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Neutron induced activity/ $^{60}\text{Co}/^{152}\text{Eu}$ /Atomic Bomb/Gamma-ray spectroscopy

Neutron-induced activities ^{60}Co and ^{152}Eu have been measured for samples collected from the Atomic-Bomb Dome locating at 161 m from the hypocenter of the Hiroshima Bomb. Specific activities $^{60}\text{Co}/\text{Co}$ and $^{152}\text{Eu}/\text{Eu}$ at the time of the detonation have been determined as $10.0 \pm 1.0 \text{ Bq mg}^{-1}$ (steel sample S4) and $80 \pm 9 \text{ Bq mg}^{-1}$ (granite sample G1), respectively. Detailed measurements of ^{60}Co and ^{152}Eu activities for samples collected from various locations of the Dome show almost no directional dependence whether the sample faced to the epicenter or not, nor vertical height dependence between 17 m height and the ground level. In addition, ^{152}Eu was not detected in the sample collected from the basement. It has been shown that the present ^{60}Co activity value, the nearest steel one to the hypocenter, as well as other short distance data are systematically lower than the calculated values based on the neutron fluence of the DS86.

INTRODUCTION

Reevaluation of Hiroshima and Nagasaki atomic-bomb dosimetry has been completed in 1987 with a final report¹⁾ of a new dosimetry system called DS86. Concerning to the evaluation of the low energy neutron fluence, ^{60}Co activation data taken by Hashizume et al.^{2,3)} both at Hiroshima and Nagasaki, systematically deviated from the calculated values by Loewe⁴⁾. Nevertheless detailed discussion on the ^{60}Co activation, the discrepancy was still in open

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question. Other ^{60}Co data taken by Saito⁵⁾, by Hoshi and Kato⁶⁾ and by Nakanishi et al.⁷⁾ were not considered in the above discussion. No detailed consideration was made on ^{152}Eu activation data in the final report.

Afterward, residual activity data on ^{60}Co and ^{152}Eu have been accumulated and a systematic deviation from the calculated activation has been revealed⁸⁾ in the ^{152}Eu activation data similarly to the ^{60}Co data. In the case of ^{60}Co measurement, data from two locations were recently added by Kimura et al.⁹⁾ and by Kerr et al.¹⁰⁾, however, ^{60}Co data were still scarce for detailed discussions on the neutron fluence.

In March 1990, the Atomic-Bomb Dome (the City of Hiroshima Commercial Display Building) located at 161 m from the hypocenter in Hiroshima City has been mended to keep it from crumbling. During the work of mending, we have collected steel and granite samples from various locations of the Dome. In this paper, new data of specific activities of ^{60}Co (half-life = 5.2719 y¹¹⁾ and ^{152}Eu (half-life = 13.2 y¹¹⁾ are given from samples of the Dome. A systematic discrepancy between the measured ^{60}Co data and the calculated activities by Mendelsohn¹²⁾ is shown in short distances from the hypocenter. The ^{152}Eu activation along vertical direction including the basement was discussed.

MATERIALS AND METHODS

Steel and granite samples

Location of the Dome is shown in Fig. 1. The coordinates of the hypocenter on the new Hiroshima City Map¹³⁾ transferred from the US Army Map¹⁴⁾ are also shown in the figure. Top and side view of the Dome and sampling locations are shown in Fig. 2. Four steel samples (S4, S8, S12, S16) were taken at 21 m above the ground level. The roof of the Dome was covered with the wood sheathing¹⁵⁾ and thin copper plates at the time of the detonation, so the steel

