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ORIGINAL PAPER

Past atmospheric Pb deposition in Lake Qinghai, northeastern Tibetan Plateau

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Abstract Two short sediment cores were recovered from sub-basins of Lake Qinghai, China and were analyzed for concentrations of Pb and 16 other elements to determine historic, regional atmospheric Pb deposition on the Tibetan Plateau. Core chronologies, dating back to the eighteenth century, were established using activities of ^{210}Pb and ^{137}Cs. The 17 elements were divided into three principal components. Variations in concentrations of PC1 elements (Al, Cr, Cu, Fe, K, Mn, Ni, and Ti) demonstrate different patterns between the two cores, and are attributed to different sediment sources in the two sub-basins. PC2 elements (Ba, Ca, Na, and Sr) may be associated with the degree of catchment weathering and/or water chemistry. Four elements (Pb, Zn, P, and Co) are related to both PC1 and PC2, and reflect a mixture of natural and anthropogenic sources. The PC3 element is Mg in the north sub-basin, and is perhaps related to aragonite precipitation and/or increased farming. Elevated Pb concentrations in uppermost sediments of both cores signify a recent regional/global increase in anthropogenic Pb release into the environment. After subtracting lithogenic Pb, derived from rock weathering and/or dust and normalized to the background immobile element Ti, results suggest that excess, anthropogenic Pb is transferred to the lake and its sediments predominantly via the atmosphere. This anthropogenic atmospheric Pb is comparable in magnitude and displays similar variation patterns in the two cores, reflecting regional atmospheric deposition and local erosion. The average anthropogenic Pb deposition rate in Lake Qinghai since the 1960s has been $\sim 12.2 \pm 3.5$ mg/m^2/a, comparable with atmospheric Pb fluxes reported for sites elsewhere in the northern hemisphere.

Keywords Atmospheric Pb · Titanium · Lake sediment · Lake Qinghai · Tibetan Plateau

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Introduction

In the last few centuries, anthropogenic activities have released increasing quantities of contaminants into local environments and have significantly affected regional-scale environments, including the Arctic and the Antarctic (e.g. Boutran et al. 1994; Braune et al. 1999; Van de Velde et al. 2005). The atmosphere has been contaminated by anthropogenic lead (Pb) in varying concentrations since ~4.5 ka BP (Patterson 1971), or even longer (Hong et al. 1994; Shotyk et al. 1998). Local to regional Pb pollution

has received considerable attention, and has been used to demonstrate anthropogenic perturbation (e.g. Nriagu and Pacyna 1988; Birch et al. 1996; Brännvall et al. 2001; Hammarlund et al. 2008). Many records of atmospheric Pb from marine/lacustrine sediments, soils, peat bogs, as well as polar ice and snow have indicated varying sources and fluxes associated with smelting of ores, metallurgy, consumption of leaded gasoline, and other fuel combustion (Nriagu 1989; Hong et al. 1994; Weiss et al. 1999).

As the highest plateau on earth, the Tibetan Plateau is a remote region with low population density and few industries, but it is still subject to anthropogenic pollution (Xiao et al. 2000; Zhang et al. 2002a; Li et al. 2006). Little information, however, is available on the region's pollution history with respect to input or accumulation because observations on the plateau have been limited. Pb in lake sediment can document the timing and relative magnitude of soil erosion and atmospheric Pb pollution at local to regional scales (Farmer 1978; Kober et al. 1999; Bindler et al. 2001; Brännvall et al. 2001; Renberg et al. 2002; Yang et al. 2002). Geographically, Lake Qinghai lies between the industrialized provinces to the east and the arid Gobi Desert to the north and west, where atmospheric dust is prevalent. Thus, the Lake Qinghai sediment offers an opportunity to explore the history of natural and anthropogenic elemental deposition on the Tibetan Plateau. Here, we present a 250-year record of atmospheric anthropogenic Pb deposition from two Lake Qinghai sediment cores and assess temporal and spatial variations in Pb input and the processes that controlled deposition.

Materials and methods

Site description

Lake Qinghai (36°32′–37°15′N, 99°36′–100°47′E) has been a large, hydrologically-closed system since at least 36 ka BP (Chen et al. 1990). The lake is located at the northeastern margin of the Tibetan Plateau, with a water surface altitude of 3,194 m above present sea level (Fig. 1). It has a surface area of about 4,260 km^2 and is fed from a high alpine catchment of more than 29,660 km^2. A cold and semi-arid continental climate, sensitive to the Asian monsoons, prevails in the entire Lake Qinghai basin. The air temperature at Lake Qinghai varies from 10.4 to 15.2°C in July and from −10.4 to −14.7°C in January, averaging ∼1.2°C (1951–2005). Due to an average water depth of 22.0 m (maximum 27.5 m),

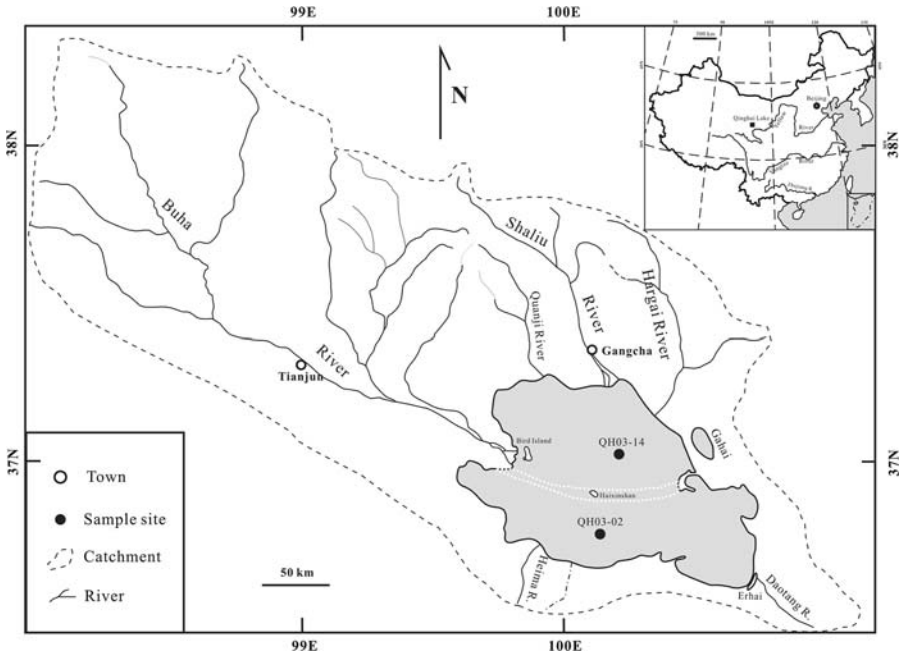

Fig. 1 Map showing the location of Lake Qinghai and core sites. *White dashed lines* show the boundary between the south sub-basin and north sub-basin (modified from An et al. 2006). *Insert map* shows the location of Lake Qinghai in China. The catchment boundary is shown as *dashed lines*

