Chapter 4   Appendix 2

THERMOLUMINESCENCE MEASUREMENT OF GAMMA RAYS

AT 1270 TO 1460 m FROM THE HYPOCENTER

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In 1963, Higashimura et al[1] reported that gamma-ray doses from the Hiroshima and Nagasaki atomic bombs could be measured by the thermoluminescence (TL) method using roof tiles exposed to A-bomb radiations. Later, this method was applied for estimating gamma-ray doses at various locations within 1000 m from the hypocenter in Hiroshima and Nagasaki.[2] These results, as well as values reported by Hashizume et al,[3] agreed with 1965 gamma-ray dose estimates (T65D)[4] suggesting that these were valid in this range both in Hiroshima and Nagasaki. Recently, however, Loewe and Mendelsohn[5,6] and Kerr[7] announced new dose estimates that lead to the view that the T65D estimates should be reappraised. According to the new estimates, the gamma-ray doses in Hiroshima would be between 1.5 and 3 times larger than the T65D values at a distance greater than 1000 m from the hypocenter, while the new estimates and the T65D estimates agree better within 1000 m.

In 1960, Kennedy[8] reported the application of TL measurements to dating of ancient pottery shards. Applying the method of Ichikawa[9] using magnetic separation for extraction of nonmagnetic quartz inclusions from the magnetic clay matrix, Fleming[10] refined the quartz inclusion method and greatly improved the accuracy of TL dating. Fleming's refinement of the quartz inclusion method calls for the use of only large quartz grains, usually about 0.1 mm diameter. Using this method, we have recently estimated gamma-ray doses from the Hiroshima bomb at distances of 1270 to 1460 m from the hypocenter. This article reports the measured values and compares them with the reassessed computed doses.

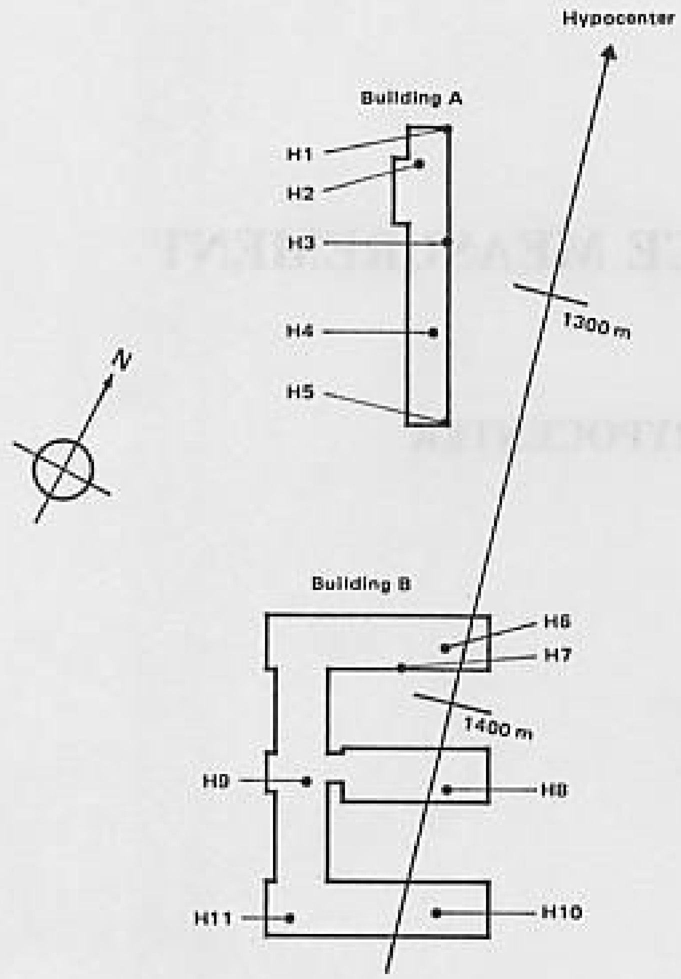

Figure 1. Location of tile samples collected from the roofs of the buildings A and B at Hiroshima University

