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Distribution of ^{90}Sr and ^{137}Cs in Annual Tree Rings of Japanese Cedar, *Cryptomeria Japonica* D.Don.

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The contents of ^{90}Sr and ^{137}Cs in two samples of Japanese cedar from Takao and Tsukui districts were determined in tree rings cut into segments representing steps of 5 years of growth. ^{90}Sr in both cedar samples and ^{137}Cs in the Tsukui cedar sample were determined after ashing and chemical isolation, while ^{137}Cs in the Takao sample was directly determined from the sample ash. The distribution of ^{90}Sr fallout in tree rings suggests that ^{90}Sr had given a rather direct effect and showed no significant translocation from sapwood to heartwood, whereas ^{137}Cs tends to concentrate in heartwood irrespective of the effect of the fallout. Average contents of ^{90}Sr and ^{137}Cs were 22 and 9.4 pCi/kg in the Takao sample (9.61 kg air dried) and were 23 and 12 in the Tsukui (4.71 kg air dried) in 1982.

INTRODUCTION

Atmospheric nuclear bomb testing, as actively performed during the 1960's, added a large quantity of artificial radionuclides to the environment. The fallout has dropped to a very low level now, with the change from atmospheric to underground testing. However, the pollution of long half-life radionuclides such as ^{90}Sr(28.8y) and ^{137}Cs(30y) accumulated in the soil and then absorbed into plants with their nutrition continues to be a physical health problem. Only sparse data are available for the artificial radionuclide content in trees[1-4]. In the present study, we determined the concentrations of ^{90}Sr and ^{137}Cs in steps of 5 annual rings of Japanese cedar to obtain the distribution patterns and average contents in 1982. We also determined stable strontium and stable cesium, together with calcium, sodium, potassium and rubidium to reveal the behavior of the radionuclides in trees more clearly.

Furthermore, since ring formation is woody plants is due to the seasonal variation of cambium activity and each tree ring in the plant can be usually assigned to the respective year

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of it's growth, a certain element incorporated with nutrition from soil may record the history of its variations of soil content in tree rings during their long growing period. Therefore, the location and amount of such kinds of elements in the annual rings may give us an indication of the environmental variations, especially anthropogenic increases of certain elements to the growing circumstances of trees. There are some arguments[5-8] whether the tree-rings maintain the history of heavy metal pollution or not. Concentrations of heavy metals, e.g. lead, in the environment started to increase from the natural level prior to 1900's AD., whereas it is very clear that the radionuclide pollution started in 1945. Therefore distribution patterns of the radionuclides in the tree rings will give some suggestive answers to settle, whether woody plants maintain the change of pollutant level in their rings over a period of time and whether woody plants can be considered long-term monitors of the environmental change or not.

MATERIAL AND METHOD

Wood samples were obtained in September 1982 from Takao and Tsuki about 48 and 53 km west of central Tokyo, respectively (Fig. 1). Cross sectional wood disks from Takao (69-years-old) and Tsukui (53-years-old) were cut at a height between 0.5m and 1m from the ground. The wood disks were cut into small segment of 5 annual rings each, and air-dried. The bark was washed and air-dried. The weight of the samples prepared ranged from 0.2 to 1.0 kg.

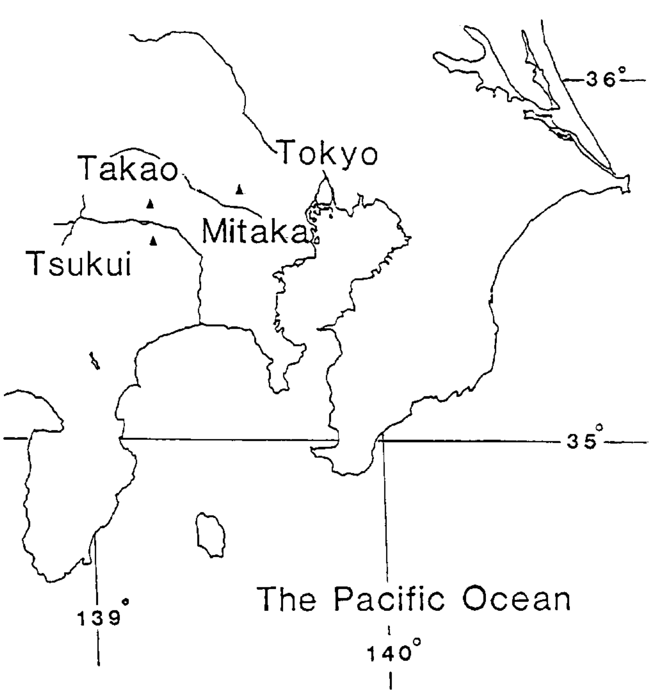

Fig. 1. Sampling sites.

