

Content of Plutonium in River Water in Japan

by

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Abstract

Content of plutonium in the water of eight main rivers in Japan was determined. The average value of the total content of plutonium is 1.4×10^{-3} pCi/l in which 0.4×10^{-3} pCi/l is contained in the suspended matter. The annual run-off plutonium is only 0.12% of the accumulated plutonium on land. This suggests that the fallout plutonium is adsorbed on soil surface firmly and it is difficult to be leached out.

Fallout plutonium-239 derived from nuclear explosion is accumulated on the earth surface. Total deposition of $^{239,240}\text{Pu}$ during the period from 1958 to 1969 is estimated to be 0.97 mCi/km² in Tokyo (MIYAKE, KATSURAGI and SUGIMURA, 1968, 1970). In ocean water, $^{239,240}\text{Pu}$ was detected both in the Pacific and the Atlantic with the concentration ranging from 0.2 to 1.2×10^{-3} pCi/l (PILLAI, SMITH and FOLSOM, 1964; MIYAKE and SUGIMURA, 1968; BOWEN, WONG and NOSHKIN, 1971).

Up to now, little is known about the content of plutonium in river water. In 1966 and 1967, water samples were collected for plutonium analyses at eight main rivers in Japan. The names of the rivers are: Kitakami, Shinano, Kiso, Mogami, Tone, Yodo, Asahi (Honshu), Yoshino (Shikoku). The locations of these rivers and sampling sites for water samples are given in Fig. 1. The sampling site on each river was selected so as to avoid industrial and urban pollution as well as inflowing sea water.

1. Method of analysis

Each 1,000 liter sample was collected and the suspended matter was separated with membrane filters (Millipore filter, HA type, 0.45μ).

The method of analysis is briefly described below. Fallout plutonium isotopes and ^{238}Pu added as a yield tracer are precipitated with ferric hydroxide. Plutonium isotopes can be retained in a column of Dowex 1 after other nuclides and carriers are eluted using successive washes of 8 M HNO₃ and concentrated HCl. Plutonium is then eluted from the resin with NH₄I dissolved in concentrated HCl and it is finally electroplated on the stainless steel plate.

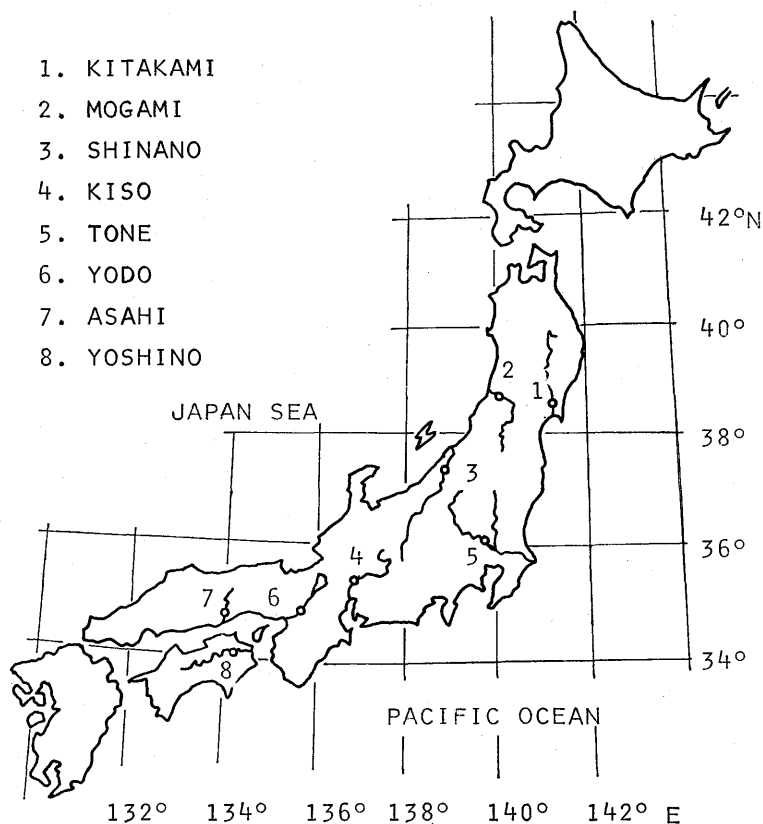


Fig. 1. Location and water sampling site of rivers in Japan.

Measurement of alpha-ray intensity of $^{239,240}\text{Pu}$ (5.15 Mev) and ^{238}Pu (5.75 Mev) is done with an alpha-ray spectrometer which consists of a silicon solid state detector and a multichannel pulse-height analyzer.