Materials and Methods

For TL dosimetry, intact tiles were collected from the roofs of the two Faculty of Science buildings at Hiroshima University. The buildings are located in the range from 1270 to 1470 m from the hypocenter. The height of the roof is 12.5 m above the ground for building A and 13.4 m above the ground for building B (Figure 1). Four tiles (one each from H1, H3, H5, and H7 in Figure 1) were taken from the railings, which rimmed the buildings, two tiles (H2 and H4) from the eaves of roofs of cottage-like structures on the roof of building A, and 10 tiles (3 from H6, 4 from H10, and 1 each from H8, H9, and H11) from the floor of the roof of building B. Tiles taken from building A were 2.6 cm thick with a smooth front surface and those from building B were 2.0 cm thick with an uneven ornamental front surface of about 0.1 cm, except H7, which was 2.0 cm thick with a smooth front surface. The location and physical characteristics of the tile samples are summarized in Table 1.

The location and distance from the hypocenter of sample H11 were estimated from the US Army Map Service map, series L902, plate number 138,449 at the Radiation Effects Research Foundation in Hiroshima. This map was previously used for determining the hypocenter location[11] and is being used in the reevaluation of A-bomb doses. The latitude and the longitude of sample H11 were located at the east-west and north-south coordinates 744.61 and 1260.14 (in yards). Distances other than that of sample H11 in Table 1 are based on actual measurements of distances between samples. Incident angles of A-bomb radiation on the surface of the tile samples were estimated using the same map and the height of the epicenter estimated by Hubbell et al.[11]

Table 1. The Location and Physical Characteristics of the Tile Samples Used for TL Measurements

Sample Number	Location	Thickness (cm)	Surface Size (cm×cm)	Texture of the Sample's Surface	Ground Distance from the Hypocenter (m)	Height above the Rooftop (cm)	Angle of the Perpendicular of the Sample's Surface to the Epicenter Line (degrees)
H 1	railing	2.6	36 × 18	smooth	1268	113	65.9
H 2	eaves	2.6	36 × 18	smooth	1277	301	27.2
H 3	railing	2.6	36 × 18	smooth	1295	113	66.4
H 4	eaves	2.6	36 × 18	smooth	1314	330	26.7
H 5	railing	2.6	36 × 18	smooth	1335	113	67.0
H 6	floor	2.0	18 × 18	uneven	1387	0	67.8
H 7	railing	2.0	23 × 6	smooth	1397	95	52.5
H 8	floor	2.0	18 × 18	uneven	1421	0	68.3
H 9	floor	2.0	18 × 18	uneven	1426	0	68.3
H 10	floor	2.0	18 × 18	uneven	1449	0	68.6
H 11	floor	2.0	18 × 18	uneven	1460	0	68.8

