

Geochemistry of major and trace elements and their environmental significances in core sediments from Bosten Lake, arid northwestern China

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ABSTRACT

Geochemical element concentrations of a 41-cm-long sediment core from Bosten Lake were analyzed with grain size, total organic carbon and total inorganic carbon, and environmental evolution over the past ~150 years was reconstructed. Based on principal component analysis (PCA) and correlation analysis of the elements, three controlling factors for vertical distribution characteristics of elements were identified, the first factor was the combined effects of terrigenous detritus and endogenous carbonate, the second controlling factor was the granularity effects of the lake sediment, the third controlling factor was the input from human activities. A first stage was from the 1870s to the 1950s, Bosten Lake was in a natural state, the deposition rates were relatively low, and the concentrations of Ca, Sr, and Ba were high. The second stage was from the 1960s to the 1990s, triggered by the inflow of agricultural return water, the sedimentation rates were clearly higher than the former stage, the concentrations of Al, Fe, and K increased notably, and Ca, Sr, and Ba decreased. The third stage comprised the period since the 2000s, the scope of human activities has been extended. Enrichment factors of Cd, Pb and P of the sediment have increased. The economic development in the basin led to an increase in pollution of the lake. Human impacts on the environmental change were embodied in the enrichment of Cd, Pb, and P, and the clear decline of biogenic Ca. Sediment geochemistry has faithfully recorded the impacts of human activities on the environmental changes of Bosten Lakes.

INTRODUCTION

Lake sediments are primarily derived from surface materials within the basin and authigenic minerals and the elements in sediments enter the lake from weathering rocks, soil erosion, and anthropogenic sources. The profiles of major and trace elements in lake sediments can carry rich information about the changes in the watershed and lake environment (Boyle *et al.*, 2004; Yang and Rose, 2005; Grosbois *et al.*, 2012). In the past few decades, human activities have played an increasingly role in the environmental changes of lake basins in the arid regions of Central Asia, causing severe damage to fragile ecosystems. In addition, many heavy metals are generally toxic to aquatic life, can be bioaccumulated and cannot

be degraded, and are a long-term human health hazard. Therefore, studies on the concentrations and changing trends of geochemical elements also have ecological significances. The analysis of sedimentary environment indicators can reconstruct the environmental history which covered the background period before human activities to the period mainly influenced by human activities (Wu *et al.*, 2013a; Ma *et al.*, 2016). This analysis is highly important for evaluating the impact of human activities in the basin, for assessing the environmental quality of the waters and guiding ecological restoration.

Bosten Lake is located in the south slope of Tianshan Mountain of the Xinjiang Uygur autonomous region, and it is the largest inland fresh water lake of China. The Bosten Lake is important for regional ecosystems, and the lake water is the resource of Kongque River, which maintains the ecosystem balance of Lop Nur region. Along with the exploitation of soil and water for the development of agriculture and manufacturing in and around the drainage basin, the ecological environment of Bosten Lake has deteriorated and caused a series of environmental problems (Fan *et al.*, 2000; Zhou *et al.*, 2001; Li and Yuan, 2002). There has been an increasing focus on modern lake environmental changes, including water quality and quantity (Zuo *et al.*, 2006; Wang *et al.*, 2013; Zhang *et al.*, 2013; Guo *et al.*, 2015; Rusuli *et al.*, 2015), and surface lake sediments (Zhang *et al.*, 2009a; Xiao *et al.*, 2010; Chen *et al.*, 2014; Liu *et al.*, 2015). Several studies have also discussed paleo-environmental changes over extended time scales (Mischke and Wünnemann, 2006; Wünnemann *et al.*, 2006; Chen *et al.*, 2008; Huang *et al.*, 2009). The spatial distribution of

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organic carbon in surface sediment and carbon burial over the past century have been used to reveal the important role in the terrestrial carbon cycle (Yu *et al.*, 2015; Yu *et al.*, 2018). However, the study on geochemical element concentrations (including heavy metals) and environmental state from lake sediment in modern times has remained a blank. The aim of this study is to distinguish the sources of the elements with multivariate statistical analysis and to describe the environmental history of Bosten Lake, which will reconstruct the effects of human activities on the lake environment from a geochemical perspective.

