

Chemical Geology 224 (2005) 223 – 236

www.elsevier.com/locate/chemgeo

Coupling between $^{210}Pb_{ex}$ and organic matter in sediments of a nutrient-enriched lake: An example from Lake Chenghai, China

G.J. Wan^{a,*}, J.A. Chen^a, F.C. Wu^a, S.Q. Xu^a, Z.G. Bai^a, E.Y. Wan^a, C.S. Wang^a, R.G. Huang^a, K.M. Yeager^b, P.H. Santschi^b

^a State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550002, China
^b Laboratory for Oceanographic and Environmental Research, Department of Marine Scienc Galveston, TX 77551, USA

Received 3 November 2004; received in revised form 29 June 2005; accepted 29 July 2005

Abstract

Sediment cores were collected from deep-water areas of Lake Chenghai, China in June 1997. The vertical profile of ¹³⁷Cs activity gives reliable geochronological results. The results also indicate that sediment accumulation rates in deep-water areas of Lake Chenghai were relatively constant in recent decades, averaging 0.43 g cm⁻² y⁻¹, despite a variable organic carbon influx. Lake Chenghai were relatively constant in recent decades, averaging 0.43 g cm⁻² y⁻¹, despite a variable organic carbon influx.
²¹⁰Pb_{eq} (=²²⁶Ra) activity was relatively constant also, with an average value of 54. $(= {}^{210}Pb_{total} - {}^{226}Ra)$ decreased exponentially, resulting in somewhat lower sediment accumulation rates (0.3 g cm⁻² y⁻¹). These lower rates are likely less reliable, as the relatively large fluctuations in $^{210}Pb_{ex}$ activities correlate closely to the organic carbon (C_{org}) content of the sediments. For example, the vertical profile of ²¹⁰Pb_{ex} activity displays peaks at mass depths of 3.7–4.7 g cm⁻² (10–12) cm) and $10-11$ g cm⁻²(25-28 cm), similar to the maxima in the vertical profile of C_{org}. This phenomenon must be related to the delivery of particulate organic matter (POM) from the water to the sediments, or to watershed soil erosion. Since the mean atomic ratios of H_{org}/C_{org} and C_{org}/N_{org} in Lake Chenghai sediments are 5.5 and 7.0, respectively, indicating that POM was predominantly derived from the remains of authigenic algae, this eliminates watershed erosion rates as a primary control on lake sedimentation rates as resolved by ²¹⁰Pb_{ex}. Sedimentation fluxes ($F(C_{org})$) of particulate organic carbon since 1970 varied between 60 to 160 g m⁻² y⁻¹ , and appeared to closely influence variations in ²¹⁰Pb_{ex} concentrations. For example, sedimentation fluxes of ²¹⁰Pb_{ex} (F(²¹⁰Pb_{ex})) showed maxima in the years 1972–1974 and 1986–1989, likely reflecting historical variations of lake biological productivity or carbon preservation.

 $© 2005 Elsevier B.V. All rights reserved.$

Keywords: $^{210}Pb_{\text{ev}}$; ^{137}Cs ; C–H–N; Sediment record; Lake Chenghai; Organic carbon

1. Introduction

210Pb associated with lake sediments is composed of both the fraction resulting from radioactive decay of ²²⁶Ra (portion in equilibrium (²¹⁰Pb_{eq})) and the fraction produced from radioactive decay of atmospheric 222 Rn

* Corresponding author. Fax: +86 851 5891609. E-mail address: gjw@ms.gyig.ac.cn (G.J. Wan).

⁽non-equilibrium, or excess portion $(^{210}Pb_{ex})$). Principle assumptions of most commonly applied ²¹⁰Pb sediment dating methods are that the flux of $2^{10}P_{\rm{ex}}$ derived from atmospheric fallout and the sedimentation rate are relatively constant, resulting in a $^{210}Pb_{ex}$ activity that decreases exponentially with depth. Over the last several decades, many researchers have reported good results using $^{210}Pb_{ex}$ dating. However, mixing of surface sediment particulates ([Edgington and Robbins](#page-11-0),

^{0009-2541/\$ -} see front matter © 2005 Elsevier B.V. All rights reserved. doi:10.1016/j.chemgeo.2005.07.025

1976; Dominik et al., 1981; Bloesch and Evans, 1982), loss of ²²²Rn from surficial sediments [\(Imboden and](#page-11-0) Stiller, 1982), post depositional remobilization of $2^{10}Pb$ (and 210 Po) at the sediment–water interface ([Wan et al.](#page-12-0), 1987; Santschi et al., 1990; Benoit and Hemond, 1991) and the correction for $^{210}Pb_{eq}$ with corresponding hori-zon samples [\(Wan et al., 1990](#page-12-0)), may lead to $\frac{210}{P}$ b_{ex} anomalies in sediment profiles, thus affecting the results of ²¹⁰Pb dating.

 $^{210}Pb_{ex}$ derived from both atmospheric deposition and watershed erosion are transferred into lake water, which is then quickly, but often not homogeneously, incorporated into lake sediments. ${}^{210}Pb_{ex}$ in lake water is brought into association with sediments along with organic particulates ([Krishnaswamy et al., 1971\)](#page-11-0). Vertical scavenging is the primary mechanism of transport for 210Pb ([Baskaran and Santschi, 2002\)](#page-11-0). Strong associations of $^{210}Pb_{ex}$ with particulate organic carbon (POC), such as observed here, agree with data from marine and terrestrial settings, indicating that the role of POC in isotope fractionation, transport and sequestration merits further investigation ([Paulsen et al., 1999](#page-12-0); Yeager and Santschi, 2003). If the flux of organic particulates is relatively constant in lake water, the transfer of $^{210}Pb_{ex}$ from lake water onto sediments will be relatively constant as well. However, when significant enrichment of nutrients in lake water occurs, this can result in changes in ecological conditions due to rapid enhancement of primary production. At Lake Chenghai, a coincident increase in the flux of algal remains to the lake bottom resulted in increased deposition and transfer of $^{210}Pb_{ex}$ to shallower oxygenated

Fig. 1. Bathymetric map of sampling sites in Lake Chenghai, SW China.

Table 1

lake sediments and possibly to remobilization from deeper, reducing sediments. For example, the accumulated amount of ${}^{210}Pb_{ex}$ in deeper-laying sediments in Lake Greifen, Switzerland, accounted for only 38% of the total influx [\(Wan et al., 1987\)](#page-12-0).

Sedimentary carbon is composed of both organic and inorganic fractions. In lakes, organic carbon mainly originates from hygrophyte and terrestrial plant detritus, whereas inorganic carbon consists of allochthonous and authigenic carbonates. The universality and polymorphism of sedimentary carbon enables it to record a considerable range of regional environmental conditions.

This paper focuses on this phenomenon and its prospective applications based on variation of ${}^{210}Pb_{ex}$ profiles in sediment cores of Lake Chenghai, which is located on the Yunnan–Guizhou plateau of China, and is a ombotrophic, nutrient-enriched lake, for which one can expect to find a clear, coupled relationship between $^{210}Pb_{ex}$ and organic matter in sediment cores.

2. Study area

Lake Chenghai ([Fig. 1,](#page-1-0) Table 1) is located approximately 20 km south of Yongsheng County, northwestern Yunnan, China (N 26° 27′~26°38′,E 100°38′~100 41′). Formed during the early Pleistocene Epoch, Lake Chenghai is a tectonic-fault depression lake, linking the Jinsha River with the Haikou River, and is part of the Yangtze River Basin. Since the middle of the Ming Dynasty (1690 A.D.), the lake water level has progressively decreased and the Haikou River has stopped flowing naturally. As a result, no water has flowed from the lake since then, and the lake has become ombotrophic. Over the course of 290 years (1690–1980) the lake water level had decreased by 39 m, with an average annual decrease of 0.134 m ([Yunnan Provincial Department of](#page-13-0) Water Conservancy et al., 1984).

This region has a temperate climate and is influenced by the southwest monsoon in the summer. Bedrock in this region is varied, including basalts, sandstones, dolomitic limestones and shales. Yunnan pine trees and other conifers, broad-leaf and mixed forests and under story dominate local vegetation. Lake water is yellow-green in color and weakly alkaline with a high hardness, and the total ion concentration in lake waters is up to 1 g L^{-1} (Table 2), approaching the lower limit ion concentration of salt lakes. The lake is eutrophic according to the definition of the Organization for Economic Cooperation and Development ([OECD, 1982\)](#page-12-0), with ortho-phosphate-P concentrations of 47 μ g/L, and nitrate-N concentrations of 220 μ g/L at the time of sampling. Abundant sunlight, favorable water temperature and aquatic chemistry create an environment conducive for the proliferation of blue-green algae. The dominant species of algae here include the Anabaena, the Microcystis and the Oscillatoria, indicating that Lake Chenghai is a moderately nutrientenriched lake ([Tao et al., 1999\)](#page-12-0).

