

STUDIES ON IN-SITU MEASUREMENT OF FALLOUT NUCLIDES DERIVED FROM THE CHERNOBYL REACTOR ACCIDENT USING HIGH-PURITY GERMANIUM DETECTOR

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1. Introduction

Fallout nuclides derived from the Chernobyl reactor accident (from now on CRA) in April 26 of 1986, were detected at Matsue in Shimane Prefecture (SW Japan located about 7,800 km East from Chernobyl reactor site) in rain water and airborne particles collected in May 4 (local time). The Shimane Prefectural Institute for Public Health and Environmental Science (SPI) started monitoring of the effect of CRA mainly by non-destructive gamma-ray spectrometry of various environmental samples collected from entire area in Shimane. Details of this observation will be given in the Science Report of SPI¹⁾. Besides the laboratory measurement of the samples collected, in-situ gamma spectrometry using high purity Ge detector was performed from May 9 to early July. This paper describes the results of in-situ measurement of the effects of CRA.

2. Measurement and Data Analysis

Two portable Ge detectors were used for in-situ measurement of gamma-ray spectrum. One is a Ge detector with efficiency of 40.5% relative to a 3"x3" NaI(Tl) detector and 1.77 keV of energy resolution (FWHM) at 1.33 MeV, and the other is a Ge detector with relative efficiency of 7.2 %. Techniques of data analysis of in-situ measurement for obtaining dose rate and/or accumulation of fallout nuclides is almost the same as the method developed by Beck et al²⁾. The Ge detector was set facing down 1 m above the ground surface, and gamma-ray spectrum was measured for 1 to 2 hours. To compare the result obtained by in-situ measurement with those by soil sampling method, surface soils of 0 - 21 cm were collected at 3 cm intervals just below the Ge detector.

About 200 measurements were made from May 9 to early July. Gamma-ray spectra were analyzed by an automatic peak search program. The dose rates for the fallout nuclides derived from CRA together with those for natural radionuclides belonging to the U- and Th-series and K-40 were calculated by multiplying the count rate of gamma-ray peaks with conversion factors obtained for each Ge detector. Depth profile of Cs-137 accumulated in surface soil before CRA was simulated by an exponential function given by $A_x = A_0 \exp(-\alpha x)$, where A_0 and A_x are the Cs-137 activity at soil surface and depth x , respectively. The α is a fitting parameter of depth profile. The α value obtained by the analysis of the soil samples ranged from 0.4 to 0.9 cm^{-1} . Detail of the calibration method of the Ge detectors will be described elsewhere¹⁾. The density of surface soil was measured to be 1.5 - 1.6 g/cm^3 , therefore, 1.6 g/cm^3 was adopted in the calculation of conversion factors.

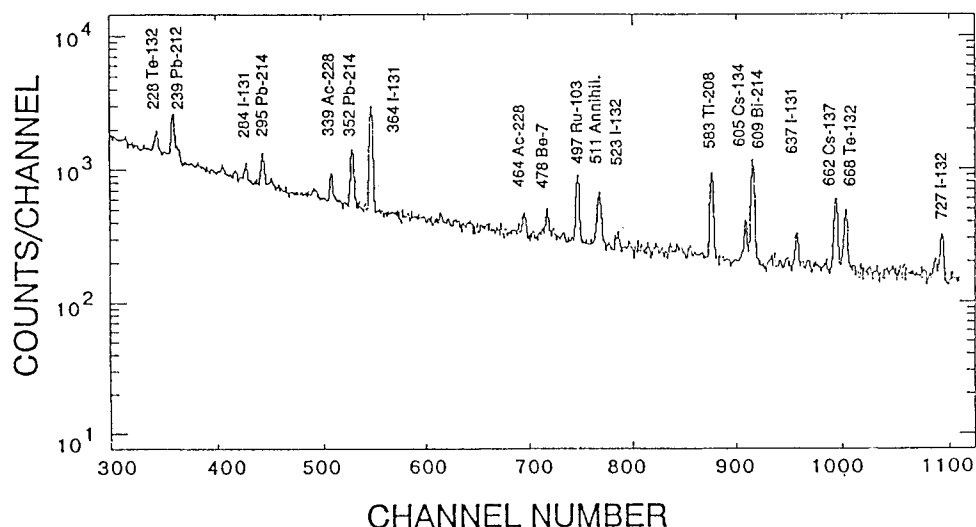


Fig. 1. In-situ gamma-ray spectrum of 200 - 700 keV region measured at 12:00 to 14:00 of May 9, 1986. Measuring time and gain setting were 120 min and 0.667 keV/ch, respectively.