METHODS

Regional setting

Bosten Lake (N41°49'~42°07'E86°26'~87°30') is located in the South of Tianshan, Xinjiang Uyghur autonomous region of China, and represent the closing point of the Yanqi Basin, which is a intramontane basin between the southern slopes of the Tianshan mountain and Taklimakan desert. The catchment area of Bosten Lake is approximately ~55,600 km² (Wang and Dou, 1998) (Fig. 1). In 2008, the water surface lied at 1050 m above sea level (asl), the maximum depth of the lake water was 14 m, the lake area was 1005 km², and the water storage was 59×10⁸ m³ (Wu *et al.*, 2013b). The lake basin has a rather steep slope near the shoreline and is quite flat in the center. The lake water is weakly alkaline and has a high

degree of hardness. The discharge of Kaidu River accounts for 84.2% of the total runoff into the lake, and the mean annual runoff is 34.12×10⁸ m³. The seasonal rivers inflow into the lake include 13 other first order rivers, such as Huangshui River, Qingshui River, Wulasitai River, Quhui River, and WuShen Tara River. The outflow river is Kongque River, which supplies water to Korla City on the way to LopNor. The climate in the basin is a typical temperate arid climate. The mean annual temperature is 8.4°C, the mean annual precipitation is 94.7 mm, and the potential evaporation is 1800 mm (Zhou *et al.*, 2014). The Yanqi basin includes four counties: Bohu county, Yanqi county, Heshuo county and Hejing county. There are approximately 26 farmland drainage ditches that enter the lake, which are the main pollution sources to the lake (Sai *et al.*, 2012).

Sample collection and analysis

To better understand the human impacts to the environmental state of Bosten Lake, a continuous sediment core with a 41-cm length was taken near the major inflow river (Kaidu River) in June 2016 (Fig. 1). A piston percussion corer, fitted with 60 mm internal diameter Perspex pipes, was used during the sampling. The core sediment had a clear water-sediment interface, which denoted that the occurrence of disturbances was absent during the sampling. The sediment core was immediately sliced into subsamples in 1-cm intervals in the field. All the samples were put into sealable

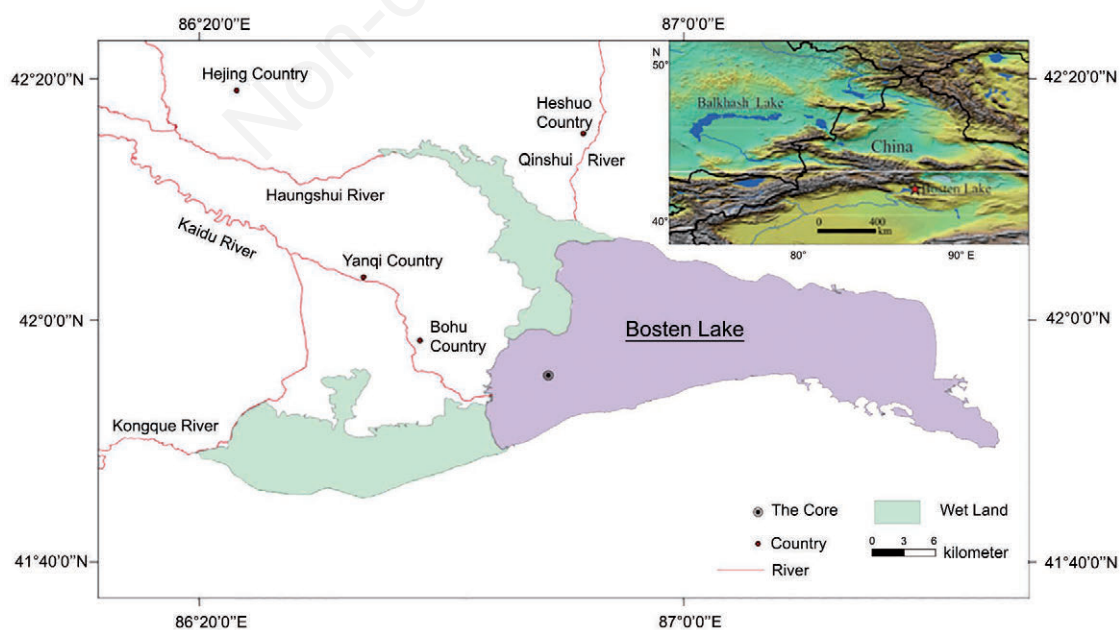


Fig. 1. Geographic Location of Bosten Lake and the sediment core.

polyethylene bags and kept in an incubator with ice bags, then transported to the laboratory and preserved in the refrigerator at a temperature of 4°C before analysis. The samples were dried in an oven at 45°C. They were first used for dating analysis and then for geochemical analysis.

An age-depth model was established with ^{210}Pb dating, which was performed using an EG&G Ortec Gamma Spectrometer, with a low-level germanium detector (EG&G ORTEC, HPGe GWL). ^{210}Pb activity was determined by gamma emission at 46.5 keV. ^{226}Ra was determined by the 295 keV and 352 keV γ -rays emitted by its daughter nuclide ^{214}Pb , after 3 weeks of storage in sealed containers to allow radioactive equilibrium.

