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Vertical Distributions of Plutonium and ^{137}Cs in Lacustrine Sediments in Northwestern China: Quantifying Sediment Accumulation Rates and Source Identifications

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We investigated the vertical distributions of $^{239+240}$Pu and ^{137}Cs activities and ^{240}Pu/^{239}Pu atom ratios in two lakes in northwestern China. The ^{240}Pu/^{239}Pu atom ratio from the Chinese nuclear test (CNT) site at Lop Nor is too complicated to identify the regional fallout source. However, radionuclides from the test site may be resolved mainly by the vertical distribution of radionuclides because there is no overlap between the global fallout peak and the local fallout peak. We analyzed Pu isotopes and ^{137}Cs in sediments from two lakes (Sugan and Shuangta) located in northwestern China, near the CNT, and Lake Sihailongwan located in northeastern China to date recent lacustrine sediments and resolve global/local fallout sources. The apparently low ^{240}Pu/^{239}Pu atom ratio of 0.103 ± 0.010 at Lake Sugan was likely representative of the fallout from the Lop Nor site. Our results also demonstrated that the $^{239+240}$Pu activity was more useful for recent chronology of lacustrine sediments, compared to ^{137}Cs. Sediment accumulation rates of 0.651, 0.058−0.061, and 0.015 g·cm^{-2}·a^{-1} for sediment cores of Lake Shuangta, Lake Sugan and Lake Sihailongwan, respectively, were obtained by using the $^{239+240}$Pu fallout peak as a discrete time marker for 1964. The respective contributions of ^{137}Cs and $^{239+240}$Pu from the CNT were estimated to be about 40 and 27% for Lake Sugan and 36 and 20% for Lake Shuangta. Plutonium isotopes in lacustrine sediments were proven to be useful for quantifying sediment accumulation rates and for source identification of the radioactive contamination.

Introduction

The Chinese nuclear weapons testing program was carried out at the Lop Nor Nuclear Test Site (hereafter referred to as the CNT, Chinese Nuclear Test Site) in northwestern China. Twenty-two atmospheric tests were conducted between 1964 and 1980 (*1*). Limited information is available on local fallout deposition following the tests. It has been reported that the tropospheric deposition of radionuclides from CNT was mainly limited to Gansu Province in northwestern China (*2*). The ^{137}Cs activity, a byproduct of weapons testing, has been used to evaluate the impact of the Chinese nuclear tests on the environment in northwestern Gansu based on soil sampling conducted in 1987−1992 (*2*). Information on the transportation and distribution of plutonium from the CNT is unknown. The vertical distribution of radionuclides in sediments of lakes in northwestern China is unique and is needed to illustrate the mechanism of transportation and deposition of radionuclides from the CNT. But first, it is necessary to establish a time sequence of the depositions.

The global fallout of radionuclides in sediments marked as 1964 has normally been employed to construct the time sequence of lacustrine sediments. The vertical distribution of radionuclides may be disturbed by a local source, which may invalidate dating results based on the global fallout peak. In this regard, the ^{240}Pu/^{239}Pu atom ratio could be a useful tool for dating of sediments since the global/local fallout sources could be resolved by isotopic compositions (*3, 4*). The ^{240}Pu/^{239}Pu atom ratio for the global fallout source has been given as 0.176 ± 0.019 (*5*). For weapons grade plutonium, the reported ^{240}Pu/^{239}Pu atom ratio ranged from 0.01 to 0.07 (*6*), while for reactor-produced plutonium the reported range was from 0.24 to 0.80 (*7, 8*); the wide range of ratios due possibly to different reactor types, age of the fuel, and burn-up etc. Moreover, the ^{240}Pu/^{239}Pu atom ratio for global fallout in early nuclear tests was higher than 0.18 (*9*). For the CNT, it becomes more complicated since the original parameters of the nuclear tests are unavailable for estimation of the ^{240}Pu/^{239}Pu atom ratio. Warneke et al. (*8*) observed a ^{240}Pu/^{239}Pu atom ratio of 0.22 in herbage archive samples collected in the 1970s and they attributed this high ^{240}Pu/^{239}Pu atom ratio to the influence of the CNT; in fact, for the 21st Chinese atmospheric nuclear test conducted on 17 November 1976, a high ^{240}Pu/^{239}Pu atom ratio of 0.224 was observed in the debris (*10*). However, Jin et al. (*11*) observed a low ^{240}Pu/^{239}Pu atom ratio of 0.16 in a soil sample collected in western Gansu.