3. Results and Discussion

In-situ gamma-ray spectrum obtained in May 9, 1986, is shown in Fig. 1. Fallout nuclides identified are Te-132 (228, 668 keV), I-131 (284, 364, 637 keV), Ru-103 (497 keV), I-132 (523, 727 keV), Cs-134 (605, 795 keV) and Cs-137 (662 keV). Results of analyses of In-situ measurements are summarized in Tables 1, and 2, and the time variations of dose (exposure) rates of Cs-137, I-131 and Ru-103 are shown in Figs. 2 to 4.

[Cs-137] Dose rate and/or accumulation of Cs-137 was calculated by assuming the depth profile of Cs-137 being exponential and/or exponential + surface distribution before and after CRA, respectively. Contribution of nuclear weapons tests is shown by a dotted line in Fig. 2. Accumulation of Cs-137 at the measuring point is order of magnitude lower than the value measured at typical uncultivated area in Shimane, because the construction of the SPI building begun in 1975. Accumulation of Cs-137 in surface soil begun since that year. As known from Fig. 2, dose rate has increased from 0.15 to 0.32 nGy/h by CRA. Appreciable increase of dose rate was not found after the initial increase during early May, because most of Cs-137 had fallen in a short time during this period. If in-situ measurements were made at uncultivated area with high amount of Cs-137, such a small increase of dose rate could not precisely be evaluated by in-situ measurement. Accumulation of Cs-137 estimated by in-situ measurement agreed well with soil sampling method within 15 %. Increase of Cs-137 accumulation by CRA was estimated to be about 70 MBq/km² at Matsue.

Table 1. Dose and accumulation of Cs-137 before and after CRA.

Measured Date	Dose (nGy/h)		Accumulation (MBq/sq.km)		Depth profile used in the calculation
	Average	Range	Average	Range	
Before CRA	0.156	[0.119 - 0.186]	110	[87 - 123]	Exponential
After CRA	0.324	[0.257 - 0.366]	180	[153 - 196]	Exponential + Surface

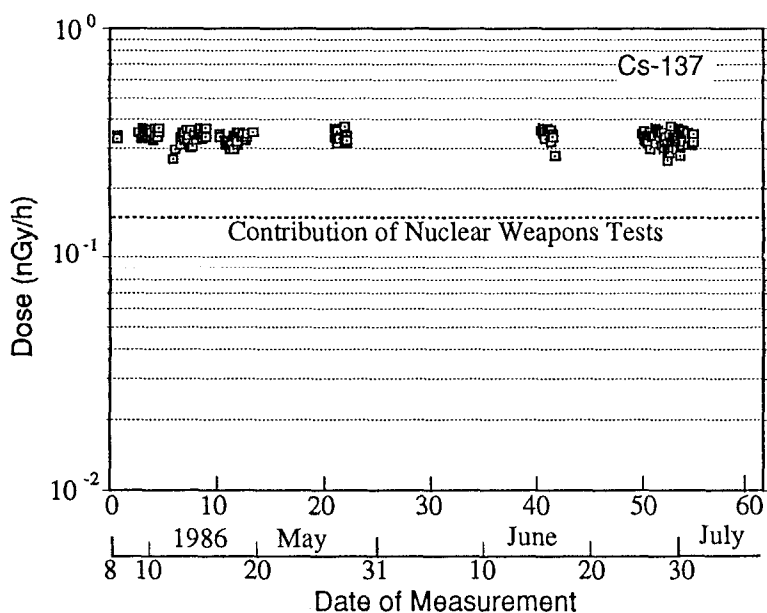


Fig. 2. Variation of dose rate due to fallout Cs-137 measured at Matsue.

[I-131] Dose rate and accumulation of I-131 at May 9 were calculated to be 0.918 nGy/h and 536 MBq/km², respectively, by assuming surface distribution ($\alpha = \infty$) on the ground. As shown in Fig. 3, apparent half-life of I-131 in an early stage (May 9 to 20) was evaluated to be 12.8 days, however, after this period, it became 7.9 days, which corresponds to the physical half-life of 8.06 d. Concentration of I-131 in the surface air measured by high volume air-sampler showed two peaks in May 8 (before the beginning of in-situ measurement) and May 13 (1/4 of that observed in May 8). Longer apparent half-life of I-131 in early stage might be explained by the delay of the deposition of I-131 (compared with other fallout nuclides), because significant fraction of I-131 was found to be in gaseous state as evidenced by the sampling with charcoal filter, and partly by the contribution of second plume observed in May 13. Integral dose due to I-131 during May 8 (when maximum air concentration was observed) through the beginning of July was estimated to be 338 nGy.