For the measurement of elemental content, the samples (~0.125 g) were first digested with $\text{HF-HNO}_3\text{-HClO}_4$ in a Berghof MWS-3 microwave digester, and then the residuals were digested with HNO_3 , and then with H_2O_2 , in a Teflon baker. Major elements (Al, Ca, Fe, K, Mg, Na) and some trace elements (Ba, Be, Mn, P, Sr, Ti, Zn and V) were measured by Inductively Coupled Plasma Atomic Emission Spectroscopy (ICP-AES), and trace elements (Cr, Co, Ni, Cu, As, Cd, Tl and Pb) were measured by Inductively Coupled Plasma Mass Spectrometry (ICP-MS). Standard solution SPEX™ (USA) was used as the standard. Quality control was assured by the analysis of duplicate samples, blanks, and reference materials (GSD-9 and GSD-11). The relative standard deviation was below 3% for ICP-AES and below 5% for ICP-MS.

Subsamples for total organic carbon (TOC) were purged with adequate HCl (~1N) to completely remove carbonate and rinsed with distilled water to achieve a neutral state and then freeze-dried. The samples (pretreated with HCl and untreated) were weighed and packed carefully in tin capsules and then measured with a CE-440 elemental analyzer for TOC and TC, respectively. Analytical precision was based on repeated samples and reference material measurements, and the standard deviation was below 5%. Total inorganic carbon (TIC) was calculated as the difference between TC and TOC.

The sediment samples used for grain-size distribution (~0.3 g) were pretreated with diluted HCl and H_2O_2 to remove carbonates and organic matters, respectively. Then the residues were washed with deionized water to achieve a neutral pH. A dispersant solution (Na_2PO_3)₆ was mixed with the residues and then dispersed by an ultrasonic oscillator for 15 min. Grain size distributions were measured by a Mastersizer 2000 Laser Grain-size Meter; the analytical precision was based on repeated measurements and the relative error was less than 1%. The parameters used in this study include the percentage compositions of clay (<4 μm), silt (4~16 μm , 16~32 μm , 32~64 μm) and sand (>64 μm).

Data analysis

Understanding the regional environmental change and pollution history must be based on an accurate chronology. Supported ^{210}Pb was derived from the decay of *in situ* ^{226}Ra , and unsupported ^{210}Pb (excess ^{210}Pb , $^{210}\text{Pb}_{\text{ex}}$) was derived from atmospheric fallout, decayed from ^{222}Rn . The $^{210}\text{Pb}_{\text{ex}}$ activity was determined by subtracting supported ^{210}Pb activity from the total ^{210}Pb activity ($^{210}\text{Pb}_{\text{tot}}$). There are two calculation models based on $^{210}\text{Pb}_{\text{ex}}$ activity: the CRS model (constant rate of ^{210}Pb supply) and the CIC model (constant initial concentration of ^{210}Pb) (Appleby, 2008). In this study, the sediment rate was calculated by the CRS model, and the average deposition rate was determined by the CIC model.

Multivariate techniques including correlation analysis and principle component analysis were used to evaluate the elemental sources, as calculated by SPSS 20.0 (SPSS Inc., USA). Spearman's correlations were used to evaluate the relationships between the grain size content, TOC, TIC and geochemical element concentrations. PCA was carried out with varimax rotation to identify the potential influencing factors of the geochemical elements. The Kaiser-Meyer-Olkin (KMO) and Bartlett tests were used to evaluate the validity of PCA.

RESULTS

Sedimentology and chronology

The core sediment consisted of relatively uniform fine-grained materials (Fig. 2). The content of grain sizes <4 μm was 11.3~23.85%, with an average of 18.03%. The content of grain sizes 4~16 μm was 34.56~45.55%, and the mean content was 40.18%. The content of grain sizes 16~64 μm ranged from 32.14% to 44.49%, and the average value was 36.73%. The content of grain sizes >64 μm was 0.22~14.14%, with a mean value of 5.05%. According to the classification criterion based on clay (<4 μm), fine silt (4~16 μm) and silt (16~63 μm), the lithology of the sediment was fine silt. There were many shells in the sediment below 23 cm.

As shown in Fig. 2, the specific activity of $^{210}\text{Pb}_{\text{tot}}$ ranged from 74.27~448.16 Bq kg^{-1} , and ^{226}Ra was between 34.97~74.13 Bq kg^{-1} . ^{210}Pb declined to nearly zero at the bottom of the core. The vertical profile of the excessive ^{210}Pb activity in the sediment cores showed an irregular serrated distribution state, and exhibited an approximately exponential decline with depth, which was similar to the profiles observed in previous studies (Chen *et al.*, 2006; Zhang *et al.*, 2009b; Liao *et al.*, 2014; Yu *et al.*, 2015).

