研究論文

152. *Analysis of Airborne Radioactivity in Japan due to Nuclear Testings*

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The level of surface airborne radioactivity in Tokyo, Japan during Apr. 1959 ~ June 1961, was investigated. In spring 1961 and 1960, a maximum peak of activity was detected with an increase of 4~5 times of the background. The half elimination time of the storage above tropopause is estimated about 300~400 days. From the γ -spectrum of the sample collected in Mar. 1959, ^{144}Ce - ^{144}Pr , ^{106}Ru - ^{106}Rh , ^{137}Cs - $^{137\text{m}}\text{Ba}$ were estimated. From the sample collected during Feb.~July 1960, ^{144}Ce - ^{144}Pr , ^{106}Ru - ^{106}Rh , ^{95}Nb , ^{103}Ru were detected, which might be ascribed to the French nuclear test effect. The cascade-sum effect of well-type scintillator was taken into consideration in estimating the crystal peak efficiency for γ -spectrum assaying.

INTRODUCTION

After the suspension of large scale nuclear testings, the level of atmospheric radioactivity has generally decreased. During the period without any major disturbances due to nuclear testings, the level and the composition of surface airborne radioactivity were investigated.

I. THE LEVEL OF ATMOSPHERIC RADIOACTIVITY

The surface airborne dust was sampled by a wet-type electrostatic dust collector located at Kokubunji** in Tokyo, Japan. The dust suspended in water was collected on the filter paper and the paper with dust was ashed and then measured by G-M counter or shielded gas-flow counter at 2 or 3 days after sampling. The results are summarized in Fig. 1. Each point indicates a datum of the sample collected with about $6 \times 10^3 \text{ m}^3/\text{hr}$ of air, and the sampling time 1~2 hr.

The curved line was drawn by taking off the fluctuation due to weather effects. And then the dotted line indicates the corrected result for the radioactive decay⁽¹⁾, assuming that the material is chiefly composed of 2~3 y aged fission products from the nuclear-test during 1957~1958. On the dotted line, the

seasonal variations can be seen. Especially the spring maximum in 1961 was detected clearly with an increase of about 4 or 5 times, the same extent as the one in 1960⁽²⁾.

In the base of these variations, a logarithmic decrease was observed to continue. This decreasing tendency may be considered to correspond to the decrease of the total amount of radioactive storage above the tropopause. From the difference of the level between two maxima in 1960 and 1961, the half elimination time may be estimated to be about 300~400 days.

II. γ -RAY SPECTRUM ANALYSIS

The γ -ray spectra of the collected dust are shown in Figs. 2 and 3. The assaying procedures of the spectrum peaks, in comparison with reasonable aged fission product nuclei are listed in Tables 1 and 2, and the results are shown in Tables 3 and 4.

Sample (A) was collected in Mar. 1959, when the activity level was in maximum increase during the nuclear test cessation, and the collected material was evaporated completely dry. Sample (B) was collected during

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Fig. 1 The results of measurement of surface airborne activity during the period spring 1959~spring 1961 by β -counting

Each point indicates one measurement.

The curved line shows smoothin gout for weather effect.

The dotted line indicates the corrected result for gross radioactive decay⁽¹⁾.

The spring maxima in 1960 and 1961 are clearly seen.

Feb.~July in 1960, at the probable time of appearance of the spring maximum. Early in the period the French nuclear test was occasionally conducted. The collected dust was treated radio-chemically by leaching with HCl and the co-precipitation with $\text{Ce}(\text{OH})_3$ and $\text{Fe}(\text{OH})_3$.

In use of the nuclide emitting more than one photon, the coincidence of all its S values in Tables 3 and 4 is considered as an evidence for the existence of such nuclide. However, actually, because of the error accruing to the peak counting rate C from the spectrum, some extent of deviation in coincidence must be considered. For mono photon emitting nucleus, no sufficient assaying method can be found only from γ -ray spectrum. But if the corresponding peak is clear without any other probable complications due to other sources, we admitted the existence of the corresponding nuclide.

