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Relationship between $^{210}Pb_{ex}$ activity and sedimentary organic carbon in sediments of 3 Chinese lakes

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ABSTRACT

This report demonstrates that organic matter was an important factor in lake sediment $^{210}Pb_{ex}$ dating. Sediment cores from lakes in central and western China with different-trophic levels were collected, and the $^{210}Pb_{ex}$ activity and total organic carbon (TOC) were measured. The Rock-Eval pyrolysis technique was used to deconvolute TOC into free hydrocarbons (S1), thermally less-stable macromolecular organic matter (S2a), kerogen (S2b), and residual carbon (RC). The results show significant correlations between TOC and $^{210}Pb_{ex}$, particularly between S2a and $^{210}Pb_{ex}$, in all the sediment cores. This indicated that the algal-derived organic component S2a may play the most important role in controlling the distribution of $^{210}Pb_{ex}$. Scavenging by algal-derived organic matter may be the main mechanism. As chronology is the key to the understanding of pollution reconstruction and early diagenesis in sediments, more attention should be paid to the influence of organic matter on $^{210}Pb_{ex}$.

1. Introduction

$^{210}Pb_{ex}$ dating is one of the most important methods for dating recent sediments (0–150 years). The primary assumptions of most applied $^{210}Pb_{ex}$ sediment-dating methods (constant rate of ^{210}Pb supply (CRS) model) are that the flux of $^{210}Pb_{ex}$ derived from atmospheric fallout and the deposition rate are relatively constant, which would lead to an exponentially decreasing trend of $^{210}Pb_{ex}$ activity with depth. ^{210}Pb dating does give reliable chronologies in many instances. However, reports show that some geochemical processes, such as mixing of surface sediments by physical or biological processes (Dominik et al., 1981; Bloesch and Evans, 1982), loss of ^{222}Rn from surficial sediments (Imboden and Stiller, 1982), and post depositional remobilization of ^{210}Pb (and ^{210}Po) at the sediment–water interface (Wan et al., 1987, 1993; Santschi et al., 1990; Benoit and Hemond, 1991) as a result of changes in acidity or redox conditions, would affect the dating results. Discrepancies were often found between $^{210}Pb_{ex}$ dating and other independent chronostratigraphic dating obtained using ^{137}Cs, ^{241}Am, ^{240}Pu/^{239}Pu, pollen, and other dating markers, and composite models are often used to correct results.

It is widely known that organic matter plays an important role in the transfer and deposition of heavy metals in lakes. However, the role of organic matter had often been neglected in models of ^{210}Pb dating. Few studies have reported the relationships between organic matter and ^{210}Pb. Krishnaswamy et al. (1971) found that $^{210}Pb_{ex}$ in lakes is brought into association with sediment along with organic particulates, and suggested that in lakes the role of organic carbon may be more important than that of inorganic matter in affecting $^{210}Pb_{ex}$ deposition in sediments. Sedimentary organic carbon (SOC) has been shown to have a strong and positive correlation with $^{210}Pb_{ex}$ in marine settings (Paulsen et al., 1999). SOC has also been shown to correlate positively and significantly with $^{210}Pb_{ex}$ in near surface (0–2 cm) samples of terrestrial sediments from the coastal plain of Texas, USA (Yeager and Santschi, 2003). Vertical scavenging is found to be the primary transport mechanism for ^{210}Pb in the shelf and slope regions of the Gulf of Mexico (Baskaran and Santschi, 2002). As described in Wan et al. (2005), if the flux of organic particulates is relatively constant in lake water, the transfer of $^{210}Pb_{ex}$ from lake water to sediments will also be relatively constant, but if the primary productivity of the lake increases significantly because of enrichment of nutrients, the increased organic particulates in the water may result in increased transfer and deposition of $^{210}Pb_{ex}$ to sediments. Total organic carbon (TOC), mainly derived from indigenous algae, was found to have an important influence on $^{210}Pb_{ex}$ activity in the sediments of a eutrophic lake, Lake Chenghai, suggesting that the rapid increase

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in the influx of particulate organic matter is an important factor facilitating the deposition of $^{210}Pb_{ex}$ by its transfer into settling sediments (Wan et al., 2005). Eutrophication has become a widespread problem in many lakes in China in recent years. The development of a better understanding of the effects of organic matter during ^{210}Pb deposition is therefore urgent. TOC has different components with different origins, and which components play more significant roles in the distribution of $^{210}Pb_{ex}$ is not clear. Whether the impacts widely existed in eutrophic lakes, the relationships between organic matter and ^{210}Pb in other trophic lakes such as oligotrophic lakes, and the mechanisms of organic compounds in impacting the $^{210}Pb_{ex}$ distribution need to be further investigated.

