Available online at www.sciencedirect.com
ScienceDirect
Environmental Pollution 152 (2008) 314–321
ENVIRONMENTAL POLLUTION
www.elsevier.com/locate/envpol

Global fallout Pu recorded in lacustrine sediments in Lake Hongfeng, SW China

Jian Zheng [a,*,1], Fengchang Wu [b,**,1], Masatoshi Yamada [a], Haiqing Liao [b], Congqiang Liu [b], Guojiang Wan [b]

[a] *Nakaminato Laboratory for Marine Radioecology, Environmental Radiation Effects Research Group, National Institute of Radiological Sciences, 3609 Isozaki-cho, Hitachinaka, Ibaraki 311-1202, Japan*

[b] *State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guanshui Road 46, Guiyang 550002, China*

Received 9 November 2006; received in revised form 4 June 2007; accepted 11 June 2007

Fallout Pu recorded in sediments can be used for quick dating of recent sediments to understand the pollution history of environmental contaminants.

Abstract

Studies on the distribution and isotope compositions of fallout Pu are important for source characterization of possible future non-fallout Pu contamination in aquatic environments, and useful for dating of recent sediments to understand the pollution history of environmental contaminants. We present the historical record of atmospheric Pu fallout reconstructed from a sediment core from Lake Hongfeng, China. The Pu activity profile was in agreement with the ^{137}Cs profile. Inventories were 50.7 Bq m^{-2} for $^{239+240}$Pu and 1586 Bq m^{-2} for ^{137}Cs. The average ^{240}Pu/^{239}Pu atom ratio was 0.185 ± 0.009, indicating that Pu originated from global stratospheric fallout rather than from direct tropospheric or close-in fallout from the Chinese nuclear testing conducted in the 1970s. Our data suggested that Lake Hongfeng would be an ideal setting for monitoring atmospheric fallout and environmental changes in this region.

Keywords: Pu isotope pollution; SF-ICP-MS; Lake Hongfeng; Lacustrine sediments; Global fallout

1. Introduction

Anthropogenic radionuclides, plutonium (^{239}Pu and ^{240}Pu) and ^{137}Cs, have been released to the environment mainly from nuclear weapons testing. Approximately 6 t of ^{239}Pu were released into the environment as a result of 541 atmospheric weapon tests (UNSCEAR, 2000). The history of atmospheric nuclear weapons' testing, and hence fallout Pu deposition, can be broadly divided into two phases: the pre- and post-moratorium weapons testing years (Buesseler, 1997). During the pre-moratorium years of 1952–1958, the high-yield thermonuclear tests conducted by the United States dominated the atmospheric pollution. Between November 1958 and September 1961, the testing moratorium between the USA and the former Soviet Union temporarily ended all new fallout inputs. The peak of global fallout deposition was in 1963 due to a series of large-scale atmospheric nuclear tests conducted by the former Soviet Union in 1961–1962. In 1963, the Limited Test Ban Treaty between the USA and the former Soviet Union put an end to the major atmospheric nuclear testing programs; since then, smaller amounts of nuclear debris have been introduced into the atmosphere by French and Chinese activities. Plutonium fallout from the 1986 Chernobyl accident has also been observed in many parts of Europe and

* Corresponding author. Tel.: +81 29 265 7130; fax: +81 29 265 9883.
** Corresponding author.
E-mail addresses: jzheng@nirs.go.jp (J. Zheng), wufengchang@vip.skleg.cn (F. Wu).
[1] These authors contributed equally to this work.

0269-7491/$ - see front matter © 2007 Elsevier Ltd. All rights reserved.
doi:10.1016/j.envpol.2007.06.027

Siberian Altai (Ketterer et al., 2004; Boulyga et al., 2003; Olivier et al., 2004). In addition, any nuclear test that is conducted at or just above the ground (i.e., surface based) will generate a substantial fraction of tropospheric (or close-in) fallout, which is deposited rapidly within the vicinity of the nuclear testing site.

Establishing accurate historical records of the distribution, inventory and source of Pu in the environment are important for environmental monitoring and radiological health protection due to its high toxicity. Existing records of fallout Pu have been derived from analyses of a variety of environmental samples, such as soils, marine and lacustrine sediments, dated corals, polar ice cores and samples from a UK herbage archive (Olivier et al., 2004). Depending on the emission source, Pu isotope ratios, in particular $^{240}Pu/^{239}Pu$ atom ratios, vary significantly in the environment, which allows weapons-grade Pu (ratios of 0.01–0.05) to be distinguished from reprocessing Pu (ratios of 0.2–0.8) and global fallout Pu (ratios of 0.17–0.19), and the transport of Pu from different sources to be studied (Borretzen et al., 2005). Plutonium has also been used for actinide transport mechanisms, air circulation and mixing-time (Kudo et al., 2000), and desertification studies (Hirose et al., 2003).

Accurate dating of lake sediments is important for many studies, such as reconstructing the pollution history of organic pollutants, and determining the historical variation of lake biological productivity and/or carbon preservation. The most commonly used radionuclides for radiometric sediment dating are ^{210}Pb and ^{137}Cs. However, ^{210}Pb sediment dating relies on the assumptions that the flux of $^{210}Pb_{ex}$ derived from atmospheric fallout and the sedimentation rate are relatively constant, which may not always be met in practical applications. Regarding ^{137}Cs dating, as indicated by Ketterer et al. (2004), one consideration is its relatively short half-life of 30.2 years; more than 60% of the original inventory of global (stratospheric) fallout ^{137}Cs has subsequently decayed, and at some point in the future it will be impractical to routinely measure fallout ^{137}Cs in environmental samples. Recently, the potential for using Pu isotopes in dating of recent aquatic sediments (<100 years age) has been considered (Ketterer et al., 2002). If ^{137}Cs and Pu activities convey essentially the same information about sedimentation processes, and combined with the specific $^{240}Pu/^{239}Pu$ isotopic fingerprint of different emission sources, it is possible to establish a reliable and long-term sediment chronology. Taking into account the long half-lives of Pu isotopes (^{239}Pu, $t_{1/2} = 2.44 \times 10^4$ yr; ^{240}Pu, $t_{1/2} = 6.58 \times 10^3$ yr), this dating technique could become a significant tool for future researchers to study the history of anthropogenic pollutants in the environment.