The bottom of the core was dated as 1868 AD, and the sediment accumulation rate was from 0.116~0.255 $\text{g cm}^{-2} \text{a}^{-1}$ based on the CRS model. The sediment deposition rates

were relatively low before the 1960s, and the average rate was $0.160 \text{ g cm}^{-2} \text{ a}^{-1}$. However, the sedimentation rate has been higher since the 1960s, and the average value was $0.229 \text{ g cm}^{-2} \text{ a}^{-1}$. The average linear deposition rate was 0.51 cm a^{-1} , determined by the CIC model. The sedimentation rate calculated in this study was higher than the former studies (Chen *et al.*, 2006; Liao *et al.*, 2014; Yu *et al.*, 2015), and the high deposition rate was primarily in agreement with the sampling location near the estuary-area of Kaidu River, where more silt and mud was deposited.

Profiles of element concentrations

Fluctuation curves of elemental content with ages are shown in Fig. 3. The concentration of Al ranged from 2.70 to 3.71 mg g^{-1} , which mainly increased from the bottom to 28 cm and then declined to the top of the core. The profile of Al was very similar to those of K, Na, Fe, Be, Ti, V, Co, Zn. The profiles of Cu, Mn, Mg, Ni, Cr, Tl were similar; the concentrations were low and fluctuated from the bottom to 28 cm and mainly increased to relatively higher concentrations to the top. The vertical distributions

of P, Pb, Cd mainly increased from the bottom to the top of the core. The concentration of Ca ranged from 144 to 188 mg g^{-1} , with high values at the bottom, began to decrease at approximately 28 cm and then maintained relatively stable values, similar to those of Ba and Sr. The concentration of As was very high in the upper 1 cm. Apart from the surface peak, the peak in As profile appeared at approximately $\sim 23 \text{ cm}$. TIC content was between 5.17% and 7.22%, and the mean content was 6.12%, similar to Ca. TOC increased from the bottom to 28 cm, declined at the depth of 20 cm and then increased to the top.

DISCUSSION

Characteristics and influencing factors of elements' content

Principal component analysis and correlation analysis were used to identify the controlling factors of major and trace elements (Morillo *et al.*, 2004; Grosbois *et al.*, 2012;

