Activation analysis for soils of Hiroshima city and estimation of gamma-ray dose rate due to neutron induced activated soil by Hiroshima atom bomb

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Abstract

For the early entrance survivors in Hiroshima and Nagasaki atomic bomb (A-bomb), radiation doses from activated materials induced by the A-bomb neutrons are dominant. For estimation of such doses, element compositions of surrounded materials such as soil and rubbles are necessary. Especially Sc density in soil is important for estimating radiation doses at the time of a few 10 days after explosion. Because ^{46}Sc which has the half-life of 84 days, is induced the A-bomb neutrons. However, few data of Sc density in soil are available in both of Hiroshima and Nagasaki cities. Purpose of this study is evaluation of Sc density in soil and the uncertainty using activation analysis.

Soil samples were taken from 11 locations within 4 km from A-bomb hypocenter at Hiroshima city. The soil samples and reference rock sample of JA-11) were activated in Kyoto University Reactor (KUR). Element compositions are relatively obtained from each identified radionuclide counting rates in soil and reference rock by Ge-detectors.

Twenty three element compositions including Al, Mn, Na and Sc are obtained by the activation analysis. The obtained element compositions are compared with values in Dosimetric System 1986 (DS86) and those are roughly the same as the reported values in DS86. Sc density in Hiroshima soil was estimated to be 5.12±0.59 (ppm). It was found the unevenness of Sc density in soils for 11 location of Hiroshima city is about 12%.

Using element compositions by the activation analysis, time variation of the exposure rate by activated soil are estimated. It was found that exposure rate in the few minute time range is dominated by ^{28}Al, in the several days by ^{24}Na, and in the a few 10 days by ^{46}Sc. This estimated result is compared with measured dose rate measured after a few months after explosion. It was found that the estimated dose rate is quite similar to the measured one.

INTRODUCTION

In dosimetric system 1986 (DS85), cumulative radiation exposure due to activated soil around hypocenter of Hiroshima atomic bomb (A-bomb) had been estimated to be 0.8 Gy at maximum[1)]. And then, the exposure from activated soil had not been updated in the reevaluated dosimetric system 2002 (DS02)[2)]. On the other hand, Imanaka et al. evaluated and updated the exposure from activated materials based on DS02 neutron fluence using thermal neutron fluence ratio (DS02/DS86) and ^{60}Co activation comparing with the results by Gritzner et al. in DS86 [3)]. It was concluded that the exposure at 1m height after 1 minute after from the Hiroshima bombing was estimated to be 4Gy/h[4)]. Tanaka et al. also

evaluated that the gamma- and beta- ray exposure in air and on skin from the induced activities of ^{24}Na, ^{56}Mn, ^{42}K and ^{46}Sc in soil according to the DS02 neutron fluence using Monte Carlo transport calculation[5]。 It shows that exposures from activated soil are estimated to be ~50mGy/h by Imanaka et al.[3] and ~40mGy/h by Tanaka et al [5] at 1 hour after the bombing, respectively. In these estimation, the element composition in soil summarized in DS86 was used.

Element composition is measured for only two locations (Hiroshima Castle and A-bomb Dome) in DS86. Variation of the composition over Hiroshima City is unknown. Before established DS86, element composition analysis for 16 locations for Hiroshima City were carried out by Hashizume et al. in 1967. However, they concentrated into Na and Mn density, and measured just one location for Sc density[6]. Sc is activated to ^{46}Sc which has relatively longer half life of 83.79d. The exposure of the early entrance survivors at 10 days or later after bombing is dominated by ^{46}Sc, therefore the Sc concentration is important parameter for the early entrance survivors. In this report, the element concentration, especially Sc concentration and it's unevenness in soil of Hiroshima City have been obtain by the activation analysis. And also, the exposure rate by the induced radioactivities in soil has been estimated.

