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$^{240}Pu/^{239}Pu$ Ratios in Sun Moon Lake Sediments: Implications for Sources of Nuclear Fallout in Taiwan

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ABSTRACT

The isotopic composition of plutonium in dated Sun Moon Lake sediments was determined by mass spectrometry in an effort to trace the source(s) and timing of nuclear fallout in Taiwan. The $^{240}Pu/^{239}Pu$ ratios in the sediment core vary by a factor of >5, with the highest ratio, 0.22, corresponding to the maximum level of nuclear fallout in 1963 and the lowest ratios, 0.04-0.06, being identified at two time horizons: 1982-83 and 1992-93. At other depth intervals, the $^{240}Pu/^{239}Pu$ ratios fall within 0.15-0.19. The data indicate that global fallout from the stratosphere is the dominant source of Pu in the sediments, whereas the tropospheric transport of low burn-up debris from Lop Nor in NW China is mostly responsible for the unusually low $^{240}Pu/^{239}Pu$ ratios.

(Key words: Plutonium, Nuclear fallout, Sun Moon Lake)

1. INTRODUCTION

Sun Moon Lake, located in the geographic center of Taiwan, is the largest natural lake on the island. Huh *et al* (1996) analyzed a sediment core collected from the lake for two radionuclides of atmospheric origin: the natural ^{210}Pb and the anthropogenic $^{239,240}Pu$. The distribution of $^{239,240}Pu$ in that core is characterized by a pronounced maximum at the 26-28 cm interval (Figure 1). Based on the ^{210}Pb-derived mean sedimentation rate over the past four to five decades, the age of the Pu maximum is consistent with the history of nuclear fallout which peaked in 1963. The inventory of $^{239,240}Pu$ in the sediment core, $0.97 \pm 0.05 mCi/km^2$, is in excellent agreement with the average cumulative $^{239,240}Pu$ fallout (0.96 ± 0.07 mCi/km^2) in the 20-30° N latitudinal band (Hardy *et al.*, 1973). The inventory of excess ^{210}Pb in the sediment core (61 dpm/cm^2) also points to a flux (1.9 $dpm/cm^2/yr$ at steady state) similar to the atmospheric ^{210}Pb flux measured at several other locations in the western Pacific margin (Huh *et al.*, 1996). These corroboratory results suggest that Sun Moon Lake sediments serves as an ideal system for the preservation of the history of atmospheric fallout in Taiwan.

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In the earlier work of Huh *et al.* (1996), Pu was measured by alpha spectrometry. However, because the energies of alpha particles emitted by ^{239}Pu and ^{240}Pu are too close to be distinguished by alpha spectrometry, the two isotopes were measured together and reported as ^{239,240}Pu. In view of this, the objective of the present work is to determine the isotopic composition of plutonium (i.e. the ^{240}Pu/^{239}Pu ratio) by mass spectrometry and infer the possible