2. Results and discussion

The results of analyses are shown in Table 1. The average value of the total content of plutonium in river water in Japan is 1.4×10^{-3} pCi/l in which 0.4×10^{-3} pCi/l is contained in the suspended matter. The above value is much less than the mean content of plutonium in rain water of 3×10^{-2} pCi/l in the years from 1966 to 1967 which was observed in Tokyo (MIYAKE, KATSURAGI and SUGIMURA, 1968, 1970).

As reported previously by MIYAKE and TSUBOTA (1963), in case of fallout ^{137}Cs and ^{90}Sr it is difficult to remove them from the soil after they have fallen on the earth. These nuclides are adsorbed tightly on the soil surface and are hardly leached out by surface water.

According to MIYAKE and TSUBOTA (1963) the rate of leaching of ^{90}Sr from the soil is 1.5% annually while that of ^{137}Cs is almost negligible. On the basis of these

Table 1. The results of analyses of $^{239,240}\text{Pu}$ in Japanese rivers.

Name of rivers	Date of sampling	Total amount of water flow at sampling site	Surface area of drainage	$^{239,240}\text{Pu}$	
				Soluble	Suspended
		($\times 10^9 \text{m}^3/\text{yr}$)	($\times 10^3 \text{km}^2$)	($\times 10^{-3} \text{pCi/l}$)	
Kitakami	Aug. 1966	10.3	7.9	0.68 ± 0.07	0.57 ± 0.06
Mogami	Aug. 1966	11.2	6.1	1.80 ± 0.10	0.57 ± 0.06
Shinano	Oct. 1966	16.2	10.1	0.23 ± 0.02	0.38 ± 0.03
Kiso	Oct. 1966	11.7	4.9	0.51 ± 0.05	0.29 ± 0.02
Tone	Nov. 1966	9.1	8.6	4.16 ± 0.20	0.21 ± 0.02
Yodo	Aug. 1967	10.1	7.2	0.07 ± 0.01	0.13 ± 0.01
Asahi	Aug. 1967	1.6	1.6	0.26 ± 0.02	0.23 ± 0.02
Yoshino	Aug. 1967	5.5	2.8	0.51 ± 0.05	0.33 ± 0.03
weighted mean				1.04	0.37

observations, they concluded that the direct contribution of rain water to the river water was about 1.7%.

Assuming that 2% of river water is derived from direct falling of rain drops on the rivers, the contribution of plutonium is 0.6×10^{-3} pCi/l which is about 40% of the plutonium content in river water. From this calculation, it can be assumed that about 0.8×10^{-3} pCi/l of plutonium in river water is derived from the drainage from the land.

It is known that the drainage surface area of these rivers is $49 \times 10^3 \text{ km}^2$ in total and the total amount of water flow is $76 \times 10^9 \text{ m}^3$ per year. Assuming that the accumulated amount of plutonium is 1 mCi/km^2 , the total amount of plutonium on land in the drainage area is estimated to be 49 Ci, while the amount of the annual flow of plutonium is 0.061 Ci which is 0.12% of the accumulated plutonium on land.

The above result of calculation suggests that the fallout plutonium is adsorbed on the soil surface firmly and it is difficult to be leached out. This means that the effect of the fallout plutonium, if any, will last longer in future.

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本邦河川水中のプルトニウム含量

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核実験により大気圏に放出されたプルトニウム-239は、地上に降下し、蓄積される。東京における全降水量は約 1 mCi/km^2 である。これが土壤に吸着されて長く地上にとどまるか、あるいは容易に溶脱されて河川水により海へ運ばれるかを明らかにすることは、将来の環境の質を予測するために重要である。

1966年から1967年にかけて、日本の主要8河川（北上川、信濃川、木曾川、最上川、利根川、淀川、旭川、吉野川）の河川水の採水を行ない、溶存および、懸濁状のプルトニウムを、 α 線波高分析法によって測定した。

8河川の河川水の平均全プルトニウム含量は、 $1.4 \times 10^{-8} \text{ pCi/l}$ で、このうち、 $0.4 \times 10^{-8} \text{ pCi/l}$ は、懸濁状であった。この結果と、降水中のプルトニウムの含量を用いて計算すると、地上に蓄積されたプルトニウムのうち、河川水によって流亡するものの割合は、年々0.12%にしか相当せず、地上に落下したプルトニウムは、長く地上に留まることが明らかになった。