Little is known about the distribution, inventory and source of Pu in the environment in China. Lin et al. (1992) investigated the distribution of $^{239+240}Pu$ activity in sediments collected from Lake Bosten, Xinjiang, China. Huh et al. (1996) observed the Pu fallout record in a subalpine lake, Lake Sun Moon. However, no information about the atom ratio of $^{240}Pu/^{239}Pu$, which is important for the identification of Pu sources in the environment, was reported in these two studies. In order to establish a reliable chronology for recent sediment dating, it is necessary to obtain more information on the distribution and history of deposition of Pu isotopes in lacustrine sediments.

In this work we present the history of atmospheric Pu fallout reconstructed using a sediment core from Lake Hongfeng in the central Guizhou Plateau of China. As Lake Hongfeng is an artificial reservoir with a known starting time of sediment accumulation (filling of the lake was completed in 1960), it provides a unique opportunity to study the sedimentation process using Pu isotopes. Pu isotopes in the sediments were analyzed with a newly developed analytical method using a sector-field ICP-MS with a high-efficiency sample introduction system (Zheng and Yamada, 2006). We focus on the source identification of Pu, the history of atmospheric Pu fallout during the past four decades in the studied area and estimation of the sedimentation rate using Pu isotopes.

2. Materials and methods

2.1. Sample collection

Lake Hongfeng (Red Maple Lake) is located ca. 30 km west of Guiyang City, Guizhou, China (26°25′–26°34′ N, 106°20′–106°26′ E). It is the largest artificial reservoir at the center of the Guizhou Plateau. Filling of lake was completed in 1960. Lake Hongfeng has a surface area of 32 km^2, a volume of 2.97×10^8 m^3, a drainage basin of 1551 km^2, a mean depth of 9.3 m, a maximum depth of about 50 m, water flux of 11.26×10^8 m^3 yr^{-1}, and a residence time of the water of 0.26 yr.

Lake Hongfeng consists mainly of two areas: the North Lake and the South Lake (Fig. 1). A sediment core was collected from the South Lake in July 2004 at a water depth of approximately 8 m using a sediment–water interface sampler designed by Yuan et al. (1993). The cored sediments were undisturbed, as indicated by the clear water–sediment interface and the preservation of fine sediment laminations. The sediment core (designated HF20040709S), with a length of 37 cm, was sectioned at 1-cm intervals *in situ*. The 1-cm long sediment samples were weighed immediately after collection, dried using a vacuum freeze dryer (Techcorp FD-3-85-MP-79-36 mT), and then re-weighed to determine mass depths and porosity. Dried samples were then ground to <0.15 mm in diameter for Pu isotope analysis.

2.2. Analytical procedure

Details of sample preparation for Pu isotope analysis have been described elsewhere (Zheng et al., 2004). In brief, the sediment samples were dried at 110 °C for at least 4 h. An aliquot of approximately 2–5 g was weighed out, and spiked with ^{242}Pu (2 pg) as yield monitor. The extraction of Pu from the sediment samples was done using 8 M HNO_3 while heating on a hot plate (180–200 °C) for at least 3 h. The separation of Pu and U, and the further purification of Pu were done by anion-exchange chromatography (AG 1-X8). The chromatographic purification was repeated twice. The chemical yield in the employed sample preparation procedure was estimated to be in the range of 59–78% with a mean of 66 ± 7%.

To determine Pu isotopes in freshwater lake sediments, a sector-field ICP-MS (Finnigan Element 2, Bremen, Germany) was used in the low resolution (LR) mode in order to utilize the maximal instrument sensitivity. An APEX-Q high-efficiency sample introduction system (Elemental Scientific Inc., Omaha, NE, USA) with membrane desolvation unit (ACM) and a conical concentric nebulizer were used as sample introduction systems. Additionally, the normal skimmer cone was replaced by a high-efficiency cone (X-cone, Thermo Finnigan) to further increase the sensitivity of SF-ICP-MS. All the measurements