3. Sampling and analytical methods

Four sediment cores were collected from Lake Chenghai in June of 1997 at a water depth of approx-

Table 2

(1) Average value based on the different water depths $(0, 5, 10, 15, 20 \text{ m}$ and boundary). Sampled $6/8/97$; air temperature: 25.5°C; analyzed 7/10/97. (2) Measured in situ.

imately 28 m using a sediment–water interface sampler designed by [Yuan et al. \(1993\)](#page-13-0) and a piston core sampler provided by the Institute of Limnology, Austrian Academy of Sciences. Sampling points were located approximately 2 km from the lake banks, tributary inlets and lake outlets [\(Fig. 1](#page-1-0)). The cored sediments were undisturbed, as indicated by the clear water–sediment interface and the preservation of fine sediment laminations. The sediment core CH970608-1, with a length of 43 cm, was sectioned at 1 cm intervals in situ using the slicing device developed by [Yuan et al.](#page-13-0) (1993). Sediment samples were weighed immediately after collection, dried using a vacuum freeze drier (Techcorp FD-3-85-MP-79-36 mT), and then reweighed to determine mass depths and porosity. Dried samples were ground to ≤ 0.15 mm in diameter for radiochemical assay.

 $137Cs$ and $226Ra$ activities were determined using gamma-spectrometry on a Canberra S-100 multi-channel spectrometer mated to a GC5019 H-P® Ge coaxial detector (efficiency 50%) or a GCW3022 H-P[®] Ge well detector (efficiency 30%). The peaks of ^{137}Cs and 226Ra used to determine their respective activities were 661.6 and 186.2 KeV, respectively. Liquid standards were used and included those supplied by the Institute of Atomic Energy, Chinese Academy of Sciences, and Isotope Products Laboratories, Gentech Industries Inc., USA (Catalog No.: 7137, and Source No.: 586-26-2 for 137Cs, Catalog No.: 7226, and Source No.: 586-26-4 for 226 Ra).

The elements C, H and N were analyzed on a Perkin Elmer PE2400 elemental analyzer, with a measurement error of $\leq 5\%$. Inorganic carbon (C_{inorg}) was measured by using the volumetric method ([Wang et al., 1996\)](#page-12-0), with an error of $\leq 10\%$.

4. Results and discussion

4.1. Particulates in the floc layer

The unconsolidated floc layer at the sediment– water interface of Lake Chenghai is approximately 1 cm thick for all sediment cores and was composed of clayey particulates. The mass depth over this interval in sediment core CH970608-1 was 0.073 g cm^{-2} , with a porosity of 97.0%. [Tables 3 and 4](#page-4-0) list the main characteristics of this horizon and the remainder of the core. In the floc layer: 1) The total carbon content is relatively low $(C_{total} = 2.14\%)$, of which C_{inorg} and C_{org} account for 0.87% and 1.26%, respectively, demonstrating that lake water composition at the boundary ([Table 2\)](#page-2-0) is oversaturated relative to carbonate sediments; 2) ^{137}Cs activity in this layer is slightly higher than that at the top of the underlying sediment layers, i.e., ≈ 7.0 vs. ≈ 5.1 Bq kg⁻¹, indicating that the clay-rich particulates from the drainage basin are an important factor, indicating recent sediment input, and that no significant particulate mixing has occurred at the sediment/water interface here; 3) ²¹⁰Pb_{total} (84.6 ± 4.2 Bq kg⁻¹) is also lower than that of the immediately underlying sediment layers $(104.2 \pm 4.7 \text{ and } 110.6 \pm 5.4 \text{ Bq kg}^{-1}),$ suggesting that this layer can display disequilibrium conditions.

4.2. Evidence from 137 Cs: average sediment accumulation rate

Investigations since the 1960s have shown that the fallout radionuclide $137Cs$ is an appropriate tracer in the study of sedimentation in lacustrine environments such as Lake Chenghai, and that it can be used to calculate average sediment accumulation rates ([Ritchie et al.](#page-12-0), 1970; Krishnaswamy et al., 1971; McHenry et al., 1973). Only two time markers developed from fallout $137Cs$ in Northern Hemisphere sediments can consistently be used for this purpose, those occurring in 1952 displaying the beginning of atmospheric bomb tests, and 1963, marking their moratorium. Since ¹³⁷Cs fallout from 1952 was less than that in 1963 and considering the isotope's half-life (\approx 30 years), this first peak is difficult to recognize ([Ritchie and McHenry, 1990\)](#page-12-0). The time marker established on the basis of fallout 137 Cs accumulated in 1963 is of major geochronological significance.

Usually, anthropogenic radionuclides that are injected into the stratosphere move back to the troposphere with a mean residence time of \sim 1 year, where they are washed out over a time scale of 1–2 weeks. The peak of annual fallout delivery for ¹³⁷Cs in the Northern Hemisphere was in 1963, just after the end of the moratorium in 1962 ([Playford et al., 1990; Agudo, 1998\)](#page-12-0). $137Cs$ generally behaves non-conservatively in the freshwater environment, being strongly adsorbed on suspended particulates. Because of this, it can move with a residence time approaching that of suspended matter ([Ritchie and](#page-12-0) McHenry, 1990).

Based on a box model of geochemical mass balance ([Imboden et al., 1980\)](#page-11-0), the residence time of suspended particles in Lake Chenghai can be calculated to be,

$$
\tau_s = \frac{h}{S} \left(C_d + \frac{1}{K_d} \right) = 1.19 \text{ yr}.
$$

where, h = average water depth (25.7 m, [Table 1\)](#page-2-0); $S = av$ erage sediment accumulation rate (0.43 \pm 0.01 g cm⁻² yr^{-1} , [Table 5\)](#page-5-0); C_d = average concentration of suspended

Table 3 Summary ²¹⁰Pb and ¹³⁷Cs data in sediment core CH970608-1 of Lake Chenghai