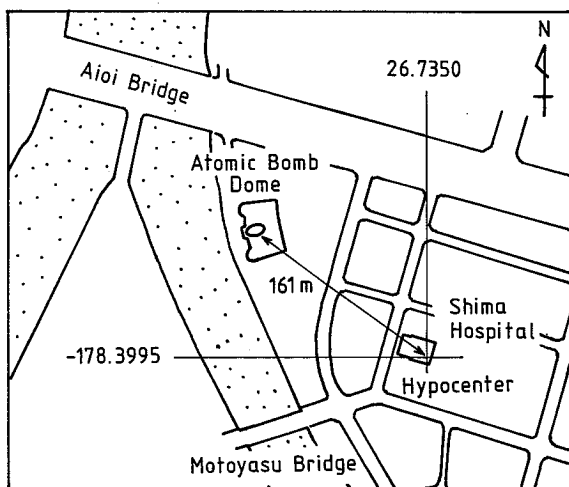


Fig. 1. Location of the Atomic-Bomb Dome in Hiroshima. The coordinates of the hypocenter on the new Hiroshima city map are also shown.

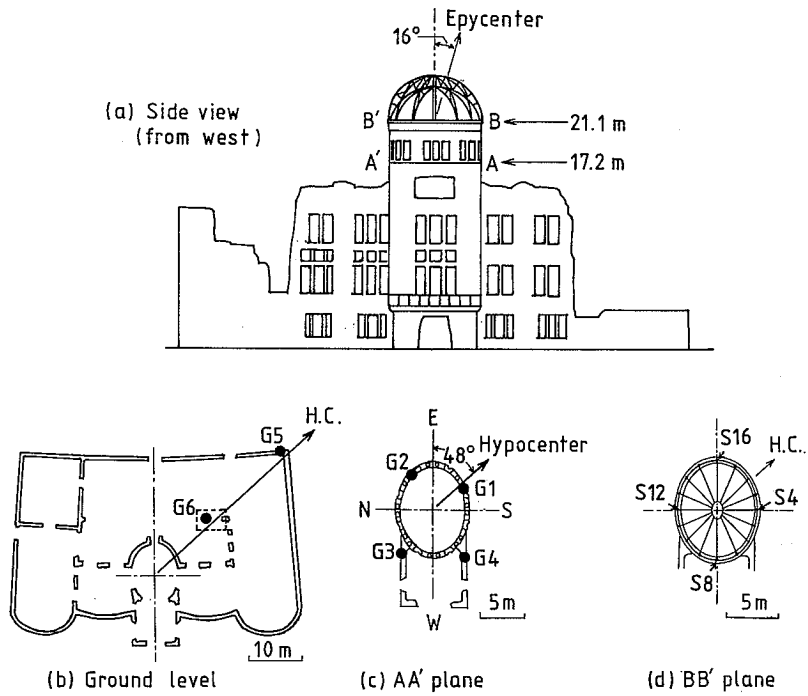


Fig. 2. Side and top views of the Atomic-Bomb Dome. Sampling locations of stone samples G1 to G6 (b and c) and steel samples S4 to S6 (d) are shown.

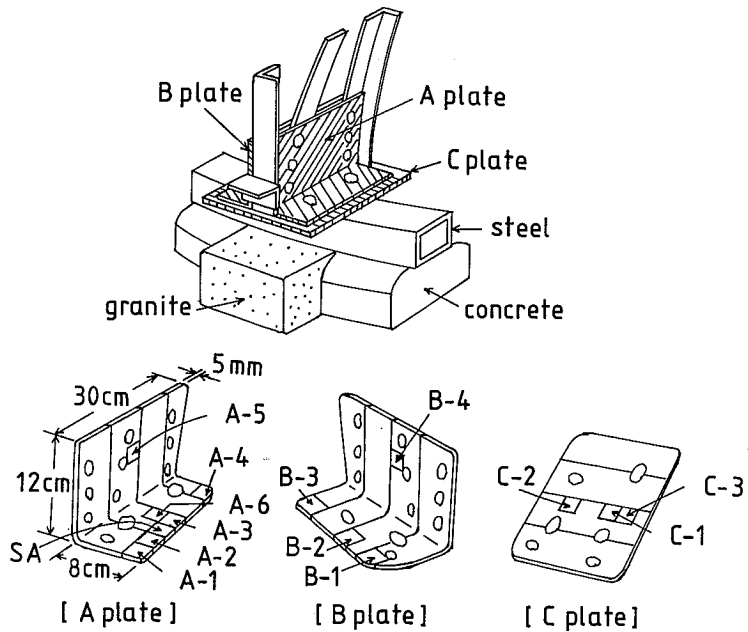


Fig. 3. Sketch of steel sample S4 composed of A, B, and C plates. Locations of small pieces for the γ ray measurement are shown for each plate.