Rock-Eval pyrolysis is a simple and rapid procedure that can automatically divide the organic carbon present into a number of groups, such as free and volatile hydrocarbons (S1), kerogen-derived hydrocarbons (S2), and residual hydrocarbons (RC). The procedure is widely used in petroleum exploration, and in recent years it has been applied in soil/sediment research (Lafargue et al., 1998; Disnar et al., 2003; Sanei et al., 2005; Stern et al., 2005; Sanei and Goodarzi, 2006; Outridge et al., 2007; Carrie et al., 2009). Carrie et al. (2009) used S1, S2, and RC in surface sediments of the Mackenzie River basin in Canada to study the different carbon distributions and origins in the region. Outridge et al. (2007) used S2 in Canadian high Arctic lake sediments to represent aquatic primary productivity, and found that a large fraction of the increased Hg concentrations observed during the 20th century could be explained by scavenging of aquatic primary productivity. However, few studies (only two, to our knowledge) have used more sensitive parameters from Rock-Eval pyrolysis to provide more robust and accurate information. The R400 parameter (the ratio between the area of the S2 peak integrated up to 400 °C normalized to its total surface), directly from Rock-Eval pyrolysis, was used as an indicator of organic matter preservation in peat (Disnar et al., 2008). Whether the S2 parameter in lake sediment could be further divided into more sensitive parameters, few studies had reported.

Three sediment cores from different-trophic lakes in China were chosen: one from a eutrophic lake, Lake Hongfeng, one from a medium-trophic lake, Lake Bosten, and one from an oligotrophic lake, Lake Shuangta. Using the Rock-Eval 6 technique, the SOC was separated into S1, S2, and RC groups. S2 was further divided into two groups: S2a (thermally less-stable macromolecular organic matter) and S2b (high molecular-weight kerogens), based on differences in pyrolysis temperature, peak position, and organic carbon origin. The main purpose of this study was to investigate the relationships between different organic components from Rock-Eval pyrolysis and $^{210}Pb_{ex}$ activities in the sediments, and to explore the different roles of organic components in affecting the $^{210}Pb_{ex}$ distribution or ^{210}Pb dating in lake sediments. The probable theoretical mechanism (model) was also discussed.

2. Sampling and methods

Three lakes with different eutrophic status were chosen in this study. Lake Hongfeng (E: 106°20′–106°27′, N: 26°26′–26°36′) is 28 km west of Guiyang City, the capital city of Guizhou Province in southwest China. It is the most important source of drinking water for Guiyang City, which has a population of more than 3 million. As pollution increased in recent decades, Lake Hongfeng had already become a eutrophic lake (Chen et al., 2007). Lake Bosten (E: 86°47′–87°26′, N: 41°22′–42°21′) is located in the Yanqi Basin of Xinjiang Province in northwest China, and has an altitude of about 1048 m and an area of 988 km^2. It is the largest inland freshwater lake in China and has a medium-trophic level (Zhou et al., 2003). Lake Shuangta (E: 96°19′–96°24′, N: 40°31′–40°35′) is located in the lower basin of the Shule River in Jiuquan City in Gansu Province in middle and upper China. This region is rainless and Lake Shuangta is an oligotrophic lake.

Three sediment cores (HF from Lake Hongfeng in 2006, BS from Lake Bosten in 2006, and ST from Lake Shuangta in 2007) were collected from the lake centers. The sediment cores were collected using a gravity corer, immediately sliced into different sections, wrapped in baked (450 °C) Al foil, and transported in ice to the laboratory, where they were stored at −20 °C until further treatment. The subsamples were lyophilized, and ground with a mortar and pestle before analyses.

Details of the ^{210}Po and ^{226}Ra analyses have been previously described by Wan et al. (1987). Briefly, the ^{210}Po activity in the sediments was analyzed by α-spectrometry on a Canberra S-100 and multi-channel spectrometer with a PIPS Si detector, and ^{226}Ra activity was determined by γ-spectrometry on a Canberra S-100 multi-channel spectrometer mated to a GCW3022 H-PR Ge well detector. The excess ^{210}Pb activity was obtained by subtracting the ^{226}Ra activity from the total ^{210}Pb activity, which was derived from ^{210}Po.

The TOC in the sediments was analyzed using a PE elemental analyzer after removal of carbonate. Reproducibility was within 3% for TOC. The organic carbon in bulk sediments was operationally characterized by Rock-Eval 6 analysis (Vinci Technologies, Rueil-Malmaison, France) at the State Key Laboratory of Organic Geochemistry of Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, allowing quantification of three types of sedimentary organic carbon based on the thermal evolution of hydrocarbons and the S1, S2, and RC carbon groups (Lafargue et al., 1998; Sanei et al., 2005; Carrie et al., 2009). Briefly, S1 and S2 were the hydrocarbons released under 300 °C and 300–650 °C during anaerobic pyrolysis, respectively, while RC was the residual carbon released from 400 to 850 °C in the oxidation oven after anaerobic pyrolysis. A standard reference material (IFP 160000, VINCI Parc d) was used in the Rock-Eval measurements.

Statistical analyses were carried out with SPSS 13.0. Concentration data were ln-transformed, and in all cases satisfied the assumptions of normality and homogeneous variances required for statistical purposes.