Depth cm	Mass depth $(g cm^{-2})$	Years a	A.D.	$137Cs^{(2)}$ (Bq kg ⁻¹)		$^{210}Pb_{eq} (= ^{226}Ra)$ $(Bq Kg^{-1})$		$Pb_{total}^{(2)}$ (Bq kg ⁻¹)		$^{210}Pb_{ex}^{(2)}(Bq Kg^{-1})$	
Floc	0.04	0.1	1997.4	7.00	±1.46	$\qquad \qquad -$		84.6	± 4.2	\equiv	
$\mathbf{1}$	0.25	0.6	1996.9	5.07	± 0.54	53.0	±1.3	104.2	±4.7	51.2	± 2.7
$\mathfrak{2}$	0.53	1.2	1996.3	5.14	± 0.61	58.8	±1.4	110.6	± 5.4	51.8	± 3.2
\mathfrak{Z}	0.78	1.8	1995.7	4.19	± 0.59	54.8	±1.5	101.4	±4.4	46.6	± 2.5
$\overline{4}$	1.24	2.9	1994.6	5.81	± 0.76	53.9	$\pm~1.8$	96.5	±4.8	42.6	± 2.6
5	1.73	4.0	1993.5	5.65	± 0.58	55.2	±1.4	93.0	± 5.8	37.8	± 2.7
6	2.11	4.9	1992.6	6.57	± 0.66	58.9	\pm 1.7	80.6	±4.4	21.7	\pm 1.7
τ	2.55	5.9	1991.6	9.75	± 0.72	50.1	\pm 1.3	75.4	±4.8	25.3	$\pm\,1.5$
8	2.98	6.9	1990.6	9.89	± 0.80	48.6	± 1.7	69.7	± 3.5	21.0	$\pm~1.0$
9	3.36	7.8	1989.7	7.92	± 0.82	60.6	± 2.1	84.4	±4.5	23.8	± 2.0
10	3.69	8.5	1989.0	2.91	± 0.48	60.2	±1.4	111.7	\pm 5.1	51.5	± 3.1
11	4.15	9.6	1987.9	2.66	± 0.59	72.4	±1.5	124.8	± 5.4	52.4	± 3.5
12	4.65	10.7	1986.8	3.82	± 0.41	54.8	$\pm\,1.1$	107.8	±4.8	53.0	± 2.7
13	5.00	11.6	1985.9	4.49	± 0.45	57.5	\pm 1.2	90.8	± 4.1	33.3	±1.9
14	5.38	12.4	1985.1	4.88	± 0.45	50.5	\pm 1.1	90.2	± 5.0	39.7	± 2.2
15	5.77	13.3	1984.2	5.29	± 0.56	67.2	\pm 1.4	78.6	± 4.1	11.4	\pm 1.4
16	6.16	14.2	1983.3	6.25	± 0.53	59.2	± 1.2	84.7	± 3.8	25.5	±1.6
17	6.55	15.1	1982.4	5.87	± 0.60	66.9	± 1.3	74.2	± 3.7	7.3	$\pm\,1.1$
18	6.95	16.0	1981.5	7.24	± 0.61	52.9	±1.4	81.3	±4.7	28.4	$\pm\,1.8$
19	7.38	17.0	1980.5	7.54	± 0.60	64.9	± 1.7	75.7	± 3.8	10.8	$\pm\,1.3$
20	7.83	18.1	1979.4	7.71	± 0.66	49.6	±1.6	77.1	± 4.3	27.5	\pm 1.6
21	8.25	19.0	1978.5	8.21	± 0.61	70.4	\pm 1.5	78.2	± 3.9	7.7	\pm 1.4
22	8.67	20.0	1977.5	9.75	± 0.76	50.4	±1.6	77.4	± 4.7	27.0	±1.7
23	9.10	21.0	1976.5	10.40	± 0.74	48.0	\pm 1.5	68.9	± 3.3	20.8	$\pm\,0.9$
24	9.56	22.1	1975.4	11.85	± 0.83	45.7	$\pm\,1.6$	74.8	± 3.8	29.0	±1.3
25	10.00	23.1	1974.4	9.89	± 0.74	53.4	±1.7	93.6	± 4.2	40.2	± 2.3
26	10.37	24.0	1973.5	4.39	± 0.56	57.6	$\pm\,1.3$	109.7	± 5.7	52.0	± 3.3
27	10.72	24.8	1972.7	4.24	± 0.52	48.5	± 1.3	92.5	± 4.1	44.0	$\pm\,2.0$
28	11.07	25.6	1971.9	5.38	± 0.55	51.3	±1.3	92.1	±4.7	40.8	± 2.3
29	11.45	26.4	1971.1	8.42	± 0.76	50.5	±1.4	71.0	± 5.0	20.5	±1.3
30	11.91	27.5	1970.0	10.44	± 0.73	45.7	\pm 1.5	71.4	±4.7	25.7	±1.3
31	12.40	28.6	1968.9	13.12	± 0.85	43.5	±1.6	33.7	\pm 1.7	\equiv	\overline{a}
32	12.88	29.8	1967.7	16.86	± 1.13	53.6	\pm 2.2	60.7	± 4.2	7.1	± 0.6
33	13.37	30.9	1966.6	17.19	± 1.20	49.4	± 2.1	36.8	± 2.4	$\overline{}$	
34	13.84	32.0	1965.5	17.16	\pm 1.01	48.5	$\pm\,1.8$	56.6	± 2.6	8.1	$\pm\,0.2$
35	14.30	33.0	1964.5	21.45	± 1.37	49.3	± 2.3	55.6	±4.5	6.3	$\pm\,0.2$
36	14.73	34.0	1963.5	19.71	± 1.20	57.9	± 2.3	57.1	± 2.8	$\overline{}$	\equiv
37	15.18	35.1	1962.4	16.04	±1.14	47.3	± 2.0	57.8	± 3.6	10.5	± 0.3
38	15.64	36.1	1961.4	14.48	± 1.17	51.6	± 2.3	55.5	± 2.8	3.9	$\pm\,0.1$
39	16.12	37.2	1960.3	14.51	±1.00	47.6	$\pm~1.8$	56.5	± 2.7	8.8	± 0.2

(1) Sampled 6/8/97; measured: $3/98$, errors expressed at 1σ . (2) Corrected to sampling time.

particles in Lake Chenghai waters $(1 \times 10^{-6} \text{ g cm}^{-3})$; $K_d =$ ¹³⁷Cs distribution coefficient between suspended particles and lake water $(5 \times 10^3 \text{ cm}^3 \text{ g}^{-1})$ [\(Santschi et](#page-12-0) al., 1986, 1988; Robbins and Edgington, 1975).

Therefore, the half-life of $137Cs$ sinking to the bottom of Lake Chenghai in association with suspended particles is $t_{1/2} = \tau_s * \ln 2 = 0.83$ yr. Accordingly, it is reasonable to assume that the retention time of $137Cs$ in association with sediments of Lake Chenghai is approximately one year. Therefore, the year 1964 is used as a dating time marker for the $137Cs$ activity maximum in lake sediments here. It is also known

that the vertical profile of $137Cs$ may be affected by diffusion and post depositional mobility in sediments. Although the mobility of $137Cs$ in these sediments appears to be limited, it can be an important factor affecting the accuracy of $137Cs$ dating ([Davis et al.](#page-11-0), 1984). Other work has shown that $137Cs$ diffusion post deposition did not significantly change the position of the accumulation maxima of $137Cs$ in sediments ([Robbins and Edgington, 1975; Wan et al.](#page-12-0), 1985, 1987).

The ¹³⁷Cs activity in sediment core CH970608-1 displays a maximum value at a mass depth of 14.30 g

Table 4 Summary C–H–N data in sediment core CH970608-1 of Lake Chenghai

Depth/cm	Mass depth/ $(g cm^{-2})$	Porosity/%	$C_{\rm total}/\%$	C_{inorg} /%	$C_{org}/\frac{6}{6}$	$\rm H_{org}/\%$	$\rm N_{org}/\%/$	H_{org}/C_{org} by atom	C_{org} / N_{org} by atom
Floc	0.04	97.0	2.14	0.87	1.26	0.45	0.34	4.2	4.4
$\mathbf{1}$	0.25	85.5	2.53	1.03	1.50	0.70	0.30	5.6	5.8
$\sqrt{2}$	0.53	91.5	2.71	1.21	1.50	0.71	0.26	5.7	6.7
3	0.78	88.7	2.72	1.22	1.50	0.72	0.20	5.8	8.9
$\overline{4}$	1.24	$\overline{}$	2.56	1.04	1.52	0.63	0.26	5.0	6.7
5	1.73	86.1	2.24	0.91	1.33	0.70	0.22	6.3	7.1
6	2.11	82.7	2.04	0.79	1.25	0.77	0.24	7.4	6.2
$\overline{7}$	2.55	81.8	1.91	0.80	1.10	0.69	0.20	7.5	6.4
8	2.98	83.4	2.00	0.77	1.22	0.85	0.21	8.3	6.9
9	3.36	85.5	2.26	1.03	1.23	0.73	0.24	7.1	$6.0\,$
10	3.69	87.3	3.15	1.46	1.70	0.63	0.34	4.4	5.9
11	4.15	\equiv	3.17	1.42	1.75	0.67	0.34	4.6	$6.0\,$
12	4.65	84.8	2.54	1.34	1.20	0.73	0.27	7.3	5.2
13	5.00	86.0	2.45	1.52	0.93	0.65	0.26	8.3	4.2
14	5.38	83.7	2.55	1.86	0.69	0.61	0.24	10.6	3.3
15	5.77	84.2	2.11	1.03	1.08	0.58	0.24	6.5	5.2
16	6.16	84.0	2.02	1.03	0.99	0.71	0.22	8.6	5.1
17	6.55	84.2	2.11	0.97	1.13	0.59	0.10	6.2	13.4
18	6.95	83.1	2.02	1.18	0.84	0.73	0.17	10.5	5.7
19	7.38	81.8	2.05	1.23	0.83	0.61	0.13	8.9	$7.2\,$
20	7.83	81.6	1.96	1.10	0.86	0.65	0.11	9.1	8.8
21	8.25	83.9	1.94	1.06	$0.88\,$	0.65	0.12	8.9	$8.7\,$
22	8.67	81.9	1.89	0.99	0.89	0.66	0.14	8.9	7.3
23	9.10	82.4	1.89	0.95	0.94	0.60	0.08	7.6	13.4
24	9.56	80.6	1.84	0.83	1.01	0.74	0.16	8.7	7.5
25	10.00	83.4	2.42	1.20	1.22	0.79	0.26	7.7	5.5
26	10.37	86.0	2.87	1.61	1.26	0.82	0.23	7.8	6.3
27	10.72	85.8	3.46	1.41	2.05	0.83	0.29	4.8	8.2
28	11.07	85.4	2.29	1.32	0.97	0.80	0.25	9.8	4.5
29	11.45	83.6	1.90	1.06	0.84	0.71	0.16	10.2	$6.0\,$
30	11.91	78.8	1.78	0.93	0.85	0.69	0.17	9.7	6.0
31	12.40	81.5	1.79	0.90	0.89	0.67	0.17	9.0	6.3
32	12.88	78.9	1.66	0.69	0.97	0.67	0.18	8.3	6.1
33	13.37	81.8	1.69	1.11	0.58	0.73	0.26	15.0	2.6
34	13.84	79.5	1.70	1.31	0.38	0.69	0.21	21.6	2.2
35	14.30	83.0	2.03	1.45	0.58	0.83	0.22	17.3	$3.0\,$
36	14.73	82.1	1.87	1.46	0.40	0.75	0.20	22.3	2.4
37	15.18	80.9	1.85	1.45	0.39	0.74	0.19	22.8	2.4
38	15.64	81.6	1.90	1.61	0.29	0.72	0.20	29.4	1.7
39	16.12	78.9	2.00	1.70	0.30	0.64	0.20	25.2	1.8

