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Vertical distributions of $^{239+240}$Pu activity and ^{240}Pu/^{239}Pu atom ratio in sediment core of Lake Chenghai, SW China

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Due to the different ^{240}Pu/^{239}Pu atom ratios from different sources of Pu in the environment, Pu isotopes have been widely used for source identification of radionuclides in sediments. In this work, using sector-field ICP-MS, we investigated Pu inventory and its isotopic composition in a lacustrine sediment core collected in Chenghai Lake, SW China. The ^{240}Pu/^{239}Pu atom ratios in this sediment core ranged from 0.166 to 0.271 with a mean of 0.195±0.021, which was slightly higher than that of global fallout. The $^{239+240}$Pu/^{137}Cs activity ratios ranged from 0.0155 to 0.0411, with a mean of 0.0215, and the $^{239+240}$Pu inventory was 35.4 MBq/km^2; both $^{239+240}$Pu/^{137}Cs activity ratio and Pu inventory were close to those values of global fallout at 20–30 °N. Three peaks were observed for both ^{137}Cs and $^{239+240}$Pu activities in the examined sediment core; they most probably indicated the maximum deposition of global fallout between 1963 and 1964, the fallout from a series of Chinese nuclear tests during the 1970s, and the deposition of resuspended Pu-bearing particles from the Chernobyl accident. Therefore, the vertical profile of Pu isotopes should provide useful time markers for rapid dating of recent sediments.

Introduction

Artificial radionuclides, ^{239}Pu ($T_{1/2}$ = 24110 yr), ^{240}Pu ($T_{1/2}$ = 6584 yr) and ^{137}Cs ($T_{1/2}$ = 30 yr) have been released to the environment mainly from nuclear weapons production and testing, reprocessing of spent nuclear power plant fuel, and satellite and reactor accidents. Depending on the emission source, Pu isotope ratios, in particular the ^{240}Pu/^{239}Pu atom ratio, vary significantly in the environment.

In recent years, studies on the distribution of Pu isotopes and their application in environmental science have been increasing considerably. For instance, Pu and its isotopes were widely used for source identification of radioactive contaminants,[1–3] for sediment dating,[4] for the study of particle scavenging process,[5,6] and for tracing soil erosion.[7] MITCHELL et al.[1] examined ^{240}Pu/^{239}Pu ratio in sediment collected at Thule (Greenland), where an accident involving the release and dispersion of Pu from fractured nuclear weapons occurred in 1968, and found a low ^{240}Pu/^{239}Pu ratio of 0.033±0.004, which is indicative of weapons-grade plutonium. MURAMATSU et al.[8] investigated ^{240}Pu/^{239}Pu atom ratios in Japanese soil samples, and they found them to be close to that of global fallout (0.18). ZHENG and YAMADA[9] reported high ^{240}Pu/^{239}Pu atom ratios of 0.24–0.28 in marine sediments collected in Sagami Bay, Japan, indicating the presence of ca. 50% Bikini Atoll nuclear test source Pu that had been transported by oceanic currents. SAYLES et al.[10] estimated the contribution of artificial radioactive contaminants from the Chernobyl accident in Ob River and Techa River sediments by measuring Pu, ^{137}Cs activities and ^{238}Pu/$^{239+240}$Pu activity ratio.

Much work has also been done on the distribution of radionuclides in marine sediments.[2,4,9] In the terrestrial environment, studies on the radioactivity contamination mainly analyzed samples obtained in the vicinity of nuclear facilities, or in the area surrounding nuclear accident sites, and these were mainly located in high latitudes of the northern hemisphere. Little is known on the distribution of Pu isotopes in lake sediments, especially in China. In this work, we report the vertical distributions of $^{239+240}$Pu activity and ^{240}Pu/^{239}Pu atom ratio in a sediment core in Lake Chenghai, Southwest China. WAN et al.[11] observed a three-peak pattern vertical distribution of ^{137}Cs in a sediment core collected in Chenghai, and they assumed the three peaks of ^{137}Cs activity corresponded to the years 1964, 1975 and 1987. We have investigated the vertical distribution of Pu isotopes in samples from the same sediment core using sector-field ICP-MS with a high efficiency sample introduction system.[12] We focus on identifying the Pu source in this sediment core and present a lake sediment record on the history of atmospheric Pu fallout in the past four decades in the studied area.