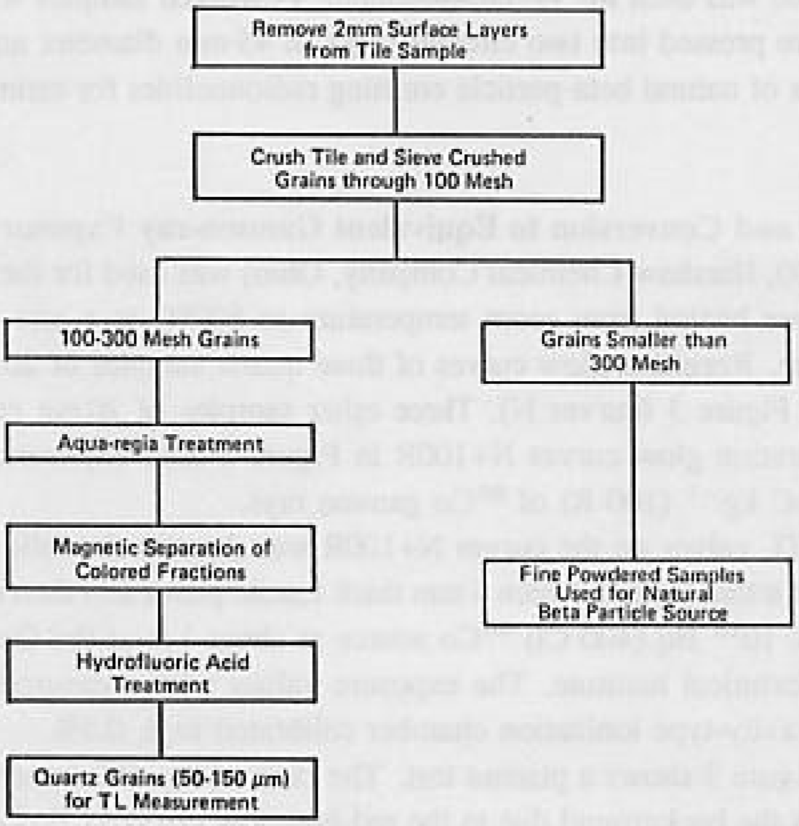

Figure 2. Schematic diagram for extraction of quartz grains from a tile and for preparation of the fine powdered samples used as the source of natural beta particles

Preparation of the Samples

To apply the quartz inclusion technique for A-bomb gamma-ray dosimetry, large quartz grains were extracted from the tiles by essentially the method described by Fleming[10] for TL dating. Figure 2 gives an outline of the method. First, a 2 mm thick layer was removed from the front surface of the tile and 1 mm thick layers from the four sides and the underside of the tile using a diamond saw. This was done to minimize the environmental effect of the surface layer of the tile and to ensure an electron equilibrium condition on the front surface of each tile sample. Then, the tile was cut into small pieces and crushed into grains by a rolling jaw crusher. The crushed grains were sieved into three groups with sizes of 100 to 200, 200 to 300, and 300 mesh or smaller. Quartz particles were extracted from the 100 to 300 mesh grains in the following way. The powdered grains were subjected to ultrasonic cleaning, first in water and then in acetone. In order to partially dissolve mineral particles, the grain samples were treated with aqua regia at room temperature for 24 hours. Of the undissolved particles, colored, magnetic particles were separated from colorless, nonmagnetic particles using an isomagnetic-type magnetic separator (Nihon Chikagaku Sha Company, Kyoto). The resultant colorless particles were treated with 10% hydrofluoric acid for one hour to remove residual nonquartz materials and to etch mildly the surface of each quartz particle.

The diameter of quartz grains extracted from the crushed particles at sizes 100 to 200 mesh (from samples H1 to H5) and those at sizes 200 to 300 mesh (from samples H6 to H11) were 74 to 150 μm and 50 to 74 μm, respectively. A major fraction of the quartz grains from each tile was used for TL measurement. Powdered samples which were smaller than 300 mesh were pressed into two circular disks of 45 mm diameter and 3 mm thickness and used as sources of natural beta-particle emitting radionuclides for estimating background exposure.

TL Measurement and Conversion to Equivalent Gamma-ray Exposure. A Harshaw TL Reader (Model 2000, Harshaw Chemical Company, Ohio) was used for the TL measurement. Quartz samples were heated from room temperature to 500°C at a rate of 20°C s^{-1} in a nitrogen atmosphere. Resultant glow curves of three quartz samples of 20 mg each from tile H10 are shown in Figure 3 (curves N). Three other samples of 20 mg each were used for obtaining the calibration glow curves N+100R in Figure 3 after exposure to the calibration exposure of 25.8 mC kg^{-1} (100 R) of ^{60}Co gamma rays.

Calibration of TL values on the curves N+100R was done in the following way. Quartz samples were put in a thin layer between 4 mm thick Lucite plates and then exposed to gamma rays from a 1.48×10^{13} Bq (400 Ci) ^{60}Co source at about 1 m at the Osaka Branch of the National Electro-Technical Institute. The exposure values were measured with a secondary national standard cavity-type ionization chamber calibrated to $\pm$ 0.5%.