We determined ^{240}Pu/^{239}Pu atom ratio in soils in Gansu Province and found that the atom ratios of ^{240}Pu/^{239}Pu in the surface soils ranged from 0.168 to 0.192 with a mean of 0.182 ± 0.008 (*12*). This result suggests that global fallout was the major source of plutonium. Therefore, until more studies on the characterization of the CNT Pu atom ratios are conducted, the use of the ^{240}Pu/^{239}Pu atom ratio for resolving of global/local plutonium sources in northwestern China may be practically difficult. Fortunately, there may be no overlap between the global fallout peak and the local fallout peak in the time-resolved records of $^{239+240}$Pu and ^{137}Cs activities. Because the global fallout peak in sediments clearly marks the period 1963−1964, the local fallout peak in sediments caused by the CNT should be at least after 1968−1969 since the large scale nuclear tests there were started from June 1967. As such the time sequence of fallout deposition can be constructed by the vertical distributions of $^{239+240}$Pu and ^{137}Cs in sediments.

The postdepositional transport of radionuclides has been the main limitation preventing their application to dating. For plutonium, the post-transportation of this radionuclide relies to a great extent on conditions in sediments and surface water, such as pH, oxidation state, and the concentration and distribution of iron, manganese, and ammonium in the

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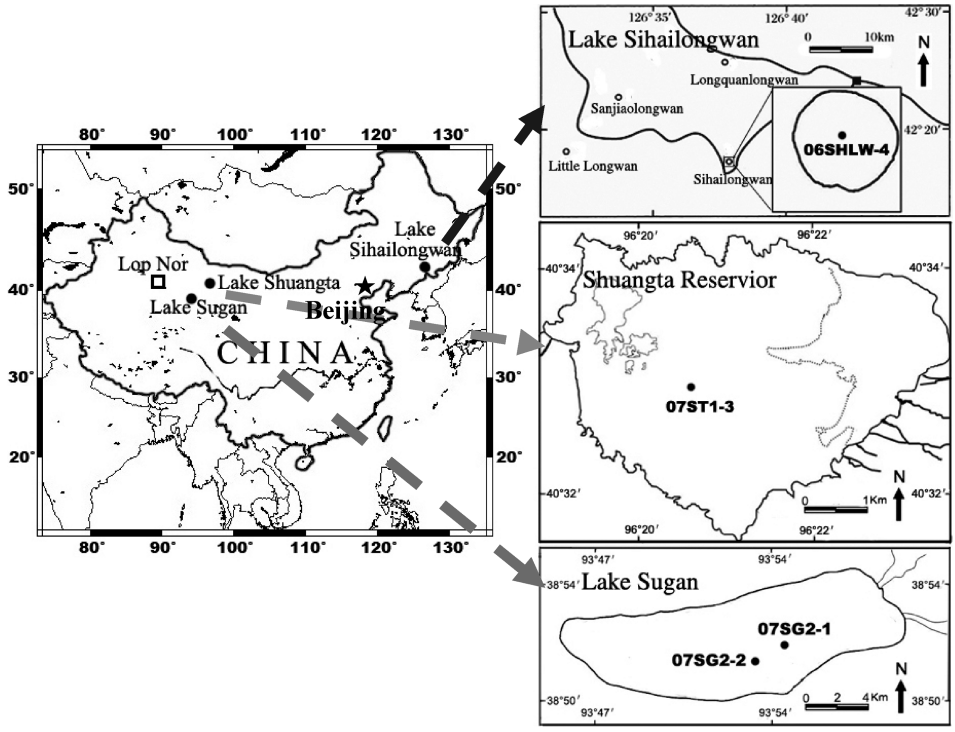

FIGURE 1. Map showing the locations of Lake Sugan, Lake Shuangta and Lake Sihailongwan, together with all sampling sites. The Chinese Nuclear Test Site at Lop Nor is also shown.

aquatic environment (*13, 14*). Livingston and Bowen (*15*) suggested that a fraction of the plutonium, accompanied by anionic complexes like HCO_3^- and organic matter, migrates to the sediment−water interface by interstitial water flow generated by sediment compaction or hydraulic ventilation and by channeling through worm burrows. Some investigators, however, proved that plutonium is not rapidly remobilized in oxic and anoxic conditions (*16, 17*). Kaplan et al. (*18*) found that slight changes in the system pH can distinctly influence the plutonium solubility. Kaplan et al. (*19*) also reported that Pu(V) which is the most mobile state for plutonium had a wide potential distribution especially in acidic environments. Though cesium seems to be a mobile element in sediments because it is soluble in pore water (*20*), the distinct accumulative peak of ^{137}Cs of 1964 has been widely applied to lacustrine sediment dating (*21*). In contrast to cesium, the postdepositional transport of plutonium is generally weaker in sediments. So, plutonium may convey more accurate information on chronology in sediments.