Soil sampling

Eleven soil samples were collected from 10 locations including Hiroshima Castle and Atomic bomb dome where the element concentration analysis were performed in DS86. The soil core (5cmϕ x 20cm) were taken using stainless steel pipes. The sapling location are shown in Fig. 1. The GPS coordinate of the sampling locations are summarized in Table 2.

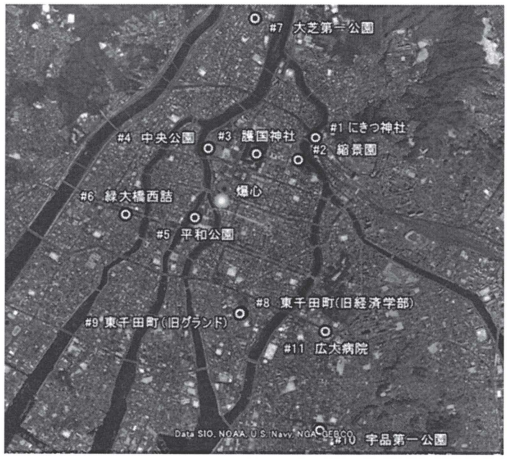

Fig. 1 Sampling locations

These samples were dried by an oven at 120 degrees for over night. The dried samples are sieved through a 2-mm mesh to remove small rocks and big plant remains. The sample for activation analysis was prepared with 10 g soil grained with mortar for uniformity in the sample. About 0.1 g grained soil was packed into about 5 mm square with polyethylene film. The neutron irradiation has been carried out at Kyoto University Reactor (KUR).

Table 1 Sampling locations and GPS coordinates

ID	Sampling location	GPS	
1	Nikitsu shrine	34°24'15.40"N	132°28'8.20"E
2	Shukeen Garden	34°24'4.50"N	132°27'59.30"E
3	Gokoku Shrine	34°24'6.20"N	132°27'35.80"E
4	Chuo Park	34°24'7.60"N	132°27'8.70"E
5	Peace Memorial Park	34°23'34.80"N	132°27'3.60"E
6	Midori Ohashi West side	34°23'34.50"N	132°26'25.10"E
7	Oshiba Daiichi Park	34°25'10.50"N	132°27'30.50"E
8	Higashi-Senda 1	34°22'51.50"N	132°27'32.50"E
9	Higashi-Senda 2	34°22'51.10"N	132°27'31.70"E
10	Ujina Daiich Park	34°21'59.50"N	132°28'19.60"E
11	Hiroshima Univ Hospital	34°22'45.10"N	132°28'53.40"E

The soils have been neutron-irradiated for 30 s, to identified short-life radionuclides. The gamma-ray spectrometry has been carried out at about 2 min and 1 hours after the irradiation using Ge-detector (EG& G ORTEC, GEM-25185) at KUR. The first measurement is mainly for ^{28}Al-concentration determination. The second measurement is mainly for ^{56}Mn and ^{24}Na--concentration determination. The soils have been neutron-irradiated for 20 min, to identified medium and long-life radionuclides. The gamma-ray spectrometry has been carried out at about 10 days and 40 days after the irradiation using Ge-detector (EG &G ORTEC, GEM-30200-P) at Hiroshima University.

The element concentrations have been relatively obtained using reference rock sample JA-1 which is andesite taken from Hakone Mountain in 1989. The reference sample is distributed by National Institute of Advanced Industrial Science and Technology (AIST) and measured element concentration at 460 Institutes in 43 nations[7].