The insert in Figure 3 shows a plateau test. The ratio of T_N (TL intensity in an unirradiated sample) minus the background due to the red-hot glow (BG) to T_{N+100R} (TL intensity in the ^{60}Co gamma-ray irradiated sample) minus T_N is plotted against heating temperature. Following Aitken and Fleming,[12] and Aitken,[13] limiting the integration to the plateau region, the ratio of the area under the glow curve N to that under $N + 100R$ minus N was used for calculating the ^{60}Co gamma-ray equivalent exposure in the sample using

$$D_T = (T_N - BG) / (T_{N+100R} - T_N) \times 25.8\,\text{mC kg}^{-1} \quad (1)$$

The relationship between TL intensity and ^{60}Co gamma-ray exposure was examined, using a portion of the quartz samples from each of the tiles. For example, Figure 4 shows the

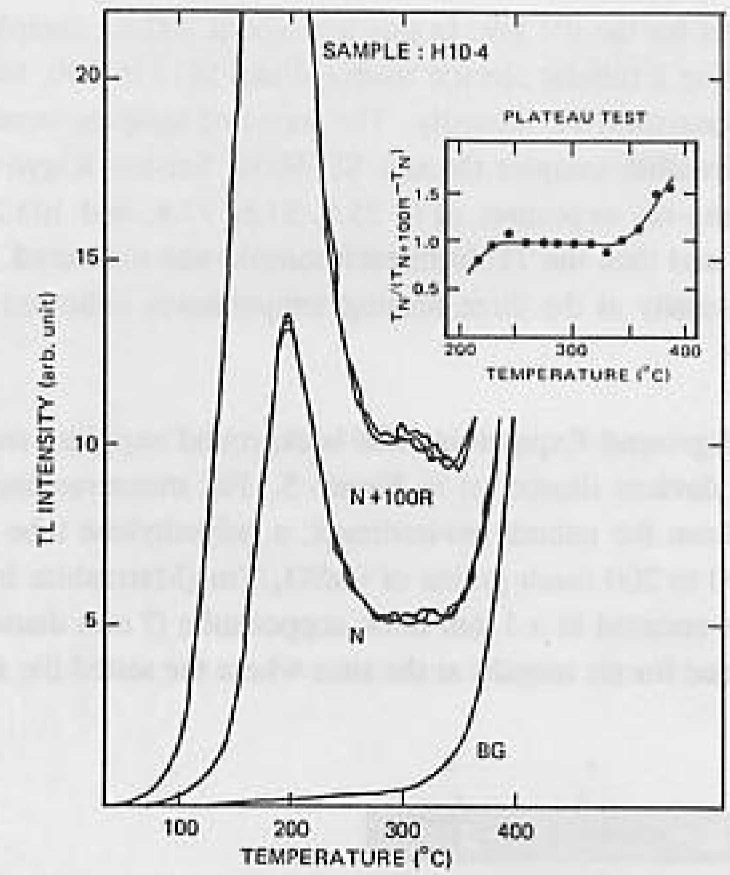

Figure 3. Thermoluminescence glow curves and a plateau test. N: TL induced by natural and A-bomb radiations. N+100R: TL induced by natural and A-bomb radiations plus ^{60}Co gamma rays for calibration. BG: incandescent glow

Figure 4. Test for linearity relation between TL intensities and exposures at three temperatures in the plateau region

results of a linearity test for the tile H9. In this test, about 300 mg samples were annealed at 500°C for one hour using a tubular electric oven (Model MTS16-740, Motoyama Company, Osaka) to eliminate preexisting TL intensity. The annealed samples were separated into four equal aliquots by a turntable sampler (Model SU-5000, Seishin Kigyo Company, Tokyo), exposed to ^{60}Co gamma-ray exposures of 0, 25.8, 51.6, 77.4, and 103.2 mC kg^{-1} (0, 100, 200, 300, and 400 R), and then the TL from each sample was measured. The relationship of exposure and glow intensity at the three heating temperatures indicated in Figure 4 exhibit linearity within ± 5%.

Measurement of Background Exposures. The background exposure from natural radiation was estimated by the devices illustrated in Figure 5. For measurement of gamma-ray and cosmic-ray exposure from the natural environment, a polyethylene tube (5 mm diameter × 30 mm) containing 100 to 200 mesh grains of $CaSO_4$:Tm (Matsushita Industrial Equipment Company, Osaka) was encased in a 1 mm thick copper tube (7 mm diameter × 10 cm). The copper tubes were placed for six months at the sites where the tested tile samples had existed.