In the present work, we report the vertical distributions of plutonium and ^{137}Cs in sediments of Lake Sugan located in northwestern Qinghai Province and Lake Shuangta in northwestern Gansu Province to illustrate the influence of close-in fallout plutonium from the CNT on these regions. Since the vertical distribution of plutonium is the main method for resolving global/local sources, Lake Sihailongwan which is an undisturbed maar (low relief volcanic crater) lake located in northeastern China was used for regional comparison. We found that the vertical distribution of plutonium paralleled that of ^{137}Cs in lacustrine sediments. The history of regional contamination of radionuclides was then constructed based on the recent chronology obtained from the vertical distribution of radionuclides in the sediments.

Experimental Section

Sample Collection. Lake Sugan, Lake Shuangta, and Lake Sihailongwan are shown in Figure 1, together with sample locations. Lake Sugan is located ca. 500 km southeast of Lop Nor (the CNT) and Lake Shuangta is located ca. 600 km east of Lop Nor. Our control Lake Sihailongwan is located 3600 km away from Lop Nor, in Jilin Province, northeastern China. Three sediment cores were collected in 2007; two, designated 2007SG2-1 and 2007SG2-2 from Lake Sugan, and one, designated 2007ST1-3, from Lake Shuangta. A fourth sediment core, 2006SHLW-4, had been earlier collected in 2006 from Lake Sihailongwan. More detailed information on the location and sampling can be found in the Supporting Information (SI).

The core sediments were undisturbed, as indicated by the clear water-sediment interface and the preservation of fine sediment laminations. Sediment samples were sectioned at 0.5 cm intervals in situ and weighed immediately after collection, dried using a vacuum freeze-dryer (Techcorp FD-3−85-MP-79-36 mT), and then reweighed to determine mass depths and porosity. Dried samples were ground to <150 μm in diameter for ^{137}Cs and Pu isotope analysis.

Analytical Procedure. Details of sample preparation for Pu isotope analysis have been described elsewhere (*22*). In brief, the extraction of Pu from sediments was done using 8 M HNO_3. Further separation and purification of Pu isotopes was conducted with a two-stage anion-exchange chromatography using AG 1-X8 and AG-MP-1 M resins. Concentrated HBr was used as the final eluent for Pu elution; the troublesome polyatomic interference $PbCl^+$ for low level Pu isotope determination by ICP-MS was eliminated. The chemical yield in the employed sample preparation procedure was estimated to be in the range of 55−91% with a mean of 71 ± 17%. The determination of Pu isotopes was conducted in the Nakaminato Laboratory of Marine Radioecology, National Institute of Radiological Sciences, Japan, with an APEX/SF-ICP-MS analytical system (*23*). The detection limits for Pu isotopes were 0.0005 mBq/g for ^{239}Pu and 0.002 mBq/g for ^{240}Pu, calculated for 1 g of sediment sample. A Pu isotope standard solution (NBS-947) with a certified ^{240}Pu/^{239}Pu atom ratio of 0.2420 was used for mass bias correction. In order to obtain precise and accurate Pu activities and ^{240}Pu/^{239}Pu atom ratios in lake sediments, the

TABLE 1. Sample Locations, $^{239+240}$Pu Inventory, ^{137}Cs Inventory and $^{239+240}$Pu/^{137}Cs Inventory Ratio in Lake Sediments

lake	approximate distance from lop nor	annual precipitation (mm)	sediment core	depth (m)	latitude(N), longitude(E)	inventory (MBq·km^{-2}) $^{239+240}$Pu	inventory (MBq·km^{-2}) ^{137}Cs [a]	$^{239+240}$Pu/^{137}Cs inventory ratio
Lake Shuangta	600 km	37.3−62.5 [b]	07ST1−3	21.0	40°32.955′, 96°20.476′	240.6 ± 5.3	7258.2 ± 529.1	0.033 ± 0.004
			07SG2−1	4.3	38°51.835′, 93°54.765′	20.2 ± 0.8	511.4 ± 52.9	0.039 ± 0.002
Lake Sugan	500 km	15.8[c]	07SG2−2	3.8	38°51.436′, 93°52.478′	24.2 ± 0.7	569.9 ± 86.1	0.042 ± 0.002
Lake Sihailongwan	3600 km	767.0 [d]	06SHLW-4	50.0	42°17.213′, 126°36.087′	62.7 ± 3.2	2503.8 ± 109.3	0.025 ± 0.003

[a] ^{137}Cs inventory was corrected to 1 July 1998. [b] cited from ref *37*. [c] cited from ref *38*. [d] cited from ref *39*.

employed analytical method was validated by the analysis of IAEA-368 and NIST-4357 ocean sediments, and NIST-4354 freshwater lake sediment. The experimentally established values of $^{239+240}$Pu activities and ^{240}Pu/^{239}Pu atom ratios were in good agreement with the certified values and those reported in the literature (*22*).