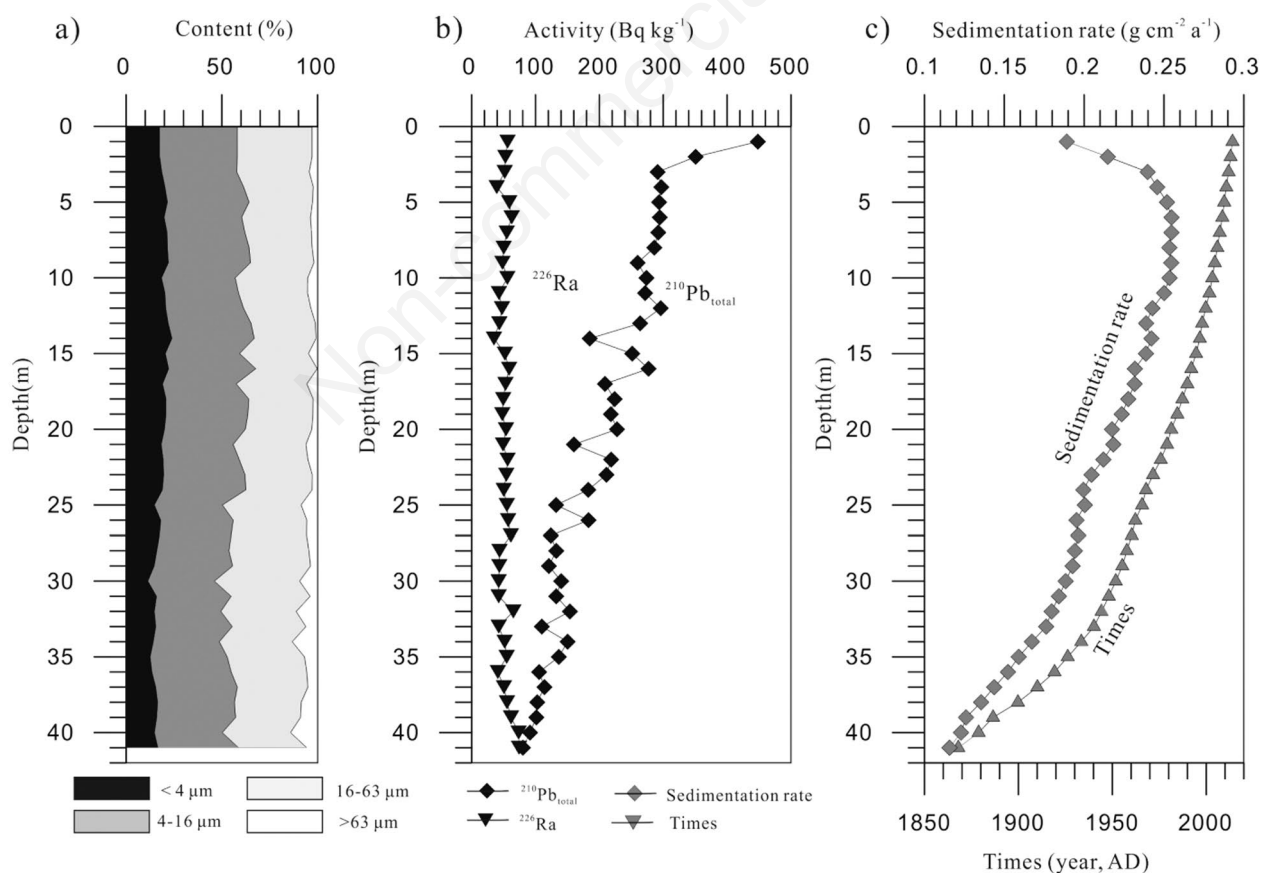


Fig. 2. a) Vertical distributions of sediment texture; b) vertical distributions of ²²⁶Ra and ²¹⁰Pb_{total}; c) ages and sedimentation rates calculated from CRS model.

Mamat *et al.*, 2014; Liu *et al.*, 2016; Ma *et al.*, 2016). The correlation analysis results were given in Tab. S1. Three principal components, which can explain 87.615% of the total variance, were extracted, and the composition matrix was shown in Fig. 4. The KMO value was 0.831, the significance of Bartlett's Sphericity was 0, which proved that the result of the analysis was valid.

The first principal component accounted for 43.249% of the total variance; Al, Be, Fe, K, Na, Ti, V, Zn, Cr, Co, Ni, and Tl had high positive loadings, Ca and Sr had high negative loadings in the component matrix. Al is a typical

lithogenic element and a major constituent of common silicate minerals (Price *et al.*, 1999). Correlation coefficients between Al and Be, Fe, K, Na, Ti, V, Zn, Cr, Co, Ni, and Tl were significantly positive. This relationship indicated that these elements primarily represent a lithogenic origin from weathering and erosion of soil parent materials in the lake watershed (Wilcke *et al.*, 1998). The significant relationship between Ca and TIC indicated that Ca originated mainly from carbonate. Between 41 and 23 cm, there were abundant biogenic shells in the sediment, which likely indicated that