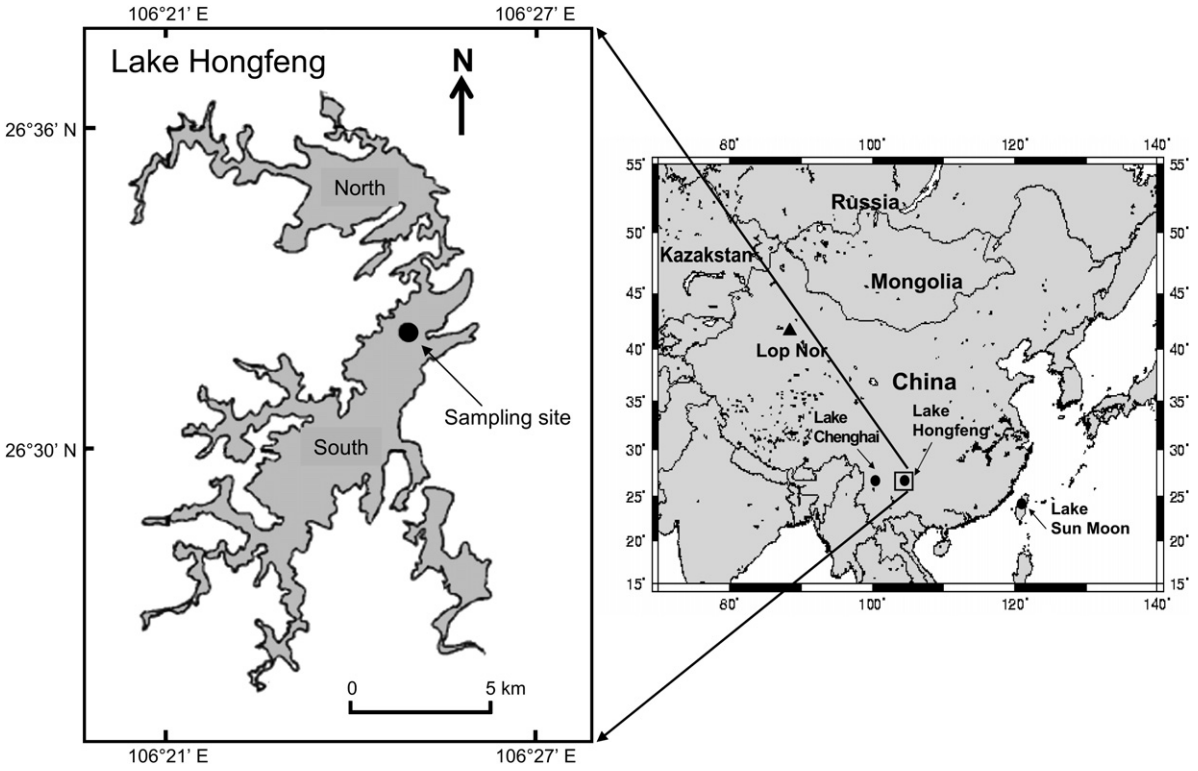

Fig. 1. Map showing the location of Lake Hongfeng and the sampling site, along with the location of two freshwater lakes, Chenghai and Sun Moon in the latitudinal band of 20–30° N. The Chinese nuclear test site at Lop Nor is also shown.

were made in the self-aspirating mode to reduce the risk of contamination by the peristaltic pump tubing. The SF-ICP-MS was optimized daily using 0.1 ng ml^{-1} U standard solution. Details on the instrument optimization and determination of Pu isotopes have been described previously (Zheng and Yamada, 2006).

^{137}Cs activity was determined using gamma-spectrometry on a Canberra S-100 multi-channel spectrometer with a GC5019 H-P® Ge coaxial detector (efficiency 50%) at the Institute of Geochemistry, Chinese Academy of Sciences. The peak of ^{137}Cs used to determine its activity was 661.6 KeV. Liquid standard (Catalog No.: 7137) supplied by the Institute of Atomic Energy, Chinese Academy of Sciences was used.

3. Results and discussion

3.1. Pu activity profile and inventory

The downcore distributions of porosity, Pu activities, ^{240}Pu/^{239}Pu atom ratios and ^{137}Cs activities obtained in the sediment core HF20040709S are summarized in Table 1. Porosity, calculated by assuming a sediment dry density of 2.45 g cm^{-3}, decreased with depth from ca. 0.96 in the surface layer to 0.53 in the bottom layer at 36–37 cm. The total $^{239+240}$Pu activities ranged from 0.058 to 2.782 mBq/g. Starting from the surface layer down to 20 cm, the total $^{239+240}$Pu activities were quite low, less than 0.1 mBq/g, compared to the average total $^{239+240}$Pu activity in soils in southern China of 0.1–0.2 mBq/g (25–45° N) (Zhu et al., 2002). The low $^{239+240}$Pu activities in the top 20 cm could be explained as follows. There is constant input of coal fly ash material coming from a coal-fire power plant located on the west bank of Lake Hongfeng, which contains an extremely low activity of Pu, and there is input of soils into the lake due to deposition, resuspension, and erosion processes. As shown in Fig. 2, the top 20 cm sediments represented the deposition since 1980 (the sediment dating is discussed in a later section in detail). Chinese and French atmospheric thermonuclear tests were carried out between 1964 and 1980. Since the 1980s, the direct atmospheric deposition of $^{239+240}$Pu has shown a marked decrease (Hirose et al., 2003). Therefore, the dilution of $^{239+240}$Pu activity in the soils by the low Pu-bearing fly ash material from the power plant has resulted in the low $^{239+240}$Pu activities in the top 20 cm sediments.

As shown in Fig. 3, the vertical distributions of $^{239+240}$Pu and ^{137}Cs activities were characterized by a pronounced peak at the 32–35 cm interval. Lake Hongfeng has a known starting time of sediment accumulation since 1960; therefore, this peak was related to the historical fallout maximum in 1963–1964. Isotopic evidence for this speculation is discussed in the next section.

Because of the variable input with time, we calculated the inventories of $^{239+240}$Pu and ^{137}Cs (decay corrected to the sampling time, 2004 July) by adding up their activity downcore. The obtained values were 50.7 Bq m^{-2} for $^{239+240}$Pu and 1586 Bq m^{-2} for ^{137}Cs, respectively. In Fig. 4, the integrated Lake Hongfeng core inventories of $^{239+240}$Pu and ^{137}Cs are compared to values obtained from other freshwater lakes in