^{137}Cs activity was determined using gamma-spectrometry on a Canberra S-100 multichannel spectrometer with a GC5019 HP-Ge coaxial detector (efficiency 50%) at the Institute of Geochemistry, Chinese Academy of Sciences.

Results and Discussion

Plutonium and ^{137}Cs in Sediments. From 1964 to 1980, 22 atmospheric nuclear tests were conducted at Lop Nor which is the only known Chinese nuclear weapons test site. In the present study, two sediment cores from Lake Sugan (07SG2-1 and 07SG2-2) and a sediment core (07ST1-3) from Lake Shuangta were collected in 2007. The sediment core 06SHLW-4, collected in 2006 from Lake Sihailongwan which is far from Lop Nor, was used for comparison. The four sediment cores were analyzed for their $^{239+240}$Pu activities and ^{240}Pu/^{239}Pu atom ratios by SF-ICP-MS, and for their ^{137}Cs activities by gamma-spectrometry. ^{137}Cs inventory, $^{239+240}$Pu inventory, and $^{239+240}$Pu/^{137}Cs inventory ratio are summarized in Table 1. The ^{137}Cs inventory radioactivity decay was corrected to 1 July 1998. The $^{239+240}$Pu and ^{137}Cs inventories of Lake Sihailongwan were 62.7 ± 3.2 and 2503.8 ± 109.3 MBq·km^{-2}, respectively. They were close to the integrated atmospheric fallout values of 58 and 2618 MBq·km^{-2} for 40°−50° N published by UNSCEAR (*24, 25*). However, the inventories of radionuclides in lake sediments in northwestern China were remarkably different from those in northeastern China. The $^{239+240}$Pu and ^{137}Cs inventories of Lake Shuangta were 240.6 ± 5.3 and 7258.2 ± 529.1 MBq·km^{-2} respectively. Both of them were obviously higher than the integrated atmospheric fallout values of 58 and 2618 MBq·km^{-2} for 40°−50° N. These unusually high inventories may be attributed to the sediment focusing process, that is, the rapid filling up of the reservoir by silt as a result of strong soil erosion, or the contribution of explosion debris from the Chinese nuclear weapons tests. Moreover, the cumulative depositions of radionuclides in different lakes located in northwestern China also varied from each other. The $^{239+240}$Pu inventory and ^{137}Cs inventory of Lake Sugan were slightly lower than those values of the integrated atmospheric fallout of 42 and 1923 MBq·km^{-2} for 30°−40° N. The $^{239+240}$Pu inventories of 07SG2-1 and 07SG2-2 sediment cores were 20.2 ± 0.8 and 24.2 ± 0.7 MBq·km^{-2}, respectively; and ^{137}Cs inventories were 511.4 ± 52.9 and 569.9 ± 86.1 MBq·km^{-2}. The low inventories of $^{230+240}$Pu and ^{137}Cs observed in Lake Sugan may be attributed to the arid climate in this region which leads to less annual precipitation and deposition (*26*).

The $^{239+240}$Pu/^{137}Cs inventory ratios in lakes in northwestern China were also different from that in northeastern China. The $^{239+240}$Pu/^{137}Cs inventory ratio of 0.025 for core 06SHLW-4 (northeastern China) was very close to the global fallout value of 0.026 ± 0.003 (*27*) in which ^{137}Cs inventory has been corrected to 1 July 1998 (Table 1). But $^{239+240}$Pu/^{137}Cs inventory ratios of 0.033, 0.039, and 0.042 for sediment cores 07ST1-3, 07SG2-1, and 07SG2-2 (northwestern China) were a little higher than the global fallout value, indicating that a contribution from a local source may have changed the composition of plutonium and ^{137}Cs in lacustrine sediments of northwestern China.