As a result of these analysis, we decided that in Sample (A), ^{144}Ce - ^{144}Pr existed, and ^{106}Ru - ^{106}Rh , ^{137}Cs - ^{137m}Ba probably existed, and in Sample (B), ^{144}Ce - ^{144}Pr , ^{106}Ru - ^{106}Rh existed, and ^{95}Nb , ^{103}Ru also probably existed. The probable effect of the French nuclear test in Sample (B) is taken into consideration in the 200 days component in η column in Table 2, with a reduction by a factor of ten considering the difference in contribution between the old (before 1958) and the new (spring 1960) nuclear tests, for example, detonation scale, the mode of spreading, decaying time *etc.*

The lack of ^{137}Cs - ^{137m}Ba peak in Sample (B) spectrum is probably due to the radiochemical treatment with the alkalimetal fraction might be lost.

The estimated nuclei printed in bracket in Tables 1 and 2, are the natural radioactive elements originated in natural soil dust.

In Sample (B) the existence of ^{95}Nb and ^{103}Ru might be attributed to the effect of the French nuclear test, and on the other hand the sufficient existence of ^{106}Ru - ^{106}Rh indicated that the spring increase in 1960 is chiefly due to the old nuclear testings.

For the measurement, the $\text{NaI}(3''\phi \times 3''h)$ well-type scintillation counter shielded by Pb 10 cm + Fe 2 cm thick layer, and 128 channel pulse height analyzer were used.

γ -ray scintillation spectrum of surface airborne dust Sample (A), collected in Tokyo, Japan, on the 3rd Mar. 1959 from $1.3 \times 10^4 \text{ m}^3$ of air as measured in Oct. 1960, with the total amount of collected material.

Fig. 2

Table 1 γ -spectrum assaying data for Sample (A) in Fig. 1

Spectrum peak (MeV)	Peak counts <i>C</i>	Estimated nucleus	γ -ray energy (MeV)	Abundance in F.P. ⁽¹⁾ η at 2y	Photon emiss. prob. ⁽³⁾⁽⁴⁾ σ (%)	Scinti. peak eff. ⁽⁵⁾ ϵ	Peak factor $H \equiv \eta \sigma \epsilon$	Peak f. to spectrum ratio ; S value $S \equiv H/C$ ($\times 10^3$)
0.12	23,000	^{147}Pm ^{144}Ce	0.121 0.134	13.5 30	0.5 10	1.00 0.99	6.8 300	$\left. \begin{matrix} 0.3 \\ 13 \end{matrix} \right\} 13$
0.32	2,500	(Ra C'')	0.3		50			
		(Ra B)	0.35					
0.42	4,700	(An)	0.4		5			
		(Ac B)	0.425		6			
0.52	8,000	e^+ ^{106}Rh	0.51 0.513	3.5	10	0.48 0.33	12	$\left. \begin{matrix} \alpha \\ 1.5 \end{matrix} \right\} 2 + \alpha$
0.66	26,000	^{106}Rh $^{137\text{m}}\text{Ba}$ ^{144}Pr	0.624 0.662 0.695	3.5 4 30	5.3 99 2	0.19 0.30 0.26	3.5 120 16	$\left. \begin{matrix} 1.3 \\ 4.6 \\ 0.6 \end{matrix} \right\} 7$

0.85	4,000	(Ac B) (Ac C'') (Ac C')	0.829 0.87 0.89		13 1 1				
1.02	120	e^+ ^{106}Rh	1.02 1.045	3.5	0.8	0.17 0.11	0.31	$\alpha' \}$ 2.6	$3 + \alpha'$
1.19	2,900	^{106}Rh	1.137	3.5	5.3	0.13	2.5	0.9	1
1.41	240	^{144}Pr	1.48	30	1	0.10	3	12	12
1.56	120	^{106}Rh	1.55	3.5	0.3	0.33	0.34	2.8	3
1.84	62	^{90}Y ^{106}Rh	1.75 1.77	5.2 3.5	0.02 0.1	0.12 0.12	0.01 0.04	0.2 0.7	1
2.23	470	^{144}Pr	2.18	30	1	0.13	3.9	8.3	8

C is the counts of each peak area in the spectrum of Sample (A) on Fig. 1.