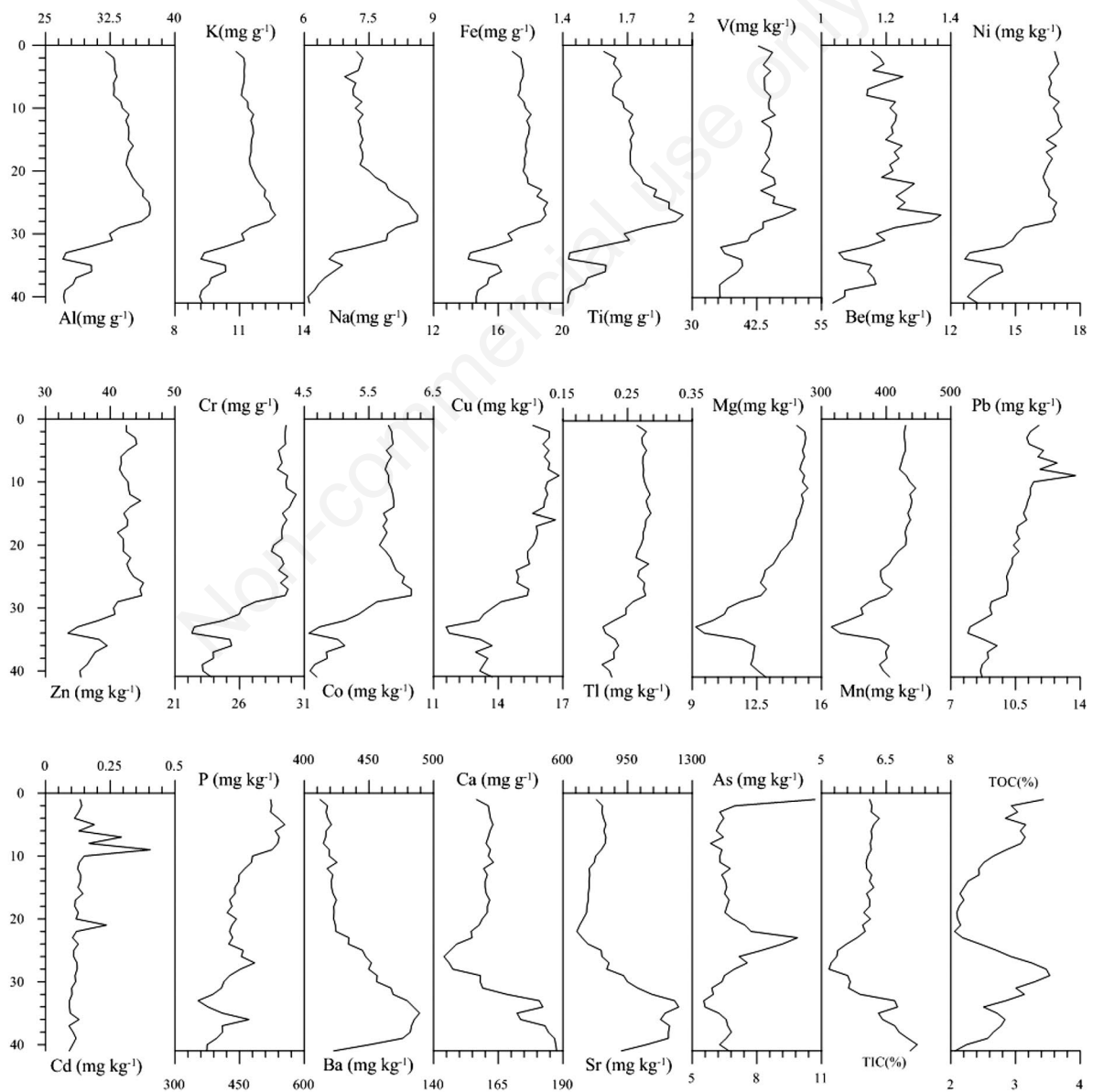


Fig. 3. Geochemical element profiles of the sediment core from Bosten Lake.

biological carbonate was one of the main sources of Ca and Sr. The carbonate in the sediment was mainly autogenous (Yu *et al.*, 2015). The geochemical behaviors of Sr and Ca in the supergene environment were largely consistent, and there was a significant correlation between them. Therefore, we regarded that Ca and Sr in the sediment core were mainly from endogenous carbonate. The negative relationship of Al, Be, Fe, K, Na, Sr, Ti, V, Zn, Cr, and Tl with Ca and TIC in the lake sediments implied dilution effects of carbonate in the sediments. The first principal component can be explained as the combined effects of terrigenous detritus and endogenous carbonate.

The second principal component accounted for 32.787% of the total variance. Mg, Mn, Cr, Cu, Tl, Pb and clay content had high positive loadings, Ba, Sr, silt and sand content had distinct negative loadings in the component matrix. There were significant positive correlations of Mg, Mn, Cr, Ni, Cu, Tl, and Pb with clay content, and negative correlations with silt and sand content, while Ba and Sr were the opposite. Ba and Sr were significantly correlated; therefore, it was determined that they were roughly homologous in the lake sediment. The second principal component can be explained as the granularity effects of the lake sediment.

The third principal component accounted for 11.580% of the total variance. Matrix loadings of P, Cd and Pb were high and positive. Under natural conditions, P was leached out from the minerals during the process of weathering, a portion was precipitated as calcium phosphate, and a portion was washed away by water. With the development of agriculture, the input of fertilizers was another important source of phosphorus. Agricultural phosphorus

impurities not only contained high Cd but also contained a certain amount of Pb and other heavy metals (Taylor, 1997; Shi *et al.*, 2008). As shown in Fig. 5, P, Pb and Cd have had higher enrichment factors during the past several decades, which indicated that they were clearly influenced by human activities. Furthermore, according to the study of Mamat *et al.* (2014), the Pb and Cd contents of the soil from Yanqi Basin were higher than the background values of Xinjiang and were obviously influenced by human activities. Zhang *et al.* (2009b) studied heavy metal contamination and found that Pb and Cd have been enriched obviously over the past several decades in the northwestern part (Huangshuigou area) of Bosten Lake. The third principal component can be explained as the impacts of human activities.

Environmental state during the past 150 years

According to the comprehensive analysis of element profiles, the environmental state of Bosten Lake during the past ~150 years can be divided into three stages as follows (Fig. 5).