Efforts have been made to study the distributions of $^{239+240}$Pu activity and ^{137}Cs activity in lake sediments. One accumulative peak which indicates the 1963 global fallout peak has usually been observed in the distribution pattern of radionuclides in lacustrine sediments and marine sediments (*3, 4, 28, 29*). The accumulative peak owing to the Chernobyl accident has been detected in sediments and soils in Europe (*8, 30, 31*). Moreover, the vertical distribution of radionuclides in the vicinity of nuclear test sites becomes more complicated. In the present work, both $^{239+240}$Pu and ^{137}Cs activities were generally presented as one fallout peak in lake sediments in northwestern and northeastern China (Figure 2). In addition, the peak shapes of the radionuclide deposition of core 07SG2-1 were nearly the same as those of core 07SG2-2. We consider that the accumulative peak in these lakes corresponds to the global fallout peak in 1963 similar to the previous studies on the distribution of radionuclides in lake sediments (*3, 4, 28, 32*). Although it is difficult to understand whether there is any other weak radionuclide peak above the 1963 global fallout peak in sediments of Lake Sihailongwan due to its very low accumulation rate of sediments, some subpeaks of $^{239+240}$Pu and ^{137}Cs activities in upper sediments were distinctly present in sediments of Lake Sugan and Lake Shuangta. These may be attributed to the input of a local radionuclides source or an inconsistent deposition history.

Krey et al. (*5*) analyzed the ^{240}Pu/^{239}Pu atom ratio in 51 soil samples collected worldwide and provided an average ^{240}Pu/^{239}Pu atom ratio of 0.176 ± 0.019 for the global fallout plutonium source. Much later, Kelley et al. (*33*) provided four proposed values for different latitude zones based on ^{240}Pu/^{239}Pu atom ratios of 54 soil samples. They are 0.180 ± 0.014, 0.178 ± 0.019, 0.173 ± 0.027, and 0.185 ± 0.047 for regions of 71−30°N, 30°N-0, 0−30°S, and 30−53°S. All lakes and reservoir studied in the present work were located in the region of 71−30°N. The vertical distribution of the ^{240}Pu/^{239}Pu atom ratio is also presented in Figure 2. Ranges of the ^{240}Pu/^{239}Pu atom ratio were 0.157−0.193, 0.160−0.192, 0.159−0.183, and 0.103−0.195 for sediment cores 06SHLW-4, 07ST1-3, 07SG2-1, and 07SG2−2, and the averages of the ^{240}Pu/^{239}Pu atom ratios were 0.182 ± 0.012, 0.178 ± 0.007, 0.174 ± 0.054, and 0.164 ± 0.026, respectively. Except for the apparently low ^{240}Pu/^{239}Pu atom ratio of 0.103 ± 0.010 in the 0.90 g·cm^{-2} segment of core 07SG2-2, all ^{240}Pu/^{239}Pu atom ratios were within the range of the global fallout value. This low ^{240}Pu/^{239}Pu atom ratio suggests that radionuclides from