(1) Sampled 6/8/97; measured: 3/98.

 cm^{-2} (or 35 cm, [Table 3\)](#page-4-0), marking the 1964 layer in the sediment and coinciding with the year of maximum global fallout (1963). Using this position (1964) as a discrete time marker, the average sediment accumula-

Table 5

Sediment accumulation rates in Lake Chenghai derived from ¹³⁷ Cs							
---	--	--	--	--	--	--	--

tion rate was determined to be 0.43 ± 0.01 g cm⁻² y⁻¹ (Table 5, [Fig. 2\)](#page-6-0).

There is a smaller but obvious secondary peak in $137Cs$ annual fallout in 1974 ([Fig. 2\)](#page-6-0). If this secondary peak of 137Cs is a useful complimentary time marker, it should be similarly considered representative of the year 1975 in terms of the deposition of lake sediments. This secondary peak in $137Cs$ activity occurs in core CH970608-1 at a mass depth of 9.56 g cm⁻² (or 24 cm) ([Table 3,](#page-4-0) [Fig. 2\)](#page-6-0), which is consistent with the year 1975. This coincides with a small global fallout peak in 1974, a result of the largest atmospheric nuclear weapon test of a non-moratorium country (China) in

Fig. 2. Vertical profile of 137Cs in sediment cores in Lake Chenghai, Lake Erhai ([Xu et al., 1999](#page-13-0)) and Lake Hongfeng ([Wan et al., 1991\)](#page-12-0) compared to global deposition of $137Cs$ in the Northern Hemisphere ([Playford et al., 1990; Agudo, 1998\)](#page-12-0) decay corrected to 1996 (by the authors).

1973, having a magnitude of 2–3 Mt ([Katsuragi, 1983\)](#page-11-0). Using the position of this secondary $137Cs$ peak (1975) as a discrete time marker for dating, the average sediment accumulation rates were determined to be 0.43 ± 0.01 g cm⁻² y⁻¹, identical to the rate calculated using the time marker of 1964, which indicates also that sedimentation rates between the years 1964 and 1975 were similar and have remained essentially constant over the last 40 years of record.

In addition, the nuclear accident at Chernobyl, in the former USSR (April 1986) resulted in the global atmospheric transportation of a significant amount of $137Cs$, which may then have been recorded in lake sediments [\(Santschi et al., 1988\)](#page-12-0). The relatively minor peak in ¹³⁷Cs activity above a mass depth of 2.98 g cm⁻² (or 8 cm) associated with the year 1990 is most likely related to the Chernobyl incident (1986).

 $137Cs$ activity in sediment core CH970608-1 displays a three-peak pattern with sediment depth [\(Table](#page-4-0) 3, Fig. 2), similar to vertical profiles of 137Cs in sediment cores of Lakes Erhai and Hongfeng, China (Fig. 2) ([Wan et al., 1991; Wan, 1999; Xu et al., 1999; Xu](#page-12-0) and Wan, 2001). The three peaks of $137Cs$ activity in these sediment cores are therefore assumed here to correspond to the years 1964, 1975 and 1987, as previously discussed.

4.3. Evidence from $^{210}Pb_{ex}$: do specific peaks reflect variable sediment accumulation?

The ²¹⁰Pb_{eq} is an equilibrium value with ²²⁶Ra measured by gamma spectrometry $(^{210}Pb_{eq} = {}^{226}Ra)$. It can be seen from [Table 3](#page-4-0) and Fig. 3 that the $^{210}Pb_{eq}$ (²²⁶Ra) profile was relatively constant $(\text{mean} = 54.3 \pm 3.2 \text{ Bq kg}^{-1})$ throughout the core. Some minor fluctuation of this value did occur, most likely due to some annual and/or seasonal variation in the supply of particulate matter to the lake. 226 Ra, an intermediate decay product of 238U, is present in the crystal lattice of clay minerals. In the particulates of surface soils, 226 Ra is for the most part present as a "residual species", remaining after soils have been leached by acidic waters ([Bai et al., 1997, 2002; Wan et](#page-11-0) al., 2001).

The $^{210}Pb_{total}$ is an equilibrium value with ^{210}Po measured by alpha spectrometry, which is corrected to sampling time (June 1997). $^{210}Pb_{ex}$ is determined by difference (i.e., $^{210}Pb_{ex} = ^{210}Pb_{total} - ^{210}Pb_{eq}$). Fig. 3 shows that ${}^{210}Pb_{\text{total}}$ and ${}^{210}Pb_{\text{ex}}$ are consistent with each other, displaying similar peak distribution patterns. The vertical profile of $^{210}Pb_{ex}$ shows obvious fluctuations. The features of interest include: 1) at depths of 3.69–4.65 g cm⁻² (or 10–12 cm) and 10.00–11.07 g cm^{-2} (or 25–28 cm), there are two distinct $^{210}\text{Pb}_{\text{ex}}$ peaks; 2) in the depth interval between these two peaks, $^{210}Pb_{ex}$ values show some minor fluctuations; 3) below a mass depth of 12.40 g cm⁻² (or 31 cm), $^{210}Pb_{ex}$ is at background or supported levels.

Use of $^{210}Pb_{ex}$ as a geochronological tool originated with [Goldberg \(1963\)](#page-11-0) who successfully dated a Greenland ice core. Then, [Krishnaswamy et al. \(1971\)](#page-11-0) and [Koide et al. \(1972\)](#page-11-0) employed ²¹⁰Pb_{ex} as a tool for determining recent chronologies in lake and coastal marine sediments, respectively. Since then, the $^{210}Pb_{ex}$ methodology has been used to date sediments in a wide variety of aquatic systems. A conceptual model for $^{210}Pb_{ex}$ dating must be translated ultimately into a means of assigning annual layers to sediment sections,

Fig. 3. Vertical profile of 210Pb in sediment core CH970608-1 of Lake Chenghai.

i.e., an age–depth relationship [\(Robbins and Herche,](#page-12-0) 1992). Conceptual models, including the constant initial concentration (CIC) and constant rate of supply (CRS) for sedimentation rate determinations using $210Pb_{ex}$ methods are discussed in detail in [Robbins](#page-12-0) (1978) and [Appleby and Oldfield \(1978\)](#page-11-0). Using the CIC model $(C=Co \times e^{-0.107z^2}, r=0.61)$ for the $^{210}Pb_{ex}$ profile, we can calculate an average sediment accumulation rate of 0.29 g cm⁻² y⁻¹, with Co=51.5 Bq kg^{-1} . This value is lower than the dating results on the $137Cs$ time-scale [\(Table 5](#page-5-0)). Using the CRS model, which is more appropriate for an ombotrophic lake, one calculates an average sediment accumulation rate of 0.32 g cm⁻² y⁻¹. The discrepancy between ²¹⁰Pbbased and bomb fallout based estimates of sediment accumulation rates have been observed before [\(Wan et](#page-12-0) al., 1987), and could be related to remobilization of $^{210}Pb_{ex}$ from the sediment, as discussed below.

Such a distinctive peak distribution could be a result of either increasing influx of $^{210}Pb_{ex}$ from waters into sediments, or a decreasing supply of particulate matter via erosion from the small drainage basin. However, the similar distribution of $^{210}Pb_{eq}$ to that of sedimentary organic carbon (SOC) suggests that this peak distribution is the result of an increase in the influx of $^{210}Pb_{ex}$ from waters into sediments, as the majority of $210Pb$ occurs in association with suspended materials in the lake (Figs. 4–6; [Tables 3, 4](#page-4-0)).