Ash % of each section of wood was calculated after carbonization and ashing below 400°C.

Analytical procedures for ^{137}Cs, ^{90}Sr and other metals were as follows:

^{137}*Cs*: the gamma-ray photopeak at 662 keV was counted with 2.5-3.0g of ash (Takao) or cesium phosphomolybdate precipitate (Tsukui) by a NaI (T1) scintillation counter equipped with a Camberra 1024 channel pulse height analyzer for (2.5-3.0) x 10^4s.

^{90} *Sr*: after dissolution of 1.0-1.5g of ash with hydrofluoric acid, perchloric acid and nitric acid containing strontium carrier, ^{90}Sr was precipitated with the carrier by adding ammonium carbonate, isolated by ion-exchange chromatography, and left for at least 3 weeks until it reached the secular equilibrium with ^{90}Y. The ^{90}Y was then milked and measured with a low background "pico β" GM counter.

Stable-Cs and Rb: cesium and rubidium contents were determined by a conventional neutron activation method, 0.5 to 1.0g of ash sample being activated for 10 min. in JRR-4 (flux: 8×10^{13}n/cm^{2}s), Tokai Research Establishment, Japan Atomic Energy Research Institute. After a cooling for 2 weeks, ^{134}Cs and ^{86}Rb were measured by a Ge(Li) detector with a Nuclear Data 4096 channel pulse height analyzer.

Stable-Sr: stable strontium was determined by the neutron activation of the oxalate precipitate from a hydrochloric acid solution of about 0.3g of ash in JRR-4 for 3 min. After cooling for 1 hour, the ^{87m}Sr produced was measured. The recovery yield of strontium in a preparation step was obtained by adding ^{85}Sr yield tracer.

Na, K and Ca: about 1.5g of ash was dissolved in 100 ml of 0.1N HCl and these metals were determined with a Jarrell-Ash Model AA-1 Mark II atomic absorption spectrometer by utilizing the standard addition method.

RESULTS

Table 1 and 2 show the concentration of elements in tree rings of Japanese cedar taken from Takao and Tsukui, respectively, air-dried weight base being adopted. For the tree disks from Takao (69-years-old), the central section from 1914 to 1947 was heartwood, the darker-colored dead part, and the outer section from 1953 to 1982 was sapwood, the lighter-colored living part. Section from 1948 to 1952 contained both, since heartwood formation does not take place symmetrically within the rings. For the 53-years-old Tsukui sample, the section from 1930 to 1950 was heartwood, from 1961 to 1982 was sapwood, and from 1951 to 1960 contained both.

The highest contents of ^{90}Sr were in 1958-1962 and 1968-1972 sections for Takao, and in 1951-1955 sections for Tsukui, excluding the bark, while no ^{90}Sr was detected in sections before 1932 for Takao and before 1940 for Tsukui.

Stable strontium contents ranged from 4 to 10 μg/g in the Takao sample and from 4 to 8 μg/g in the Tsukui sample.

Calcium concentration ranged from 400 to 1600 and from 600 to 1200 μg/g in the Takao and Tsukui samples, respectively. Strontium and calcium, both alkaline-earth metals, seem to show the same behavior. ^{137}Cs concentration in heartwood was noticeably higher than in the