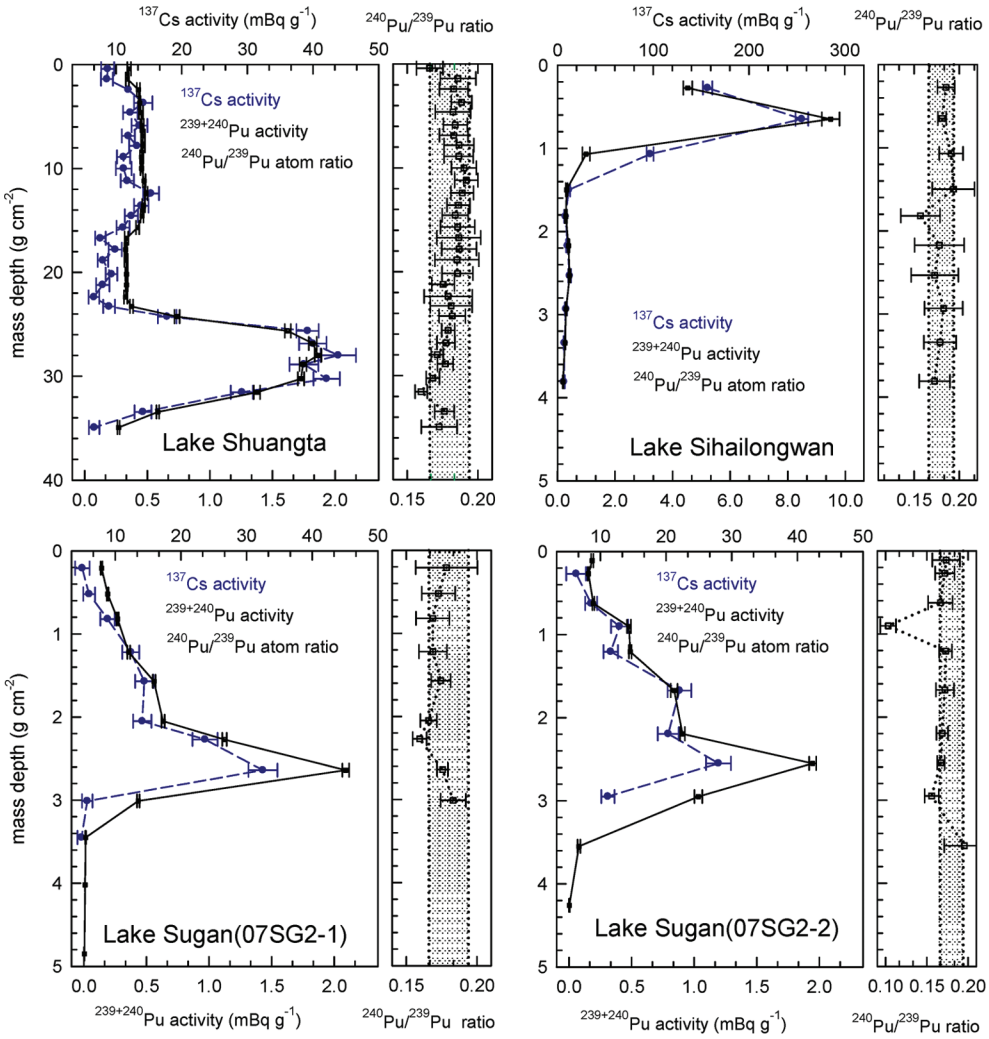

FIGURE 2. Vertical distributions of ^{137}Cs, $^{239+240}$Pu activity and ^{240}Pu/^{239}Pu atom ratio in Lake Sugan, Lake Shuangta, and Lake Sihailongwan. (Shading in the figure indicates the ^{240}Pu/^{239}Pu atom ratio range of the global fallout).

the local source, such as the close-in fallout of the CNT had been deposited in Lake Sugan. Unfortunately, not enough ^{240}Pu/^{239}Pu atom ratios could be obtained to resolve the local/global fallout plutonium source. Moreover, the low ^{240}Pu/^{239}Pu atom ratio appeared in a layer very close to the surface. Chronology information is necessary to know whether debris of the Chinese nuclear tests was directly deposited in Lake Sugan or was transported to Lake Sugan due to its later resuspension.

Dating Lacustrine Sediments by Plutonium Accumulative Peak. The global fallout peak of ^{137}Cs activity has been widely employed to date lacustrine and marine sediments (*28, 34*). Due to its short half-life (30.17 y), about 70% of the ^{137}Cs deposited in sediments has disappeared through its radioactive decay. Precise determination of ^{137}Cs becomes the main barrier to the application of ^{137}Cs in recent chronology measurements for sediments. Plutonium, an artificial radionuclide with a long half-life, also can be used to study the recent chronology of lakes because sediments preserve the $^{239+240}$Pu accumulative peak the same as they do the ^{137}Cs accumulative peak. However, due to their different chemical properties, whether the ^{137}Cs and Pu isotopes convey essentially the same information about sedimentation processes and chronology is a major point that should be elucidated first. Some studies have been done to evaluate the comparability of sedimentation processes in freshwater sediments between $^{239+240}$Pu and ^{137}Cs by correlation between $^{239+240}$Pu activity and ^{137}Cs activity in freshwater lakes (*3, 35*). $^{239+240}$Pu and ^{137}Cs activities in sediments of Lake Sugan, Lake Shuangta and Lake Sihailongwan had high correlations as shown in SI Figure 1S, in which the radioactive decays of ^{137}Cs activities were corrected to the deposition time assuming the activity peak represented 1964. This result demonstrated that the $^{239+240}$Pu and ^{137}Cs activities convey the same information of chronology for these three lakes.