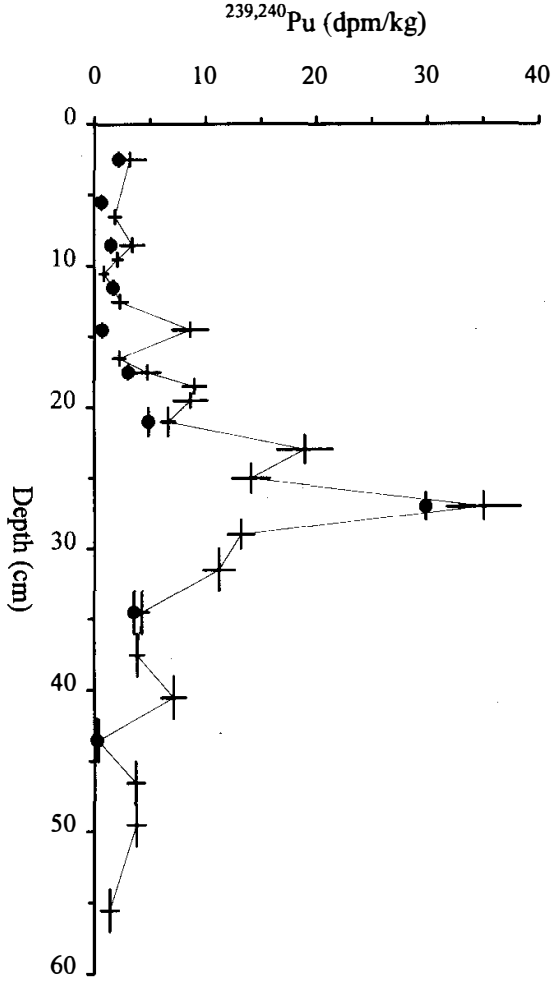

Fig. 1. Profile of ^{239,240}Pu in Sun Moon Lake sediments. Crosses represent measurements made by alpha spectrometry(from Huh *et al.*,1996). Results obtained by mass spectrometry(solid circles) are also plotted for comparison.

sources of Pu and, by extension, of other anthropogenic nuclides, such as ^{137}Cs and ^{90}Sr, in Taiwan.

2. METHOD

Nine of the samples previously analyzed for ^{239,240}Pu by alpha spectrometry were selected here for Pu isotopic analysis by mass spectrometry. With ^{244}Pu (NIST SRM-996) being added as the yield determinant and Nd as the carrier, the samples were digested using HCl+HF followed by HNO_3+$HClO_4$. Upon total dissolution and evaporation to incipient dryness, the samples were re-dissolved in a mixture of HCl + hydroxylamine (about 70 ml) and adjusted to a concentration of ca. 0.9M H^+/1M H_3NOH^+. The solution was heated to boiling point to ensure that

Pu was reduced to Pu(III). Three c.c. of diethyl oxalate were added to the sample, while being mixed vigorously to bring about the homogeneous precipitation of $Nd_2(C_2O_4)_3$ carrying Pu(III). The precipitate was digested with HNO_3+$HClO_4$ to remove oxalate, and the solution was evaporated to dryness and converted to 1.2M HCl. NH_4OH was then added to form $Nd(OH)_3$ precipitate. The precipitate was dissolved in ca. 5 ml of 7.2M HNO_3, and the solution was passed through an anion column (AG1x8, 100-200 mesh) of 10-ml capacity. Subsequent to the passage of the sample solution, the column was washed with 7.2M HNO_3 followed by 9M HCl to remove impurities. Plutonium was then eluted down the column into a 5-ml Teflon beaker using 1.2M HCl.

These ion-exchange procedures described here were repeated using a micro-column of 0.5-ml capacity, with the final 1.2M HCl solution of ca. 500 μl dripped onto a Teflon pad with a depression in the center. The solution was evaporated to dryness and converted to a ca. 10-μl drop of 8M HNO_3. A nominally 150-μm diameter anion exchange resin bead was introduced into the 10 μl drop using a tungsten needle. After the pad was set on a vibration platform for several hours, the resin bead was removed and placed onto the mass spectrometer filament for isotopic analysis at the Battelle Pacific Northwest Laboratories.

3. RESULTS AND DISCUSSION

The activities of the $^{239,240}Pu$ and $^{240}Pu/^{239}Pu$ atom ratios determined by mass spectrometry (MS) are listed in Table 1. For comparison, the $^{239,240}Pu$ activities previously measured by alpha spectrometry (AS) are also listed. Although a generally good correspondence exists between the two sets of data (Figure 2) , the superiority of the MS data over the AS data is obvious. First, the potential for any uncertainties with the AS data is far greater than that with the MS data. Second, the MS data are systematically lower than the AS data, reflecting lower blank levels for the former. For one sample analyzed by AS (namely, the 14-15 cm depth interval), the data had to be nullified due to serious contamination. In short, the AS method requires much larger samples and poses greater difficulty in rendering clean chemistry.