the lake becomes thermally stratified (hypolimnion <6°C, epilimnion 12–15°C) in summer and is covered by ice from late October to early April (Yan et al. 2002). The mean annual precipitation (1951–2005) is 337 mm/a, but evapo-transpiration is three to four times higher than precipitation (Sun et al. 1991).

The lake is divided into two nearly equal sub-basins by a NNW-trending piggy-back horst, from which an island (Haixinshan) emerges (An et al. 2006). Detrital input to the lake is provided mainly by five major rivers (Buha River, Shaliu River, Quanji River, Hargai River, and Heima River) and by atmospheric deposition. Our quantification of dissolved inputs suggested that the atmosphere contributes 36–57% of the total dissolved cations to river waters (Zhang et al. 2009). The impact of human influences on the Lake Qinghai basin is very limited and sediments remain undisturbed after deposition, which is confirmed by irregular laminations in both sediment cores. Such conditions are expected to prevent remobilization of metals after they are scavenged and trapped in the sediments.

Sample collection

In July 2003, two sediment cores (QH03-02, QH03-14) (Fig. 1) were recovered from the deepest points of two sub-basins in Lake Qinghai using a UWITEC gravity corer. Both retrieved cores preserved an intact sediment-water interface. Core QH03-02 (36°40′47.5″N, 100°07′19.5″E), collected from the south sub-basin at a water depth of 24.7 m, was 28.5 cm long. Core QH03-14 (37°00′59.9″N, 100°13′24.1″E) was 27.0 cm long and was from the north sub-basin, where the water depth is 26.5 m. Sediments are dominated by irregularly laminated silty clay and carbonates, indicating little bioturbation or other post-depositional disturbance. The uppermost 2–3 cm of sediment is yellow–grey clay, and the underlying 3–4 cm is grey–black clay. Sediment below 7 cm is black clay. Both sediment cores were sectioned on-site at 0.5-cm intervals. All samples were stored in a freezer at −4°C prior to chemical analysis.

Sediment dating

Samples were air-dried and weighed, and then ground to a fine powder <38 μm in an agate mortar for dating and chemical analyses. Both sediment cores were dated by measurements of ^{210}Pb and ^{137}Cs and application of the constant rate of supply (CRS) dating model (Appleby 2001). ^{210}Pb inventories were used to correct for the effects of sediment focusing. Both ^{210}Pb and ^{137}Cs activities were measured on a multi-channel γ-ray spectrometer (PerkinElmer, GWL-120-15) at the State Key Laboratory of Lake Sciences and Environment, Chinese Academy of Sciences.

Chemical analysis

Approximately 0.3 g of ground sediment was digested in 2 mL concentrated HNO_3 and 1 mL $HClO_4$ at a temperature of 90–190°C for 16 h. The residue was then dissolved in 2 mL of 4 M HCl and diluted to 10 mL with deionised water. The concentrations of 17 elements (Al, Ba, Ca, Co, Cr, Cu, Fe, K, Mg, Mn, Na, Ni, P, Pb, Sr, Ti, and Zn) in bulk sediments were analyzed using a Leeman inductively coupled plasma atomic emission spectrometer (ICP-AES) at the State Key Laboratory of Lake Sciences and Environment, Chinese Academy of Sciences. The precision of our analysis was ∼10% at the 95% confidence level. Recovery rates for elements in the international standard reference materials (NBS1645, 1646 and 2704) were around 95%.

Results

Core chronology

Excess ^{210}Pb ($^{210}Pb_{ex}$) activity shows an exponential decrease with depth in both core profiles (Fig. 2), further confirming the absence of bioturbation in these sediments. The 1986 ^{137}Cs peak is clearly visible at core depths of 2.0–2.5 cm (QH03-02) and 3.5–4.0 cm (QH03-14), respectively, but the 1963 peak is indistinct in both profiles. The exponential decrease of $^{210}Pb_{ex}$ in both sediment cores (Fig. 2) suggests that bulk sediment accumulated at a relatively constant rate in the respective sub-basins. Estimated linear mass bulk sedimentation rates were generated for both cores using the CRS model (Table 1). Core chronologies are shown on the sediment profiles in Figs. 6 and 7.

Average mass accumulation rates inferred from the ^{137}Cs and ^{210}Pb dates are 65.7 and 63.2 mg/cm^2/a

