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Measurements of ¹³⁴Cs and ¹³⁷Cs in urine and estimation of the internal dose of an adult exposed to the Chernobyl Accident

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Abstract To estimate the internal dose of a Chinese visiting scholar internally contaminated because of the Chernobyl Accident, the contents of ¹³⁴Cs and ¹³⁷Cs in urine were measured using a Ge(Li) γ -spectrometer. The internal doses were calculated based on data from the ICRP Publications. The effective doses from ¹³⁴Cs and ¹³⁷Cs were estimated to be 61 μ Sv and 98 μ Sv, respectively. The sum of 159 μ Sv was lower than the total effective dose (310 μ Sv), from the inhalation and ingestion of natural radionuclides. The dose of ¹³¹I was also reviewed referring to the UNSCEAR 2000 Report. The equivalent effective dose of ¹³¹I was estimated to be 2.9 mSv, 18 times more than the amount of ¹³⁴Cs and ¹³⁷Cs. Therefore, it is considered that the earlier estimation of internal doses of ¹³¹I is important in evaluating radiation injuries from a nuclear reactor accident.

Keywords Gamma spectrometer, ¹³⁴Cs, ¹³⁷Cs, Urine, Internal dose, Chernobyl Accident **CLC numbers** X838, X591

1 Introduction

In the analysis of a nuclear accident emergency, rapid determination of the quantity of radioactive materials present in the human body is necessary for estimating the internal doses. Following that, corresponding measures could be taken, including decontamination of the radioactive materials and elimination enhancement of radionuclides from the human body.

Usually, a whole-body counter can be used to detect radionuclides directly and estimate internal doses. Besides, radioanalysis of urine can be done to obtain the contents of radionuclides in urine. Then, based on the excretion equations of radionuclides, the initial intake of the radionuclides can be calculated to estimate their internal doses according to the publications of the International Commission on Radiological Protection (ICRP). In radioanalysis, the following instruments can be used: γ spectrometer, liquid scintillation counter, low background α/β counter, α spectrometer, inductively coupled plasma mass spectrometery (ICP-MS), and so on. Among these methods, high-resolution gamma spectroscopy provides fast, accurate, nondestructive isotopic analysis of radionuclides in bioassay samples and environmental samples. Therefore, it is often used in the activities of radiation emergency response.

In this study, using a Ge(Li) gamma spectrometer, a urine analysis was made for a visiting Chinese scholar, who worked at Kiev city (150 km from the Chernobyl Nuclear Power Plant) from late April to middle August in 1986 (just after the Chernobyl Accident). After 297 d, 351 d and 387 d of the accident, the contents of ¹³⁴Cs and ¹³⁷Cs in his urine samples were determined separately, and his internal doses from ¹³⁴Cs and ¹³⁷Cs were estimated.

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In the early phase of the accident, ¹³¹I was the major contributor to the internal dose, therefore, the dose of ¹³¹I was also reviewed by referring to the UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation) 2000 Report.^[1,2]

2 Material and methods

2.1 Calibration of Ge(Li) gamma spectrometer

The detector of the S-80 Ge(Li) gamma spectrometer (Canberra Company, USA) was housed within a shield, which was constructed of "old lead" with a thickness of 10 cm. The lead was graded with 3 mm copper and 5 mm Perspex. The volume of the measuring cup was 660 mL. Its energy resolution and relative efficiency were 2.0 keV and 20%, respectively. The integrated background was 1.6 cps (50 keV – 2 MeV).

Standard solutions of ²²Na, ⁵⁴Mn, ⁵⁷Co, ⁶⁰Co, ⁶⁵Zn, ⁸⁸Y, ¹⁰⁹Cd, ¹³³Ba, ¹³⁷Cs, ²¹⁰Pb, and ²⁴¹Am from Shanghai Institute of Measurement and Testing Technology were used for energy calibration and efficiency calibration. From these radionuclides, the full energy peak efficiencies for energies of 46.5, 59.5, 88.0, 122.1, 136.5, 276.4, 302.8, 356.0, 383.8, 661.7, 834.8, 898.0, 1115.5, 1173.2, 1274.5, 1332.5, and 1836.1 keV were obtained. The energy efficiency calibration curve is shown in Fig.1.



Fig.1 The energy efficiency calibration curve.