samples were almost directly exposed to the neutrons. Four granite cores of 6.8 cm dia. \times 30 cm leng. (G1, G2, G3, G4) were taken at 17 m above the ground level and one granite core (G5) was taken at the ground level. One concrete core (G6) of the same size was taken from the ceiling of the basement. As shown in Fig. 3, the steel sample (the base plate of the arch) was composed of three plates with 5 mm thick; i.e., two L-shaped plates and one flat plate, which were named as A, B, and C, respectively.

Sample preparation and elemental analysis

Since the Dome was near to the hypocenter, γ rays of ^{60}Co induced in steel samples were directly measurable with a low-background Ge detector. To determine the specific activity $^{60}\text{Co}/\text{Co}$, disk sample (10.5 mm dia. \times 1.6 mm thick., 1.133 g) and three stick samples (4.0 mm² \times 15 mm, 2.8 mm² \times 15 mm, 2.6 mm² \times 15 mm, total 1.101 g) were obtained from the plate SA of S4 sample shown in Fig. 3. To investigate local deviation of ^{60}Co activity, many pieces (25 mm \times 25 mm \times 1 mm) were cut from the upper surface of the sample S4 as shown in Fig. 3. The depth profile of ^{60}Co activity in steel plates were measured from the samples S4-A-3, S4-A-6, S4-C-1 and S4-C-3, where the reverse side of 1 mm thickness was taken for the samples of S4-A-6 and S4-C-3. Same size pieces were cut from samples S8, S12 and S16 at similar positions to S4-A-1. All steel sample were etched with 3M HCl, washed with water and dried prior to the measurement.

Elemental analyses in the steel samples have been done by the Kawasaki-Steel Techno-Research Co. (K-TEC) by means of the atomic absorption method. The concentration of Co was determined for each steel plate. In addition to Co content (0.26 ± 0.02 mg/g) for steel plate S4-A, Cu and Ni contents were also determined as 0.26 ± 0.02 mg/g and 0.27 ± 0.02 mg/g, respectively, to estimate the yield of ^{60}Co through the fast neutron reactions of $^{63}\text{Cu}(n,\alpha)^{60}\text{Co}$ and $^{60}\text{Ni}(n,p)^{60}\text{Co}$. According to the cross sections for these reactions mentioned in ref. 4, yields of ^{60}Co were estimated negligibly small compared to the thermal neutron activation.

Granite core samples were cut into orbicular plates of 2 cm thickness. The surface plates were grained to less than 100 mesh and 30 g of the stone powder was packed in a plastic vessel. Concentration of the stable Eu in stone samples were determined by means of the neutron activation analysis. A couple of 4 g powder sample were taken and 50 μg Eu was added to one of them. Neutron activation was performed at the research reactor of Kinki-University under the thermal neutron flux of $10^7 \text{ n}\cdot\text{cm}^{-2}\cdot\text{s}^{-1}$.

Since the ^{152}Eu activity level was very low for the sample G6, Eu was chemically enriched referring to the alkali fusion method¹⁶⁾. Enriched sample of 3.45 g was obtained out of 45 g original sample. The Eu content in the enriched sample was determined by means of the neutron activation analysis. In this case, $^{152\text{m}}\text{Eu}$ (half-life=9.3 h¹¹⁾) was activated by using the ^{252}Cf neutron source at the Research Institute for Nuclear Medicine and Biology of Hiroshima University.