Experiments have been carried out to simulate the transportation of plutonium in sediments. Kaplan et al. (*18, 19, 36*) found that changes of pH and oxidation state made the plutonium mobile in certain sediments. Moreover, plutonium partitioning to colloidal and particulate matter would finally cause the remigration of plutonium in soil (*13, 14*). Plutonium transport in sediment would cause its redistribution, further influencing its application in recent chronology for lakes. It is unknown whether dating results of plutonium chronology are credible because it is difficult to estimate the extent of plutonium remigration in lacustrine sediments. However, the influence of remigration of radiocesium on its recent chronology has been discussed by Wan et al. (*34*). They found that the global fallout peak of ^{137}Cs was applicable to measuring recent chronology of sediments though there was a weak remigration and redistribution of ^{137}Cs in sediments. So we employed ^{137}Cs as a reference to study the mobility of plutonium in sediments. The factor for interzone percentage ($F_{(Pu/Cs)}$) of cumulative deposition density of plutonium vs ^{137}Cs in ranges which are close to the

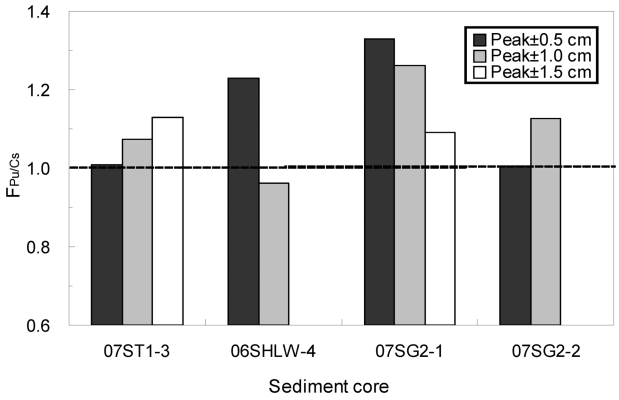

FIGURE 3. Factor for interzone cumulative deposition density of $^{239+240}$Pu vs ^{137}Cs in sediment cores 07ST1-3, 06SHLW-4, 07SG2-1, and 07SG2−2. Peak ± 0.5 cm, Peak ± 1 cm and Peak ± 1.5 cm in the figure denote the boundary of the interzone.

peak was used to evaluate the validity of recent chronology as described by plutonium; the cumulative deposition density here is the interzone inventory (MBq·km^{-2}). The model is described as follows:

$$F_{(\mathrm{Pu/Cs})i} = \frac{\sum_{p-i}^{p+i} (D_{(\mathrm{Pu})x}) / I_{\mathrm{Pu}}}{\sum_{p-i}^{p+i} (D_{(\mathrm{Cs})x}) / I_{\mathrm{Cs}}} \quad (1)$$

where I_{Pu} and I_{Cs} are the inventories of $^{239+240}$Pu and ^{137}Cs, respectively; $\sum_{p-i}^{p+i} D_{(Pu)x}$ is the sum of cumulative deposition densities of $^{239+240}$Pu in layers between $p_{peak} - i$ and $p_{peak} + i$ cm; $\sum_{p-i}^{p+i} D_{(Cs)x}$ is the sum of cumulative deposition densities of ^{137}Cs in layers between $p_{peak} - i$ and $p_{peak} + i$ cm. $\sum_{p-i}^{p+i} (D_{(Pu)x}) / I_{Pu}$ and $\sum_{p-i}^{p+i} (D_{(Cs)x}) / I_{Cs}$ means the interzone percentages of plutonium and ^{137}Cs in layers between $p_{peak} - i$ and $p_{peak} + i$ cm, respectively. There are three probabilities for $F_{(Pu/Cs)}$: (1) if the mobility of Pu is lower than that of ^{137}Cs, $F_{(Pu/Cs)} > 1$; (2) if the mobility of Pu is equal to that of ^{137}Cs, $F_{(Pu/Cs)} = 1$; and (3) if the mobility of Pu is higher than that of ^{137}Cs, $F_{(Pu/Cs)} < 1$.

Since the fallout process for plutonium and ^{137}Cs is the same, the interzone percentage should be theoretically the same. Additionally, the interzone percentage can decrease due to lose of radionuclides. So, $F_{(Pu/Cs)}$ can be applied to evaluate the relative velocity of their postdeposition. As shown in Figure 3, we found that the majority of $F_{(Pu/Cs)}$factors were greater than 1.0 for $^{239+240}$Pu vs ^{137}Cs in sediment cores, indicating that the mobility of plutonium in lakes is evidently lower than that of ^{137}Cs. Therefore, $^{239+240}$Pu is also applicable to measuring recent chronology of lacustrine sediment, and is even more useful than ^{137}Cs.