Experimental

Chemical and reagents

All commercial chemicals were of analytical-reagent grade and were used without further purification. Nitric acid, HCl, $HClO_4$, NH_4I, H_2O_2, and $NaNO_2$ were obtained from Kanto Chemicals (Tokyo, Japan).

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Plutonium-242 (CRM 130, Plutonium Spike Assay and Isotopic Standard, New Brunswick Laboratory, USA) was used to spike the samples. The standard solution of ^{242}Pu was diluted to obtain stock solutions with a concentration of 10 ng/ml (1.5 Bq/ml) in 1N HNO_3 solution. The anion-exchange resin used in this study was AG 1-X8 (100–200 mesh, Bio-Rad).

Instrumentation

To determine the Pu isotopes in fresh water lake sediments, a sector-field (SF) ICP-MS (Finnigan Element 2, Bremen, Germany) was used in the low resolution (LR) mode in order to get maximal instrument sensitivity. An APEX-Q high-efficiency sample introduction system (Elemental Scientific Inc., Omaha, NE, USA) with a 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) further increasing the sensitivity of the SF-ICP-MS. All the measurements 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. The details on the instrument optimization have been described previously.[12]

Sample collection and analytical method

Lake Chenghai is located ca. 20 km south of Yongsheng County, northwestern Yunnan, China (26°27'–26°38'N, 100°38'–100°41'E). 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. Due to a progressive decrease of lake water level since the middle of the Ming Dynasty (1690 A. D.), the lake has become ombotrophic. Lake water is weakly alkaline with high hardness and its total ion concentration is up to 1 g/l,[11] approaching the lower limit ion concentration of salt lakes. In addition, abundant sunlight, favorable water temperature and aquatic chemistry create an environment conducive for the proliferation of blue-green algae, making Lake Chenghai a moderately nutrient-enriched lake. The sediment core was collected from Lake Chenghai in June 1997 at a water depth of approximately 28 m using a sediment–water interface sampler designed by YUAN et al.[13] The basic physical and chemical characteristics of Lake Chenghai can be found in the literature.[11] The sampling point was located approximately 2 km from the lake banks, tributary inlets and lake outlets. 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. 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 ground to <0.15 mm in diameter for Pu isotope analysis.

Details of sample preparation and Pu atom ratio analysis have been described elsewhere.[14] In brief, the sediment samples were dried at 110 °C for at least 4 hours. An aliquot of approximately 1–5 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 8M HNO_3 while heating on a hot plate (180–200 °C) for at least 3 hours. The separation of Pu and U, and the further purification of Pu were done by anion-exchange chromatography (AG 1-X8). 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%.

Results and discussion

The results on ^{137}Cs activity, $^{239+240}$Pu activity, and ^{240}Pu/^{239}Pu atom ratio obtained in the sediment core CH970608-1 are summarized in Table 1. The sedimentation rate in Lake Chenghai has been ca. 1 cm/year in recent years. The total $^{239+240}$Pu activities and ^{137}Cs activities ranged from 0.092 to 0.469 mBq/g, and from 2.66 to 21.45 mBq/g, respectively. The ^{240}Pu/^{239}Pu atom ratios and $^{239+240}$Pu/^{137}Cs activity ratios ranged from 0.164 to 0.271, and from 0.016 to 0.041, respectively. The inventory of $^{239+240}$Pu obtained in CH970608-1 core was 35.4 MBq/km^2 and in good agreement with the integrated atmospheric fallout of 36 MBq/km^2 for 20°–30° N published by UNSCEAR.[15]