[Ru-103] As shown in Fig. 4, apparent half-life of Ru-103 was calculated to be 36.8 days, which is a little shorter than the physical half-life of 39.6 days. This may be explained by the downward migration of Ru-103 due to the rain fall during this period. Dose rate and accumulation of Ru-103 estimated for May 9 are 0.305 nGy/h and 140 MBq/km², respectively. Integral dose due to Ru-103 was calculated to be 436 nGy, which is little higher than that of I-131.

Dose rate due to Cs-137 measured after 8 months of CRA was 0.28 nGy/h, which is 4/5 of that measured at Ibaraki Prefecture (0.35 nGy/h), located 700 km East of Shimane.

Table 2. Dose, accumulation, integral dose and apparent half-life of fallout nuclides.

Nuclide	May 9, 1986		Integral Dose (nGy)	Apparent Half-life
	(nGy/h)	(MBq/sq.km)		
Cs-137	0.318	177	0.32 x hour	>1000d
I-131	0.918	536	338	12.8 d (until May 20) 7.9 d (after May 20)
Ru-103	0.305	140	436	36.7 d

References

- 1. K. Terai et al. Ann. Rep. Shimane Pref. Inst. Public Health and Env. Sci., in preparation.
- 2. H. L. Beck et al, HASL 258 (1972).

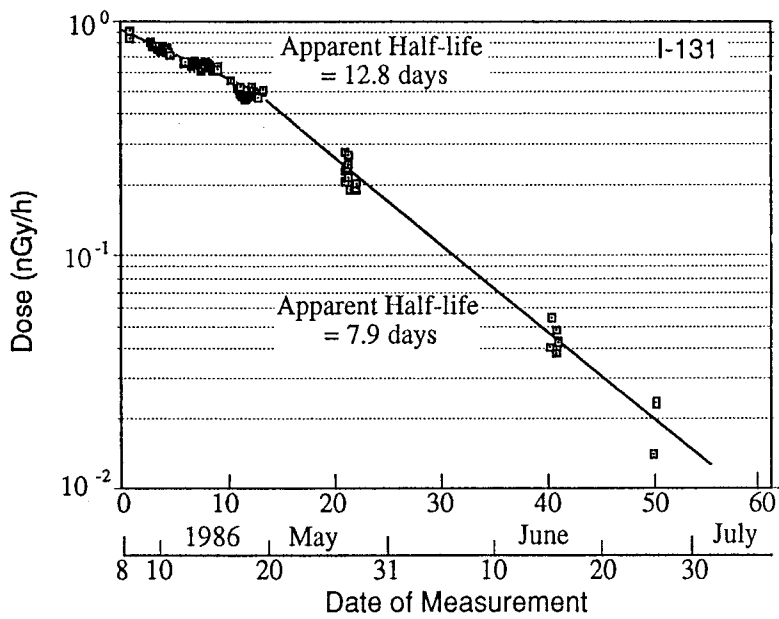


Fig. 3. Variation of dose rate due to fallout I-131 measured at Matsue.

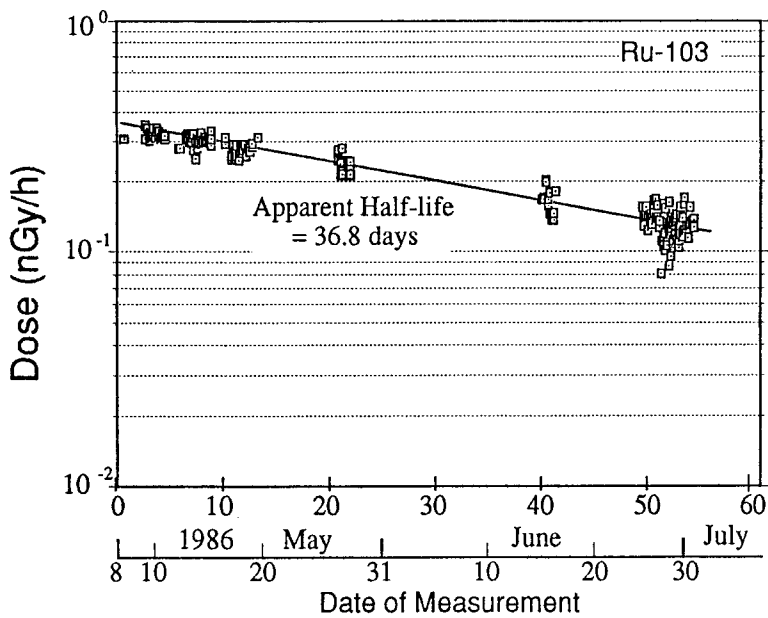


Fig. 4. Variation of dose rate due to fallout Ru-103 measured at Matsue.