Table 1
Pu data in sediment core HF20040709S of Lake Hongfeng, China

Depth (cm)	Mass depth (g cm^{-2})	Porosity	A.D.	^{240}Pu/^{239}Pu	$^{239+240}$Pu activity (mBq/g)	^{137}Cs activity[a] (mBq/g)
0–1	0.095	0.956	2004.6	0.193 ± 0.021	0.058 ± 0.003	–
1–2	0.254	0.950	2003.9	0.183 ± 0.021	0.063 ± 0.004	–
2–3	0.371	0.952	2003.4	0.192 ± 0.017	0.060 ± 0.003	–
3–4	0.473	0.954	2002.9	0.170 ± 0.022	0.058 ± 0.005	–
4–5	0.560	0.956	2002.5	0.213 ± 0.023	0.070 ± 0.005	–
5–6	0.674	0.952	2002.0	0.191 ± 0.018	0.068 ± 0.004	–
6–7	0.806	0.944	2001.4	0.186 ± 0.021	0.080 ± 0.005	6.8 ± 2.7
7–8	0.946	0.943	2000.8	0.171 ± 0.030	0.069 ± 0.006	–
8–9	1.079	0.941	2000.4	0.162 ± 0.027	0.071 ± 0.006	–
9–10	1.192	0.947	1999.9	0.191 ± 0.018	0.079 ± 0.008	–
10–11	1.317	0.949	1999.3	0.189 ± 0.023	0.056 ± 0.003	–
11–12	1.453	0.938	1998.7	0.182 ± 0.015	0.076 ± 0.003	–
12–13	1.612	0.928	1997.9	0.191 ± 0.015	0.079 ± 0.004	–
13–14	1.779	0.921	1997.1	0.207 ± 0.015	0.080 ± 0.004	–
14–15	2.048	0.888	1995.9	0.183 ± 0.010	0.087 ± 0.003	4.3 ± 1.3
15–16	2.347	0.870	1994.5	0.182 ± 0.019	0.093 ± 0.005	6.7 ± 1.4
16–17	2.646	0.861	1993.1	0.188 ± 0.009	0.093 ± 0.003	6.2 ± 1.8
17–18	2.971	0.863	1991.6	0.195 ± 0.010	0.092 ± 0.003	4.0 ± 1.2
18–19	3.297	0.865	1990.1	0.183 ± 0.016	0.085 ± 0.004	5.4 ± 0.8
19–20	3.603	0.858	1988.7	0.187 ± 0.009	0.107 ± 0.003	4.8 ± 1.1
20–21	3.944	0.850	1987.1	0.186 ± 0.016	0.130 ± 0.007	6.7 ± 1.5
21–22	4.296	0.842	1985.5	0.182 ± 0.018	0.143 ± 0.008	4.6 ± 1.1
22–23	4.728	0.837	1983.5	0.187 ± 0.013	0.150 ± 0.005	5.6 ± 1.1
23–24	5.083	0.850	1981.9	0.182 ± 0.010	0.180 ± 0.005	8.1 ± 1.2
24–25	5.409	0.856	1980.4	0.186 ± 0.019	0.200 ± 0.015	7.1 ± 1.5
25–26	5.749	0.851	1978.8	0.184 ± 0.015	0.240 ± 0.012	12.4 ± 1.3
26–27	6.128	0.835	1977.1	0.181 ± 0.008	0.308 ± 0.008	10.6 ± 1.5
27–28	6.510	0.834	1975.3	0.177 ± 0.009	0.366 ± 0.012	17.1 ± 1.6
28–29	6.877	0.839	1973.6	0.172 ± 0.013	0.492 ± 0.037	17.1 ± 1.7
29–30	7.286	0.829	1971.7	0.186 ± 0.009	0.677 ± 0.029	29.3 ± 2.5
30–31	7.679	0.833	1969.8	0.184 ± 0.007	0.806 ± 0.026	29.8 ± 3.2
31–32	8.055	0.825	1968.1	0.189 ± 0.010	1.206 ± 0.075	45.6 ± 2.8
32–33	8.471	0.823	1966.2	0.194 ± 0.007	2.002 ± 0.062	44.5 ± 3.2
33–34	8.815	0.833	1964.1	0.193 ± 0.006	2.782 ± 0.108	54.0 ± 3.2
34–35	9.270	0.797	1962.8[b]	0.174 ± 0.009	1.037 ± 0.048	39.4 ± 2.8
35–36	10.15	0.630	1961.5[b]	0.176 ± 0.012	0.464 ± 0.019	15.5 ± 1.5
36–37	11.36	0.527	1960.3[b]	0.182 ± 0.014	0.211 ± 0.009	5.1 ± 1.2
Mean				0.185 ± 0.009		

[a] Decay corrected to sampling time (July 9, 2004).
[b] dating according to mean sedimentation rate.

China located in the same latitudinal band (20–30° N). For $^{239+240}$Pu inventory, Huh et al. (1996) reported a value of 35.9 Bq m^{-2} in Lake Sun Moon (23°52′ N, 120°55′ E); a similar value of 35.4 Bq m^{-2} was observed in Lake Chenghai (26°27′–26°38′ N, 100°38′–100°41′ E) (Zheng et al., in press). These values were in good agreement with the integrated atmospheric fallout of 36 Bq m^{-2} for 20–30° N published by UNSCEAR (2000). The Pu inventory observed in Lake Hongfeng core had a higher value of 50.7 Bq m^{-2}, which accounted for ca. 140% of the integrated atmospheric fallout value. Similarly, a higher ^{137}Cs inventory was found in Lake Hongfeng (Fig. 4). This inventory enrichment could be attributed to: (1) a sediment focusing effect due to the lake-wide redistributions of radionuclides (Ketterer et al., 2002; Wan et al., 2005); and (2) intensive soil erosion in the lake's watershed. Therefore, Lake Hongfeng received fallout not only from the direct atmospheric deposition but also from previously deposited material, for instance, soils transported to the lake via soil erosion.

3.2. ^{240}Pu/^{239}Pu atom ratio and ^{137}Cs/$^{239+240}$Pu activity ratio: sources of Pu in Lake Hongfeng sediments

The high inventories of ^{137}Cs and $^{239+240}$Pu in Lake Hongfeng core also raised a question, whether there are other sources besides the global fallout. Two possible emission sources for Pu in the environment since the 1970s were Chinese nuclear tests and the Chernobyl accident. Chinese thermonuclear bomb tests were conducted eight times, namely, on 6th (June 1967), 8th (December 1968), 10th (September 1969), 11th (October 1970), 15th (June 1973), 16th (June 1974), 21st (November 1976), and 26th (October 1980) (Yamada et al., 2005). As stated in the introduction, depending on the emission source, the atom ratio of ^{240}Pu/^{239}Pu varies significantly in the environment, which allows the use of ^{240}Pu/^{239}Pu ratio as a geochemical tracer to identify the source of Pu and study the transport of Pu from different sources in the environment (Buesseler and Sholkovitz, 1987; Zheng and Yamada,

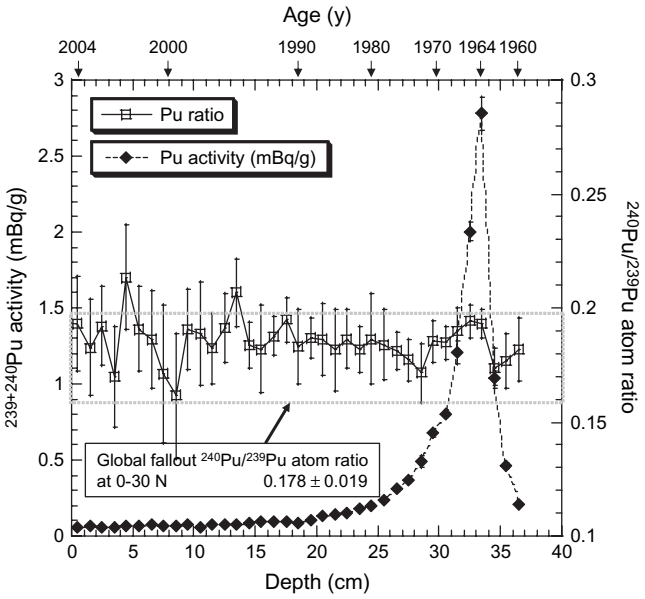

Fig. 2. Vertical profiles of $^{239+240}$Pu activity and ^{240}Pu/^{239}Pu atom ratio in the core (HF20040709S) collected in Lake Hongfeng. The dashed horizontals show the range of global fallout ^{240}Pu/^{239}Pu atom ratios reported by Kelley et al. (1999).