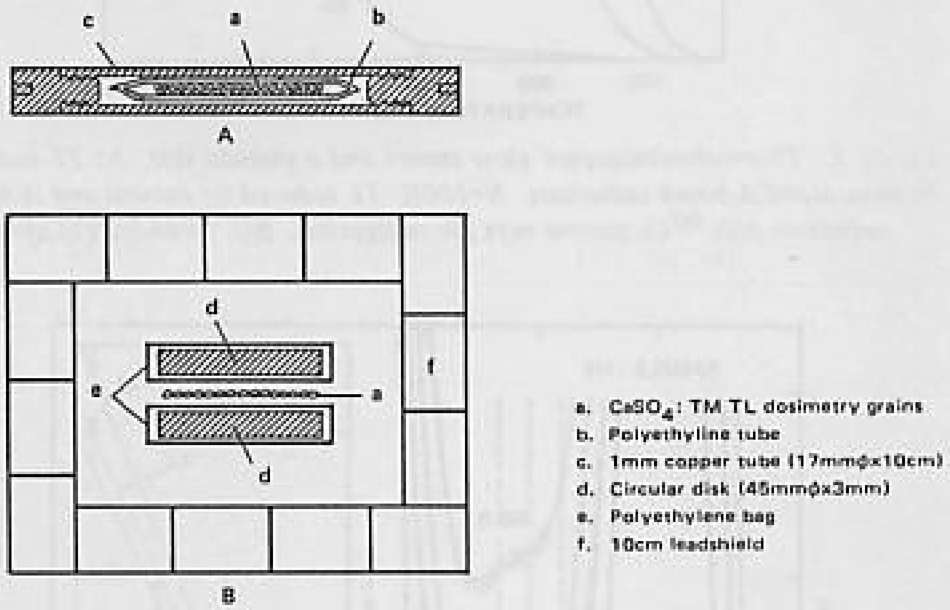

Figure 5. Cross section diagram of the devices used for evaluation of background exposure of natural gamma rays from outside the tile samples (A) and of natural beta particles within the tile samples (B)

For measurement of beta-particle exposures from natural radionuclides inside tile samples, residual parts of crushed samples at sizes smaller than 300 mesh were used as the source of natural beta rays (Figure 2). An appropriate amount of the fine crushed tile samples were encased in an aluminum ring, placed on a hard metal plate, and then pressed at a pressure of 62.5 Pa into a circular disk-like form. The two disks thus prepared were each covered with a polyethylene sheet of 3.5 mg cm^{-2} thickness to absorb alpha particles from natural radionuclides within the disks. The $CaSO_4$:Tm grains were spread in a thin, uniform layer between the two disks (Figure 5). This natural beta-particle dosimetry set was encased in a box with 10 cm lead walls and the sample left in place for one month.

Attenuation of natural beta particles in the polyethylene sheet used for alpha-particle attenuation was estimated from an empirical curve. The curve was constructed from the decrease in TL intensity in $CaSO_4$:Tm grains that were sandwiched together between polyethylene sheets and between two geochemical reference rock samples (JG-1 granodiorite, Geological Survey, Japan)[14] with increasing thickness of the polyethylene sheets. The sheets were inserted between each of the standard samples and the thin layer of dosimeter grains. The attenuation factor obtained for the polyethylene sheet was 1.13.[15]

Results

The data obtained for natural background exposures are listed in Table 2. The values for beta-particle dose ("exposure") rate listed in Table 2 were calculated from TL values of $CaSO_4$:Tm grains by making correction for attenuation in the polyethylene shielding-sheet (Figure 2). Samples H1 to H5 were taken from building A, which was built in 1938, and samples H6 to H11 from building B, which was built in 1931 (Figure 1). Assuming that the age of the tile samples taken from these buildings is equal to the age of the buildings (Table 2), the total background exposure was obtained by multiplying the measured background exposure rate by the tile age.

Table 2. Background Radiation Exposures for Ornamental Tiles from Hiroshima University

Tile Samples	Exposure Rate		Age (y)	Total Background Exposure ($mC \cdot kg^{-1}$)
	Beta Particle[a] ($mC \cdot kg^{-1} \cdot y^{-1}$)	Gamma Ray[b] ($mC \cdot kg^{-1} \cdot y^{-1}$)		
H1 - H 5	0.064 ± 0.002[b]	0.021 ± 0.001[d]	45 ± 1	3.8 ± 0.2
H6 - H11	0.070 ± 0.002[c]	0.023 ± 0.001[e]	52 ± 1	4.8 ± 0.2

[a]The annual rates are estimated from experimental exposures induced during 1 to 6 months.
[b]From six tiles.
[c]From four tiles.
[d]From three locations.
[e]From three locations.