Table 1. Element Concentration in Tree Ring Sample: *Japanese Cedar* from Takao

year*	ash %	^{90}Sr (pCi/kg) airdry)	Sr	Ca	^{137}Cs (pCi/kg) airdry)	Cs (ng/g) airdry)	K	Na	Rb
			(μg/g airdry)				(μg/g airdry)		
1914-1922	1.2	ND	9.2 ± 1.3	900	11 ±2	3.0 ±0.3	4600	47	4.5 ±0.3
1923-1927	0.95	ND	6.1 ± 0.7	700	8.3 ±1.0	2.5 ±0.2	4100	57	2.5 ±0.2
1928-1932	1.1	ND	7.0 ± 1.0	780	17 ±2	3.2 ±0.2	4700	52	3.9 ±0.3
1933-1937	1.2	4.9± 3.3	6.5 ± 1.1	700	12 ±1	2.1 ±0.4	3700	51	2.7 ±0.2
1938-1942	0.93	11 ± 3	3.8 ± 0.6	500	12 ±1	3.0 ±0.2	3500	46	2.6 ±0.2
1943-1947	0.80	11 ± 5	4.5 ± 0.5	480	14 ±1	2.9 ±0.2	3700	38	2.7 ±0.2
1948-1952	0.59	12 ± 3	4.9 ± 0.5	430	8.5 ±0.9	2.1 ±0.4	2500	34	0.95 ±0.06
1953-1957	0.35	27 ± 4	5.4 ± 0.6	430	4.6 ±0.8	1.2 ±0.1	2000	19	0.78 ±0.06
1958-1962	0.60	45 ± 3	9.6 ± 1.6	880	5.8 ±1.0	1.7 ±0.4	1900	33	0.74 ±0.05
1963-1967	0.49	23 ± 3	8.1 ± 0.9	930	8.6 ±0.8	1.5 ±0.2	1500	36	0.89 ±0.07
1968-1972	0.45	39 ± 2	6.7 ± 0.7	720	5.3 ±0.8	1.0 ±0.2	1300	32	0.87 ±0.07
1973-1977	0.47	27 ± 3	8.2 ± 1.1	690	2.4 ±0.8	1.3 ±0.2	1400	37	0.96 ±0.08
1978-1982	0.83	40 ± 3	13 ± 2	1600	6.6 ±1.0	3.5 ±1.0	1900	42	0.81 ±0.05
bark	1.6	48 ±20	116 ±17	4200	15 ±2	17 ±2	1900	88	1.9 ±0.1

* Approximate year of growth of the tree ring as estimated by counting the distinctly visible rings in the tree section.

ND: 'Not Detectable' by the analytical procedures used.

Table 2. Element Concentration in Tree Ring Sample: *Japanese Cedar* from Tsukui

year*	ash %	^{90}Sr (pCi/kg) airdry)	Sr	Ca	^{137}Cs (pCi/kg airdry)	Cs (ng/g) airdry)	K	Na	Rb
			(μg/g airdry)				(μg/g airdry)		
1930-1935	0.93	ND	6.4±0.7	910	29 ± 6	5.1± 1.2	3500	38	3.1 ±0.2
1936-1940	0.84	ND	4.1±0.4	750	17 ± 3	4.4± 1.0	3100	34	2.7 ±0.2
1041-1945	0.81	11 ±4	4.9±0.5	700	19 ± 4	5.0± 1.1	3100	33	2.4 ±0.2
1946-1950	0.79	22 ±4	5.5±0.6	780	7.0 ± 2.1	2.1± 0.8	3200	32	2.1 ±0.2
1951-1955	0.58	30 ±2	3.4±0.4	640	8.4 ± 1.9	2.1± 0.7	2100	24	1.7 ±0.4
1956-1960	0.37	23 ±2	5.0±0.5	660	7.2 ± 2.1	1.4± 0.2	1000	17	0.88 ±0.07
1961-1965	0.36	23 ±5	5.8±0.7	780	11 ± 3	1.2± 0.4	1000	16	0.73 ±0.06
1966-1970	0.32	24 ±1	5.0±0.6	790	12 ± 2	0.88±0.14	790	15	0.56 ±0.05
1971-1975	0.33	23 ±1	7.0±0.8	1100	7.2 ± 1.9	1.1± 0.2	930	27	0.62 ±0.05
1976-1982	0.45	25 ±2	5.2±0.6	1200	12 ± 3	2.2± 0.3	960	21	0.73 ±0.07
bark	2.1	130 ±9	25 ±5	7200	38 ±10	25 ± 2	1800	97	1.9 ±0.2

* Approximate year of growth of the tree ring as estimated by counting the distinctly visible rings in the tree section.