3. Results and discussion

3.1. Relationships between $^{210}Pb_{ex}$ and TOC in lakes

The relationships between $^{210}Pb_{ex}$ and TOC in the three studied lakes are shown in Fig. 1. The TOCs were 2.17%–6.45%, with a mean of 4.47%, for Lake Hongfeng, 1.12%–2.18%, with a mean of 1.71%, for Lake Bosten, and 0.09%–1.23%, with a mean of 0.58%, for Lake Shuangta. The TOC was highest in Lake Hongfeng and lowest in Lake Shuangta, which agreed with the trophic levels of the three lakes, i.e., eutrophic in Lake Hongfeng, medium trophic in Lake Bosten, and oligotrophic in Lake Shuangta. In the eutrophic Lake Hongfeng, in which organic particulates were quite abundant, a significant positive correlation was found between $^{210}Pb_{ex}$ and TOC, with correlation coefficients of 0.723 ($p < 0.001$). The situation was quite similar to that in the eutrophic Lake Chenghai (Wan et al., 2005), in which the $^{210}Pb_{ex}$ also showed strong positive relationship with sedimentary organic carbon, and suggested the $^{210}Pb_{ex}$ activity in lake sediment could be largely influenced by the TOC. The results in Lake Chenghai further supported that organic matter in eutrophic lakes has an important impact on the $^{210}Pb_{ex}$ activity in

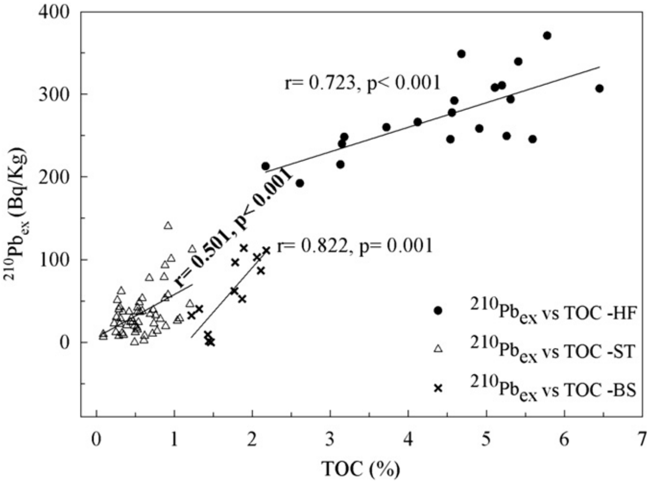

Fig. 1. Relationships between $^{210}Pb_{ex}$ and TOC in 3 Chinese lakes (HF: Lake Hongfeng; BS: Lake Bosten; ST: Lake Shuangta).

sediments. Therefore, more attentions should be paid to ^{210}Pb dating in eutrophic lakes. In the medium-trophic Lake Bosten, which was less rich in organic particulates than Lake Hongfeng was, a significant positive correlation was also found between $^{210}Pb_{ex}$ and TOC, with correlation coefficients of 0.822 ($p = 0.001$). Interestingly, in the oligotrophic Lake Shuangta, where there were relatively less organic particulates, $^{210}Pb_{ex}$ was also correlated significantly with TOC, with correlation coefficients of 0.501 ($p < 0.001$). The results suggested that no matter the lake trophic level is eutrophic, medium trophic, or oligotrophic, organic matter all played an important role in controlling the $^{210}Pb_{ex}$ activity in sediments.

3.2. Rock-Eval pyrolysis of sediments

TOC represents the sum of various organic compounds, and these compounds may play completely different roles in the distribution of $^{210}Pb_{ex}$ in sediments. To investigate the roles of different organic components, the Rock-Eval 6 technique was used to separate the organic carbon into several portions. Traditional Rock-Eval pyrolysis separates organic carbon into several groups, in which S1, S2, S3CO (CO produced during pyrolysis), S3CO$_2$ (CO$_2$ produced during pyrolysis) and RC groups are the most relevant in soil/sediment research (Sanei et al., 2005; Sanei and Goodarzi, 2006; Outridge et al., 2007; Carrie et al., 2009). For example, Carrie et al. (2009) used S1, S2, and RC to study the organic carbon distribution and origins in Canadian Mackenzie River basin. However, it is not clear whether these parameters could be divided into more sensitive and meaningful parameters with different sources in lake sediments because no such studies have been carried out before.

Fig. 2b–d show the typical pyrolysis curves of the sediments in three lakes. In the general analysis of mineral samples, in high-temperature pyrolysis (300–650 °C), there is one main peak corresponding to kerogen pyrolysis and all other peaks are negligible. However, the pyrolysis curves of the three sediments all show two obvious peaks in the S2 group; one can be defined as S2a, and the other as S2b. The organic components of S1, S2a, S2b, and RC obtained by pyrolysis are discussed in the present study.