The $^{240}Pu/^{239}Pu$ atom ratios exhibit the surprisingly large range of 0.04 to 0.221, with the highest ratio corresponding to the Pu peak at 26-28 cm. This finding is fully consistent with the historical trend according to which the highest $^{240}Pu/^{239}Pu$ ratios in the atmosphere of the Northern Hemisphere occurred in 1963 as a result of the most intensive high-yield tests in that year (Noshkin and Gatrousis, 1974). Furthermore, the increase in the $^{240}Pu/^{239}Pu$ ratio with time prior to 1963 also conforms with the increase in high-yield tests during that period. What are more intriguing are the low $^{240}Pu/^{239}Pu$ ratios of 0.04 at 2-3 cm and 0.06 at 11-12 cm which , based on ^{210}Pb chronology, represent the time horizons of 1982-83 and 1992-93, respectively (Huh *et al.*, 1996). Such unusually low ratios are typical of fuels reprocessing Pu (i.e., Pu extracted from spent fuels), and of results from low burn-up tests. Although Taiwan had made an attempt to develop nuclear weapons during the 1970s and 1980s, the plan was later abolished. Hence, the possibility of any disposal of fuels reprocessing Pu at Sun Moon Lake is non-existent. This means that it is more likely that such low $^{240}Pu/^{239}Pu$ ratios at Sun Moon Lake were derived from low burn-up tests. The most well known site for such tests is the Nevada test site (NTS), the fallout from which had the very low average $^{240}Pu/^{239}Pu$ ratio of

Table 1. Activities of ^{239,240}Pu and ^{240}Pu/^{239}Pu atom ratios in Sun Moon Lake sediments.

	^{239,240}Pu (dpm/kg)		^{240}Pu/^{239}Pu
Depth (cm)	by Alpha Spec.	by Mass Spec.	atom ratio
2-3	3.24+1.45	2.22+0.12	0.041+0.003
5-6		0.671+0.051	0.153+0.025
6-7	1.89±0.55		
8-9	3.44+1.09	1.52+0.08	0.152+0.017
9-10	2.10+0.45		
10-11	0.86+0.39		
11-12	1.76+0.45	1.67+0.04	0.060+0.005
12-13	2.30+0.73		
14-15	8.63+1.6	0.690+0.021	0.167+0.011
16-17	2.29+0.57		
17-18	4.82+1.17	3.08+0.07	0.173+0.009
18-19	9.03+1.06		
19-20	8.68+1.53		
20-22	6.65+0.62	4.87+0.07	0.167+0.005
22-24	19.0+2.5		
24-26	14.1+1.7		
26-28	35.1+3.3	29.9+0.3	0.221+0.005
28-30	13.2+1.2		
30-33	11.2+1.4		
33-36	4.29+0.64	3.59+0.06	0.191+0.007
36-39	3.58+0.64		
39-42	7.14+1.09		
42-45	0.34+0.34	0.243+0.033	0.163+0.046
45-48	3.73+0.78		
48-51	3.82+0.80		
54-57	1.39+0.80		

Alpha spectrometry data are from Huh *et al.* (1996).
Errors quoted are one standard deviation based on counting statistics.

0.03 (Beck and Krey, 1983; Hicks and Barr, 1984). Moreover, the influence of this was discerned thousands of kilometers away, such as in the North Atlantic (Buesseler *et al.*, 1985), the Gulf of Mexico (Scott *et al.*, 1983) and probably even in Greenland (Koide *et al.*, 1985). A 1953 NTS test showed that even though the debris was confined to the tropopause, the fallout reached Taiwan (Machta and Hubert, 1956). Despite this, a dilemma is raised in attributing the low ^{240}Pu/^{239}Pu ratios at Sun Moon Lake to the NTS fallout: the Nevada above-ground testing ceased in 1970, whereas the low ^{240}Pu/^{239}Pu ratios at Sun Moon Lake occurred after 1980.