ND: 'Not Detectable' by the analytical procedures used.

sapwood for both Takao and Tsukui samples. The stable cesium concentration in the heartwood was also higher than in the sapwood. Other alkali metals determined, sodium, potassium and rubidium, showed the same trend as cesium. The metal concentrations found in the latest, most outer, tree rings of both samples, 1978 to 1982 for Takao and 1976 to 1982 for Tsukui, were usually higher than those found in the older adjacent sapwood rings. Furthermore, the bark gave the highest values in ^{90}Sr, ^{137}Cs, sodium, calcium and strontium readings.

DISCUSSION

At first, atmospheric nuclear bomb testings closely related to the radioactive fallout in Japan are briefly summarized. The first nuclear bomb detonations were in 1945. The number of tests increased gradually from 1951 and reached about 100 a year in 1957, corresponding to 174 Mton of TNT for 1945-1958. From 1958 to September of 1961, nuclear testing was abolished for a short period, but then actively resumed again, going up to over 130 a year in 1962, estimated to be total 337 Mt TNT (1961-1962). From the late 1960' to the early 1970's, experiments of a few kt class nuclear bombs were carried out in China, and the last atmospheric testing, the Chinese 26th, was carried out in 1980. The change from atmospheric to under ground testing resulted in a gradual decrease in radioactive fallout, leaving two fallout peaks at around 1959 and 1963.

Specific activities of ^{90}Sr and ^{137}Cs obtained from the values in Table 1 and 2 are shown in Figs. 2-5 as a function of tree ring age, together with ^{90}Sr/Ca and ^{137}Cs/K ratios. Curves of

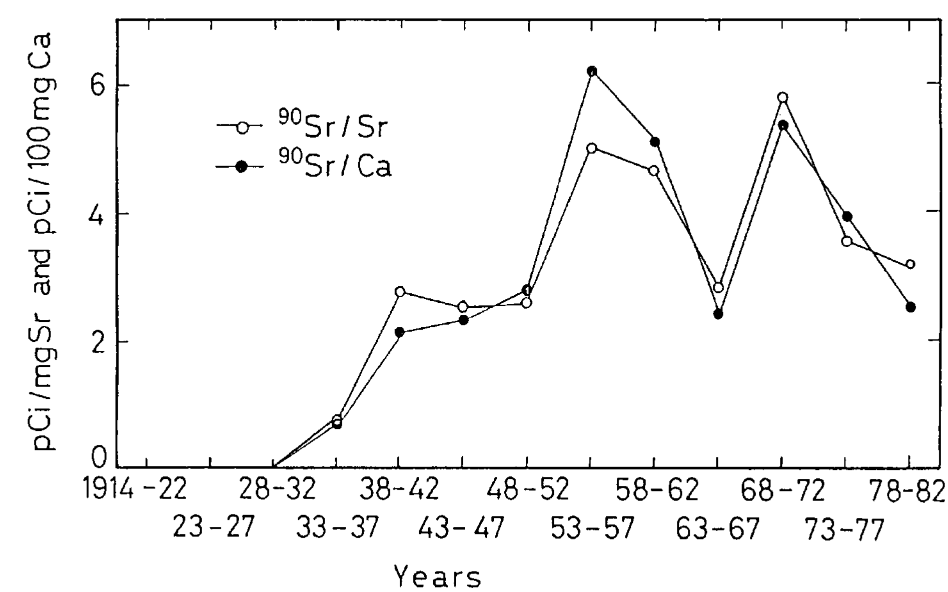

Fig. 2. Relations of ^{90}Sr/Sr and ^{90}Sr/Ca to tree ring age for Takao sample.