SOC has been shown to strongly and positively correlate with ²¹⁰Pb in marine settings [\(Paulsen et al.,](#page-12-0) 1999). SOC has also been shown to correlate positively and significantly with $^{210}Pb_{ex}$ in near surface (0–2 cm) samples of terrestrial sediments from the coastal plain of Texas ([Yeager and Santschi, 2003\)](#page-13-0). This same relationship is found in Lake Chenghai, when comparing the values of $^{210}Pb_{ex}$ and ^{137}Cs , decay-corrected to the

Fig. 4. ²¹⁰Pb_{ex} and ¹³⁷Cs profiles in sediment core CH970608-1 of Lake Chenghai.

Fig. 5. Positive relationship between the POC concentration and reactive 210Pb and a negative relationship between the POC fraction and reactive ¹³⁷Cs.

time of deposition, with SOC, a strong positive relationship is evident between $^{210}Pb_{ex}$ and SOC that is the opposite of that for $137Cs$ (Fig. 5). Such relationships suggest uneven deposition patterns for these two nuclides, likely due to post-depositional redistribution patterns of sediments and radionuclides that have also been described in other lakes ([Brunskill et al., 1984](#page-11-0); Schuler et al., 1991; Wieland et al., 1991, 2001), which will be discussed in more detail below.

The $^{210}Pb_{ex}$ peak distribution in these sediments is in good agreement with SOC (Fig. 6). In order to examine a prospective relationship between organic sedimentation and the accumulation of $^{210}Pb_{ex}$, it is necessary to investigate the sources and year-by-year variation of organic matter content in the sediments of Lake Chenghai.

4.4. Radionuclide inventories

The calculated $137Cs$ inventory in this core from Lake Chenghai is 243 ± 4 mBq cm⁻², and that of

Fig. 6. Vertical profiles of C_{org} and $^{210}Pb_{ex}$ in sediment core CH970608-1 from Lake Chenghai.

²¹⁰Pb_{ex} is 697 \pm 8 mBq cm⁻² (Table 6). When compared to the expected lake inventories from atmospheric fallout, i.e., $40-80$ mBq cm^{-2} for 137 Cs ([Playford et al.,](#page-12-0) 1990; Agudo, 1998) and $161-323$ mBq cm^{-2} for $^{210}Pb_{ex}$ ([Lee et al., 2004\)](#page-11-0), both ^{137}Cs and $^{210}Pb_{ex}$ are enriched here (Table 6). Similar discrepancies between sedimentation rates determined by ${}^{210}Pb_{ex}$ and other radionuclides (e.g., 239,240 Pu, 90 Sr and 137 Cs) and varve counting was previously documented for another eutrophic and seasonally anoxic lake, Lake Greifen in Switzerland ([Wan et al., 1987\)](#page-12-0). However, a different pattern of radionuclide enrichment was found for Lake Greifen, where the $^{210}Pb_{ex}$ inventory in that lake was below that expected from atmospheric deposition. It appears, therefore, that Lake Chenghai is different in that low 210Pb-derived sediment accumulation rates are not coupled to low sediment inventories, possibly due to counteractive processes such as sediment focusing effects related to particle deposition.

This pattern of enrichment and depletion of fallout radionuclides in Lake Chenghai can be compared to that from Lake Erhai, both in West Yunnan and Lake Hongfeng in Central Guizhou, China (Table 6). In Lake Erhai, a similar pattern of ${}^{210}Pb_{ex}$ inventory depletion is observed, where $137Cs$ is close compared to atmospheric fallout, while in Lake Hongfeng, the inventories of both radionuclides are enriched compared to expected atmospheric fallout. Lake-wide redistributions of radionuclides leading to sediment focusing effects appears to be a common feature in both small (e.g., [Brunskill et](#page-11-0) al., 1984) and large (e.g., [Edgington and Robbins,](#page-11-0) 1976) lakes.

4.5. Carbon in sediments of Lake Chenghai

4.5.1. Organic and inorganic carbon

Carbon is an important component in lake sediments, and is composed of organic and inorganic fractions. Due to the universality and polymorphism of sedimentary carbon, it is capable of recording a wealth

Fig. 7. C_{org}, C_{inorg} and C_{total} profiles in sediment core CH970608-1 from Lake Chenghai.

of regional environmental information, however, these same characteristics can pose some additional difficulties in interpretation.

For sediment core CH970608 of Lake Chenghai, organic and inorganic carbon concentrations are well correlated above 12.88 g cm^{-2} (or 32 cm) but show an inverse relationship below that depth, likely due to cessation of early diagenesis ([Table 4](#page-5-0) and Fig. 7). More importantly, at depths of $3.69-4.65$ g cm⁻² (or 10–12 cm) and 10.00–11.07 g cm⁻² (or 25–28 cm) the carbon concentration peaks mirror the distribution pattern of $^{210}Pb_{ex}$, indicating a close relationship between $^{210}Pb_{ex}$, SOC and carbonate.

4.5.2. Sedimentary inorganic carbon sources

Sedimentary inorganic carbon consists mainly of autochthonous and allochthonous carbonates. Allochthonous carbonates are produced by weathering of parent rocks and/or soils in the drainage basin and are then transported into the lake by runoff. Autochthonous carbonates include those derived from inorganic precipitation, carbonate shells of organisms and carbonates produced from organic carbon by post depositional diagenesis.

Calcite precipitation requires super saturation with respect to calcite in solution. Two sets of factors may

Table 6

Expected versus measured inventories of $137Cs$ and $210Pb_{ex}$ for Lakes Chenghai, Erhai and Hongfeng, China

Lake	Radionuclide	Expected inv.	Reference	Measured inv.	References	Calculated inv. from simulation
L. Chenghai (West Yunnan)	^{137}Cs	$40 - 80$	(1)	$243 + 4$	This work	$\approx 106 + 2$
	$^{210}Pb_{ex}$	$161 - 323$	(2)	$697 + 8$	This work	$\approx 161 + 2$
L. Erhai (West Yunnan)	^{137}Cs	$40 - 80$	(1)	$52 + 3$	(3)	$\approx 114 + 6$
	$^{210}Pb_{ex}$	$32 - 161$	(2)	305 ± 5	(4)	$\approx 486 + 7$
L. Hongfeng (Central Guizhou)	^{137}Cs	$80 - 160$	(1)	$358 + 5$	(3)	\approx 394 + 6
	$^{210}Pb_{ex}$	484-645	(2)	$1070 + 19$	(5)	\approx 737 + 13

References: (1) [Agudo, 1998; Playford et al., 1990](#page-11-0); (2) [Lee et al., 2004;](#page-11-0) (3) [Bai et al., 2002; Wan et al., 2001;](#page-11-0) (4) [Xu et al., 1999](#page-13-0); (5) [Wan et al.,](#page-12-0) 1990.

cause super saturation. One is biological; algae assimilate $CO₂$ during photosynthesis, leading to a rise of pH and corresponding decrease in $[H⁺]$, which increases the ionic activity product (IAP) and degree of super saturation, eventually inducing calcite precipitation. In short, photosynthesis of hydrophytes assimilates $CO₂$, shifting the equilibrium of the reaction towards the right, inducing calcite precipitation on the outside of the shells of organisms, providing direct evidence for biologically induced calcite formation ([Hodell et al](#page-11-0)., 1998; Kelts and Hsu, 1978; Robbins and Blackwelder, 1992). The other processes capable of super saturation are physical–chemical; including temperature changes, evaporation and condensation of water and solution and release of $CO₂$. For such reactions, temperature is the most obvious variable ([Hodell et al., 1998; Kelts and](#page-11-0) Hsu, 1978; Robbins and Blackwelder, 1992; Wigley and Plummer, 1976): (1) Increased temperature decreases the solubility and dissolution equilibrium constant of calcite, thereby increasing the super saturation degree and promoting calcite precipitation. (2) During warmer years, the thermal stratification of the lake occurs earlier and its duration is longer, resulting in greater photosynthetic pumping of $CO₂$ from the epilimnion, which is favorable for the formation of super saturation conditions. (3) Also during warmer years, blooming phytoplankton populations not only increase calcite super saturation as a result of greater photosynthetic removal of $CO₂$, but also act as nucleation sites for calcite crystallization, accelerating precipitation. (4) More $CO₂$ escapes from the water because of its decreasing solubility at higher temperature, helping to drive calcite precipitation. (5) Finally, in warmer years, water evaporation increases the concentrations of +Ca²⁺, CO₃ and HCO₃ in the water, a condition that also contributes to the formation of super saturation conditions.