The first stage was from the 1870s to the 1950s. Bosten Lake was in a natural state in this stage. Experts and scholars basically formed a consistent view that the ecological environment of Bosten Lake was rarely influenced by human activities before 1958 (Zhou *et al.*, 2001). Trace element concentrations in sediments were mainly driven by the watershed weathering and unpolluted natural atmospheric deposition. The element concentrations in this stage can be regarded as natural background levels. The concentrations of Ca, Ba and Sr were high, while concentrations of other elements such as Al, K and Fe were relatively low. The deposition rates were low, as calculated in this stage. During the mid-1950s, the hydrochemistry type was $\text{HCO}_3\text{-Ca}^{2+}\text{-Mg}^{2+}$, in accordance with the water type in the basin (Li and Yuan, 2002; Xiao *et al.*, 2015). Mollusks were abundant before the 1960s (Li *et al.*, 1987), which was also evidenced by the great number of biogenic shells and high Ca concentrations in the sediment core.

The second evolution stage was between the 1960s and the 1990s. Agricultural activities and land expansion had been developed in the basin and the lake received return water for the treatment of saline-alkali soils (Zhou *et al.*, 2001). The concentrations of Al, Fe, Mn, *etc.* have been greatly increased compared to the former stage. Along with the agricultural return water from Yanqi Basin, which was constantly discharged into the lake, the water type of the lake has changed to $\text{SO}_4^{2-}\text{-Cl-Na}^+$, which was consistent with the chemical type of drainage water (Zhou *et al.*, 2001; Li and Yuan, 2002), and finally led to the mass mortality of mollusks (Li *et al.*, 1987), as evidenced by the disappearance of biogenic shells in the sediment core. Ca, Sr and Ba exhibited abrupt decreases

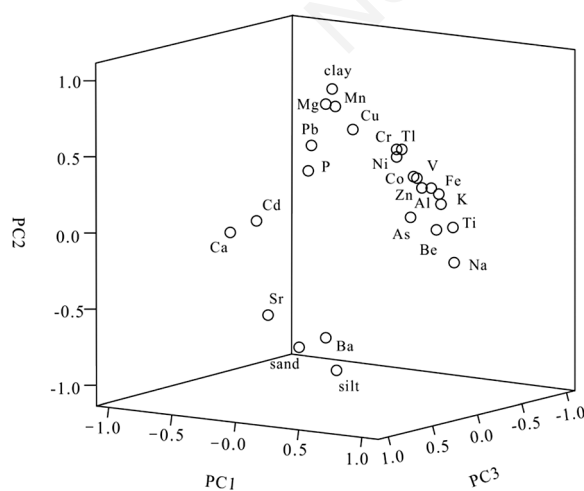


Fig. 4. PCA scores plot of geochemical elements in the sediment core of Bosten Lake.

after the 1960s, which can probably be attributed to the decrease of biogenic carbonate. As the basins from arid regions are vulnerable to soil erosion (Battarbee *et al.*, 1985), the sediment deposition rates were obviously higher in this stage. The obvious peak of As in this period was probably because of the early diagenetic effect of the lake sediment (Carbonell-Barrachina *et al.*, 1999).

The third stage was since the 2000s. The impacts of human activities on the lake evolution have been strengthened since the previous two stages. Economics has led to rapid development in this stage (Fig. 5). The lake water has been polluted, eutrophicated, and salinized, and the water quality was between type III-IV (Wu *et al.*, 2013b). The increasing accumulations of heavy metals (Cd, Pb) and P with higher EFs in this stage were in accordance with the increase of agriculture, industrial pollution and domestic wastewater inputs (Yuan and Yang, 2008; Sai *et al.*, 2012). The decline of sedimentation rate during recent years was likely because the saline-alkali soil treatment methods have greatly improved compared to the former stage (Chen *et al.*, 2007) and the government have been conscious of the importance of regional environmental protection and promulgated relevant regulations (Wang *et al.*, 2004), thus leading to the decrease of detrital matter in the lake.

In arid region, during the past centuries, especially since 1950s, the obvious increase of water consumption and the imported pollutants have led to a series of

environmental problems, like lake shrinkage, pollutants input, eco-environment degeneration and so on (Micklin 1988; Fan *et al.*, 2000; Lioubimtseva and Henebry, 2009). Lake Chaiwopu and Lake Ebinur in northwest China were two lakes which were greatly influenced by human activities during the past century. Through the study of sediment geochemistry of Lake Chaiwopu, the author found that due to a large number of water exploration, the enrichment of Ca and Mg were replaced by the enrichment of Na, which was in accordance with the transition of water type, furthermore, Pb and P has been enriched during the past decades (Liu *et al.*, 2016). In Lake Ebinur, the declined of water level and condensed of water salinity were in agreement with the accumulation of Na and Mg and the reduction of Ca and Sr, while the heavy metals have not enriched in the sediment core (Ma *et al.*, 2016; Liu *et al.*, 2017). Sediment geochemistry has faithfully recorded the impacts of human activities on the environmental changes of arid lakes.