$^{90}Sr/Sr$ and $^{90}Sr/Ca$ (Fig. 2 and 4) are very similar to each other as was expected from a similar distribution pattern of strontium and calcium. Curves of $^{137}Cs/Cs$ and $^{137}Cs/K$ likewise resemble each other, as seen in Fig. 3 and 5. ^{137}Cs was detected through all the sections down to 1914 core for Takao and the 1930 core for Tsukui, giving rather smooth curves of the specific activities. The annual rings of Japanese cedar are generally quite distinguishable because of their relatively rapid annual growth, especially between spring and summer. They are of a very simple cross section, and hence the assignment of the age to each ring may be as accurate as ± 1 year. The Takao sample had the sapwood of about 30 years and the Tsukui sample had that of about 20 years. It may be reasonable to assume that the samples had had the sapwood of almost the same respective duration in 1959, the time that gave the first peak of annual ^{137}Cs deposition in and around Tokyo. The assumption leads to that, in 1959, Takao and Tsukui sample should have had the sapwood-heartwood boundary at 1930 and 1940, respectively. It is quite obvious, therefore, the ^{137}Cs was translocated into the heartwood across the sapwood-heartwood boundary. Katayama *et. al.*[2] reported very similar results and suggested that alkali metals may contribute much to the heartwood formation of this species. Brownridge[1] also showed the all-through distribution pattern of ^{137}Cs but no correlation between the advent of nuclear weapon testings and the concentration of ^{137}Cs in the xylem of five species in the U.S.A..

As for ^{90}Sr, a very clear contrast to ^{137}Cs was found; ^{90}Sr was not detected in the sections before 1932 for Takao and before 1940 for Tsukui, though ^{137}Cs was found all through the sections of both samples, as mentioned above. The fact, that no ^{90}Sr was detected in the sections of core through 1932 for Takao and the sections of core through 1940 for

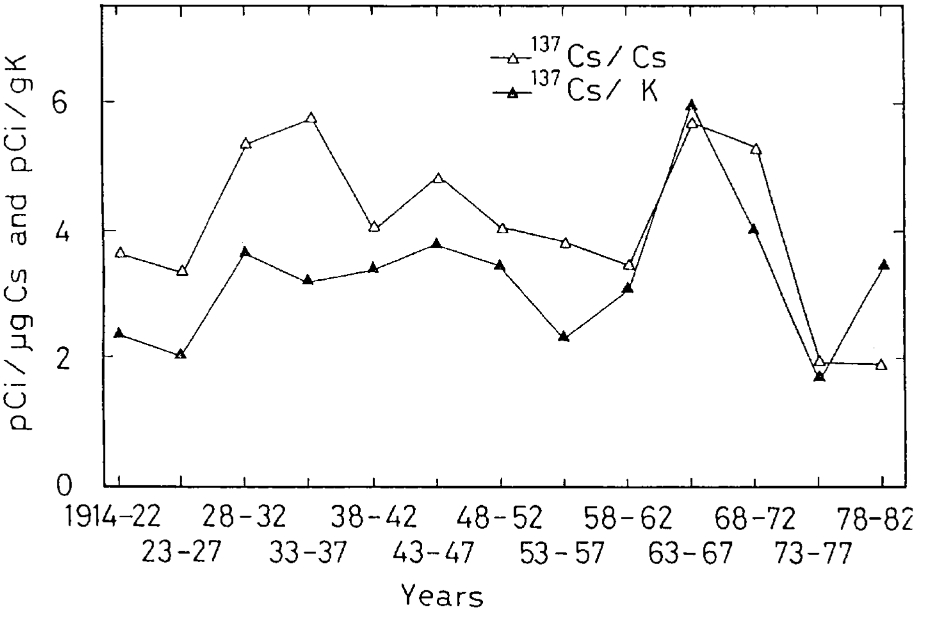

Fig. 3. Relations of $^{137}Cs/Cs$ and $^{137}Cs/K$ to tree ring age for Takao sample.