Gamma-ray measurement

Two low-background Ge spectrometers were used for the γ -ray measurement. One was a 124 cm³ coaxial type Ge detector¹⁷⁾ (the CX detector, the relative efficiency: 28%) with a 20 cm

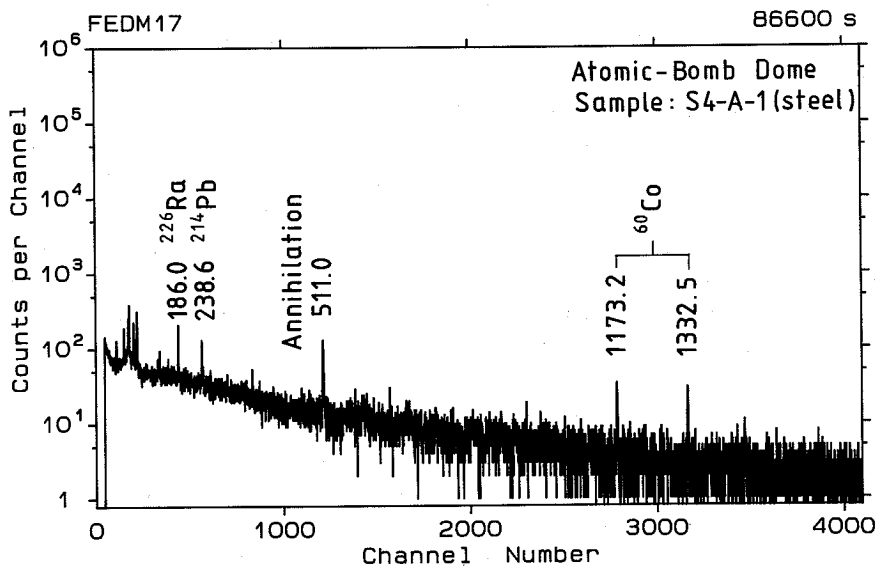


Fig. 4. Typical γ -ray spectrum of steel sample S4-A-1 obtained from the Dome measured with a 124 cm^3 coaxial Ge detector. The ^{60}Co activity was induced by neutrons of the bomb.

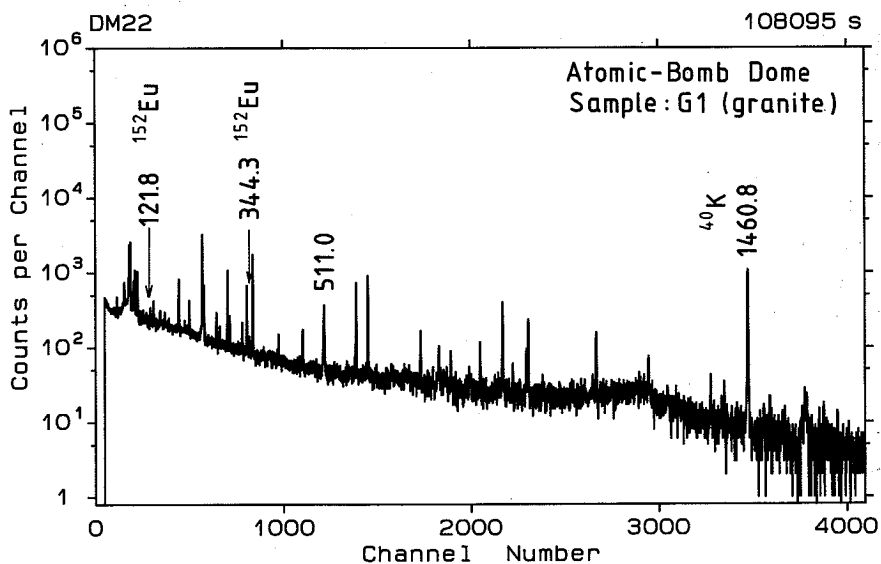


Fig. 5. Typical γ -ray spectrum of granite sample G1 obtained from the Atomic-Bomb Dome measured with a 124 cm^3 coaxial Ge detector. The ^{152}Eu activity was induced by neutrons of the bomb.

thick lead shielding and the other is a 120 cm³ well type Ge detector¹⁸⁾ (the WL detector, the relative efficiency: 23%) with a 25 cm thick lead shielding and an anticoincidence system to reduce the cosmic-ray background.