Vertical profiles of $^{239+240}$Pu and ^{137}Cs activities

Figures 1b and 1c show the vertical profiles of ^{137}Cs activity, $^{239+240}$Pu activity and ^{240}Pu/^{239}Pu atom ratio; for comparison, the annual deposition of ^{137}Cs in the northern hemisphere is also plotted (Fig. 1a). There was a significant downcore variation of ^{137}Cs and $^{239+240}$Pu activities and ^{137}Cs and $^{239+240}$Pu activities showed a similar distribution pattern. In the examined sediment core, for both ^{137}Cs and $^{239+240}$Pu activities, three peaks were observed at 8 cm, 24 cm and 35 cm depths, respectively. Their peak activities for ^{137}Cs were 9.89 mBq/g, 11.85 mBq/g and 21.45 mBq/g, and for $^{239+240}$Pu activities, 0.231 mBq/g, 0.232 mBq/g and 0.469 mBq/g, respectively. Based on the ^{210}Pb dating results (Table 1), the peak at 35 cm corresponded to a deposition time of 1964 which was in good agreement with the annual maximum deposition of ^{137}Cs in the northern hemisphere (Fig. 1a),[16] indicating that the peak at 35 cm resulted from the global fallout in 1963–1964.

Table 1. ^{137}Cs and $^{239+240}$Pu activities, ^{240}Pu/^{239}Pu atom ratios and $^{239+240}$Pu/^{137}Cs activity ratios in sediment core from Lake Chenghai[a]

Depth, cm	Mass depth, g·cm^{-2}	A.D.	^{137}Cs,[b] mBq·g^{-1}	$^{239+240}$Pu, mBq·g^{-1}	^{240}Pu/^{239}Pu, atom ratio	$^{239+240}$Pu/^{137}Cs	Pu inventory, MBq·km^{-2}
1	0.25	0.6 1996.9	5.07±0.54	0.114±0.007	0.182±0.017	0.023	0.285
2	0.53	1.2 1996.3	5.14±0.61	0.143±0.016	0.206±0.019	0.028	0.400
3	0.78	1.8 1995.7	4.19±0.59	0.172±0.012	0.197±0.020	0.041	0.430
4	1.24	2.9 1994.6	5.81±0.76	0.134±0.019	0.165±0.020	0.023	0.616
5	1.73	4.0 1993.5	5.65±0.58	0.131±0.014	0.201±0.019	0.023	0.642
6	2.11	4.9 1992.6	6.57±0.66	0.212±0.019	0.195±0.011	0.032	0.806
7	2.58	5.9 1991.6	9.75±0.72	0.199±0.022	0.223±0.016	0.020	0.876
8	2.98	6.9 1990.6	9.89±0.80	0.231±0.02	0.271±0.011	0.023	0.993
9	3.36	7.7 1989.8	7.92±0.82	0.138±0.017	0.171±0.024	0.017	0.524