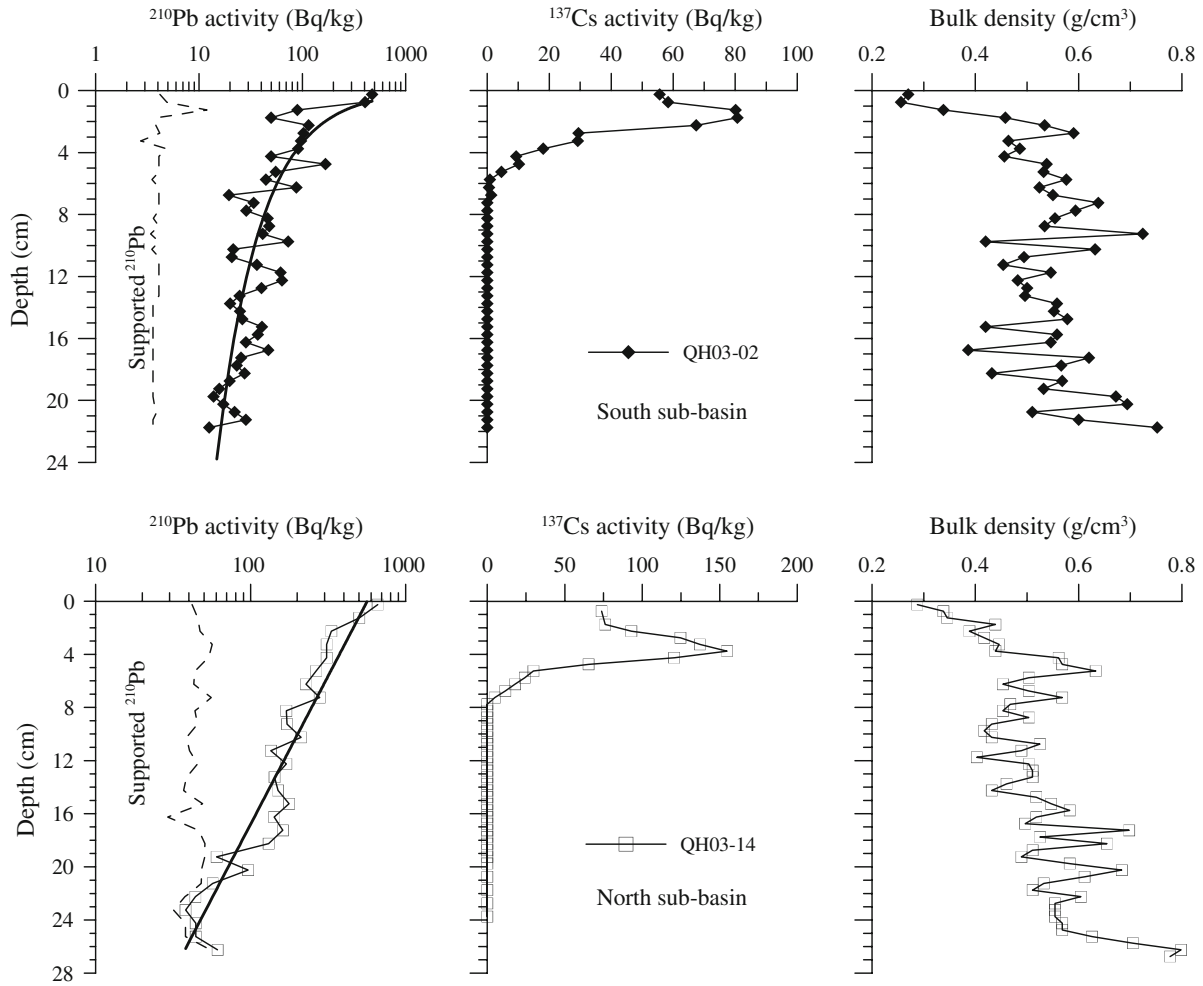

Fig. 2 Fallout radionuclide concentrations and bulk density in cores QH03-02 and QH03-14 from Lake Qinghai, showing total and supported ^{210}Pb, ^{137}Cs and bulk density of the sediments

Table 1 Core descriptions and linear mass sedimentation rates and fluxes

Core	QH03-02	QH03-14
Location	South sub-basin	North sub-basin
Latitude/Longitude	36°40′47.5″N/100°07′19.5″E	37°00′59.9″N/100°13′24.1″E
Water depth (m)	24.7	26.5
Core length (cm)	28.5	27.0
^{210}Pb age (a) (AD)	1750	1780
Sedimentation rate[a] (mm/a)	0.5–2.2 (1.13)	0.61–2.50 (1.21)
Sedimentation flux[a] (mg/cm^2/a)	12.8–147.5 (65.7)	34.5–125.9 (63.2)

[a] Numbers in parentheses are mean values

for QH03-02 and QH03-14, respectively. The ^{210}Pb dates are in good agreement with the ^{137}Cs dates. For example, at 6.50–6.75 cm depth (QH03-02) and 7.00–7.25 cm depth (QH03-14), where the first detection of ^{137}Cs activity is assumed to correspond to AD 1952 (Benoit and Rozan 2001), the CRS model yielded dates of AD 1951.6 for QH03-02 and AD 1953.2 for QH03-14. The mean linear sedimentation

Table 2 Loading results of principal component analyses (PCA) for 17 elements in sediment cores QH03-02 and QH03-14 from Lake Qinghai

Core	QH03-02			QH03-14		
PC	Factor 1	Factor 2	Factor 3	Factor 1	Factor 2	Factor 3
Al	**0.894**	0.044	–0.301	**0.943**	–0.099	–0.147
Ba	**0.628**	**–0.738**	–0.006	0.014	**–0.839**	0.258
Ca	0.421	**–0.681**	**0.526**	–0.389	**–0.583**	**0.665**
Co	**0.730**	0.249	0.273	**0.774**	–0.079	–0.019
Cr	**0.943**	–0.011	–0.139	**0.913**	–0.140	0.103
Cu	**0.701**	–0.410	0.144	**0.580**	–0.119	0.264
Fe	**0.928**	0.130	–0.255	**0.844**	–0.293	0.231
K	**0.899**	–0.037	–0.386	**0.870**	–0.240	–0.119
Mg	**–0.868**	–0.036	0.254	–0.139	0.365	**0.864**
Mn	**0.901**	–0.303	–0.029	**0.791**	–0.023	0.213
Na	–0.122	**0.827**	0.269	–0.016	0.516	0.108
Ni	**0.938**	0.152	0.042	**0.886**	0.142	0.271
P	**0.669**	0.415	0.499	0.144	**0.736**	0.489
Pb	**0.617**	0.519	0.445	0.065	**0.684**	–0.112
Sr	0.284	**–0.845**	0.373	**–0.601**	–0.517	0.508
Ti	**0.777**	0.390	–0.418	**0.853**	–0.202	–0.300
Zn	**0.685**	0.407	0.462	**0.607**	0.533	0.351

High loadings in each PC are bolded and negative are underlined. The elements related to more than one PC are highlighted by grey shadow

rates of 1.13 mm/a (QH03-02) and 1.21 mm/a (QH03-14) are close to the rates estimated by our previous measurements (Zhang et al. 2002b). The Buha River, supplying >50% of the water and sediment, feeds into the north sub-basin, resulting in a slightly faster sedimentation rate at the QH03-14 site than at the QH03-02 site, in agreement with the study by Xu et al. (2006). The two cores are dated back to AD 1750 (QH03-02) and AD 1780 (QH03-14).