The basis for judging the saturation state with respect to calcite is to compare the IAP with the equilibrium constant (K_e) . One convenient method for computation of IAP ([Stumm and Morgan, 1996\)](#page-12-0) is:

$$
IAP = (Ca^{2+})(CO_3^{2-})
$$

= $(r_{Ca^{2+}} \times [Ca^{2+}]r_{HCO_3^-} \times k_2 \times Alk)/(H^+)$ (1)

Where $+r_{Ca^{2+}}$ and r_{HCO_3} are respectively the activity coefficients of Ca^{2+} and HCO_3^- , $[Ca^{2+}]$ is the $+Ca^{2+}$ concentration, k_2 is the second dissociation constant for H_2CO_3 , Alk is the alkalinity of the water, and (H^+) is the H^+ concentration.

The super saturation index of water (IAP/k_e) can be calculated by Eq. (1). Using hydro-chemical data from

Lake Chenghai ([Table 2\)](#page-2-0), results show that the super saturation indexes vary between 1.4–2.3, suggesting that lake water is super saturated with respect to calcite and precipitation can be expected. Previous studies have revealed that sedimentary inorganic carbon in deep water sediments of Lake Chenghai are dominated by the autochthonous carbonate fraction ([Wang et al., 1989](#page-12-0); Wang, 1993; Chen et al., 2002). Supply of terrestrial clastic debris diminishes while the carbonate content in sediments increases with increasing water depth and greater distance from the banks of Lake Chenghai ([Wang et al., 1989\)](#page-12-0), supporting the contention that sedimentary carbonates are derived from autochthonous calcite precipitation instead of allochthonous carbonates derived from catchment erosion. In deep water (≈ 30) m), the calcite content in sediments of Lake Chenghai reaches 10% ([Wang, 1993\)](#page-12-0).

4.5.3. Sources of organic carbon in sediments

Sedimentary organic carbon in lakes mainly originates from a combination of hygrophyte and terrestrial plant detritus. The C–N–H composition of organic matter in lacustrine sediments is closely related to their sources. Organic matter is commonly classified into algal, herbaceous, woody and/or coaly matter according to its structure. Generally, algal organic matter is easily oxidized and is thus seldom preserved, followed by herbaceous organic matter. Woody and coaly types are the most stable. Because of its relatively high content of saturated organic compounds, algal organic matter tends to be hydrogen rich (as indicated by its H_{org}/C_{org} ratio). Herbaceous organic matter is composed mostly of aromatics, with relatively low hydrogen content (and H_{org}/C_{org} signature). Woody organic matter consists predominantly of polycyclic aromatic compounds and so has a lower hydrogen content (and $H_{\text{org}}/C_{\text{org}}$ signature). Coaly organic matter is essentially carbonaceous residue (oxidized or altered plant materials) with the lowest hydrogen content (and H_{org}/C_{org} signature). When the H_{org}/C_{org} ratio of sedimentary organic matter is low, its source cannot be judged directly because low values for this ratio could be attributed to either a predominance of woody or coaly organic matter or to the oxidation of an originally hydrogen-rich algal organic matter fraction. When the $H_{\text{org}}/C_{\text{org}}$ ratio of sedimentary organic matter is high, its source can be identified easily. Sedimentary organic matter with H_{org}/C_{org} ratio values greater than 1.7 usually originates from phytoplankton. However, values of 4–25, likely indicating residual water content, prevent conclusive interpretations. The C_{org}/N_{org} ratio of sedimentary organic matter can be used effectively to

reveal the sources of organic matter. Hygrophyte, such as algal organic matter, is enriched in protein with a $C_{\text{org}}/N_{\text{org}}$ ratio of less than 10, while allochthonous terrestrial organic matter is enriched in humus, which has a C_{org} / N_{org} ratio of 20–30 ([Talbot and Livingstone](#page-12-0), 1989; Dean, 1999).

The organic matter associated with Lake Chenghai sediments has a relatively high H_{org}/C_{org} value (mean is 7.6), which might include associated water. The total range in the ratio value from the sediment core is 4.2– 10.6. The other elemental signatures [\(Table 4](#page-5-0) and Fig. 8), however, also suggest that sedimentary organic matter in the deep water of Lake Chenghai is derived mostly from phytoplankton. Organic matter associated with sediments of Lake Chenghai has a relatively low C_{org} / N_{org} value (mean is 6.7). The total range in the ratio value from the sediment core is 3.3–13.4. The elemental signatures [\(Table 4](#page-5-0) and Fig. 8) therefore suggest that sedimentary organic matter in the deep water of Lake Chenghai is derived specifically and predominantly from the remains of indigenous algae.

4.6. Depositional fluxes of organic carbon

[Fig. 6](#page-7-0) shows the vertical profiles of organic carbon (C_{org}) and ²¹⁰Pb_{ex} in sediment core CH970608-1. All three parameters are distributed similarly with depth.

In the study of recent sedimentation at Lake Erhai, China, researchers reported sedimentary carbon data, which has been used to develop a three-stage evolutionary model of organic matter, i.e., "deposition–de-composition–accumulation" ([Wan et al., 2003\)](#page-12-0). At Lake Chenghai, the C_{org} content within the top 4 cm of core CH970608-1 (mass depth of 1.24 g cm^{-2}) is essentially constant, with an average value of 1.50%, demonstrating that organic matter in the sediments was at steady state in the \approx 3 years represented by that

Fig. 8. Depositional fluxes of $^{210}Pb_{ex}$ and C_{org} over the past 30 years in the sediments of Lake Chenghai.

interval ([Fig. 6\)](#page-7-0). However, below 15.64 g cm^{-2} (or 38) cm) or \approx 36 years of sedimentation, C_{org} again shows a relatively constant value, with an average of 0.30%, which marks the entry of organic matter into the accumulation stage, according to the [Wan et al. \(2003\)](#page-12-0) model. This approach assumes that during the time "in between" $(3-36 \text{ yr})$, organic matter was in the decomposition stage, i.e., $C_{org} = 1.50^{(-0.112Z')}$; where $Z' = 15.64 - 1.24$ g cm⁻²=14.4 g cm⁻².

Therefore, the calculated decomposition rate constant, λ , of organic matter in the lake sediments during early diagenesis, $(\lambda = 0.11 \text{ cm}^2 \text{ g}^{-1} \times \text{S} = 0.11 \text{ cm}^2$ g^{-1} × 0.43 g cm⁻²) is 0.05 y⁻¹, where S is the average rate of sediment accumulation in Lake Chenghai, the residence time $(\tau = 1/\lambda)$ is 20.6 yr. and the half life $(t_{1/2} = [\ln 2]/\lambda = [\ln 2] \times \tau)$ of C_{org} in the sediments is 14.2 yr.

From modeling results discussed above and the average accumulation rate of sediments at Lake Chenghai, we have calculated the sedimentation fluxes $(F(C_{\text{org}}))$ of organic carbon as $S \times C_{\text{org}}$ since approximately 1970 (above 30 cm, mass depth of 12 g cm^{$^{-2}$}) (Fig. 8). Values of $F(C_{\text{org}})$ range from 60 to 160 g m⁻² y^{-1} , averaging 100 g m⁻² y⁻¹.

5. Summary and conclusions

[Fig. 6](#page-7-0) clearly shows that the profiles of C_{org} and $^{210}Pb_{ex}$ in sediment core CH970608-1 display similar distribution patterns. When the fluxes of $^{210}Pb_{ex}$ $(F({}^{210}Pb_{ex}))$ are calculated and compared with those of organic carbon ($F(\mathrm{C}_{org})$), they significantly correlate (Fig. 8), exhibiting peak values during the periods of 1972–1974 and 1986–1989.

Carbonate contents of Lake Chenghai sediments show periodic fluctuations, which could be a result of climatic temperature changes. A warm climate can create the conditions by which photosynthesis increases with a contemporaneous bloom in phytoplankton, thereby increasing the importance of autochthonous carbonates as an organic carbon sink ([Chen et al.](#page-11-0), 2000). Lake Chenghai, as a closed and eutrophic lake, has undergone major changes in terms of primary productivity and eutrophication in response to external forces during the past several centuries, as determined from sediment pigments ([Wu et al., 2002\)](#page-12-0). As an example, the pigment ratio (Osc/Myx, Oscillaxanthin (Osc) and Myxoxanthophy II (Myx)) peak was present at approximately 11 cm of depth (collected May, 1999, [Wu et al., 2002\)](#page-12-0). Based on variations in inorganic C and O isotopic compositions of sediments in Lake Chenghai, [Wang et al. \(2002\)](#page-12-0) traced climatic changes as well

as photosynthesis variance in lake waters over several decades in this catchment, whereby these parameters changed with a periodicity of 11–12 years.