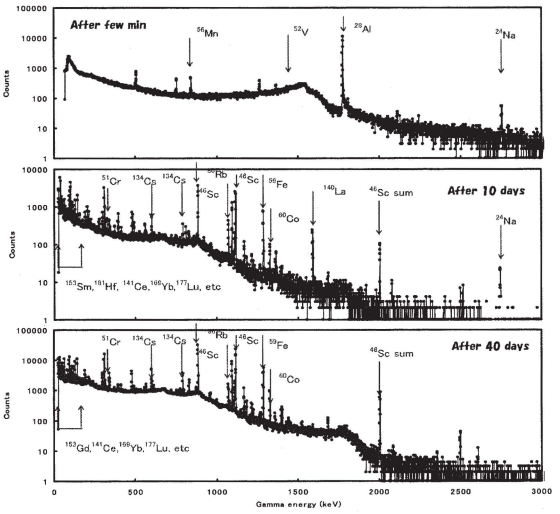

Fig. 2 Example of gamma-ray spectrum

Results

The gamma-ray spectra for #3 (Gokoku Shrine) are shown in Fig.2. Short-life radionuclides of ^{28}Al, ^{56}Mn, ^{52}V and ^{24}Na are identified in Fig 2 (a). Medium- and long-life

Table 2 Averaged values of element cencetration in soil and DS86 values[1)]

element	Z	Concentration (ppm)		DS86 value (ppm) Hiroshima Castle	DS86 value (ppm) A-bomb Dome
Na	11	19300	±20%	16000	12400
Al	13	63300	±14%	71000	64900
Sc	21	5.12	±12%	5	5
V	23	21.4	±26%	22.3	25.3
Cr	24	20	±65%	20.5	27.3
Mn	25	517	±13%	467	587
Fe	26	17100	±16%	17700	20600
Co	27	4.13	±23%	3.7	3.8
Rb	37	137	±12%	230	225
Sr	38	45	±38%	88	70
Zr	40	105	±40%	41	35
Nb	41	2.0	±60%	7	5
Sb	51	1.87	±11%	1.4	0.8
Cs	55	4.4	±14%	5	5
La	57	27.4	±23%	23	21
Ce	58	88	±24%	40	36
Sm	62	3.7	±27%	3.4	3
Eu	63	0.81	±15%	0.9	0.9
Gd	64	2.23	±44%		
Lu	71	0.133	±33%		
Hf	72	4.17	±19%	4	4
Th	90	14.3	±30%	13.3	9.9
U	92	0.85	±18%	2.8	2.6

radionuclides of ^{153}Sm, ^{181}Hf, ^{141}Ce, ^{169}Yb, ^{177}Lu, ^{85}Sr, ^{51}Cr, ^{140}La, ^{134}Cs, ^{95}Zr, ^{134}Cs, ^{46}Sc, ^{86}Rb, ^{233}Pa, ^{152}Eu, ^{153}Gd, ^{60}Co and ^{59}Feare identified in Fig. 2 (b) and (c). Counting rate ratio of soil samples to reference samples decay-corrected at just irradiation finished are multiplied to the known element concentrations. The 23 element concentrations including Al, Mn, Na and Sc, for 11 soil samples are obtained and the averaged values over 11 samples is listed in Table 2. The concentration of main components such as Al, Mn, Na, Sc and Fe are varied about 11-15%, the other trace elements show 20-30% unevenness. Present values of element concentrations are consistent with DS86 values of Hiroshima Castle and A-bomb Dome shown in Table 2.

Sc, Mn and Nacocentration and their unevenness

Obtained element concentrations of Sc, Mn and Na are shown by scatter plots in Fig. 3. Figure 3(a) correlation of Mn-Na concentration and (b) correlation of Sc-Na concentration are shown. And also, element concentration by Hashizume et al[6)] and DS86 values[1)] are shown in the same figures. There are no correlation in both of the Sc-Na and Mn-Na. In Fig. 3(a) and (b), the Na and Sc concentration shows consistency with those of DS86 and Hashizume et al. within their unevenness, however the Mn concentration shows a half of Hashizume's values. The unevenness of Na, Mn and Sc concentration are 19300±3900 (±20%) ppm, 517±68 (±13%) ppm and 5.12±0.59 (±12%) ppm, respectively.