10	3.69	8.5 1989.0	2.91±0.48	0.106±0.007	0.203±0.021	0.036	0.350
11	4.15	9.6 1987.9	2.66±0.59	0.103±0.013	0.202±0.019	0.039	0.474
12	4.65	10.7 1986.8	3.82±0.41	0.095±0.006	0.164±0.018	0.025	0.475
13	5.00	11.5 1986.0	4.49±0.45	0.145±0.011	0.192±0.018	0.032	0.508
14	5.38	12.4 1985.1	4.88±0.45	0.096±0.012	0.195±0.011	0.020	0.365
15	5.77	13.3 1984.2	5.29±0.56	0.124±0.008	0.188±0.026	0.023	0.484
16	6.16	14.2 1983.3	6.25±0.53	0.097±0.018	0.202±0.027	0.016	0.378
17	6.55	15.1 1982.4	5.87±0.60	0.111±0.013	0.195±0.025	0.019	0.433
18	6.95	16.0 1981.5	7.24±0.61	0.126±0.014	0.199±0.020	0.017	0.504
19	7.38	17.0 1980.5	7.54±0.60	0.147±0.021	0.192±0.022	0.020	0.632
20	7.83	18.0 1979.5	7.71±0.66	0.134±0.015	0.203±0.019	0.017	0.603
21	8.25	19.0 1978.5	8.21±0.61	0.169±0.025	0.178±0.021	0.021	0.710
22	8.67	20.0 1977.5	9.75±0.76	0.166±0.012	0.181±0.029	0.017	0.697
23	9.10	21.0 1976.5	10.40±7.40	0.206±0.035	0.188±0.012	0.020	0.886
24	9.56	22.0 1975.5	11.85±0.83	0.232±0.027	0.210±0.011	0.020	1.067
25	10.00	23.0 1974.5	9.89±0.74	0.168±0.014	0.210±0.014	0.017	0.739
26	10.37	23.9 1973.6	4.39±0.56	0.092±0.023	–	0.021	0.340
27	10.72	24.7 1972.8	4.24±0.52	0.104±0.018	–	0.025	0.364
28	11.07	25.5 1972.0	5.38±0.55	0.093±0.015	0.184±0.014	0.017	0.326
29	11.45	26.4 1971.1	8.42±0.76	0.150±0.018	0.199±0.021	0.018	0.570
30	11.91	27.4 1970.1	10.44±0.73	0.192±0.019	0.195±0.022	0.018	0.883
31	12.40	28.6 1968.9	13.12±0.85	0.247±0.034	0.219±0.027	0.019	1.210
32	12.88	29.7 1967.8	16.86±1.13	0.283±0.015	0.193±0.022	0.017	1.358
33	13.37	30.8 1966.7	17.19±1.20	0.310±0.027	0.189±0.019	0.018	1.519
34	13.84	31.9 1965.6	17.16±1.01	0.405±0.016	0.175±0.016	0.024	1.904
35	14.30	32.9 1964.6	21.45±1.37	0.469±0.046	0.198±0.017	0.022	2.157
36	14.73	33.9 1963.6	19.71±1.20	0.395±0.039	0.186±0.019	0.020	1.699
37	15.18	35.0 1962.5	16.04±1.14	0.378±0.029	0.193±0.021	0.024	1.701
38	15.64	36.0 1961.5	14.48±1.17	0.386±0.025	0.174±0.023	0.027	1.776
39	16.62	38.3 1959.2	14.51±1.00	0.391±0.032	0.179±0.017	0.027	3.832
40	17.32	39.7 1957.8	5.41±0.84	0.141±0.019	0.200±0.026	0.026	0.860