Steel samples were measured mainly with the CX detector at the position close to the detector's end-cap. Typical γ -ray spectrum for steel sample S4-A-1 is shown in Fig. 4. Two γ -ray peaks of ⁶⁰Co are clearly seen in the figure. To determine the specific activity ⁶⁰Co/Co, both the CX and the WL detector were used. The γ -ray detection efficiency for ⁶⁰Co was determined using reference sources of QCR11 (produced by Amersham Int.) for the CX detector and NES-100S (produced by DuPont NEN Products) for the WL detector. The efficiency of the steel sample for the CX detector was determined from the measured dependences of the efficiency along the detector axis and the radial direction. In the case of the WL detector, it was confirmed that the efficiency in the well was constant. Self-absorption correction for the reference source and steel samples were negligible. The efficiencies of both detectors are given in Table 1.

Granite samples G1-G5 were measured with the CX detector. Typical γ -ray spectrum for sample G1 is shown in Fig. 5. Efficiency calibration for ¹⁵²Eu was performed with a standard sample contained a known amount (4.47 Bq) of ¹⁵²Eu. The enriched sample of G6 was pressed into a polypropylene tube and was measured with the WL detector.

RESULTS

Specific activity ⁶⁰Co/Co for the steel sample S4-A is given in Table 1. Since measured values with two different detectors agree well with each other, an average value was adopted for the specific activity of the sample. Local deviations of ⁶⁰Co activation in steel plates S4-A, S4-B and S4-C were investigated comparing ⁶⁰Co γ -ray counting rates corrected for the Co content in the steel plate and the results were given in Table 2. Local deviations of ⁶⁰Co activation for steel samples S4 to S16 were investigated in the same manner and the results were given in Table 3. The depth profile of ⁶⁰Co activity induced in steel plates is shown in Fig. 6.

Specific activity ¹⁵²Eu/Eu for stone samples G1 to G6 are given in Table 4. Since γ -rays of ¹⁵²Eu were not detected for the sample G6, only the upper limit was given in the table.

Table 1. Specific activity ⁶⁰Co/Co in steel sample S4-A obtained from the Atomic-Bomb Dome

Sample*	Co content (ppm)	Ge detector		⁶⁰ Co/Co [†] (Bq mg ⁻¹)	
		Type	Efficiency** (cps/Bq)	Measured	Average
S4-A, #1	260±20	coaxial	0.0407±0.00017	10.1±1.6	10.0±1.0
S4-A, #2	260±20	well	0.0782±0.00014	9.8±1.3	

*) The ground and the slant range were 163±15 m and 602±15 m and 602±21 m, respectively, and the height was 21 m above the ground. Sample #1: disk of 10.5 mm dia. ×1.6 mm thick., sample #2: three sticks of 4 mm²×15 mm, 2.8 mm²×15 mm, 2.6 mm²×15 mm.

***) Effective efficiency for 1173 keV+1332 keV peak counts.

†) Corrected at the time of the detonation.

Table 2. Counting rates of ^{60}Co 1173 keV + 1332 keV peaks for the steel sample S4

Steel plate (Co content)	No.	H/V*	^{60}Co counting rate (cps/mgCo)
S4-A (260 ± 20 ppm)	1	H	0.51 ± 0.05
	2	H	0.53 ± 0.06
	3	H	0.46 ± 0.06
	4	H	0.45 ± 0.05
	5	V	0.47 ± 0.05
S4-B (230 ± 20 ppm)	1	H	0.44 ± 0.06
	2	H	0.44 ± 0.06
	3	H	0.52 ± 0.07
	4	V	0.44 ± 0.05
S4-C (80 ± 6 ppm)	1	H	0.27 ± 0.05
	2	H	0.37 ± 0.06

*) H: horizontal plate, V: vertical plate.