S1, the free hydrocarbons, originates from the pyrolysis of mainly low molecular-weight amorphous carbon (Sanei et al., 2005; Outridge et al., 2007). RC, called residual carbon or dead carbon, is the final thermostable fraction of organic carbon from algae; it is also contributed from the organic carbon content of terrestrial cellulose and black carbon (Outridge et al., 2007; Carrie et al., 2009), which are rich in condensed aromatic carbon. The S2 compounds in sediments generally correspond to the highly aliphatic biomacromolecular structure of algal cell-walls (Sanei et al., 2005), and can be used to represent the aquatic primary productivity in high Arctic lakes (Outridge et al., 2007), or to discuss algal-derived organic carbon distributions in basins (Carrie et al., 2009). S2b, which is typical kerogen-derived hydrocarbons, is mainly derived from high molecular-weight kerogens, and S2a consists mainly of thermally less-stable macromolecular organic matter such as aliphatic-rich humic acid and kerogen precursors (Peters, 1986). Only one main peak is observed in high-temperature pyrolysis of algae (Fig. 2a); the peak position is similar to that of S2a in sediments, suggesting that S2a may originate from algae and may correspond to the highly aliphatic biomacromolecular structure of algal cell-walls in sediments (Sanei et al., 2005). The pyrolysis results suggest that S2a was mainly derived from algae in the three sediments and that S2a may be a better indicator of algal productivity in lakes.

Table 1 shows the average distribution of the organic components in TOC in the sediments of the studied lakes. RC was the major TOC component, accounting for 53.3%–85.1% in the three cores; S1, S2a, and S2b were the minor components, accounting for 4.0%–8.0%, 4.3%–9.8%, and 3.7%–20.5%, respectively. The distributions of organic components in the sediments are similar to those reported in soil in the Guangzhou areas (Song et al., 2002), showing that kerogen and black carbon were the major constituents of organic carbon. As Fig. 3 shows, S2a in Lake Hongfeng was the highest among the three lakes, with a mean value of 4.56 ± 1.87 mg HC/g; S2a in Lake Bosten was lower than that in Lake Hongfeng and higher than that in Lake Shuangta, with a mean value of 1.54 ± 0.35 mg HC/g; S2a in Lake Shuangta was the lowest, with a mean value of 0.21 ± 0.10 mg HC/g. The results were also consistent with the lake trophic levels, i.e., eutrophic for Lake Hongfeng, medium trophic for Lake Bosten, and oligotrophic for Lake Shuangta, which further confirmed that S2a could also be an indicator of the algal productivity.

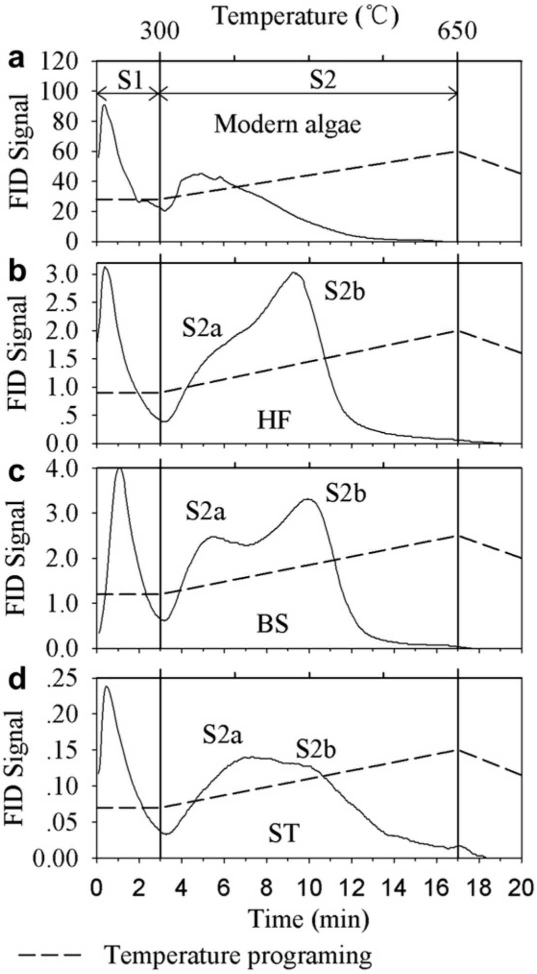

Fig. 2. Formation of hydrocarbon peaks (S1 and S2) during pyrolysis stage of modern algae and sediments (HF: Lake Hongfeng; BS: Lake Bosten; ST: Lake Shuangta).

Table 1
Average distribution of organic components in TOC in lake sediments.

%	HF ($N = 20$)	BS ($N = 12$)	ST ($N = 53$)
S1	4.6 ± 1.2	8.0 ± 1.1	4.0 ± 3.8
S2a	9.8 ± 2.5	9.0 ± 0.9	4.3 ± 2.9
S2b	8.7 ± 3.0	20.5 ± 2.8	3.7 ± 2.9
RC	73.1 ± 5.0	53.8 ± 2.7	85.1 ± 13.1
TOTAL[a]	96.2 ± 2.2	91.3 ± 1.8	97.3 ± 4.2

[a] Sum percentage of S1, S2a, S2b and RC in TOC.