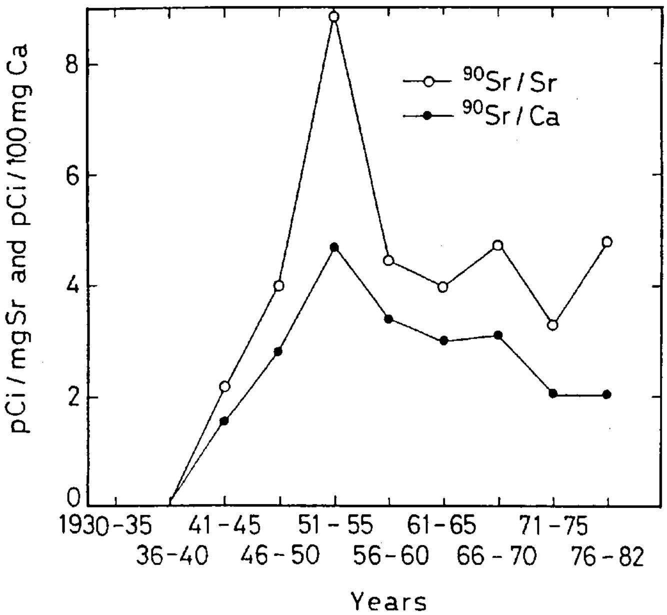

Fig. 4. Relations of $^{90}Sr/Sr$ and $^{90}Sr/Ca$ to tree ring age for Tsukui sample.

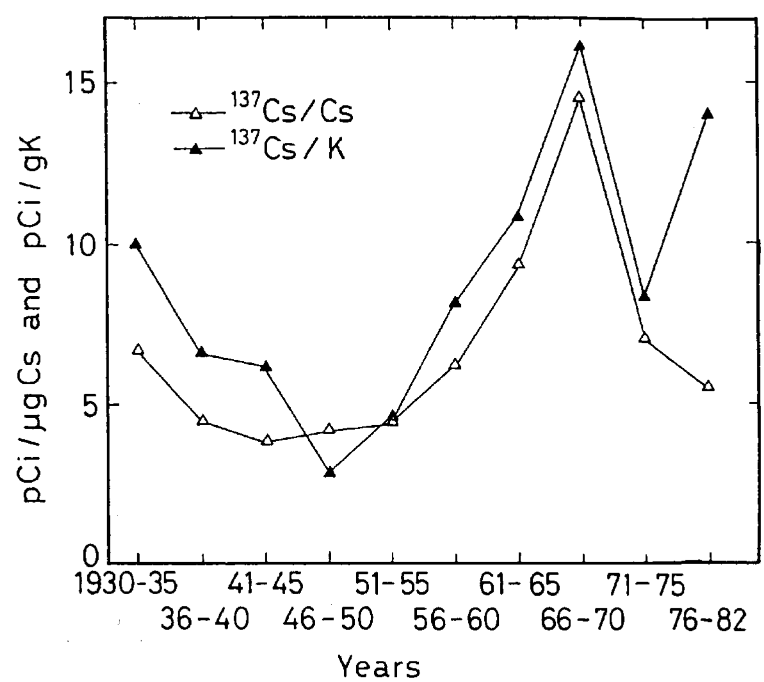

Fig. 5. Relations of $^{137}Cs/Cs$ and $^{137}Cs/K$ to tree ring age for Tsukui sample

Tsukui, suggests that no significant translocation of ^{90}Sr from sapwood into heartwood had taken place, because 1930 and 1940 are the estimated respective sapwood-heartwood boundary period in 1959. While the existence of ^{90}Sr in the sections of 1933-1947 for Takao and of 1941-1945 for Tsukui, i.e. the sections existing before the first nuclear bomb test, could be elucidated by the fact that ^{90}Sr seems to be absorbed or to pass through the living sapwood and become fixed to some extent, added to the alkaline earth elements fixed already at the time of sapwood cell formation.

Several peaks are found in the specific activity/year relations, as seen in Fig. 2-5. Some of them appear to coincide with the radioactive fallout peaks in the Tokyo area from a yearly viewpoint, but it could be said that only two examples, presented here, are not sufficient for a detailed discussion. We are continuing the experiments further, and will discuss the details, i.e., the correlations between the radioactivity peaks in trees and those in fallout, total inventory, radiation dose and the relationships of the specific activities in trees to those in soil in a separate paper. In conclusion, the average contents of ^{90}Sr and ^{137}Cs in both samples were calculated to be 22 and 9.4 for Takao and 23 and 12 pCi/kg for Tsukui, respectively, in 1982. It has hereby been concluded that strontium and other similar elements may record the environmental variation in the tree rings to the extent as mentioned above, but not for cesium and like elements, by using Japanese cedar.

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