η at 2 y shows the activity abundance ratio in the 2 y old fission products.

σ is each γ -photon emission probability of the nucleus containing conversion effect.

ε indicates the intrinsic peak efficiency of 3 cm R NaI⁽⁶⁾ crystal (real size was 3" ϕ $\times$ 3" h well-type), with the cascade-sum effect correction.

γ -ray scintillation spectrum of surface airborne dust Sample (B) collected in Tokyo, Japan during Feb.~July 1960, as measured in Oct. 1960, after radiochemical treatment.

Fig. 3

Table 2 γ -spectrum assaying data for Sample (B) in Fig. 2

Spectrum peak (MeV)	Peak counts C	Estimated nucleus	γ -ray energy (MeV)	Abundance in F.P. ⁽¹⁾ η		Photon emiss. prob. ^{(3) (4)} σ (%)	Scinti. peak eff. ⁽⁵⁾ ϵ	Peak factor $H \equiv \eta \sigma \epsilon$	Peak f. to spectrum ratio ; S value $S \equiv H/C$ ($\times 10^3$)	
				at 2y	at 200 d					
0.12	53,000	¹⁴⁷ Pm	0.12	13.5		0.5	1	6.8	} 5.9 6	
		¹⁴⁴ Ce	0.134	30		10	0.99	300		
		¹⁴¹ Ce	0.145		0.2	30	0.99	6		
0.23	6,600	⁹⁵ Zr	0.235		1.4	3	0.86	3.7	5.6 } $\bar{6} + \beta$ β	
		(Th B)	0.239							
0.33	3,200	¹⁴⁰ La (Ra B+C'')	0.328 0.3 0.35		<i>f</i>	17	0.67	11 <i>f</i>	3.4 <i>f</i> 3 <i>f</i>	
0.49	2,200	¹⁴⁰ La	0.49		<i>f</i>	21	0.43	9 <i>f</i>	4.1 <i>f</i> 4 <i>f</i>	
		¹⁰³ Ru	0.498		0.4	90	0.42	14	6.4 6	
0.52	1,900	e ⁺	0.51				0.48		α } 6.3 } 6 + α	
		¹⁰⁶ Rh	0.513	3.5		10	0.33	12		
0.64	1,100	¹⁰³ Ru	0.61		0.4	6	0.33	0.7	0.6 } 3.2 } 4	
		¹⁰⁶ Rh	0.624	3.5		5.3	0.19	3.5		
		^{137m} Ba	0.662	4		99	0.3	120		110
0.73	1,700	¹⁴⁴ Pr	0.695	30		2	0.26	16	9.4 } 21 } 30	
		⁹⁵ Zr	{ 0.724	}	1.4	49	0.27	36		
			{ 0.757			49	0.25			
0.79	1,400	⁹⁵ Nb	0.768		2.5	100	0.24	60	43 40	
		¹⁴⁰ La	0.815		<i>f</i>	19	0.23	4.4 <i>f</i>		3.1 <i>f</i> 3 <i>f</i>
0.87	1,300	(Ac B)	0.829			13			} α' }	
		(AcC'+C'')	0.87 0.89			2				
1.11	70	e ⁺	1.02				0.17		α' } 4.3 } 4 + α'	
		¹⁰⁶ Rh	1.045	3.5		0.8	0.11	0.31		
1.18	550	¹⁰⁶ Rh	1.137	3.5		5.3	0.13	2.5	} 4.7 5	
		⁹¹ Y	1.19		1	0.4	0.17	0.07		
1.55	270	¹⁴⁴ Pr	1.48	30		1	0.1	3	} 12 12	
		¹⁰⁶ Rh	1.55	3.5		0.3	0.33	0.34		
1.64	170	¹⁴⁰ La	1.60		<i>f</i>	41	0.13	5.3 <i>f</i>	31 <i>f</i> 30 <i>f</i>	
		¹⁰⁶ Rh	1.77	3.5		0.1	0.12	0.04	2.4 2	
2.27	490	¹⁴⁴ Pr	2.18	30		1	0.13	3.9	8 8	

The column signatures are the same as in Table 1.