Exposure from activated soil

Exposure from activated soil are estimated using the obtained element concentration. Induced radioactivities in 1 g soil by A-bomb neutrons can be calculated using the element concentration, DS02 thermal neutron fluence and activation cross sections. However, to estimate the exposure, shielding factor and scattering factor (called conversion factor, here) in soil and air are needed. In this estimation, results by Tanaka's Monte Carlo calculation [5] are used for such factor. The factor of induced radioactivity of ^{24}Na to dose in air by ^{24}Na are calculated and applied to all induced radioactivities of ^{56}Mn, ^{46}Sc, ^{60}Co, ^{134}Cs, ^{59}Fe, ^{42}K, ^{51}Cr, ^{28}Al and ^{233}Pa. The conversion factor for each radionuclide should be different from that of ^{24}Na. Therefore this estimate of exposure rate is approximation. Time variation of each exposure rate by radionuclide is calculated to be decreased by each half-life. Figure 4 shows the time variation of exposure rate at around the hypo center of Hiroshima A-bomb. There are few measured data after bombing. There are only 4 measured data at one to three month after bombing by Miyazaki and Masuda[8] and by Manhattan Engineering District[9]. For comparison with our estimation, these data also plotted in the same figure. The estimation and measured data show the good agreement.

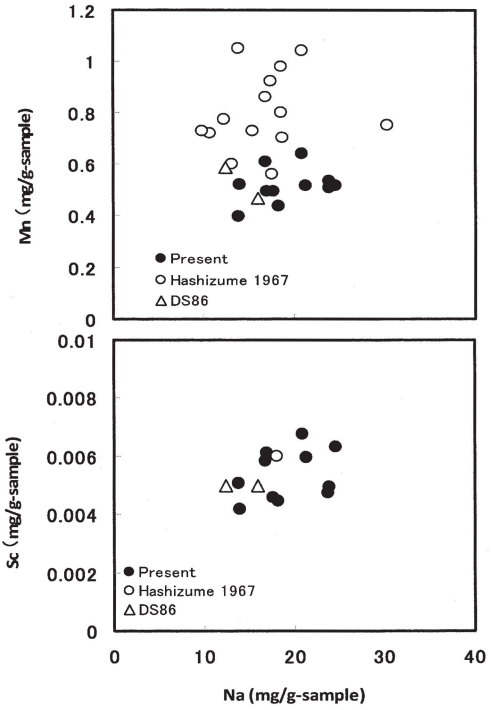

Fig. 3 (a) Mn-Na and (b)Sc-Na correlation

The exposure rate at a few minute after bombing is estimated to be 7-5Gy/h(due to ^{28}Al), at a few hours to be 0.1－0.02Gy/h (due to ^{56}Mn and ^{24}Na) and a few month to be 1mGy/h (due to ^{46}Sc). From these results, it was ascertained that exposure rate dominated by ^{28}Al during a few minute, by ^{24}Na in a few minute to about 10 days and by ^{46}Sc in a few 10 days to 1 year.

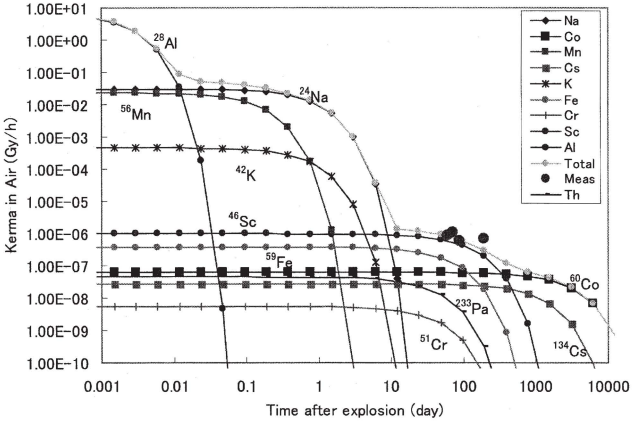

Fig. 4 Time variation of exposure rate estimated by element concentrations.