 $^{210}Pb_{ex}$ derived from atmospheric fallout adsorbs onto particle surfaces and therefore does not find its way into the lattice of minerals (Krishnaswamy et al., 1971). Previous studies showed that 95.5% and 96.4% of all lead species were associated with POC in the sediments of Lakes Erhai and Hongfeng, China, respectively, whereas only 4.5% and 3.6% of total lead was concentrated in carbonates, ([Wan et al., 1993\)](#page-12-0). A primary assumption of $210Pb$ sediment dating is the constant flux of $^{210}Pb_{ex}$, which is derived from the atmosphere and then transferred into sediments in lake water. When the sedimentation rate is relatively constant, the $^{210}Pb_{ex}$ activity in sediments decays at a first order rate after incorporation into the sedimentary record via organic carbon. If the removal flux of particulate organic matter in the lake water increases, $^{210}Pb_{ex}$ can be scavenged from the water proportionately (as long as enough is still available) and then transferred to sediments that are being deposited on the lake bottom. It is reasonable to conclude that the rapid increase in the influx of particulate organic matter is an important factor, facilitating the deposition of $^{210}Pb_{ex}$ by its transfer into settling sediments. While the increase in the flux of particle bound $^{210}Pb_{ex}$ makes it less than ideal for geochronology, due to the violation of the constant flux assumption, the enhancement of the sedimentation influx of $^{210}Pb_{ex}$ is, to some extent, indicative of historical changes in the primary productivity of Lake Chenghai.

Acknowledgements

This work was supported by the National Natural Science Foundation of China (Grant No. 40373038), a project of the National Key Project for Basic Research (2002CB412300), and the U.S. National Science Foundation (Grant No. NSF 82-50). The authors express their gratitude to Dr. Laodong Guo for help. [LW]

References

- Agudo, E.G., 1998. Global distribution of 137Cs inputs for soil erosion and sedimentation studies. Use of $137C$ s in the Study of Soil Erosion and Sedimentation. IAEA-TECDOC-1028, pp. 117 – 121.
- Appleby, P.G., Oldfield, F., 1978. The calculation of ²¹⁰Pb dates assuming a constant rate of supply of unsupported 210Pb to the sediment. Catena $5, 1-8.$
- Bai, Z.G., Wan, X., Wan, G.J., Wang, C., Huang, R., 1997. Geo-
chemical speciation of ⁷Be ¹³⁷Cs ²²⁶Ra and ²²⁸Ra in soils of the

karst region, southwestern China and their erosion trace. Acta Sci. Circumstantiae 17 (4), 407 – 411 (in Chinese).

- Bai, Z.G., Wan, G.J., Huang, R.G., Liu, D.S., 2002. A Comparison on accumulation characteristics of 7 Be and 137 Cs in lake sediments and surface soils in western Yunnan and central Guizhou, China. Catena 49, 253-270.
- Baskaran, M., Santschi, P.H., 2002. Particulate and dissolved ²¹⁰Pb activities in the shelf and slope regions of the Gulf of Mexico waters. Cont. Shelf Res. 22 (10), 1493 – 1510.
- Benoit, G., Hemond, H.F., 1991. Evidence for diffusive redistribution of Pb in lake sediments. Geochim. Cosmochim. Acta 55, 1963 – 1975.
- Bloesch, J., Evans, R.D., 1982. Lead-210 dating of sediments compared with accumulation rates estimated by natural markers and measured with sediment traps. Hydrobiologia 92, 578 – 586.
- Brunskill, G.J., Ludlam, S.D., Peng, T.H., 1984. Mass balance for ¹³⁷Cs in water, varved and non-varved sediments. Chem. Geol. $44, 101 - 117.$
- Chen, J., Wan, G.J., Huang, R., 2000. Recent climatic changes and the chemical records in Chenghai Lake. Mar. Geol. Quat. Geol. 20 (1), 39 – 42 (in Chinese).
- Chen, J., Wan, G.J., Wang, F., Zhang, D.D., Huang, R., Zhang, F., Schmidt, R., 2002. Environmental records of carbon in recent lake sediments. Sci. China, Ser. D: Earth Sci. 45 (10), 875 – 884.
- Davis, R.B., Hess, C.T., Norton, S.A., Hanson, D.W., Hoagland, K.D., Anderson, D.S., 1984. ¹³⁷Cs and ²¹⁰Pb dating of sediment from soft-water lakes in New England (U.S.A.) and Scandinavia, a failure of 137 Cs dating. Chem. Geol. 44, 151-185.
- Dean, W.E., 1999. The carbon cycle and biogeochemical dynamics in lake sediments. J. Paleolimnol. 21 (4), 375 – 393.
- Dominik, J., Mangini, A., Muller, G., 1981. Determination of recent deposition rates in Lake Constance with radioscopic methods. Sedimentology 28, 653-677.
- Edgington, D.N., Robbins, J.A., 1976. Patterns of deposition of natural and fallout radionuclides in sediments of Lake Michigan and their relation to limnological processes. In: Nriagu, J.O. (Ed.), Environmental Biogeochemistry. Ann Arbor, pp. 705 – 729.
- Goldberg, E.D., 1963. Geochronology with ²¹⁰Pb. Symposium on Radioactive Dating, 122–130. International Atomic Energy Agency (IAEA), Vienna, Austria.
- Hodell, D.A., Schelske, C.L., Fahnenstiel, G.L., Robbins, L.L., 1998. Biologically induced calcite and its isotopic composition in Lake Ontario. Limnol. Oceanogr. 43 (2), 187 – 199.
- Imboden, D.M., Stiller, M., 1982. The influence of radon diffusion on the ²¹⁰Pb distribution in sediments. J. Geophys. Res. 87, 557–565.
- Imboden, D.M., Tschopp, J., Stumm, W., 1980. Die Rekonstruktion fruherer Stofffrachten in einem See mittels Sedimentuntersuchungen. Schweiz. Z. Hydrol. 42 (1), 1 – 14.
- Katsuragi, Y., 1983. A study of ⁹⁰Sr fallout in Japan. Papers Meteor. Geophys. 33 (4), 277-291.
- Kelts, K., Hsu, K.J., 1978. Freshwater carbonate sedimentation. In: Lerman, A. (Ed.), Lakes: Chemistry, Geology, Physics. Springer Verlag, Berlin, pp. 295 – 323.
- Koide, M., Soutar, A., Goldberg, E.D., 1972. Marine geochronology with 210Pb. Earth Planet. Sci. Lett. 14, 442 – 446.
- Krishnaswamy, S., Lal, D., Martin, J.M., Meybeck, M., 1971. Geochronology of lake sediments. Earth Planet. Sci. Lett. 11, $407 - 414$
- Lee, H.N., Wan, G.J., Zheng, X., Sanderson, C.G., Josse, B., Wang, S., Yang, W., Tang, J., Wang, C., 2004. Measurements of Pb-210

and Be-7 in China and their analysis accompanied with global model calculations of Pb-210. J. Geophys. Res. –Atmos. 109 (D22203). [doi:10.1029/2004JD005061.](doi:10.1029/2004JD005061)