Sediment geochemistry

Principal component analysis (PCA) was carried out for the element concentrations in both sediment cores to explore element associations and heavy metal origins. Three principal components (PCs) were extracted from 17 elements. The percentages of the total variance explained were 55.3% (PC1), 20.8% (PC2) and 10.6% (PC3) for QH03-02, and 42.3% (PC1), 22.8% (PC2) and 9.8% (PC3) for QH03-14.

The PCA results show that the first principal component (PC1) has high loadings of Al, Cr, Cu, Fe, K, Mn, Ni, and Ti (hereafter referred to as PC1-elements) and PC2 has Ba, Ca, Na, and Sr (PC2-elements) (Table 2). Elements Pb, Zn, P, and Co have relatively high loadings for both PC1 and PC2. The exception is Mg, which is negatively correlated with PC1-elements in QH03-02 and is an exclusive element in PC3 for QH03-14.

Down-core distributions of the PC1-elements all show similar patterns, except for Mg (Fig. 3). All these elements are relatively immobile, mainly derived from catchment rocks and atmospheric dust deposition. The large differences in patterns of all these elements between the two cores (Fig. 3) indicate different sediment sources between the two sub-basins (An et al. 2006). On the other hand, variations in element concentrations with core depth are closely tied to rock weathering and/or deposition conditions associated with regional erosion and human disturbance. Low values for most PC1-elements at around 11–14 cm and 5 cm in the QH03-02 core are attributed to extensive farming activities on the southern shore of the lake from around AD 1900 to 1920 and during the 1950s (LIGCAS 1994). A relatively constant input of Mg occurs throughout much of the recent history for both cores, with two large depositional events recorded in core QH03-02. High Mg in the topmost sediments indicates aragonite precipitation associated with global warming (Li et al. 2007). Currently, the lake is saturated with respect to carbonates.

PC2-elements (Ba, Ca, Na, and Sr: Fig. 4) are relatively mobile. These elements are associated with the degree of catchment weathering and/or water chemistry. High strontium in sediments indicates strong weathering under humid conditions (Jin et al. 2006). Variations of Na, Ba, and Ca concentrations may be a function of dilution by erosional inputs of other minerals. Increased Na in the topmost sediments indicates increased salinity related to a dramatic decline in lake level (Li et al. 2007).

Elements Pb, Zn, P, and Co vary in the two cores (Fig. 5) and have relatively high loadings for both PC1 and PC2. These elements are from multiple sources, indicating possible anthropogenic contamination of these sediments. In particular, Pb and P concentrations increased from bottom to top in both cores, especially in the uppermost part of the cores. Nevertheless, patterns of Pb and P increase differ between the two cores. We attribute these distinct patterns to respective differences in sedimentation conditions and/or sediment sources associated with erosion and human disturbance between the two sub-basins, reflected by variations in sediment geochemistry (Figs. 3, 4). However, except for the periods during which there was extensive farming on the southern shore, temporal variations of total Pb and Ca concentrations are comparable between the two sediment cores (Fig. 6). The comparable profiles provide a means (1) to develop reliable chronologies using horizons of known age, and (2) to reconstruct a record of human pollutant inputs.

Calculation of anthropogenic Pb

Similarities between variations in Pb and immobile elements before AD 1950 suggest that some Pb was

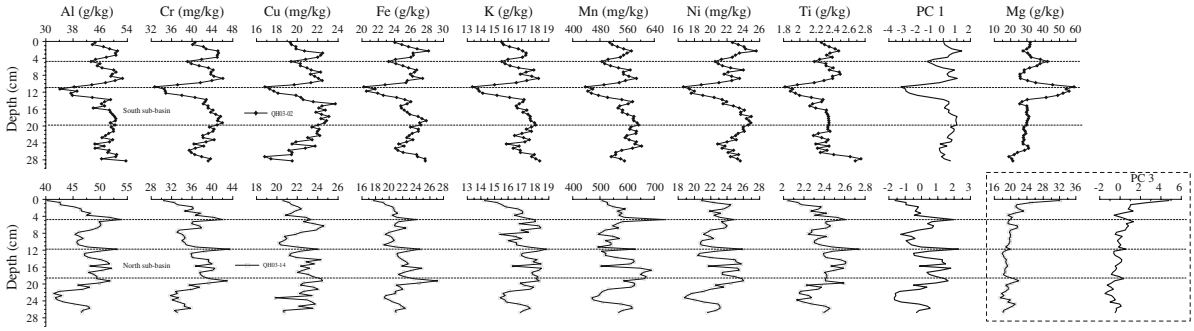

Fig. 3 Concentrations of PC1-elements (Al, Cr, Cu, Fe, K, Mn, Ni, and Ti) versus sediment depth in cores QH03-02 and QH03-14. These elements are mainly associated with catchment weathering products. *Dashed lines* show correlative variations in these elements. Variation of Mg in core QH03-02 is negatively correlated with the other PC1-elements, but Mg in QH03-14 is related to the single factor (PC3) alone