Summary

In order to estimate exposure from activated soil by Hiroshima atomic bomb, element concentration in soil of Hiroshima City are analyzed by the activation analysis at Kyoto University Reactor. Eleven soil samples were collected from Hiroshima City within 4 km from hypocenter.

As a result of the activation analysis, 23 element concentations including Al, Mn, Na and Sc are obtained. Sc concentration relate to exposure rate at about 10 days after bombing due to lack of data in DS86 is obtained. Sc concentration and unevenness are obtained to be 5.12±0.59 (±12%) ppm.

Mn concentration is a half of value by Hashizume et al [5], however Mn and Na concentration are consist with DS86 values. The averaged value of Mn and Na concentrations over 11 soil samples are Mn: 517±68 (±13%) ppm and Na: 19300±3900 (±20%) ppm, respectively.

The exposure rate at a few minute after bombing is estimated to be 7-5Gy/h(due to ^{28}Al), at a few hours to be 0.1－0.02Gy/h (due to ^{56}Mn and ^{24}Na) and a few month to be 1mGy/h (due to ^{46}Sc). From these results, it was ascertained that exposure rate dominated by ^{28}Al during a few minute, by ^{24}Na in a few minute to about 10 days and by ^{46}Sc in a few 10 days to 1 year. The estimated exposure rate is the good agreement with by Miyazaki and Masuda[8] and by Manhattan Engineering District[9].

Reference

1) Roesch, W. C., Eds. (198s U. S.—Japan Joint Reassessment of Atomic Bomb Radiation Dosimetry in Hiroshima and Nagasaki. Vols. 1 and 2. Radiation Effects Research Foundation.

2) Young, R. W. and Kerr, G. D., Eds. (2005) Reassessment of the Atomic Bomb Radiation Dosimetry for Hiroshima and Nagasaki–Dosimetry System 2002. Vols. 1 and 2. Radiation Effects Research Foundation .

3) Gritzner ML, Woolson WA (1987) Calculation of doses due to atomic bomb induced soil activation. In: Roesch WC (ed) Reassessment of atomic bomb radiation dosimetry—dosimetry system 1986, vol 2, chap 6, App 2. Radiation Effects Research Foundation, Hiroshima, pp 342–351.

4) Imanaka, T., Endo, S., Tanaka, K. and Shizuma, K., (2008) Gamma-ray exposure from neutron-induced radionuclides in soil in Hiroshima and Nagasaki based on DS02 calculations, Radiat Environ Biophys 47:331–336.

5) Tanaka, K., Endo, S., Imanaka, T., Shizuma, K., Hasai, H. and Hoshi, M. (2008) Skin dose from neutron-activated soil for early entrants following the A-bomb detonation in Hiroshima: contribution from β and γ rays. Radiat. Environ. Biophys. 47: 323–330.

6) Hashizume, T., Maruyama, T., Shiragai, A., Tanaka, E., Izawa, M., Kawamura, S. and Nagaoka, S. (1967) Estimation of the air dose from the atomic bombs in Hiroshima and Nagasaki. Health Phys. 13: 149-161.

7) Imai, N., Terashima, S., Itoh, S. and Ando, A.(1995) 1994 compilation of analytical data for minor and trace elements in seventeen GSJ geochemical reference samples, "igneous rock series", Geostandards Newsletter 19: 135-213 .

8) Miyazaki Y, Masuda T (1953) Radioactivity in Hiroshima city and its suburbs caused by the atomic bomb (II), vol 1. In: Collection of survey reports on the atomic bomb disasters, Japan Society for the Promotion of Science, Tokyo, pp 35–38 (in Japanese).

9) Pace N, Smith RE (1959) Measurement of the residual radiation intensity at the Hiroshima and Nagasaki atomic bomb sites, ABCC TR 26–59. Radiation Effects Research Foundation, Hiroshima.