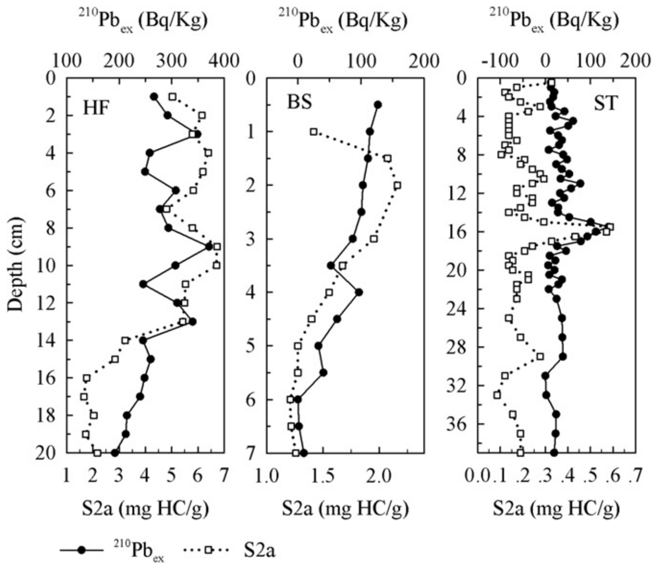

Fig. 3. $^{210}Pb_{ex}$ and S2a profiles in studied sediment cores (HF: Lake Hongfeng; BS: Lake Bosten; ST: Lake Shuangta).

3.3. Relationships between $^{210}Pb_{ex}$ and organic carbon components

The correlations between temporal variations in organic carbon components in the sediment cores and $^{210}Pb_{ex}$ are shown in Table 2. In Lake Hongfeng, the correlations between $^{210}Pb_{ex}$ and S1, S2a, S2b, and RC are all significant ($p < 0.01$), with $r = 0.656$, 0.725, 0.759, and 0.662, respectively, indicating that free hydrocarbons, thermally less-stable macromolecular hydrocarbons, kerogens, and residual carbon all significantly influenced the distribution of $^{210}Pb_{ex}$ in HF. In Lake Bosten, S1, S2a, and RC significantly correlated with $^{210}Pb_{ex}$, with $r = 0.908$, 0.750, and 0.816, respectively ($p < 0.01$), but S2b had no significant correlation with $^{210}Pb_{ex}$. In Lake Shuangta, significant positive correlations were found between $^{210}Pb_{ex}$ and S2a, S2b, and RC, with $r = 0.748$, 0.737, and 0.458, respectively ($p < 0.01$), but no significant correlation was found between $^{210}Pb_{ex}$ and S1. The common characteristic of the three sediment cores is that there are significant positive correlations between $^{210}Pb_{ex}$ and S2a and RC. This suggests that the S2a and RC compounds play more important roles in the distribution of $^{210}Pb_{ex}$ activity in sediments. As previously discussed, S2a consists of thermally less-stable macromolecular organic matter derived mainly from algae, and could represent algal productivity in lakes. It is therefore algal-derived organic matter that significantly controls the $^{210}Pb_{ex}$ activity in sediments. The relationship between TOC and $^{210}Pb_{ex}$ could therefore be significantly reduced in sediments containing high proportions of terrestrial organic matter. In eutrophic Lake Hongfeng, the abundant algae would be the main source of S1, S2b, and RC components, so the S1, S2b, and RC components all significantly correlated with $^{210}Pb_{ex}$; in medium-trophic Lake Bosten and oligotrophic Lake Shuangta, whose algal productivities were not as high as that of Lake Hongfeng, the increased proportions of terrestrial organic matter to S2b in Lake Bosten and to S1 in Lake Shuangta may be the main reason for the decrease in the correlations of these components with $^{210}Pb_{ex}$. Although RC showed significant correlations with $^{210}Pb_{ex}$ in all three lakes, the r value (0.458) in Lake Shuangta was obviously lower than that in Lake Hongfeng and Bosten, as the organic particulates obviously decreased; but the correlation coefficients between S2a and $^{210}Pb_{ex}$ were quite similar in the three lakes (Table 2). The results suggested that S2a was the most important organic component in controlling the $^{210}Pb_{ex}$ activity in sediments.

Table 2
Correlations between $^{210}Pb_{ex}$ and organic carbon components in the three lake sediment cores.

	$^{210}Pb_{ex}$ (Bq/Kg)		
	HF ($N = 20$)	BS ($N = 12$)	ST ($N = 53$)
TOC (%)	0.723**	0.822**	0.501**
S1 (mg HC/g)	0.656**	0.908**	0.197
S2a (mg HC/g)	0.725**	0.750**	0.748**
S2b (mg HC/g)	0.759**	0.429	0.737**
RC (%)	0.662**	0.816**	0.458**

Note: * = significant at $p < 0.05$, ** = significant at $p < 0.01$.