Using the $^{239+240}$Pu fallout peak as a discrete time marker of 1964, we obtained mean sediment accumulation rates of 0.651, 0.061, 0.058, and 0.015 g·cm^{-2}·a^{-1} for sediment cores 07ST1-3, 07SG2-1, 07SG2-2, and 06SHLW-4. We also established the time sequence of sediments using the calculated sediment accumulation rates combined with mass depth. As shown in Table 1, the annual precipitation was significantly different in the lake areas. Around Lakes Sugan and Shuangta, the annual precipitation ranged from 15.8 to 62.5 mm (*37*, *38*); however, for Lake Sihailongwan, which has the lowest sediment accumulation rate among the investigated lakes, the annual precipitation was 767 mm (*39*), suggesting that sediment accumulation rate did not necessarily correlate to the annual precipitation.

The Net ^{137}Cs and $^{239+240}$Pu Inventories from CNT. The ^{240}Pu/^{239}Pu atom ratio is the best means to resolve local/global plutonium sources since plutonium from different sources has dissimilar ^{240}Pu/^{239}Pu atom ratios (*4*). But the ^{240}Pu/^{239}Pu atom ratio is too complicated to be used to calculate the net inventory from the CNT. Ren et al. (*26*) reported that 40% of the ^{137}Cs in a lake located in the southwestern part of Dunhuang County was from the CNT, and that the contributions from the CNT (about 70%) were mainly deposited in the period from 1969 to 1975. In the present work, we also estimated the contributions from the CNT based on the time sequence of sediments. For an easy comparison, cumulative deposition density of ^{137}Cs and $^{239+240}$Pu in each layer, and the annual deposition of ^{137}Cs in the northern hemisphere were converted to abundance relative to the peak value. As shown in SI Figure 2S, the cumulative deposition densities of ^{137}Cs and $^{239+240}$Pu upon the peak of 1964 are higher than the global fallout. The first Chinese nuclear test using plutonium was conducted in December 1968 (*40*). Considering the transportation time in the atmosphere and the settling time in the water column, deposition in the sediment by radionuclides from the CNT should be from 1969 or later. In the present work, we took 1970 as the dividing point and estimated the net ^{137}Cs and $^{239+240}$Pu inventories from the CNT by eliminating deposition of global fallout. The contribution of the net ^{137}Cs and $^{239+240}$Pu inventories from the CNT in sediment cores 07SG2-1, 07SG2-2, and 07ST1-3 are summarized in Table 2. As shown there, 43.4% of the ^{137}Cs in core 07SG2-1 and 41.7% of the ^{137}Cs in core 07SG2-2 were from the CNT, similar to the results of the lake in Dunhuang County obtained by Ren et al. (*26*). The contributions of the net ^{137}Cs and $^{239+240}$Pu inventories from the CNT to Lake Shuangta were lower than those to Lake Sugan, indicating that the contribution of the CNT has an inverse correlation to the distance between sample locations and the test site. The net ^{137}Cs and Pu inventories from CNT obtained in the present study were a preliminary estimation, because in addition to recording direct atmospheric deposition of fallout radionuclides, a sediment core will also record the ongoing, continuous deposition of fallout previously deposited in the watershed, as surface soils (containing

TABLE 2. Contributions of the Net ^{137}Cs and $^{239+240}$Pu Inventories from the CNT

sediment core	radionuclide	inventory from CNT (MBq·km^{-2})[a]	percent of CNT (%)
07SG2-1	^{137}Cs	221.9	43.4
	$^{239+240}$Pu	5.3	26.3
07SG2-2	^{137}Cs	211.7	41.7
	$^{239+240}$Pu	6.6	27.2
07ST1-3	^{137}Cs	2218.1	30.6
	$^{239+240}$Pu	48.2	20.0

[a] ^{137}Cs inventory was corrected to 1 July 1998.

fallout) are eroded and this material enters the lake and is then deposited. In addition, it should be noted that we employed an acid leaching technique for the determination of Pu isotopes in lake sediments, which could underestimate the contribution of the CNT because some refractory Pu might be present, as has been demonstrated in the study of Pu from the Nevada Test Site (*41*). More accurate estimation of the CNT contribution can be expected from future studies of soils and lake sediments in an area closer to the Lop Nor test site.

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Supporting Information Available

Information on the locations of the lakes investigated and the sediment core sampling. Figure 1S: Correlations between $^{239+240}$Pu activity and ^{137}Cs activity (radioactive decay corrected to the time of deposition) in Lake Shuangta, Lake Sugan and Lake Sihailongwan. Figure 2S: Abundance of annual deposition of ^{137}Cs in the northern hemisphere and abundance of cumulative deposition density of ^{137}Cs and $^{239+240}$Pu in each layer of sediments. This material is available free of charge via the Internet at http://pubs.acs.org.

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