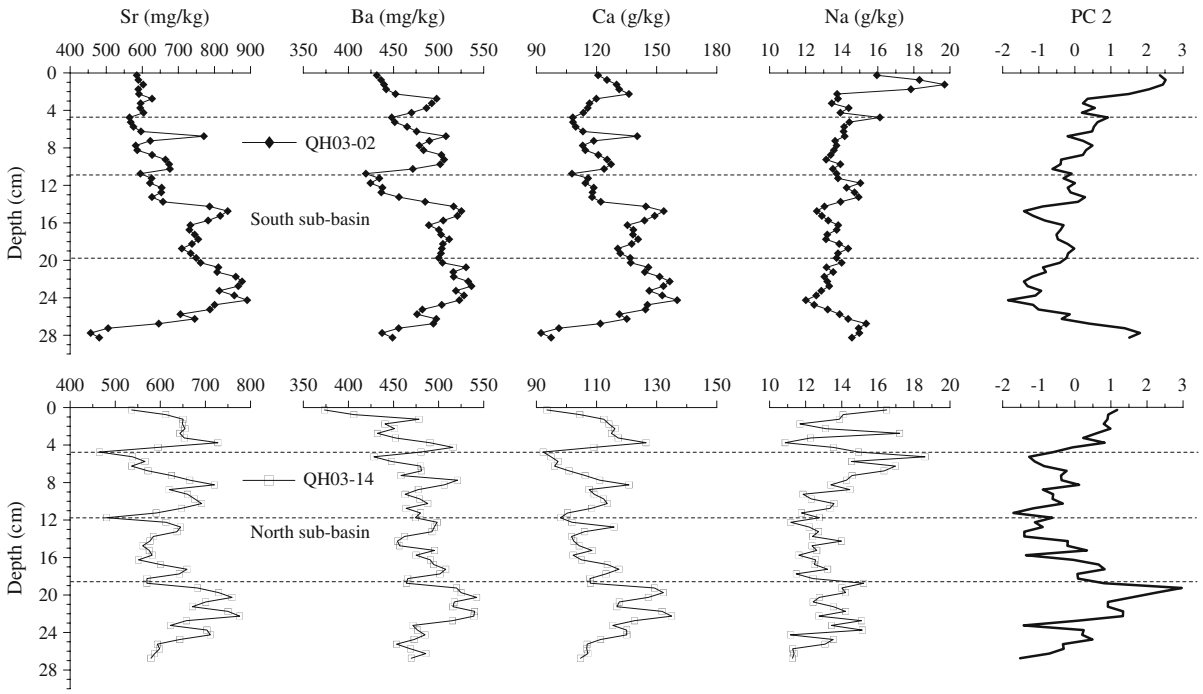

Fig. 4 Concentrations of four relatively mobile PC2-elements (Sr, Ba, Ca, and Na) versus depth in sediment cores QH03-02 and QH03-14. *Dashed lines* show correlative variations in these elements

supplied from the catchment. To account for variations in the contribution of lithogenic or 'background' Pb to the total Pb concentrations in both sediment cores, Pb concentrations were normalized to Ti, a conservative element with little or no anthropogenic input (Norton and Kahl 1987; Boyle 2001). Using Ti as the lithogenic reference element, the concentration of lithogenic Pb attributable to rock weathering and atmospheric dust was estimated as follows:

$$\mathrm{Pb}_{\mathrm{lithogenic}} = [\mathrm{Ti}]_{\mathrm{sample}} \times ([\mathrm{Pb}]/[\mathrm{Ti}])_{\mathrm{background}} \quad (1)$$

where $([Pb]/[Ti])_{background}$ is taken from the average composition of middle Holocene sediments in Lake Qinghai where [Pb] = 10.9 mg/kg, [Ti] = 2.15%. Then, the anthropogenic Pb was calculated as

$$\mathrm{Pb}_{\mathrm{anthropogenic}} = [\mathrm{Pb}]_{\mathrm{total}} - [\mathrm{Pb}]_{\mathrm{lithogenic}} \quad (2)$$

Thirdly, the calculated anthropogenic Pb concentration was converted to accumulation rate (AR) (μg/cm^2/a) as follows:

$$\mathrm{AR}_{\mathrm{Pb\,anthropogenic}} = [\mathrm{Pb}]_{\mathrm{anthropogenic}} \times R \times \rho \quad (3)$$

where R is sedimentation rate and ρ is dry bulk density of the sediment. Finally, the Pb enrichment factor (EF) was calculated as:

$$\mathrm{EF} = ([\mathrm{Pb}]/[\mathrm{Ti}])_{\mathrm{sample}} \Big/ ([\mathrm{Pb}]/[\mathrm{Ti}])_{\mathrm{background}} \quad (4)$$

Lithogenic Pb, anthropogenic Pb, and Pb EFs and fluxes for both cores were calculated (Fig. 7). The lithogenic Pb is correlated well with PC1 in the respective cores. Although the EFs are >1, calculated anthropogenic Pb exceeds the lithogenic Pb only in the topmost sediments in both sediment cores.

Discussion

The geochemistry of the Lake Qinghai sediment preserves valuable information concerning catchment weathering of source rocks, and atmospheric and anthropogenic inputs of elements to the basin. As one of the International Key Protection wetlands and a Key National Nature Reserves area, there are no large factories or mining operations within the Lake