[a] Sampling time June 8, 1997; ^{137}Cs measuring time Dec. 1997–June 1998; $^{239+240}$Pu measuring time Feb. 2006.
[b] Data from WAN et al. (2005); ^{137}Cs radioactivity decay corrected to sampling time.

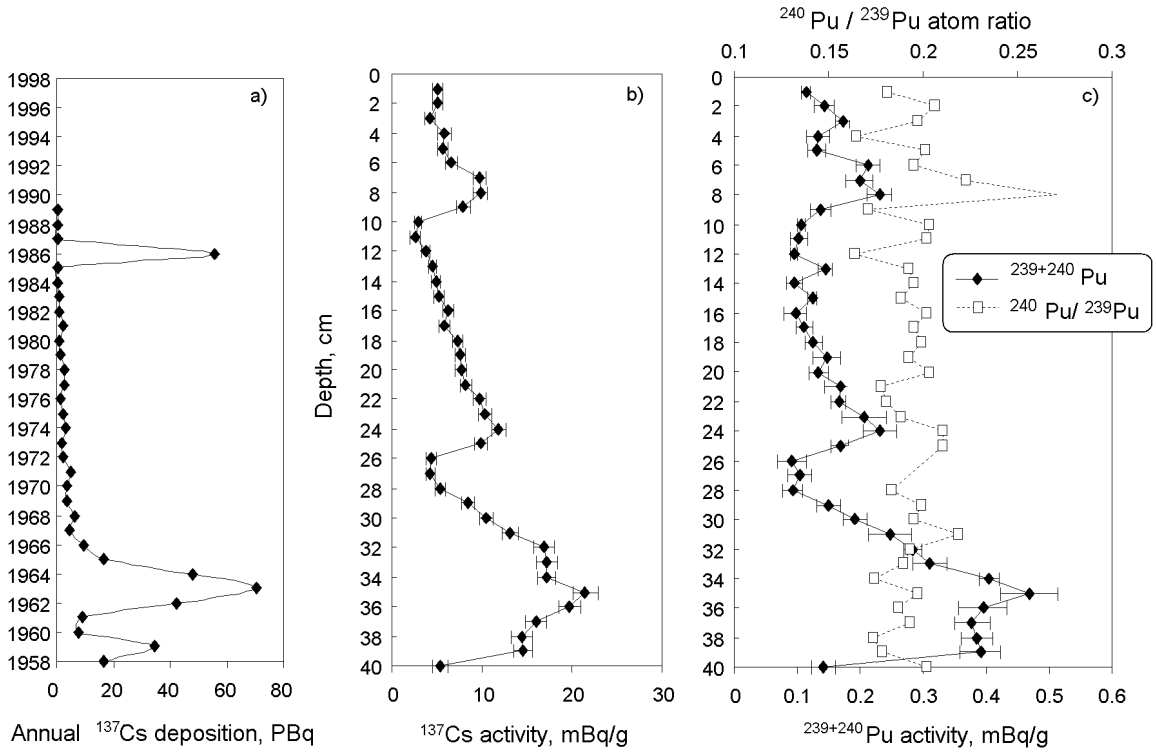

Fig. 1. Annual deposition of ^{137}Cs in the northern hemisphere (a), and vertical profiles of ^{137}Cs activity (b), and vertical profile of $^{239+240}$Pu activity and ^{240}Pu/^{239}Pu atom ratio (c) in Lake Chenghai sediment core samples

As shown in Fig. 1a, after the 1963 maximum deposition, the global fallout ^{137}Cs decreased year by year, and there was no any deposition peak from the late 1960s till the Chernobyl accident in 1986, while a peak for both ^{137}Cs and $^{239+240}$Pu activities appeared for the 20–24 cm layer from the Lake Chenghai sediment core. The peak maximum was seen at 24 cm, corresponding to the deposition in 1975 based on ^{210}Pb dating. WAN et al.[17] have reported a 1975 ^{137}Cs activity deposition peak in Lake Hongfeng sediment collected in 1988 (HF8801). A similar 1975 ^{137}Cs activity deposition peak was observed in sediment core from Bosten Lake, Xingjing, China,[18] reflecting the influence of Chinese above-ground nuclear tests. Therefore, we thought it is plausible to attribute the deposition peak of ^{137}Cs and $^{239+240}$Pu activities at 24 cm to the influence of Chinese nuclear tests during the late 1960s to 1970s.

In the northern hemisphere, the contamination of the radioactive nuclides resulting from the Chernobyl accident has been observed in many countries. Considering the fact that there were no above-ground nuclear tests in China since 1980, the peak of ^{137}Cs and $^{239+240}$Pu at 8 cm in Chenghai Lake sediment core may be explained by the deposition of Chernobyl source radionuclides.

Vertical profile of ^{240}Pu/^{239}Pu atom ratios

As summarized in Table 1 and shown in Fig. 1c, the lowest value of ^{240}Pu/^{239}Pu atom ratio, 0.164±0.018, appeared at depth 12 cm, and the highest value of 0.271±0.011 was observed at depth 8 cm. The mean value of ^{240}Pu/^{239}Pu atom ratios in Lake Chenghai sediment core (CH970608-1) was 0.195±0.021, which was slightly higher than that of global fallout (0.178±0.019, KELLEY et al.[19]) at the latitudinal band of 0–30 °N.

Chinese above-ground level nuclear tests started from 1964. Thermonuclear bomb tests were conducted 8 times from 1967 to 1980.[20] In order to evaluate the possible effect of Chinese nuclear tests on the distribution of radionuclides in the investigated sediment core, we divided the sediment core into several layers based on the results of ^{210}Pb dating: (1) 1–17 cm (1996–1982); (2) 18–31 cm (1981–1968); (3) 33–39 cm (1966–1959); and (4) 40 cm (1957). Figure 2 shows the average value of ^{240}Pu/^{239}Pu atom ratios in these four layers, they were 0.197±0.130, 0.197±0.028, 0.187±0.011 and 0.200±0.026, respectively.