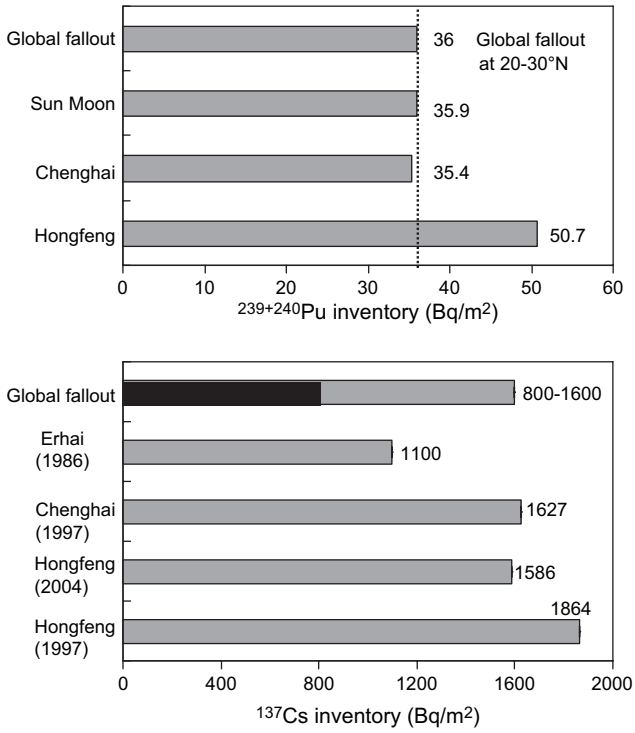

Fig. 4. Comparison of $^{239+240}$Pu and ^{137}Cs inventories obtained in Lake Hongfeng and in Lakes Chenghai, Erhai and Sun Moon. Inventory data for Lakes Sun Moon, Erhai and Chenghai were adopted from Huh et al. (1996), Wan et al. (2005) and Zheng et al. (in press), respectively.

2004; Oktay et al., 2000). Krey et al. (1976) reported the mass ratio of ^{240}Pu/^{239}Pu in global fallout to be 0.18 ± 0.02, based on a worldwide program of sampling conducted at 21 sites in 1970 and 1971 between 30° N and 60° N. More recently, Kelley et al. (1999) provided regional baseline data on the ^{240}Pu/^{239}Pu atom ratio from analysis of Pu isotopes in archived soil samples collected in the 1970s from 54 locations around the world. The regional average composition of global fallout Pu in the 0–30° N latitudinal band was 0.178 ± 0.019 (atom ratio $\pm 2\sigma$ error). Plutonium resulting from the Chernobyl accident has been characterized with a high ^{240}Pu/^{239}Pu atom ratio of 0.408 (Muramatsu et al., 2000). The characteristic ^{240}Pu/^{239}Pu atom ratio for the Chinese thermonuclear bomb tests remains unknown so far; however, Warneke et al. (2002) suggested a value higher than that of global fallout for the Chinese nuclear tests.

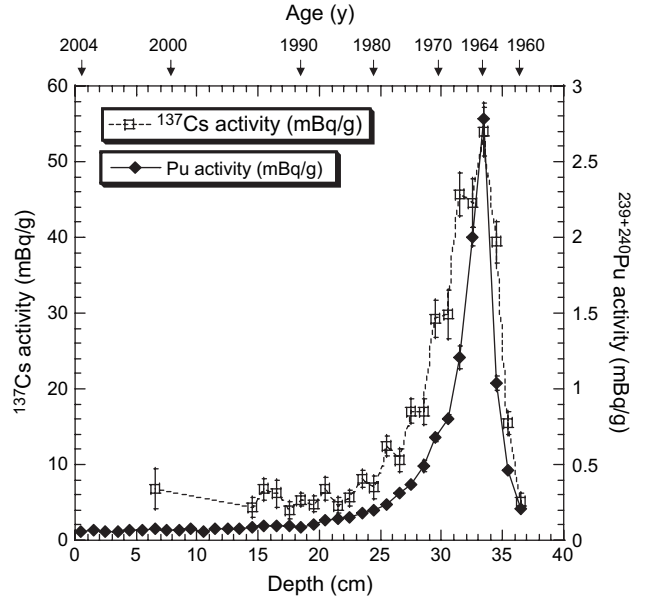

Fig. 3. Vertical profiles of $^{239+240}$Pu and ^{137}Cs activities in core HF20040709S.

The ^{240}Pu/^{239}Pu atom ratios in Lake Hongfeng core ranged from 0.162 to 0.213. As shown in Fig. 2, the vertical distribution of ^{240}Pu/^{239}Pu atom ratios did not have a significant variation, they fell well within the global fallout value range. The mean of the ^{240}Pu/^{239}Pu atom ratio in HF20040709S core was 0.185 ± 0.009, clearly indicating the source of Pu as global fallout.