η at 200 d is the η value for 200 d old fission products, with a reduction by a factor of ten due to the difference in contribution of the different tests.

f in the column of η at 200 d expresses a trace amount.

Table 3 γ -spectrum assaying result for Sample(A)

Estimated nucleus	γ -ray energy	<i>S</i> -value	Accompanying nucleus	Peak clarity
¹⁴⁴ Ce	0.134	13	¹⁴⁷ Pm	○
¹⁴⁴ Pr	0.695		^{137m} Ba	
	1.48	12	None	
	2.18	8	None	○
⁶ Ru- ¹⁰⁶ Rh	0.513	2 + α	e ⁺	
	0.624		^{137m} Ba	
	1.045	3 + α'	e ⁺	
	1.137	1	None	
	1.555	3	None	
	1.77	1	⁹⁰ Y	
Cs- ^{137m} Ba	0.662	7	¹⁰⁶ Rh ¹⁴⁴ Pr	○

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Table 4 γ -spectrum assaying result for Sample (B)

Estimated nucleus	γ -ray energy	S-value	Accompanying nucleus	Peak clarity
^{95}Zr	0.235	$6 + \beta$	Th B	
	0.724	} 30	^{144}Pr	
	0.757			
^{95}Nb	0.768	40 ¹	None	○
^{144}Ce	0.134	6	^{147}Pm ^{141}Ce	
^{144}Pr	0.695	(10)	^{95}Zr	
	1.48	12	^{106}Rh	
	2.18	8	None	
^{106}Ru - ^{106}Rh	0.513	$6 + \alpha$	e^+	
	0.624	4	^{103}Ru ^{137m}Ba	
	1.045	$4 + \alpha'$	e^+	
	1.137	5	^{91}Y	
	1.55		^{144}Pr	
	1.77	2	None	
^{140}La	0.328	3 f	Ra B+C''	
	0.49	4 f	^{103}Ru	
	0.815	3 f	^{95}Nb	
	1.60	30 f	^{106}Rh	
^{103}Ru	0.498	6	None	○
	0.61		^{106}Rh	
^{137}Cs - ^{137m}Ba	0.662	110	^{106}Rh	

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核実験による大気中放射能の解析

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大気中塵埃放射能の濃度を東京国分寺で1959年4月から1961年6月まで測定した結果、60年および61年の春季増大(約4~5倍)を検出した。圏界面上放射能蓄積量の拡散降下による減少半減期は300~400日程度と得られた。 γ 線スペクトル測定解析によって、1959年3月採集の試料から ^{144}Ce - ^{144}Pr 、 ^{106}Ru - ^{106}Rh 、 ^{137}Cs - ^{137m}Ba を、1960年2月~7月採集の試料から ^{144}Ce - ^{144}Pr 、 ^{106}Ru - ^{106}Rh 、 ^{95}Nb 、 ^{103}Ru を検出した。

スペクトル解析に用いたシンチレータピーク効率にはウェル型 NaI の cascade-sum 効果を考慮した。

正 誤 表 (Errata) Vol. 3, No. 7

		誤		正
p. 524 (28)	右段(8)式	$= -k_4(T_f - T_c)$	を	$= -k_4(T_{f'} - T_c)$
p. 528 (32)	左段(15)式および 右段(16)式の右辺	$= \frac{n_0 \delta \bar{T}_c(s)}{\delta n(s)} \times$	を	$= \frac{n_0 \delta \bar{T}_c(s)}{\delta n(s)} -$
p. 530 (34)	左段下から8行目	おては	を	おいては