A comparison of $^{210}Pb_{ex}$ and S2a in the sediment cores is shown in Fig. 3. In Lake Hongfeng, below a depth of 16 cm, S2a was quite low and $^{210}Pb_{ex}$ was also at its lowest level; at depths of 16–9 cm, S2a increased from 1.76 to 6.72 mg HC/g, and $^{210}Pb_{ex}$ also increased significantly from 248 to 371 Bq/kg, with fluctuations, in this part of the core; at depths of 9 cm to the sediment surface, both S2a and $^{210}Pb_{ex}$ were at relatively high levels. In Lake Bosten, $^{210}Pb_{ex}$ increased slowly from a depth of 7 cm to the surface, and the S2a curve also slowly increased along with the $^{210}Pb_{ex}$ curve (Fig. 3). In Lake Shuangta, $^{210}Pb_{ex}$ fluctuated consistently with S2a below depths of 24 cm; the $^{210}Pb_{ex}$ had a large peak at depths from 18 cm to 14 cm, with a maximum value at a depth of 16 cm, and the S2a curve also had a large peak at the same depths and with the same characteristics; at depths of 12–8 cm, variations in the ^{210}Pb curve were similar to those in the S2a curve (Fig. 3). Overall, the $^{210}Pb_{ex}$ fluctuations were all quite similar as the S2a fluctuations in the 3 lakes, which indicated that, whether the S2a curve varied slowly or suddenly, S2a was important in controlling $^{210}Pb_{ex}$ distribution in sediments.

As S2a is mainly derived from algae, it suggested that the distribution of $^{210}Pb_{ex}$ in sediments was much influenced by algal productivity. The higher the algal productivity, the larger the $^{210}Pb_{ex}$ distribution in the sediments would be. Vertical scavenging was found to be the primary mechanism of transport for ^{210}Pb in the continental shelf and slope regions of the Gulf of Mexico (Baskaran and Santschi, 2002). $^{210}Pb_{ex}$ in lake water is transported into sediments in association with organic particulates (Krishnaswamy et al., 1971). Wan et al. (2005) found that the rapid increase in sedimentary organic matter derived predominantly from the remains of indigenous algae in Lake Chenghai appeared to closely influence $^{210}Pb_{ex}$ distribution in sediments. Therefore, vertical scavenging by algae may be the main mechanism of transport of $^{210}Pb_{ex}$ to sediments in lakes. The primary assumption of ^{210}Pb sediment-dating (CRS model) is a constant flux of $^{210}Pb_{ex}$; the $^{210}Pb_{ex}$ is derived from the atmosphere and then transferred into lake sediments. When algal productivity increases, scavenging of $^{210}Pb_{ex}$ by algae would also increase, and this would lead to relatively high $^{210}Pb_{ex}$ activity in sediments.

3.4. Models of the relationships between scavenging by algal productivity and $^{210}Pb_{ex}$ in sediment cores

To quantitatively investigate the relationship between algal productivity and $^{210}Pb_{ex}$ in sediments, and to determine the influence of algal-derived organic matter on ^{210}Pb dating, we tried to interpret the mechanism using a corrected model of the organic matter. The basic equation of ^{210}Pb dating is

$$^{210}Pb_{ex} = (^{210}Pb_{ex})_0 \times e^{-\lambda t} \quad (1)$$

where $^{210}Pb_{ex}$ is the excess ^{210}Pb in the sediment, $(^{210}Pb_{ex})_0$ is the excess ^{210}Pb in the surface sediment, λ is the ^{210}Pb radioactive decay constant, and t is the age in years. In the CRS model, $(^{210}\mathrm{Pb}_{\mathrm{ex}})_0$ is assumed to be constant; it is derived from atmospheric fallout and then transmitted to the sediment surface. If the sedimentary accumulation rate is constant, Eq. (1) transforms to

$$h = -(s/\lambda) \times \ln(^{210}\mathrm{Pb}_{\mathrm{ex}}) + \left[(s/\lambda) \times \ln(^{210}\mathrm{Pb}_{\mathrm{ex}})_0\right] \tag{2}$$

where h is the sediment depth (or cumulative mass depth) and s is the average sediment accumulation rate (or mass accumulation rate). If $(^{210}\mathrm{Pb}_{\mathrm{ex}})_0$ is constant (CRS model), $\ln(^{210}\mathrm{Pb}_{\mathrm{ex}})$ would show a linear correlation with h. However, the earlier discussion (part 3.3) suggests that scavenging by algal productivity would significantly influence $(^{210}\mathrm{Pb}_{\mathrm{ex}})_0$, so $(^{210}\mathrm{Pb}_{\mathrm{ex}})_0$ is not constant. Here, S2a was used to represent the algal productivity, and the following hypothesis regarding the relationship between $(^{210}\mathrm{Pb}_{\mathrm{ex}})_0$ and S2a was made when the algal productivity was within a certain range in actual lakes:

$$(^{210}\mathrm{Pb}_{\mathrm{ex}})_0 = a \times (\mathrm{S2a})^n + b \tag{3}$$

where a, b, and n are constants (which may be different in different lakes with different algal productivities; this needs further investigation). For $^{210}\mathrm{Pb}_{\mathrm{ex}}$ in lake water brought into sediments in association with organic particulates (Krishnaswamy et al., 1971), b is defined as zero. Combining Eqs. (2) and (3), we obtain the following multiple linear equation:

$$\ln(^{210}\mathrm{Pb}_{\mathrm{ex}}) = n \times \ln\mathrm{S2a} - (\lambda/s) \times h + \ln a \tag{4}$$