2.2 Determination of ¹³⁴Cs and ¹³⁷Cs in urine

To determine the ¹³⁴Cs and ¹³⁷Cs in the urine, the Ge(Li) γ spectrometer was used to measure the urine directly without an extensive sample preparation procedure. Twenty-four-hour urine was collected, followed by the transfer of 460 mL into the measuring

cup to be counted in the γ spectrometer. Net peak area counts of the 602, 662, and 796 keV gamma rays emitted from ¹³⁴Cs and ¹³⁷Cs were used to calculate their respective activities.

3 Results and discussion

3.1 Results of ¹³⁴Cs and ¹³⁷Cs

The contents of ¹³⁴Cs and ¹³⁷Cs measured in the subject's urine are summarized in Table 1. The following fractional excretion equation^[3] was used to calculate the excretion fraction of the day when the urine was collected:

$Y_{\rm s}(t) = 0.1 {\rm e}^{-0.693t} + 0.005 {\rm e}^{-0.006t}$

The total excretion through urine was obtained by dividing the contents in urine by the fraction. And the fraction of total excretion by urine was 0.8.^[4] Besides, the decay correction for ¹³⁴Cs was also considered. Thus, the initial intakes of ¹³⁴Cs and ¹³⁷Cs of this subject are estimated in Table 2.

Table 1 The contents of 134 Cs and 137 Cs in the subject's 24-hour urine

Days after the	Volume of urine	¹³⁴ Cs	¹³⁷ Cs
accident/d	/L	/Bq•L ⁻¹	/Bq•·L ⁻¹
297	1.51	1.47	3.21
351	1.63	0.64	2.53
387	1.75	0.38	1.59

Table 2 The estimated initial intakes of ^{134}Cs and ^{137}Cs for the subject

Days after the	Fraction of urinary	Initial intake /Bq	
accident/d	excretion for Cs (Y_s)	¹³⁴ Cs ¹³⁷ Cs	
297	8.415×10^{-4}	4332 7205	
351	6.086×10^{-4}	2952 8462	
387	4.904×10^{-4}	2402 7086	
Average		3229 7584	

For ¹³⁴Cs and ¹³⁷Cs, dose coefficients recommended by the ICRP Publication, $72^{[5]}$ were 1.9×10^{-8} Sv•Bq⁻¹ and 1.3×10^{-8} Sv•Bq⁻¹, respectively. The dose coefficient is the committed effective dose per unit intake of radioactive material into the body (Sv•Bq⁻¹).

On the basis of the average intake of 134 Cs and 137 Cs in Table 2, the committed effective doses from 134 Cs and 137 Cs are estimated to be 61 μ Sv and 98 μ Sv, respectively, with a total of 159 μ Sv. The value is lower than the world-averaged total effective dose (310 μ Sv) from the inhalation and ingestion of natural radionuclides. Of the 310 μ Sv, 170 μ Sv is from 40 K,

and the other 140 μ Sv is from the long-lived radionuclides in uranium and thorium series.^[6]

Without performing the elimination enhancement of radionuclides for the subject, the contents of ¹³⁴Cs and ¹³⁷Cs in his urine were below the minimum detection limit of the measuring system on Feb 11, 1989 (1022 d after the accident).

3.2 Dose review of ¹³¹I

In the later phase of the Chernobyl Accident, ¹³⁴Cs and ¹³⁷Cs were the major contributors to health problems, but in the early phase of the accident, radioactive iodine played a major role in impacting human health. To estimate the internal dose for radioactive iodine, the initial intakes of ¹³¹I were estimated from the initial intakes of ¹³⁴Cs and ¹³⁷Cs.

UNSCEAR 2000 Report^[6] provided the data to estimate the principal radionuclides released in the Chernobyl Accident. According to the data, based on the ratio of ¹³¹I to ¹³⁴Cs (33.4) and the ratio of ¹³¹I to ¹³⁷Cs (19.9), as well as the evaluated intakes of ¹³⁴Cs (3229 Bq) and ¹³⁷Cs (7584 Bq), the intakes of ¹³¹I were estimated to be 107849 Bq and 150922 Bq, respectively. Furthermore, based on the estimated average of ¹³¹I intake $(1.3 \times 10^5 \text{ Bq})$ and the dose coefficient for ¹³¹I (2.2 × 10⁻⁸ Sv•Bq⁻¹)^[5], the effective dose from ¹³¹I was estimated to be 2.9 mSv.

Among the total effective doses of 3.1 mSv from ¹³⁴Cs, ¹³⁷Cs, and ¹³¹I, the internal dose from ¹³¹I is 18

times higher than the sum of ¹³⁴Cs and ¹³⁷Cs. Therefore, the earlier estimation of internal doses of ¹³¹I is important in evaluating the radiation injuries of a nuclear reactor accident.

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