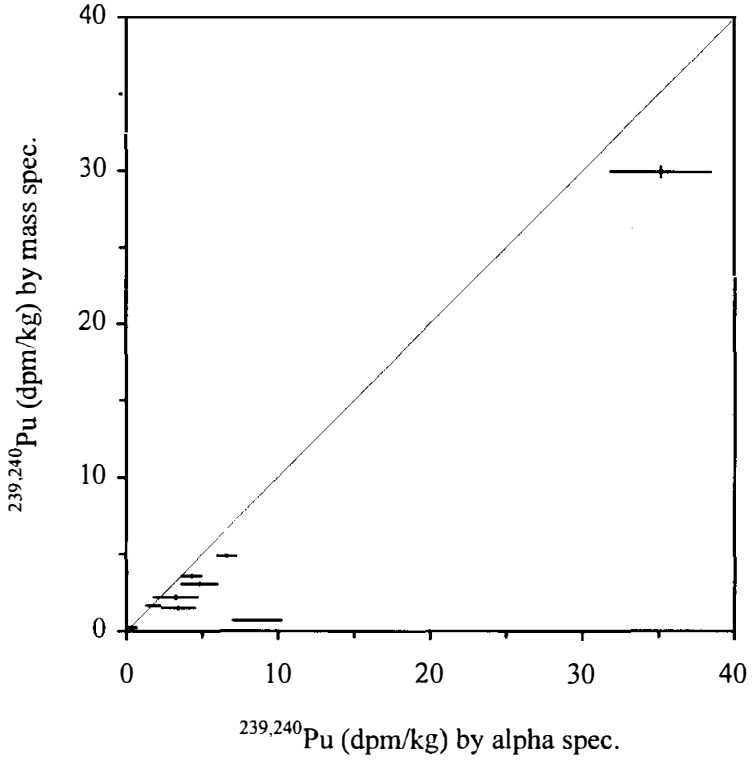

Fig. 2. Correlation between ^{239,240}Pu data measured by mass spectrometry and alpha spectrometry.

It follows that, with fuels reprocessing activities in Taiwan and the NTS fallout both ruled out, the most probable source that gave rise to the abnormally low ^{240}Pu/^{239}Pu ratios in the Sun Moon Lake sediments is related to Chinese nuclear tests. Since 1964, the People's Republic of China (PRC) has conducted 45 nuclear tests. Though the ^{240}Pu/^{239}Pu ratios from PRC tests were highly uncertain and rarely studied, they were in general much lower than the mean ratio of 0.184 from global fallout. It is in fact especially worth noting that the ^{240}Pu/^{239}Pu ratio in the debris from the fifth Chinese nuclear test of December 28, 1966, at Lop Nor, did not exceed 0.02 (Mamuro *et al.*, 1967) – the lowest ratio ever reported. That particular test had a TNT-equivalent yield of only several hundred kilotons (Carter and Moghissi, 1977), however, the fallout was transported by the Jet Stream and reached Northern California within five days (Potter *et al.*, 1967). Because of the southeastward extension of the Mongolian high pressure system, nuclear fallout from northwest China could have impacted Taiwan much more profoundly. It is highly possible that some atmospheric tests conducted by the PRC during the 1980s and 1990s led to similar low ^{240}Pu/^{239}Pu ratios. The debris could have been transported in the troposphere and carried to Taiwan, especially in the Monsoon season when the southern part of the Westerlies may have been entrained in the northeast winds. The occasional deposition in Taiwan of "mud rain" carrying loess from NW China attests to this transport mechanism (Lin, F., personal communication).

Other than the anomalous ratios discussed above, the ^{240}Pu/^{239}Pu ratios measured at other depths above 26 cm fall within 0.15-0.18, similar to the values observed in the stratosphere of

the Northern Hemisphere (Hardy, 1978). Therefore, as with most places around the world, global fallout is the dominant source of Pu in Taiwan most of the time.

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