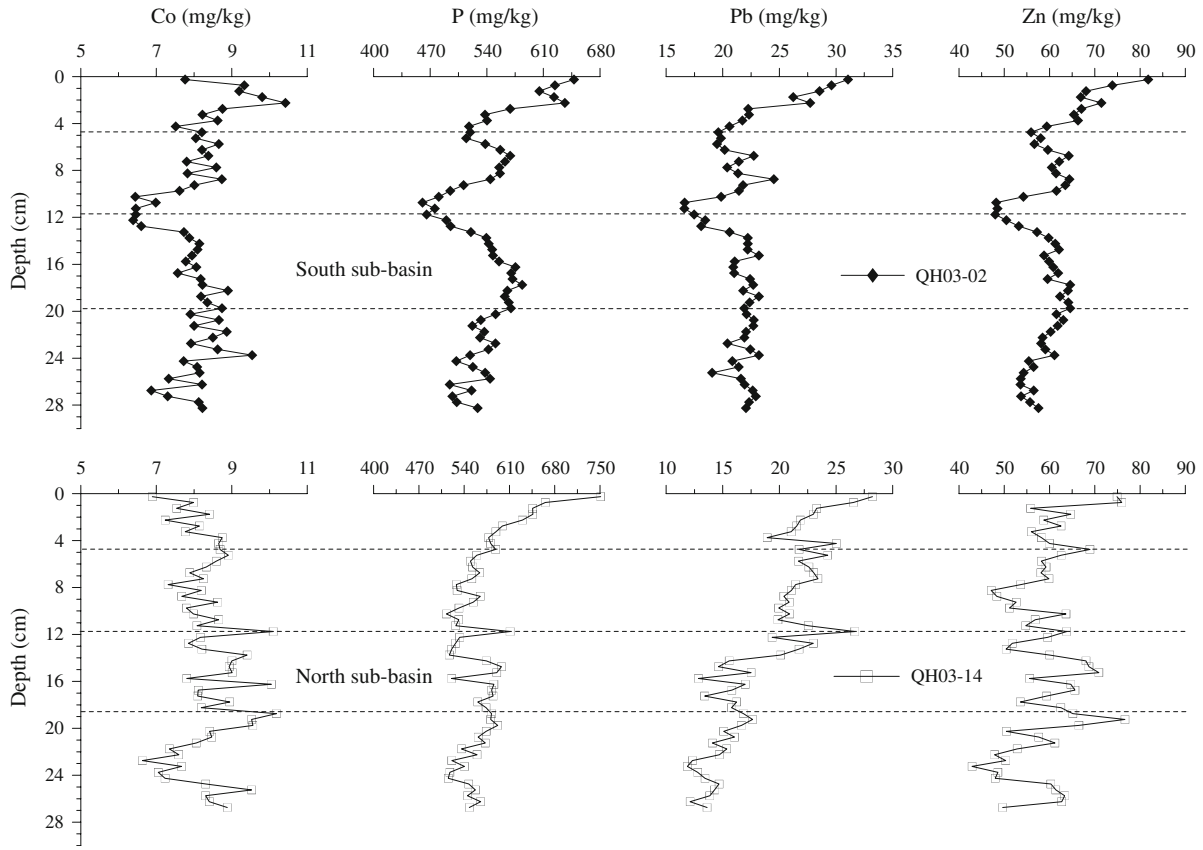

Fig. 5 Concentrations of elements (Co, P, Pb, and Zn) versus depth in sediment cores QH03-02 and QH03-14, which are related to two or more principal components (PCs). *Dashed lines* show correlative variations in these elements

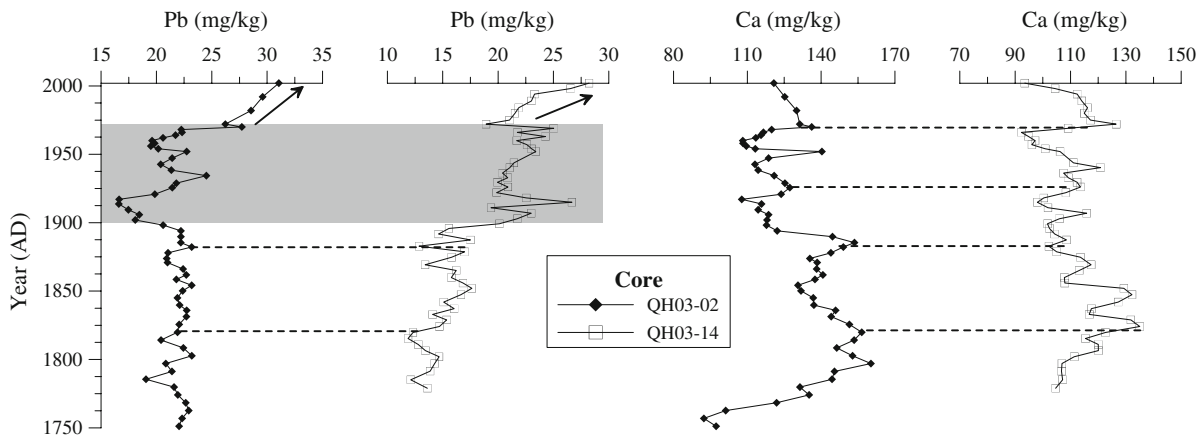

Fig. 6 Total Pb and Ca concentrations in cores QH03-02 and QH03-14. *Dashed lines* show comparable variations of Ca and Pb in the lower parts of cores from the two sites, but not for middle part (*shadowed*), which may reflect the influence of local processes (see "Discussion" in the text). *Arrows* indicate the periods with increasing Pb