CONCLUSIONS

Principal component analysis combined with correlation analysis was used to identify the controlling factors of geochemical elements in Bosten lake sediments. The first controlling factor was the combination effect of terrigenous detritus and endogenous carbonate, supported by the high positive scores of Al, Be, Fe, K, Na, Ti, V, Zn,

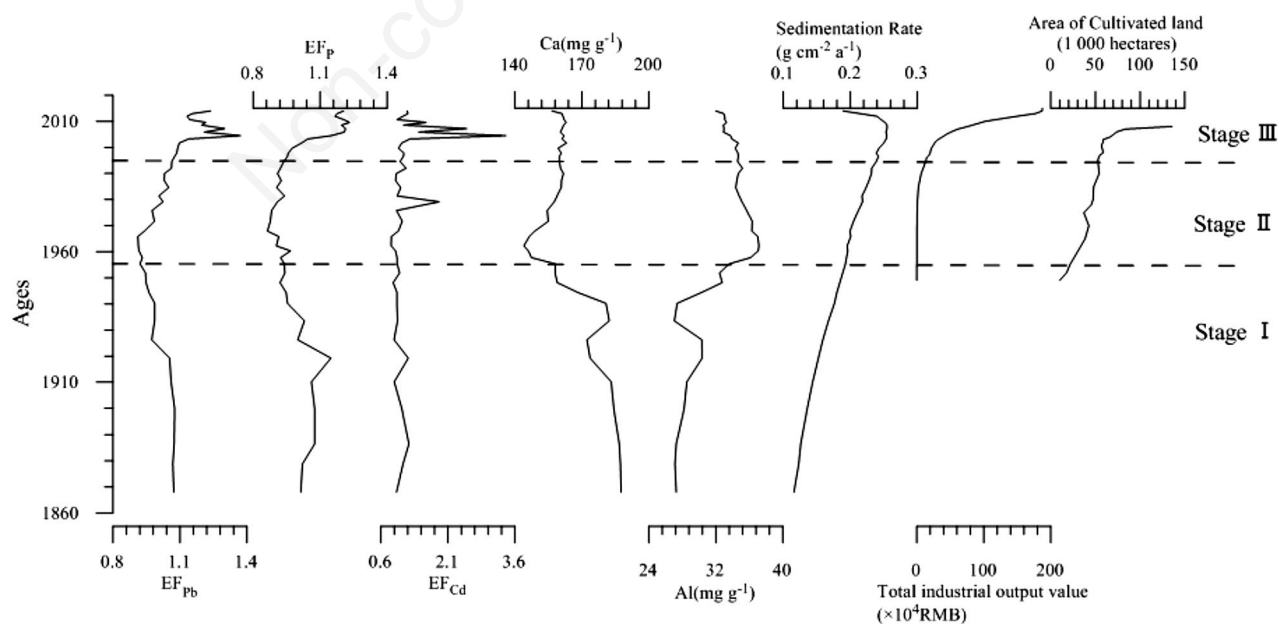


Fig. 5. Evolution stages of Bosten Lake during the past 150 years. The background values from elemental profiles were calculated as the average concentration before 1960, and Al was used as the reference element in EF calculations (Schropp *et al.*, 1990; Ma *et al.*, 2016). The total industrial output value and the Area of Cultivated Land were extracted from the Xinjiang Statistical Yearbook.

Cr, Co, Ni, Cu, Tl and the high negative scores of Ca and Sr. The second controlling factor was the granularity effects of the lake sediment, supported by the high positive scores of Mg, Mn, P, Cr, Ni, Cu, Tl, Pb and clay content, and the high negative scores of Ba, Sr, silt and sand content. The third controlling factor was the inputs of human activities, as evidenced by the high positive scores of Pb, Cd and P.

Based on a comprehensive analysis of geochemical elements, the environmental state of Bosten Lake during the past ~150 years can be divided into three stages. The first stage was from the 1870s to the 1950s, the lake was in a natural state, and the deposition rate was relatively low. Ca, Sr, and Ba concentrations were relatively high, and biogenic shells were abundant in the sediment. The second stage was between the 1960s and the 1990s, the agricultural return water caused high deposition rates of the sediment in this stage, the concentrations of Ca, Ba, and Sr had declined and biogenic shells had disappeared due to the increased salinity. The third stage comprised the period since the 2000s, with the rapid development of society and economy in the basin, the enrichments of P, Cd, and Pb in the lake sediment were probably derived from human inputs. The deposition rates have begun to decline in recent years of this stage.

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