensification of erosional activity is reflected in the Xing Yun sediment record as the input of both Pb and Hg remained high during the Ming Dynasty (Fig. 4), which (though it spanned only ~300 years), accounts for 28%, 28%, and 32% of the total P, Pb, and Hg deposited in the lake, respectively (Fig. 5).

Substantial biogeochemical shifts also occurred in Xing Yun shortly after the transition from the Ming to Qing Dynasty (1644 AD). The rate of carbon burial increased and coincided with a decrease in extractable Al, suggesting decreased erosion and landscape stabilization (Fig. 4). Changes in the C/N and N/P ratios and decreases in $\delta^{13}\text{C}_{\text{org}}$ and $\delta^{15}\text{N}_{\text{org}}$ may indicate a reduction of eutrophic conditions due to agricultural and water management changes. This is reflected in the shift to lower $\delta^{18}\text{O}_{\text{carb}}$ values, and presumably higher lake levels, approximately 75 years after the shifts in other variables. Once again, the rapid nature of this transition and the lack of a clear connection to any change in ASM strength (Wang et al., 2005) suggest that this rise in lake level was likely driven by human activities. The total pollution index for P, Pb, and Hg reached the highest levels during this time period, indicating the intensification of land use change and/or metal resource extraction. The Qing Dynasty accounts for 31%, 34%, and 39% of total P, Pb, and Hg, respectively, deposited in the lake (Fig. 5).

Post-Qing sediments (1911 AD-present) are characterized by the highest $\delta^{13}\text{C}_{\text{org}}$ (–25‰) and $\delta^{15}\text{N}_{\text{org}}$ (7‰) values, increases in %C and %N, and high inputs of Al, P, Pb, and Hg (Fig. 4). Xing Yun entered a unique state during the 20th century; however, in the context of entire sediment record, the total pollution index for P, Pb, and Hg is relatively low compared to previous time periods (Fig. 5). The 20th century accounts for only 18%, 18%, and 16% of total P, Pb, and Hg, respectively, deposited in the lake. Thus, while the rate of current change in Xing Yun water and sediment quality is unprecedented, the current level of pollution is not, indicating that modern increases in eutrophication and water quality degradation are part of a much longer legacy of human alteration to the Xing Yun lake catchment system.

5. Conclusions

The 3500-year, multi-proxy lake sediment record presented here reveals that substantial alterations to Xing Yun Lake and its

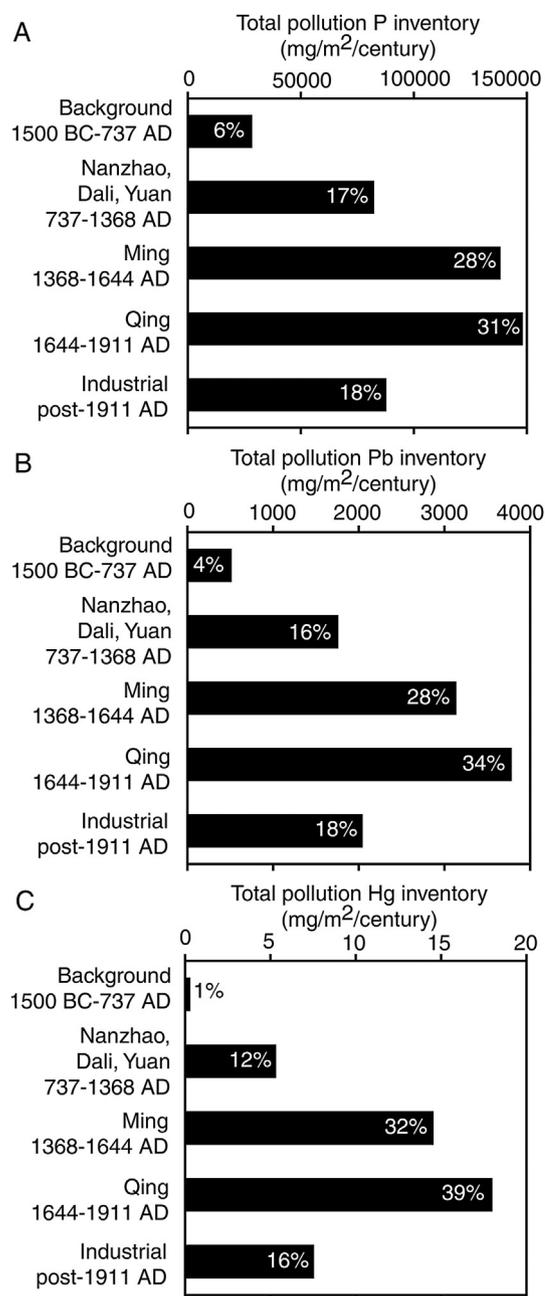


Fig. 5. Total pollution inventories for A – phosphorus (P), B – lead (Pb), C – mercury (Hg). Total P, Pb, and Hg inventories were calculated as the product of dry sediment inventory (g/m^2) and element concentration (mg/g). Concentrations within core intervals lacking data were estimated using linear interpolation between measurements. Intervals were summed and divided by time period (in centuries).

watershed co-occurred with several cultural and dynastic transitions and that the impacts of human modification on the lake and its watershed were greater than those of climate change. The record implies that each group of people altered the lake-catchment system in a different way and to a different degree. Furthermore, the often abrupt nature of these transitions suggests that anthropogenic manipulations were rapid, occurring within years to decades. Although water quality deterioration was thought to be a relatively recent phenomenon, this study indicates that human actions have profoundly affected the lake for >1500 years. The role of climatic change and variability in the strength of the ASM may have partially contributed to observed changes, but we find that the

primary driver of shifts in lake level and trophic status over the past 2000 years was anthropogenic activity. The differences among the various organic matter and geochemical proxies illustrate that, at times, nutrient loading associated agricultural activities varied independently from erosional inputs associated with other land use changes.

Our temporally higher resolution study reveals anthropogenically driven changes that earlier investigations on lakes in the Kunming region did not adequately identify due to either limited chronological constraint (Sun et al., 1986; Brenner et al., 1991; Whitmore et al., 1994b) or a lack of focus on the short-term, abrupt nature of change (Hodell et al., 1999). This study underscores the need for additional lake sediment studies in regions characterized by a historically complex and variable relationship between humans and the environment to provide context for current environmental issues.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.quascirev.2014.05.019>.

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