Table 3. Local deviation of ^{60}Co 1173 keV + 1332 keV peak counting rate for four steel samples.

Steel sample	Distance (m)		Height (m)	Co content (ppm)	Counting rate (cps/mgCo)
	ground	slant			
S4-A-1	163 ± 15	602 ± 21	21	260 ± 20	0.51 ± 0.05
S8-A-1	169 ± 15	604 ± 21	21	250 ± 20	0.47 ± 0.05
S12-A-1	168 ± 15	603 ± 20	21	250 ± 20	0.52 ± 0.08
S16-A-1	163 ± 15	602 ± 20	21	260 ± 20	0.50 ± 0.06

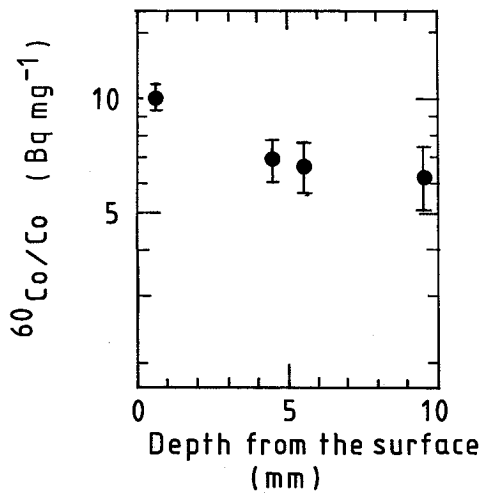
**Fig. 6.** Depth profile of the specific activity $^{60}\text{Co}/\text{Co}$ for total 10 mm of steel plates S4-A and S4-C.

Table 4. $^{152}\text{Eu}/\text{Eu}$ in stone samples obtained from the Atomic-Bomb Dome

Sample	Height (m)	Distance (m)		Eu content (ppm)	$^{152}\text{Eu}/\text{Eu}^\dagger$ (Bq mg $^{-1}$)
		ground	slant		
G1	17	161±15	585±21	0.88±0.06	80±9
G2	17	165±15	586±21	0.78±0.06	73±12
G3	17	173±15	589±21	0.73±0.06	86±13
G4	17	168±15	587±21	0.84±0.04	94±10
G5	0.35	137±15	596±21	0.57±0.05	86±13
G6*	-1.2	150±15	600±21	3.3 ±0.5	<2.1**

*) Eu enriched sample.

***) Upper limit.

†) Corrected at the time of the detonation.

DISCUSSION

1. Local deviation of ^{60}Co and ^{152}Eu activities

Specific activity $^{60}\text{Co}/\text{Co}$ were measured in detail for steel sample S4. It is shown that the ^{60}Co activities are almost same for plates A and B as shown in Table 2. The ^{60}Co activity for the plate C is about 3/5 of those of A or B plates since it was shielded with the 5 mm thick steel plate (A and B). The depth profile shown in Fig. 6 also represents that the ^{60}Co activity for upper plate is obviously higher than the lower plate.

The ^{60}Co activities for four steel samples S4 to S16 were almost same in accord with the result for ^{152}Eu activities of granite samples G1 to G4, nevertheless, steel samples S8 and S12 and stone samples G2 and G3 were not faced to the epicenter of the bomb. These results indicate that thermal neutron fluences were almost the same magnitude at sampling locations around the Dome. Similar results have been obtained in the case of depth profile measurement of ^{152}Eu for the Motoyasu-bridge pillar¹⁹⁾ located at 132 m from the hypocenter. It was shown that the ^{152}Eu activity was almost equal at the north, west, south and east surfaces of the pillar, where the west surface did not look the epicenter. According to the neutron cross section data²⁰⁾, the (n,γ) cross section of ^{151}Eu is dominant for thermal neutrons (5800 b). In contrast, the cross section of ^{59}Co for neutron energies 0.5–1 keV is about 100 b in average, which is higher than 37 b for thermal neutrons. This indicates that the ^{60}Co activity data reflects rather epithermal region than thermal region of the incident neutron energy spectrum. According to the present results, it could be said that almost no directional dependence exists for the thermal- and epithermal-energy neutrons at short distances from the hypocenter.