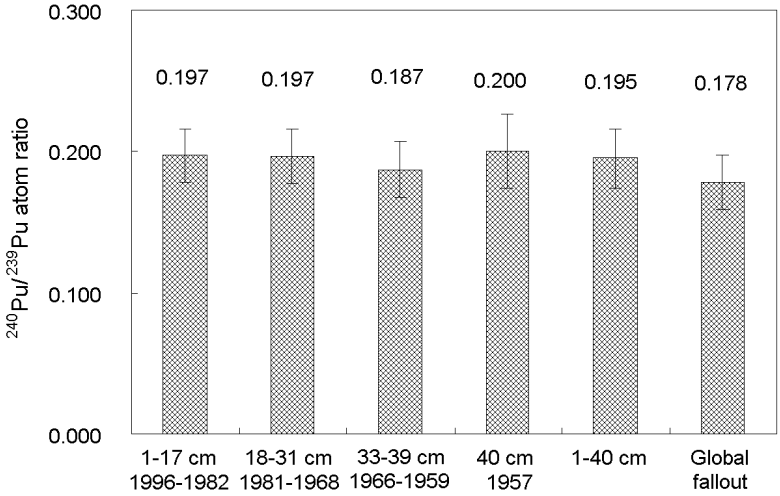

Fig. 2. Comparison of ^{240}Pu/^{239}Pu atom ratios in different layers in Lake Chenghai sediment core samples

In Lake Chenghai sediment core, we observed a value of 0.200±0.026 at the 40 cm layer, corresponding to the deposition in 1957. This result was consistent with that reported by KOIDE et al.[21] They indicated that before the 1960s, the global fallout value of ^{240}Pu/^{239}Pu atom ratio was higher than 0.18 (ca. 0.20–0.22).

In the layer from 33 to 39 cm, the mean of ^{240}Pu/^{239}Pu atom ratios was 0.187±0.011, which was close to the global fallout value of 0.178±0.019 for the latitudinal bands of 0–30 °N,[19] indicating that the source of ^{137}Cs and Pu isotopes was the stratospheric global fallout without contamination from other sources.

The mean of ^{240}Pu/^{239}Pu atom ratios in the layer from 18 to 31 cm was 0.197, higher than that of the global fallout value of 0.18. Based on ^{210}Pb dating, this layer corresponded to the deposition years from 1968 to 1981, the period of Chinese nuclear tests. In addition, the peak of $^{239+240}$Pu activity appeared at 24 cm, which coincided with the year when the largest thermonuclear test was conducted. Thus, the sediments in this layer to some extent reflected the deposition of radionuclides resulting from Chinese nuclear tests. Due to the lack of information about Pu isotopic composition of Chinese nuclear tests, however, at this stage, we could not make a quantitative evaluation on the contribution of Chinese nuclear tests to the radioactivity contamination in Lake Chenghai sediments.

Similarly, relatively high ^{240}Pu/^{239}Pu atom ratios were observed in the layer from 1 to 17 cm, the mean was also 0.197±0.130. This layer corresponded to the deposition from 1982 to 1996. During this period, there were no above-ground level nuclear tests anywhere in the world. The only source of Pu with high ^{240}Pu/^{239}Pu atom ratio was the Chernobyl accident in 1986; plutonium resulting from the Chernobyl accident has been characterized with a high ^{240}Pu/^{239}Pu ratio of 0.408.[22] A high value of 0.271±0.011 for ^{240}Pu/^{239}Pu atom ratio was observed at a depth of 8 cm. However, the ^{210}Pb dating indicated a deposition year of 1990 for this depth. We speculated that there was no direct atmospheric deposition of Chernobyl source Pu in Lake Chenghai sediments in 1986 when the accident took place, but the later re-suspension and deposition of deposited Pu-bearing particles could contribute to the observed high ^{240}Pu/^{239}Pu atom ratio in Lake Chenghai sediments.