If algal productivity played an important role in the $^{210}\mathrm{Pb}_{\mathrm{ex}}$ distribution in sediments, $\ln(^{210}\mathrm{Pb}_{\mathrm{ex}})$ would show a significant linear correlation with lnS2a. The linear correlation coefficients between $\ln(^{210}\mathrm{Pb}_{\mathrm{ex}})$ and lnS2a in the three lake sediment cores were 0.735 ($p < 0.01$) in HF, 0.643 ($p < 0.05$) in BS, and 0.558 ($p < 0.01$) in ST. All three lake sediment cores show significant linear correlations between $\ln(^{210}\mathrm{Pb}_{\mathrm{ex}})$ and lnS2a, suggesting that algal productivity played an important role in the $^{210}\mathrm{Pb}_{\mathrm{ex}}$ distribution in the sediments. To estimate the effects of algal productivity and those of radioactive decay on the $^{210}\mathrm{Pb}_{\mathrm{ex}}$ distribution in sediments separately, multiple linear regressions were performed using $^{210}\mathrm{Pb}_{\mathrm{ex}}$, S2a, and the cumulative mass depth (h, g/cm^2) in the three cores. The results were as follows:

$$\ln(^{210}\mathrm{Pb}_{\mathrm{ex}}) = 0.207 \times \ln\mathrm{S2a} - 0.021 \times h + 5.205 \quad \mathrm{HF} \tag{5}$$

$$\ln(^{210}\mathrm{Pb}_{\mathrm{ex}}) = 0.545 \times \ln\mathrm{S2a} - 1.128 \times h + 5.311 \quad \mathrm{BS} \tag{6}$$

$$\ln(^{210}\mathrm{Pb}_{\mathrm{ex}}) = 1.154 \times \ln\mathrm{S2a} - 0.007 \times h + 5.325 \quad \mathrm{ST} \tag{7}$$

The adjusted complex correlation coefficients were 0.701 ($p = 0.001$), 0.744 ($p = 0.016$), and 0.544 ($p < 0.001$) for HF, BS, and ST, respectively, suggesting that $\ln(^{210}\mathrm{Pb}_{\mathrm{ex}})$ had a significant linear correlation with lnS2a and h in all three cores. For Lake Hongfeng (Eq. (5)), the p values for inclusion of lnS2a and h were 0.081 and 0.705, respectively. Only lnS2a was close to the significant level in this model and h was far from significant level. For Lake Shuangta (Eq. (7)), lnS2a was also the only significant factor in the model (for inclusion, $p < 0.001$), and h was not significant (for inclusion, $p = 0.359$). The results suggested that algal productivity was the dominant controller in the $^{210}\mathrm{Pb}_{\mathrm{ex}}$ profiles of Lakes Hongfeng and Shuangta, and this may be the main reason why the $^{210}\mathrm{Pb}_{\mathrm{ex}}$ profiles did not show exponentially decreasing trends in Lakes Hongfeng and Shuangta (Fig. 3). In Lake Bosten, although both lnS2a and h show significant linear correlations with $\ln(^{210}\mathrm{Pb}_{\mathrm{ex}})$ ($r = 0.643$, $p = 0.016$ for lnS2a, $r = -0.800$, $p = 0.002$ for h), h was the only significant factor in the Eq. (6) model (for inclusion of h, p was 0.050, and for inclusion of lnS2a, p was 0.798). This implied that the $^{210}\mathrm{Pb}_{\mathrm{ex}}$ decay effect was the dominant controller in the $^{210}\mathrm{Pb}_{\mathrm{ex}}$ distribution in Lake Bosten (for $t = h/s$, and s assumed to be constant). Therefore, the $^{210}\mathrm{Pb}_{\mathrm{ex}}$ profile of Lake Bosten showed a decreasing trend, which was different from the profiles for Lakes Hongfeng and Shuangta.