As given in Table 4, the $^{152}\text{Eu}/\text{Eu}$ specific activity of the granite sample G5 obtained at the ground level is almost equal to those of samples G1 to G4 obtained at 17 m above the ground level. The sample G5 is nearest to the hypocenter among the stone samples in the ground range, however, the slant range is almost same with others. This result indicates that the activation of Eu at the same slant range never depends whether the sample is located on the ground or 17 m

above the ground level, i.e., the reflection of neutrons from the ground is negligible. Gamma-rays of ^{152}Eu were not detected from the sample G6 obtained in the basement. Only the upper limit is given in Table 4. The ratio of activities in basement to the ground level G5 is less than 1/50.

2. Specific activity of ^{60}Co against distance from the hypocenter

Measured ^{60}Co data including the present work are summarized in Table 5. Saito's data in the table were obtained correcting the ^{60}Co decay at the time of the detonation of the bomb to the original data⁵⁾. The ^{60}Co activity of the Fukoku Seimei Building by Hoshi and Kato⁶⁾ in the table is the value of the iron ring faced to the epicenter. From six ^{60}Co activity data given for Aioi bridge by Hoshi and Kato⁶⁾, three data for plate A (vertical) and three data for plate B (horizontal) were respectively averaged. The ^{60}Co data by Hashizume et al.²⁾ in the table are only for the iron rings located on rooftop for comparison with other data (their data of embedded rebar in concrete were not given here). These ^{60}Co data are shown in Fig. 7 against ground range in comparison with the calculation by Mendelsohn¹²⁾ assuming the bomb yield to be 15 kiloton²¹⁾. This calculation has been performed using the Monte Carlo transport code on a model of Co activation in a free-field over the ground.

Table 5. Summary of specific activity $^{60}\text{Co}/\text{Co}$ data in Hiroshima

Location	Distance (m)		Material	$^{60}\text{Co}/\text{Co}^*$ (Bq mg ⁻¹)	Authors	ref.
	Ground	Slant				
Shima Hospital	0±20	580±15	rooftile	12.50±0.99	Nakanishi et al.	7
Motoyasu Bridge	128±20	594±16	granite	12.8±5.1		
A-Bomb Dome	163±15	602±21	steel plate	10.0±1.0	Present work	
A-Bomb Dome			water trough	3.96±0.28	Saito	5
			water trough	4.05±0.06		
Sumitomo Bank	250	614	iron ring	6.0±1.3	Hoshi, Kato	6
Aioi Bridge	300	652	steel plate A	2.9±0.3		
			steel plate B	1.8±0.2		
Fukoku Seimei	330	651	iron ring	5.7±0.6	Kimura et al.	9
Chugoku Electric Co.	687	883	handrail	0.440±0.063		
				0.423	Kerr et al.	10
Yokogawa Bridge	1295	1415	steel plate D	0.00515±0.00080	Kimura et al.	9
			steel plate C	0.00257	Kerr et al.	10
			steel plate D	0.0056		
Honkawa School	373	696	iron ring	4.4**	Hashizume et al.	2
Fukuromachi School	441	719	iron ring	3.3**		
Kirin Beer Hall	650	862	iron ring	0.98**		
Kodo School	727	921	iron ring	0.53**		
City Hall	997	1147	iron ring	0.11**		

*) Corrected at the time of the detonation.