Variation of $^{239+240}$Pu/^{137}Cs activity ratios

The vertical profile of $^{239+240}$Pu/^{137}Cs activity ratio in Lake Chenghai sediments is shown in Fig. 3. We found that the lowest $^{239+240}$Pu/^{137}Cs activity ratio was 0.016 and the highest was 0.041. The mean of $^{239+240}$Pu/^{137}Cs activity ratio in this sediment core was 0.0215, very close to the value of 0.021 for global fallout.[15] It was interesting to note that the layer from 17 cm to 32 cm, corresponding to the period of Chinese above-ground level nuclear tests, generally showed lower $^{239+240}$Pu/^{137}Cs activity ratios with a mean of 0.019, suggesting that $^{239+240}$Pu/^{137}Cs activity ratios in sediments may reflect the deposition of radionuclides from different sources.

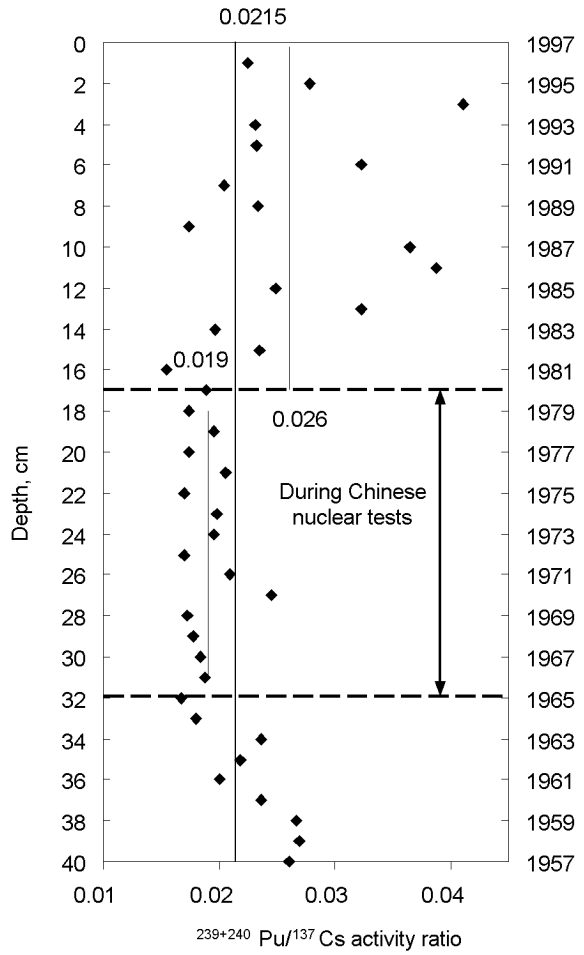

Fig. 3. Vertical profile of $^{239+240}$Pu/^{137}Cs activity ratio in Lake Chenghai sediment core samples

Conclusions

In this work, we used high-resolution sector field ICP-MS to investigated the vertical profiles of Pu isotopes in Lake Chenghai sediment core samples. The following conclusions can be drawn:

(1) $^{239+240}$Pu inventory (35.4 MBq/km^2) in Lake Chenghai sediments was close to the integrated atmospheric fallout of 36 MBq/km^2 for 20–30 °N published by UNSCEAR.[15] The mean of $^{239+240}$Pu/^{137}Cs activity ratios was similar to that of global fallout, the mean of ^{240}Pu/^{239}Pu atom ratios, however, was slightly higher than that of global fallout.

(2) Three peaks were observed for both ^{137}Cs and $^{239+240}$Pu activities in the examined sediment core; based on the characteristic ^{240}Pu/^{239}Pu atom ratio and $^{239+240}$Pu/^{137}Cs activity ratio and the ^{210}Pb dating, they indicated the maximum deposition of global fallout between 1963 and 1964, the series of Chinese nuclear tests during the 1970s, and the deposition of resuspended Pu-bearing particles from the Chernobyl accident. Therefore, the vertical profile of Pu isotopes could be useful as time marker for rapid dating of recent sediments.

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