Because the $^{210}\mathrm{Pb}_{\mathrm{ex}}$ activities in Lakes Hongfeng and Shuangta were predominantly influenced by algal productivity, and did not show any exponentially decreasing trends, the core dating can not be calculated using the CRS model. The results of ^{210}Pb dating in Lake Bosten using the CRS model (stepwise sedimentary rate model) are shown in Fig. 4. Above a depth of 3.5 cm, the mean sedimentary accumulation rate was 0.0416 g/cm^2/y; below a depth of 3.5 cm, the mean sedimentary accumulation rate was 0.0089 g/cm^2/y, with a date of 1782 AD at a depth of 7 cm. Although the ^{137}Cs profile (data not shown here) in Lake Bosten was too disordered for dating, the ^{137}Cs activity (2.80 Bq/kg) at a depth of 6.5 cm indicates that the date would not be earlier than 1950 AD. Therefore, the probable date at a depth of 7 cm would not be earlier than 1940 AD. Based on the MLR models of Eqs. (4) and (6), the BS core age was also calculated (Fig. 4), and the mean sedimentary accumulation rate was found to be 0.0282 g/cm^2/y, with a date of 1899 AD at a depth of 7 cm. The result was 52.2% less than that using the CRS model, and much closer to the probable result. This suggested that algal productivity still had much influence on the ^{210}Pb-dating results (CRS model), although the $^{210}\mathrm{Pb}_{\mathrm{ex}}$ decay effect predominantly controlled the $^{210}\mathrm{Pb}_{\mathrm{ex}}$ distribution in Lake Bosten. In the lower part of the sediment core, where the algal productivity was relatively low (Fig. 3), the differences between the CRS and MLR models were much larger than those in the upper part (Fig. 4), where algal productivity was much higher. This indicated that when the concentration of algal-derived organic matter was relatively low, variations in algal-derived organic matter would have a larger influence on the ^{210}Pb dating than they do at higher concentrations. Based on Eqs. (3)–(7), n in Eq. (3) was found to decrease with increasing lake trophic levels. This may also suggest that the $^{210}\mathrm{Pb}_{\mathrm{ex}}$ activity was more influenced by algal-derived organic matter variations in low trophic lakes than it was in high trophic lakes. In oligotrophic lakes, in which there were relatively few organic particulates, an increase in the amount of organic particulates would associate a higher percentage of $^{210}\mathrm{Pb}_{\mathrm{ex}}$ with the sediment, but in a eutrophic lake, in which organic particulates were relatively abundant, the same increase in the amount of organic particulates would associate a relatively lower percentage of $^{210}\mathrm{Pb}_{\mathrm{ex}}$ with the sediment.

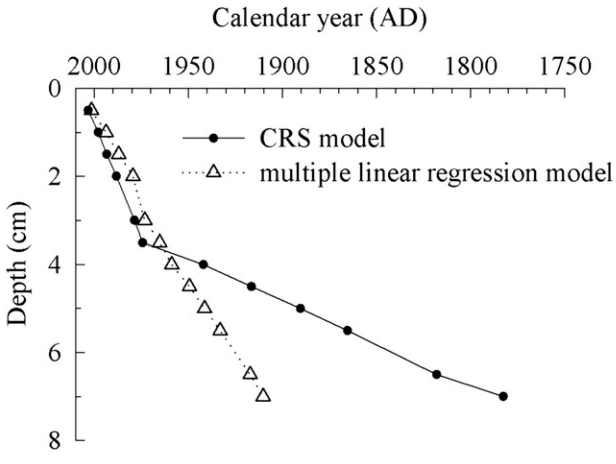

Fig. 4. ^{210}Pb dating using the CRS model and multiple linear regression model in Lake Bosten.

4. Conclusions

In eutrophic Lake Hongfeng, medium-trophic Lake Bosten, and oligotrophic Lake Shuangta, significant positive correlations were found in each case between TOC and $^{210}Pb_{ex}$, indicating that TOC plays an important role in the distribution of $^{210}Pb_{ex}$ in sediments, regardless of the trophic levels of the lakes. With a view to determining the probable different roles of the organic components, Rock-Eval pyrolysis was used to separate the TOC into S1, S2a, S2b, and RC components. Among the different organic components, S2a, which was found to be a better indicator of algal productivity, was the most important factor in controlling the $^{210}Pb_{ex}$ activity in sediments. Vertical scavenging by algal-derived organic matter may be the primary mechanism. The MLR models (models corrected for algal-derived organic matter) proposed in the present study give good interpretations of the relationships between $^{210}Pb_{ex}$ activity in sediments and algal-derived organic matter, and the $^{210}Pb_{ex}$ decay effect, and also of the $^{210}Pb_{ex}$ profiles in the three lakes. Although the corrected model may not actually determine the influence of organic components on ^{210}Pb dating, it would help in the further investigation of the relationships and help to find more accurate models. Lake eutrophication has become a widespread and serious problem in recent years. Our observation suggested that the accuracy of ^{210}Pb dating in sediments would not only be largely influenced by the eutrophicating process, but also by the oligotrophic status, which has relatively small algal-productivity variations. It is very important, and also urgent, to quantitatively describe the impact of organic matter on $^{210}Pb_{ex}$ in sediments and to develop a more accurate model for the sediment dating. As chronology is the basis of many studies into environmental pollution, much more attention should be paid to the influence of organic matter on $^{210}Pb_{ex}$ in lake sediments.

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