**) These absolute values were cited in ref. 10. The original values were given in ref. 2.

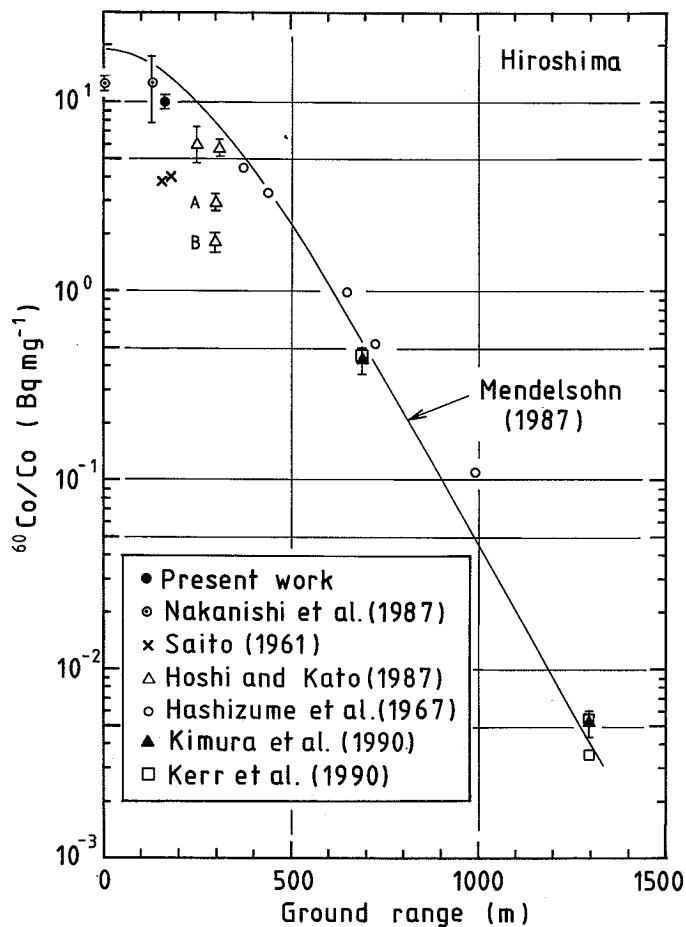


Fig. 7. Specific activity $^{60}\text{Co}/\text{Co}$ against ground range from the hypocenter. Symbols of the present work and other authors are indicated in the figure and each value is given in Table 5. The solid line is a calculation by Mendelsohn¹²⁾.

At short distances within 300 m, present value of ^{60}Co is in consistent trend with those data of nonmetallic samples of Shima Hospital (rooftile) and Motoyasu Bridge (granite) by Nakanishi et al.⁷⁾ and iron ring data of Sumitomo Bank and Fukoku Seimei Building by Hoshi and Kato⁶⁾. It is noted that these measured ^{60}Co data are systematically lower than the calculation. This result agrees with those in the case of $^{152}\text{Eu}/\text{Eu}$ specific activity measurements⁸⁾. As shown in Fig. 7, Saito's data⁵⁾ were lower than the neighboring data. They have estimated thermal neutron fluences of the Hiroshima and the Nagasaki bomb for the first time, however, lack of detailed information on sample makes further discussions difficult. Two data of Aioi bridge marked A and B in the figure are 1/2 to 1/3 lower than those of Fukoku Seimei Building located at almost the same ground range. A detailed calculation of Co activation including the geometry of the sample surroundings would be necessary to explain the discrepancy.

As shown in Fig. 7, recent data by Kimura et al.⁹⁾ and by Kerr et al.¹⁰⁾ agree well with each other, but show different trend from iron ring data by Hashizume et al. Although the calculation by Mendelsohn seems to agree with the recent ^{60}Co data, a detailed calculation by Kerr et al.¹⁰⁾ including the sample surroundings on the Co activation gives a slight lower value for the handrail sample of Chugoku Electric Co. and a factor of two lower value for the steel plates of Yokogawa Bridge. It is necessary to accumulate more ^{60}Co data at 500–1500 m to discuss the discrepancy between the measured data and the calculation. The systematic discrepancy between the measured data and the calculation at short distances as well as those at long distances must be consistently explained to investigate whether systematic errors exist in the neutron transport calculation and the source-term calculation of the Hiroshima bomb.

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