The activity ratio of ^{137}Cs/$^{239+240}$Pu has been another indicator of radionuclides originating from global fallout in the environment. Hodge et al. (1996) reported that soils that contained primarily global fallout radionuclides exhibited a mean activity ratio of ^{137}Cs/$^{239+240}$Pu of 34 ± 4 (factoring in the radioactive decay of ^{137}Cs as of 1 July 1998) based on studies in 155 sites throughout the Northern Hemisphere. Therefore, by measuring the ^{137}Cs and $^{239+240}$Pu activities in soil, it can be determined whether there is a second source for Pu in that sample. Cizdziel et al. (1999) have reported Pu anomalies in attic dust and soil collected at locations surrounding the Nevada Test Site based on this ratio. We calculated the ^{137}Cs/$^{239+240}$Pu activity ratio in HF20040709S core based on the inventories of ^{137}Cs and $^{239+240}$Pu shown in Fig. 4. By using the ^{137}Cs inventory corrected for radioactive decay

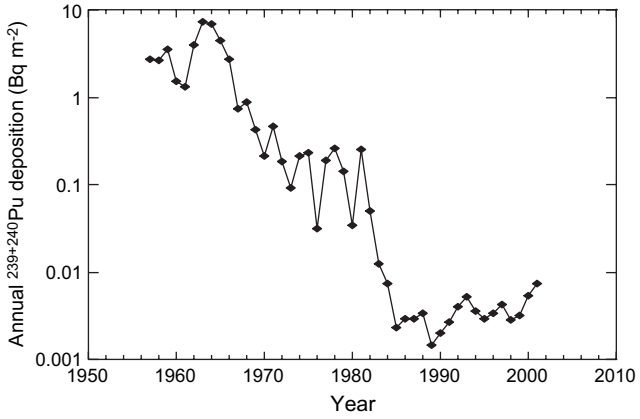

Fig. 5. Annual $^{239+240}$Pu deposition observed at Tsukuba, Japan during 1957–2001 (adopted from Hirose et al., 2003).

to July 2004 and June 1997, respectively, we found the ^{137}Cs/$^{239+240}$Pu ratios were 31.3 and 36.7. This result provided further evidence for the identification of global fallout Pu in Lake Hongfeng core.

Hirose et al. (2003) investigated the annual $^{239+240}$Pu deposition in Tsukuba, Japan, during the period from 1957 to 2001 (Fig. 5). They found that temporal variations of the $^{239+240}$Pu deposition in the years from 1957 to 1980 were controlled by the stratospheric fallout due to the atmospheric nuclear weapon testing, and after the 26th Chinese nuclear test in October 1980, a marked increase of $^{239+240}$Pu deposition occurred in the spring of 1981. However, as can be seen in Fig. 2, for $^{239+240}$Pu activity, no peak could be observed in the top 29 cm, corresponding to the deposition time of 1970 to the present. In addition, as discussed above, both the atom ratio of ^{240}Pu/^{239}Pu and the activity ratio of ^{137}Cs/$^{239+240}$Pu indicated that global fallout was the source of radionuclides present in this core. Therefore, we could conclude that there was no observable contribution of Pu from Chinese nuclear tests through tropospheric or close-in fallout or from the Chernobyl accident in April 1986.

3.3. Sedimentation rate: estimation from Pu isotopes

Investigations since the 1960s have shown that the fallout radionuclide, ^{137}Cs, is an appropriate tracer in the study of sedimentation in lacustrine environments (Krishnaswamy et al., 1971; Wan et al., 2005). However, as discussed in the introduction, due to its short half-life (ca. 30 years), the determination of ^{137}Cs activity will be difficult in the future. Therefore, interest in using Pu as an alternative tracer for the study of sedimentation in lacustrine environments has increased considerably (Ketterer et al., 2002, 2004). A major concern for the application of Pu isotopes for sedimentation studies is whether the ^{137}Cs and Pu isotopes convey essentially the same information about sedimentation processes and chronology because of their different chemical properties. As can be seen in Fig. 3, the ^{137}Cs and $^{239+240}$Pu activity profiles were very similar in the Lake Hongfeng core. Each nuclide exhibited a maximum activity in the 32–35 cm horizon, which could be interpreted as spanning the 1963–1965 time frame. We further corrected the radioactive decay of ^{137}Cs activity to the deposition time assuming the activity peak in the 33–34 cm interval represented 1964. As shown in Fig. 6, the $^{239+240}$Pu and ^{137}Cs activities in Lake Hongfeng sediments showed a high correlation ($R^2 = 0.878$). A similarly high correlation was obtained in a sediment core collected in Lake Chenghai ($R^2 = 0.881$). Therefore, the ^{137}Cs and Pu activities provided the same information about sedimentation processes in freshwater sediments. This was also the conclusion reached by Ketterer et al. (2002), who studied ^{137}Cs and $^{239+240}$Pu profiles in sediments from Old Woman Creek (Huron, OH, USA).

The peak in weapons testing during 1962 produced a peak annual $^{239+240}$Pu and ^{137}Cs deposition during 1963 in the Northern Hemisphere and during 1964 in the Southern Hemisphere (Hamilton et al., 1996). Wan et al. (2005) estimated the half-life of ^{137}Cs sinking to the bottom of Lake Chenghai in association with suspended particles using a box model of geochemical mass balance. They concluded that it was reasonable to assume that the retention time of ^{137}Cs in association with