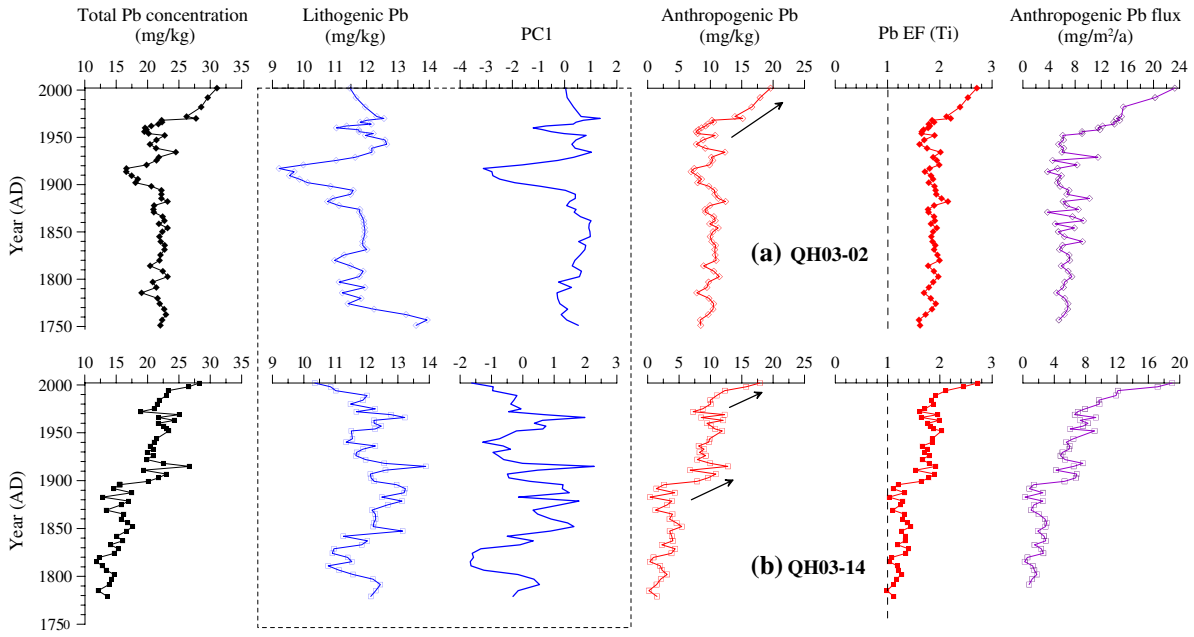

Fig. 7 Profiles of total Pb concentration, lithogenic and anthropogenic Pb, Pb EF and atmospheric flux, calculated using Ti as a reference element, relative to local background, for cores **a** QH03-02 and **b** QH03-14 from Lake Qinghai. Calculation of lithogenic and anthropogenic Pb, and Pb EFs, is described in the text. The first principal components (PC1) for the respective cores are shown for comparison with lithogenic variations. *Arrows* indicate increasing anthropogenic Pb toward the sediment surface, and during 1880–1900 in the northern sub-basin

Qinghai catchment. Because Lake Qinghai is located at a transition between arid and semiarid areas, aeolian dust is introduced to the lake annually in dry deposition, rain, and snow. Regional pollutants enter the lake via direct atmospheric deposition or runoff from the catchment.

PCA results indicate that variability in element concentrations in the two sediment cores can be expressed by three principal components that together explain 87% (QH03-02) and 75% (QH03-14) of the total variance. PC3 explains mainly the Mg variance in QH03-14. Therefore, there are two major processes controlling elemental compositions in recent Lake Qinghai sediments. Because of the different source rocks in the Lake Qinghai sub-basins, the geochemistry and sedimentation rates in the south sub-basin differ from those of the north sub-basin, as indicated by both PC1- and PC2-elements (Figs. 3, 4). PC1-elements are related to the catchment source rocks and PC2-elements are closely associated with catchment weathering, sedimentation processes and water chemistry. Supplying up to 80% of the surface runoff to Lake Qinghai (Yan et al. 2002), the north sub-basin received more sediments, mainly from the Buha and Shaliu Rivers (Fig. 1), and atmospherically-deposited nuclides, than the south sub-basin (Xu et al. 2006). This resulted in a slightly higher sedimentation rate and radionuclide (^{210}Pb and ^{137}Cs) contents in QH03-14 than in QH03-02 (Fig. 2).

Among the analyzed elements, four (Pb, Zn, P, and Co) are related to both PC1 and PC2. These elements are typical anthropogenic contaminants, and thus reflect an anthropogenic contribution to the lake. The correlation between Fe and trace metal concentrations has been used to distinguish between natural levels of trace elements and anthropogenically-enriched elements (Presley et al. 1992; Tkalin et al. 1996). Good correlation between Fe and trace metal concentrations is expected for unpolluted sediments (Presley et al. 1992). The correlation coefficient matrix between Fe and some metals (Al, Co, Cr, Cu, Ni, Pb, and Zn) is presented in Table 3. Low correlation coefficients between Fe and Pb, Zn, Co indicate a possible contaminant source for these three elements. Previous studies have shown close relationships between Pb, Zn, and Cu in sewage sludge (Alloway 1990). In the Lake Qinghai sediment, relationships between Pb, Zn, and Cu are not strong

Table 3 Correlation (Pearson) coefficient matrices between metal concentrations in sediment cores (QH03-02 and QH03-14) from Lake Qinghai

Core	QH03-02							QH03-14						
Element	Al	Co	Cr	Cu	Fe	Ni	Pb	Al	Co	Cr	Cu	Fe	Ni	Pb
Co	0.571							**0.714**						
Cr	**0.857**	0.653						**0.895**	0.677					
Cu	0.570	0.403	**0.761**					0.502	0.283	0.518				
Fe	**0.892**	0.656	**0.903**	0.539				**0.782**	0.674	**0.846**	0.585			
Ni	**0.828**	0.694	**0.879**	0.591	**0.892**			**0.763**	0.649	**0.794**	0.563	**0.718**		
Pb	0.469	0.621	0.459	0.172	0.529	0.654		0.116	0.018	0.082	−0.125	−0.174	0.119	
Zn	0.494	0.658	0.625	0.475	0.552	0.679	**0.848**	0.457	0.436	0.474	0.315	0.455	0.666	0.269

Bold and underline values (>0.700) mean a strong positive relationship between two elements

(Table 3), especially for Cu, which belongs to the PC1-elements that are mainly derived from local crustal sources. This further suggests different sources and geochemical behaviors of Pb (and Zn) in the sediments of Lake Qinghai. Pb (and Zn) in the lake sediments may reflect atmospheric inputs from anthropogenic sources in the region.

Normalized to Ti, the anthropogenic component in both sediment cores is lower than the natural contribution until the 1960s, though anthropogenic sources are responsible for part of the Pb input to the lake even at the bottom of both cores (Fig. 7). Lithogenic Pb concentrations generally vary with PC1 in the sediment cores (Fig. 7), indicating that correction for the lithogenic contribution is appropriate. The two sub-basins, with nearly equal area, would be expected to display similar calculated accumulations if only direct atmospheric contamination were deposited. Fluctuations in anthropogenic Pb accumulation before the 1960s, however, differ between the two sediment cores. This might be attributed to differences between the sub-basins with respect to (1) catchment runoff, (2) topographic features, and/or (3) local farming activities. Some of the anthropogenic atmospheric contribution may come from the catchment, where it is initially deposited, but later transported to the lake. The southern part of the Lake Qinghai catchment is characterized by relatively steep and narrow terrain. This landform favors rapid material transport into the lake. Such input includes contamination from atmospheric deposition, and shows relatively constant flux and EF of anthropogenic Pb to the QH03-02 site before the 1960s (Fig. 7). Decreased inputs of anthropogenic Pb around AD 1900–1920 and in the 1950s are attributed to dilution by relatively higher bulk sedimentation rates, in turn caused by greater erosional inputs. Higher erosion was a consequence of extensive farming on the southern shore of the lake during these periods (LIGCAS 1994). Although the Buha and Shaliu Rivers transport substantial sediment to the north sub-basin, extensive flat plains and foothills surrounding the north sub-basin lead to in situ trapping and preservation of atmospheric contamination after deposition. This led to relatively low flux of anthropogenic Pb before AD 1900 when farming was limited, which is also reflected by low total P in the sediments (Fig. 7). A large increase in anthropogenic Pb from 1900 to 1920 might be attributable to soil tillage and increased rates of soil erosion, coincident with gradually increasing population and enhanced agricultural activity in the towns of Tianjun and Gangcha (LIGCAS 1994). This increase raised the anthropogenic Pb flux in the north sub-basin to a level similar to that in the south sub-basin (Fig. 7). This flux was maintained until the 1960s, and was followed by a dramatic increase in Pb accumulation recorded in both sediment cores. Increased population and farming activities may have been responsible for the gradual increase in total P (and Mg) concentrations in QH03-14 since the 1900s (Figs. 3, 5, 8).