- McHenry, J.R., Ritchie, J.C., Gill, A.C., 1973. Accumulation of fallout cesium-137 in soils and sediments in selected watersheds. Water Resour. Res. 9, 676-686.
- OECD, 1982. Eutrophication of Waters, Monitoring, Assessment and Control. Organization for Economic Cooperation and Development, Paris.
- Paulsen, S.C., List, E.J., Santschi, P.H., 1999. Modeling variability in 210Pb and sediment fluxes near the Whites Point Outfalls, Palos Verdes Shelf, California. Environ. Sci. Technol. 33, 3077 – 3085.
- Playford, K., Lewis, G.N.J., Carpenter, R.C., 1990. Radioactive fallout in air and rain: results to the end of 1989. Report AEA-EE-0227, DOE/HMIP/PR/91/042, Harwell Laboratories, UK, 21.
- Ritchie, J.C., McHenry, J.R., 1990. Application of radioactive fallout cesium-137 for measuring soil erosion and sediment accumulation rates and patterns: a review. J. Environ. Qual. 19, 215-233.
- Ritchie, J.C., Clebsch, E.E.C., Rudolph, W.K., 1970. Distribution of fallout and natural gamma radionuclides in litter, humus, and surface mineral soils under natural vegetation in the Great Smoky Mountains, north Carolina-Tennessee. Health Phys. 18, 479 – 491.
- Robbins, J.A., 1978. Geochemical and geophysical applications of radioactive lead. In: Nriagu, J.O. (Ed.), The Biogeochemistry of Lead in the Environment. Elsevier/North-Holland Biomedical Press, Amsterdam, pp. 285–383.
- Robbins, L.L., Blackwelder, P.L., 1992. Biochemical and ultrastructural evidence for the origin of whitings: a biologically induced calcium precipitation mechanism. Geology 20, 464 – 468.
- Robbins, J.A., Edgington, D.N., 1975. Determination of recent sedimentation rates in Lake Michigan using Pb-210 and Cs-137. Geochim. Cosmochim. Acta 39, 285 – 304.
- Robbins, J.A., Herche, L.R., 1992. Models and uncertainty in ²¹⁰Pb dating of sediments. International Association of Theoretical and Applied Limnology, Proceedings, Congress in Barcelona, 25(1): 217–222. Verh. Internat. Verein. Limnol. Stuttgart 1993, E. Schweizerbart'sche Verlagsbuchhandlung.
- Santschi, P.H., Nyffeler, U.P., Anderson, R.F., Schiff, S.L., O'Hara, P., 1986. Response of radioactive trace metals to acid–base titration in controlled experimental ecosystems: evaluation of transport parameters for application to whole-lake radiotracer experiments. Can. J. Fish. Aquat. Sci. 43, 60-77.
- Santschi, P.H., Bollhalder, S., Farrenkothen, K., Lueck, A., Zingg, S., Sturm, M., 1988. Chernobyl radionuclides in the environment: tracers for the tight coupling between atmospheric, terrestrial, and aquatic geochemical processes. Environ. Sci. Technol. 22 (5), 510 – 516.
- Santschi, P.H., Hoehener, P., Benoit, G., Brink, M.B., 1990. Chemical processes at the sediment–water interface. Mar. Chem. 30, $269 - 315$.
- Schuler, C.H., Wieland, E., Santschi, P.H., Sturm, M., Lück, A., Farrenkothen, K., Bollhalder, S., Beer, J., Bonani, G., Hofmann, H.J., Suter, M., Wölfli, W., 1991. A multi-tracer study of radionuclides in Lake Zurich, Switzerland 1. Comparison of atmospheric and sedimentary fluxes of ⁷Be, ¹⁰Be, ²¹⁰Pb, ²¹⁰Po and ¹³⁷Cs. J. Geophys. Res. 96 (C9), 17051–17065.
- Stumm, W., Morgan, J.J., 1996. Aquatic Chemistry, Chemical Equilibria and Rates in Natural Waters. A Wiley-Interscience Publication. John Wiley & Sons, New York, p. 1022.
- Talbot, M.R., Livingstone, D.A., 1989. Hydrogen index and carbon isotopes of lacustrine organic matter as lake level indicators. Palaeogeogr. Palaeoclimatol. Palaeoecol. 70, 121-137.
- Tao, W., Xia, F., Jing, C., 1999. On environmental issues of Lake Chenghai and its management strategy. Resources and Environment in the Yangtze Basin 8 (2), 210-214 (in Chinese).
- Wan, G.J., 1999. ¹³⁷Cs dating by annual distinguish for recent sedimentation: samples from Erhai Lake and Hongfeng Lake. Quat. Sci. 19 (1), 73-80 (in Chinese).
- Wan, G.J., Santschi, P.H., Farrenkothen, K., Sturm, M., Stumm, W., 1985. The distribution and dating of 137 Cs for recent sediments in Lake Greifen, Switzerland. Acta Sci. 3 (3), 361-365 (in Chinese).
- Wan, G.J., Santschi, P.H., Sturm, M., Farrenkothen, K., Lueck, A., Werth, W., Schuler, C., 1987. Natural $(^{210}Pb, ^{7}Be)$ and fallout $({}^{137}Cs, {}^{239,240}Pu, {}^{90}Sr)$ radionuclides as geochemical tracers of sedimentation in Greifensee, Switzerland. Chem. Geol. 63, 181-196.
- Wan, G.J., Huang, R., Wang, C., Rong, J., 1990. A variance of vertical profile for ${}^{210}Po_{ex}$ at the top of sediment in Hongfeng Lake. Chin. Sci. Bull. 35 (22), 1910-1914.
- Wan, G.J., Lin, W., Huang, R., Chen, Z., 1991. Dating characteristics and erosion traces of 137 Cs vertical profiles in Lake Hongfeng sediments. Chin. Sci. Bull. 36 (8), 674-677.
- Wan, G.J., Liu, J., Li, B., 1993. The isotopic character and the remobilization of lead at the top of sediment in Erhai. Chin. Sci. Bull. 38 (2), 139-142.
- Wan, G.J., Bai, Z., Liu, T., Wang, S., Chen, J., Huang, R., 2001. The differentials of fallout 137Cs between western Yunnan and central Guizhou: Implication for the barrier effect of Qinghai-Xizang uplift on global atmospheric pollutants in Yunnan-Guizhou plateau. Quat. Sci. 21 (5), 407 – 415 (in Chinese).
- Wan, G.J., Bai, Z., Qing, H.R., Mather, J.D., Huang, R.G., Wang, H.R., Tang, D.G., Xia, B.H., 2003. Geochemical records in recent sediments of Lake Erhai: implications for environmental changes in a low latitude-high altitude lake in southwest China. J. Asian Earth Sci. 21 (5), 489-502.
- Wang, Y., 1993. Lacustrine carbonate chemical sedimentation and climatic–environmental evolution: a case study of Qinghai Lake and Daihai Lake. Oceanologia et Limnologia Sinica 24 (1) , $31 - 36$ (in Chinese).
- Wang, Y., Hu, W., Zhang, X., 1989. Carbonate sediments in lakes of Yunnan, China. Oceanologia et Limnologia Sinica 20 (2), $122 - 130$ (in Chinese).
- Wang, H., Wan, G.J., Chen, Y., 1996. Determination of organic carbon in lacustrine sediment with a PE2400 Element Analyzer. Chongqing Environ. Sci. 18 (5), 36-38 (in Chinese).
- Wang, F., Wan, G.J., Liu, C., Xu, S., 2002. The correlation of inorganic C, O isotopic values for Lake Chenghai sediments and its environmental implications. Chin. J. Geochem. 21 (2), 186 – 192 (in Chinese).
- Wieland, E., Santschi, P.H., Beer, J., 1991. A multi-tracer study of radionuclides in Lake Zurich, Switzerland: 2. Residence times, removal processes, and sediment focusing. J. Geophys. Res. 96 (C9), 17067 – 17080.
- Wieland, E., Lienemann, P., Bollhalder, S., Lück, A., Santschi, P.H., 2001. Composition and transport of settling particles in Lake Zurich: importance of lateral pathways. Aquat. Sci. 63 (2), $123 - 149.$
- Wigley, T.M., Plummer, L.N., 1976. Mixing of carbonate waters. Geochim. Cosmochim. Acta 40, 989 – 995.
- Wu, J., Jiang, X., Xia, W., Pan, H., Yin, Y., 2002. Climate and primary productivity over the last 500 years at Chenghai Lake, Yunnan. Mar. Geol. Quat. Geol. 22 (2), 95 – 98 (in Chinese).
- Xu, S., Wan, G.J., 2001. Vertical distribution of 137Cs and 210Pb and their dating in modern sediments of Chenghai Lake, Yunnan province. Geol.-Geochem. 29 (1), 28-31 (in Chinese).
- Xu, J., Wan, G.J., Wang, C., Huang, R., Chen, J., 1999. Vertical distribution of ²¹⁰Pb and ¹³⁷Cs and their dating in recent sediments of Lugu Lake and Erhai Lake, Yunnan province. J. Lake Sci. 11 (2), 110-116 (in Chinese).
- Yeager, K.M., Santschi, P.H., 2003. Invariance of isotope ratios of lithogenic radionuclides: more evidence for their use as sediment source tracers. J. Environ. Radioact. 69 (3), 159-176.
- Yuan, Z.Q., Wu, D.S., Huang, R.G., Chen, Z.L., Wu, F.C., Wan, G.J., 1993. A sampling device for collecting samples near the lacustrine sediment–water interface. Environ. Sci. 14 (1), 70-73 (in Chinese).
- Yunnan Provincial Dept. of Water Conservancy and Hydropower Electricity and Yunnan Provincial Hydrological Station,, 1984. Earth's Surface Water Resources in Yunnan Province, pp. 15 – 16 (in Chinese).