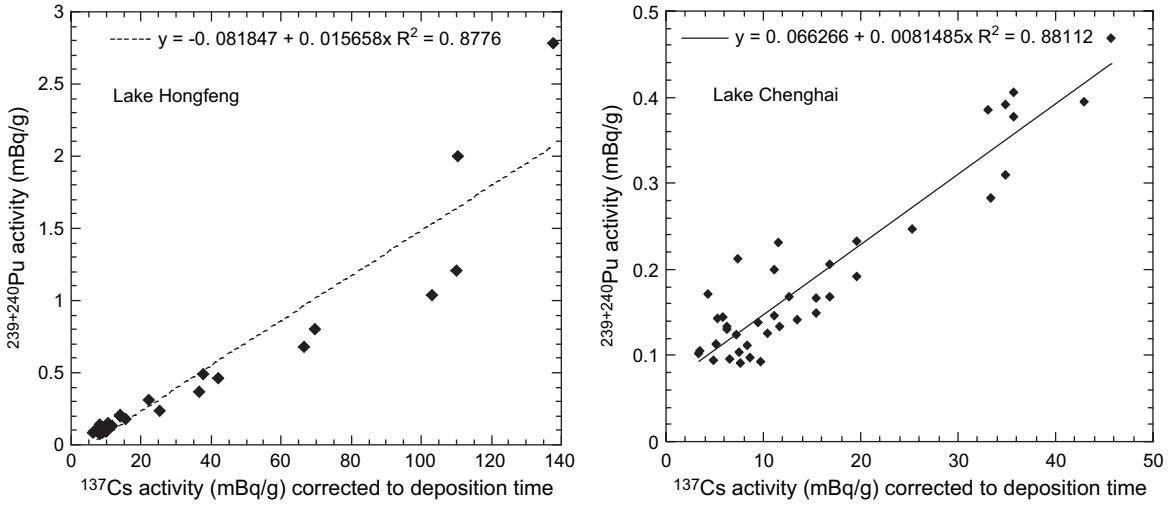

Fig. 6. Correlations between $^{239+240}$Pu activity and ^{137}Cs activity (radioactive decay corrected to the time of deposition) in Lake Hongfeng and Lake Chenghai.

sediments was approximately 1 year. Therefore, we could use the year 1964 as the dating time marker for the ^{137}Cs and $^{239+240}$Pu activity maximum in sediments in Lake Hongfeng.

The ^{137}Cs and $^{239+240}$Pu activities in sediment core HG20040709S displayed a maximum at a depth of 34 cm (Table 1), 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, we obtained a mean sedimentation rate of 0.84 cm yr^{-1} and a mean sediment accumulation rate of 0.217 g cm^{-2} yr^{-1}. Through field section and indoor treatment, we know that the total depth (*in situ*) of this sediment core (HF20040709S) was 37 cm (mass depth 11.36 g cm^{-2}) and underneath was yellow parent soil at the bottom of the lake. This fact indicated that the bottom sediment at the 37 cm horizon represented the starting sedimentation in 1960, which was in good agreement with the dating result (Table 1 and Fig. 2) using the obtained mean sedimentation rate.

It has been known that the vertical profile of ^{137}Cs may be affected by diffusion and post-depositional mobility in sediments. However, many studies have shown that ^{137}Cs diffusion post-deposition did not significantly change the position of the accumulation maxima of ^{137}Cs in sediments (Robbins and Edgington, 1975; Wan et al., 1987). In Lake Hongfeng, the history of the man-made lake is well documented (i.e., filled about 1960 just before the peak of global fallout), and the sedimentation rate is relatively high (0.84 cm yr^{-1} as estimated in the present work), which limited the possible post-depositional mobilization of ^{137}Cs and resulted in the coincidence of ^{137}Cs and Pu isotope vertical profiles. However, whether ^{137}Cs and Pu isotopes always have the same vertical distribution profiles, that is to say, to convey the same information about sedimentation processes as a common phenomenon, more investigations from different lake sediments with a variety of sedimentary rates are needed.

4. Conclusions

From the viewpoint of environmental radioactivity monitoring, a need still exists to construct a global distribution pattern (inventory and isotope compositions) for nuclear fallout, especially for Pu, so that possible future environmental inputs of non-fallout Pu can be identified and its impact can be quickly evaluated. On the other hand, the fallout nuclides, such as ^{137}Cs and Pu showed good potential to be geochemical tracers for quick dating of recent sedimentation in aquatic environment. In this work, probably, for the first time, we obtained a sediment core record for ground level Pu activity, inventory and ^{240}Pu/^{239}Pu atom ratio for northern temperate latitudes in China. An average ^{240}Pu/^{239}Pu atom ratio of 0.185 ± 0.009 was obtained in the sediment core collected in Lake Hongfeng, which indicated that global fallout was the source of these radionuclides. We found that the ^{137}Cs and Pu isotopes conveyed essentially the same information about sedimentation processes and chronology in Lake Hongfeng. It is our conviction that this study presented a reliable short-term (<50 year) sediment chronology for Lake Hongfeng sediments which is important for the study of pollution history of environmental contaminants, such as heavy metals and POPs in aquatic environments.

Acknowledgements

We would like to thank Dr. M. Kusakabe and Dr. T. Aono for their many valuable suggestions and fruitful discussions during this study. This work was jointly supported by the Director Research Fund, National Institute of Radiological Sciences, Japan (E16460-00000010), NSF of China (40525011, 40632011), and the Chinese Academy of Sciences (kzcx2-yw-102) and CAS international cooperation project.

References

Borretzen, P., Standring, W.J.F., Oughton, D.H., Dowdall, M., Fifield, L.K., 2005. Pu and U atom ratios and concentration factor in Reservoir 11 and Asanov Swamp, Mayak PA: an application of accelerator mass spectrometry. Environmental Science & Technology 39, 92–97.

Boulyga, S.F., Zoriy, M., Ketterer, M.E., Becker, J.S., 2003. Depth profiling of Pu, ^{241}Am and ^{137}Cs in soils from southern Belarus measured by ICP-MS and α and γ spectrometry. Journal of Environmental Monitoring 5, 661–666.

Buesseler, K.O., Sholkovitz, E.R., 1987. The geochemistry of fallout plutonium in the North Atlantic: II. ^{240}Pu/^{239}Pu ratios and their significance. Geochimica et Cosmochimica Acta 51, 2623–2637.

Buesseler, K.O., 1997. The isotopic signature of fallout plutonium in the North Pacific. Journal of Environmental Radioactivity 36, 69–83.

Cizdziel, J., Hodge, V., Faller, S., 1999. Resolving Nevada test site and global fallout plutonium in attic dust and soils using ^{137}Cs/$^{239+240}$Pu activity ratios. Health Physics 77, 67–75.

Hamilton, T.F., Millies-Lacroix, J.C., Hong, G.H., 1996. ^{137}Cs (^{90}Sr) and Pu isotopes in the Pacific Ocean: sources and trends. In: Guegueniat, P., Germain, P., Metivier, H. (Eds.), Radionuclides in the Oceans (RADOC 96-97) Inputs and Inventories. Les Editions de Physique. Les Ulis, France, pp. 29–58.