A dramatic increase in anthropogenic Pb deposition since the 1960s is consistent with the regional/global trend in Pb emissions to the atmosphere (Nriagu 1989; Weiss et al. 1999). In Lake Qinghai, increased Pb accumulation could be a result of (1) a gradual increase in atmospheric Pb emission from industrial sources and fuel combustion, especially in south Asia, (2) the Qing-Zang highway, constructed in the early 1950s, which increased Pb emission via combustion of leaded gasoline used by trucks, and (3) increased erosion into

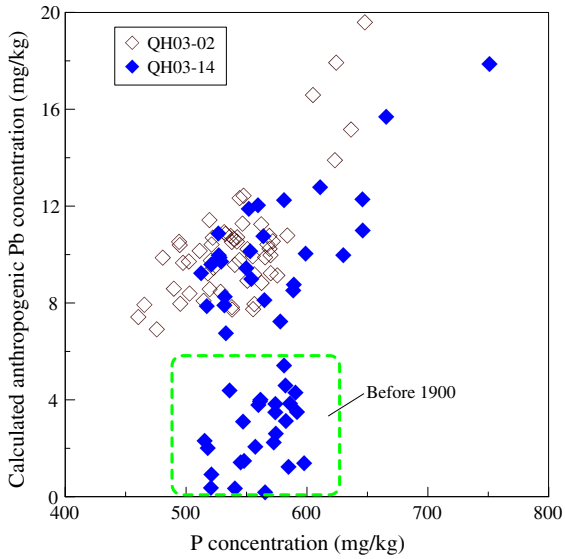

Fig. 8 Relationship between the calculated anthropogenic Pb and total P in Lake Qinghai sediments. Sediments before AD 1900 in core QH03-14 stand out owing to their low P concentration. At that time, farming was limited in the northern and Buha tributaries of the catchment (see "Discussion" in the text)

the lake, resulting from large-scale land reclamation and cultivation within the Lake Qinghai catchment since the 1960s. These processes were further tested with the enrichment factors (EFs) of anthropogenic Pb in both sediment cores (Fig. 7). The EFs are higher than unity from the bottom of both cores. Notably, EFs of anthropogenic Pb in both cores are similar and vary synchronously since the beginning of the twentieth century, further indicating a common atmospheric source. The low EF for QH03-14 before the 1900s is attributed to the flat topography and consequent limited soil erosion in the north sub-basin.

Fluxes of anthropogenic Pb, calculated using Ti as a reference element, show average accumulation rates (ARs) of Pb for QH03-02 and QH03-14 were 6.6 ± 1.1 and 3.5 ± 2.1 mg/m^2/a before the 1960s, and 13.1 ± 3.5 and 10.5 ± 3.0 mg/m^2/a since then, respectively (Fig. 7). On average, ~12.2 ± 3.5 mg/m^2 of anthropogenic Pb has been deposited annually in Lake Qinghai since the 1960s. This flux is comparable to values reported in Europe from peat bogs, for the period before the Industrial Revolution (8.5–15.5 mg/m^2/a), and to lake sediments in Lochnagar (Scotland) (19.7 mg/m^2/a), both of which were thought to receive pollutants solely from atmospheric deposition (Weiss et al. 1999; Yang et al. 2002). The AR of anthropogenic Pb in modern Lake Qinghai is lower than atmospheric Pb fluxes (8.7–40.1 mg/m^2/a) to the water surface in the Qingdao area, an industrial region in East China (Qi et al. 2005). This indicates limited local Pb contribution to Lake Qinghai. To our knowledge, there is only one report on atmospheric Pb deposition for the Tibetan Plateau (Huo et al. 1999), a 40-a record from an ice core. Due to differences between lakes and snow (or ice) with respect to transport and deposition processes, direct comparison of anthropogenic Pb concentrations or EFs from lake sediments with those from an ice core is not appropriate. Nevertheless, a comparison of temporal variation in anthropogenic Pb in ice cores and lake sediments is useful to assess regional pollution trends. Similar to our record, Pb concentrations in the Dapusuo ice core also show a significant, gradual increase between AD 1957 and 1997 (Huo et al. 1999).

Conclusions

Lake Qinghai is hydrologically closed and has no large factories or mining operations within its catchment. Thus, atmospheric deposition is the principal source of metal pollution to Lake Qinghai. We used the site-specific Pb/Ti ratio in local rock to separate anthropogenic Pb from lithogenic Pb, and interpreted excess Pb as coming from atmospheric contributions. Concentrations and fluxes of atmospheric anthropogenic Pb in the Lake Qinghai sediments increased gradually over the past two centuries, coincident with increased emissions of atmospheric contaminants. Though the two Qinghai sub-basins display different sedimentation rates and sediment geochemistry due to different rock weathering and inflow associated with local topography, the calculated atmospheric Pb accumulation is similar between the two sub-basins, whose flux since the 1960s is comparable with the average atmospheric Pb flux in the Northern Hemisphere. We suggest that the anthropogenic Pb budget in Lake Qinghai is influenced mainly by regional atmospheric pollution, but modified by local processes such as farming and consequent soil erosion.

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