Hirose, K., Igarashi, Y., Aoyama, M., Kim, C.K., Kim, C.S., Chang, B.W., 2003. Recent trends of plutonium fallout observed in Japan: plutonium as a proxy for desertification. Journal of Environmental Monitoring 5, 302–307.

Hodge, V., Smith, C., Whiting, J., 1996. Radiocesium and plutonium: still fixed together in background soils after more than thirty years. Chemosphere 32, 2067–2075.

Huh, C.A., Chu, K.S., Wei, C.L., Liew, P.M., 1996. Lead-210 and plutonium fallout in Taiwan as recorded at a subalpine lake. Journal of Southeast Asian Earth Sciences 14, 373–376.

Kelley, J.M., Bond, L.A., Beasley, T.M., 1999. Global distribution of Pu isotopes and ^{237}Np. Science of the Total Environment 237/238, 483–500.

Ketterer, M.E., Watson, B.R., Matisoff, G., Wilson, C.G., 2002. Rapid dating of recent aquatic sediments using Pu activities and ^{240}Pu/^{239}Pu as determined by quadrupole inductively coupled plasma mass spectrometry. Environmental Science & Technology 36, 1307–1311.

Ketterer, M.E., Hafer, K.M., Jones, V.J., Appleby, P.G., 2004. Rapid dating of recent sediments in Loch Ness: inductively coupled plasma mass spectrometric measurements of global fallout plutonium. Science of the Total Environment 322, 221–229.

Krey, P.W., Hardy, E.P., Pachucki, C., Rourke, F., Coluzza, J., Benson, W.K., 1976. Mass isotopic composition of global fallout plutonium in soil. In: Transuranium Nuclides in the Environment. IAEA; Internal Atomic Energy Agency, Vienna, pp. 671–678. IAEA-SM-199/39.

Krishnaswamy, S., Lal, D., Martin, J.M., Meybeck, M., 1971. Geochronology of lake sediments. Earth and Planetary Science Letters 11, 407–414.

Kudo, A., Zheng, J., Yamada, R., Tao, G., Sasaki, T., Sugahara, M., 2000. Global transport rates and future prediction of hazardous materials: Pu

and Cs-from Nagasaki to Canadian Arctic. Water Science & Technology 42, 163–169.

Lin, R., Wei, K., Cheng, Z., 1992. Distributions of ^{210}Pb, ^{228}Th, ^{239,240}Pu and ^{3}H and their implications in sediment core from Bosten Lake, Xinjiang, China. Geochimica 1, 63–69 (in Chinese).

Muramatsu, Y., Ruhm, W., Yoshida, S., Tagami, K., Uchida, S., Wirth, E., 2000. Concentrations of ^{239}Pu and ^{240}Pu and their isotopic ratios determined by ICP-MS in soils collected from the Chernobyl 30-km zone. Environmental Science & Technology 34, 2913–2917.

Oktay, S.D., Santschi, P.H., Moran, J.E., Sharma, P., 2000. The ^{129}Iodine bomb pulse recorded in Mississippi River Delta sediments: results from isotopes of I, Pu, Cs, Pb and C. Geochimica et Cosmochimica Acta 64, 989–996.

Olivier, S., Bajo, S., Fiefield, L.K., Gaggeler, H.W., Papina, T., Santschi, P.H., Schotterer, U., Schwikowski, M., Wacker, L., 2004. Plutonium from global fallout recorded in an ice core from the Belukha glacier, Siberian Altai. Environmental Science & Technology 38, 6507–6512.

Robbins, J.A., Edgington, D.N., 1975. Determination of recent sedimentation rates in Lake Michigan using Pb-210 and Cs-137. Geochimica et Cosmochimica Acta 39, 285–304.

UNSCEAR, 2000. Sources and Effects of Ionizing Radiation. United Nations Scientific Committee on the Effects of Atomic Radiation Exposures to the Public from Man-made Sources of Radiation. United Nations, New York.

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. Chemical Geology 63, 181–196.

Wan, G.J., Chen, J.A., Wu, F.C., Xu, S.Q., Bai, Z.G., Wan, E.Y., Wang, C.S., Huang, R.G., Yeager, K.M., Santschi, P.H., 2005. Coupling between $^{210}Pb_{ex}$ and organic matter in sediments of a nutrient-enriched lake: an example from Lake Chenghai, China. Chemical Geology 224, 223–236.

Warneke, T., Croudace, I.W., Warwick, P.E., Taylor, R.N., 2002. A new ground-level record of uranium and plutonium isotopes for northern temperate latitudes. Earth and Planetary Science Letters 203, 1047–1057.

Yamada, Y., Yasuike, K., Komura, K., 2005. Temporal variation of carbon-14 concentration in tree-ring cellulose for the recent 50 years. Journal of Nuclear and Radiochemical Sciences 6, 135–138.

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. Environmental Science 14, 70–73 (in Chinese).

Zheng, J., Yamada, M., 2004. Sediment core record of global and Bikini close-in fallout Pu in Sagami Bay, Western Northwest Pacific margin. Environmental Science & Technology 38, 3498–3504.

Zheng, J., Yamada, M., 2006. Inductively coupled plasma-sector field mass spectrometry with a high-efficiency sample introduction system for the determination of Pu isotopes in settling particles at femtograms levels. Talanta 69, 1246–1253.

Zheng, J., Yamada, M., Wang, Z.L., Aono, T., Kusakabe, M., 2004. Determination of plutonium and its isotopic ratio in marine sediment samples using quadrupole ICP-MS with the shield torch system under normal plasma conditions. Analytical and Bioanalytical Chemistry 379, 532–539.

Zheng, J., Liao, H.Q., Wu, F.C., Yamada, M., Fu, P.Q., Liu, C.Q., Wan, G.J. A study of the vertical distributions of $^{239+240}$Pu activity and ^{240}Pu/^{239}Pu atom ratio in sediment core of Lake Chenghai, SW China. Journal of Radioanalytical and Nuclear Chemistry, in press.

Zhu, Y.G., Yang, J.C., Zhu, Y.I., 2002. Radiological research in China: current status and perspectives. In: Borretzen, P., Jolle, T., Strand, P. (Eds.), Proceedings from the International Conference on Radioactivity in the Environment, Monaco, pp. 595–599.