The measured values of TL intensities of quartz grains extracted from 16 tiles taken from the two buildings at Hiroshima University are summarized in Table 3. The TL intensities are given in terms of equivalent exposure after calibration by irradiation of each quartz sample with an appropriate ^{60}Co gamma-ray exposure. The equivalent exposures of TL intensities in tile samples in Table 3 are mean values of triplicate TL intensities with the standard deviation estimated from variation in the triplets. The location of the 16 tile samples is indicated in Figure 1. The distance from the hypocenter to sites where these tiles were located are given in the second column of Table 3. Exposure for natural background is from Table 2. The last column gives net exposure in $0.258\,mC\,kg^{-1}$ (R).

Discussion

Sixteen tiles from the roof of the two buildings of Hiroshima University were collected as the best materials available for TL dosimetry of gamma rays from the Hiroshima A-

Table 3. TL Measurements of Radiation Exposures in Ornamental Tiles Exposed to the Hiroshima A-bomb at Distances of 1270 to 1460 m from the Hypocenter

Sample Number	Ground Distance (m)	Total Exposure[a] (mC·kg⁻¹)	Background Exposure (mC·kg⁻¹)	Net Exposure (mC·kg⁻¹)	(R)
H 1	1268	43.1 ± 2.1	3.8 ± 0.2	39.2 ± 2.3	152 ± 8.8
H 2	1277	44.4 ± 3.9	3.8 ± 0.2	40.5 ± 4.1	157 ± 15.8
H 3	1295	43.3 ± 5.7	3.8 ± 0.2	39.5 ± 5.9	153 ± 22.8
H 4	1314	34.6 ± 6.5	3.8 ± 0.2	30.7 ± 6.7	119 ± 25.8
H 5	1335	31.5 ± 4.1	3.8 ± 0.2	27.6 ± 4.3	107 ± 16.8
H 6-1	1387	28.1 ± 4.4	4.8 ± 0.2	23.2 ± 4.6	90 ± 17.9
H 6-2	1387	27.6 ± 1.3	4.8 ± 0.2	22.7 ± 1.5	88 ± 5.9
H 6-3	1387	30.2 ± 1.5	4.8 ± 0.2	25.3 ± 1.7	98 ± 6.9
H 7	1397	26.3 ± 3.1	4.8 ± 0.2	21.4 ± 3.3	83 ± 12.9
H 8	1421	24.3 ± 1.5	4.8 ± 0.2	19.4 ± 1.7	75 ± 6.9
H 9	1426	21.9 ± 1.5	4.8 ± 0.2	17.0 ± 1.7	66 ± 6.9
H 10-1	1449	26.1 ± 3.4	4.8 ± 0.2	21.2 ± 3.6	82 ± 13.9
H 10-2	1449	24.8 ± 1.8	4.8 ± 0.2	19.9 ± 2.0	77 ± 7.9
H 10-3	1449	23.0 ± 2.8	4.8 ± 0.2	18.1 ± 3.0	70 ± 11.9
H 10-4	1449	24.8 ± 1.8	4.8 ± 0.2	19.9 ± 2.0	77 ± 7.9
H 11	1460	18.1 ± 1.5	4.8 ± 0.2	13.2 ± 1.7	51 ± 6.9

[a]Mean values of triplicate TL intensities with the standard deviation estimated from variations in the triplets.

bomb for the following reasons: (1) the buildings are loacted between 1270 and 1470 m from the hypocenter without any shielding between the epicenter and the buildings, (2) the collected tile samples can be accurately located, (3) the tile samples collected had not been significantly heated by the explosion of the bomb or by other causes, (4) the recorded dates of construction of the buildings can be used as good estimates of the dates when the tiles were kiln fired as the tiles used would have been produced shortly before construction of the buildings, and (5) buildings A and B are old, but still exist, and many tiles were collected from an open area of the floor of the roof, the railing of the roof, and the eaves of a small structure on the roof, but not from the walls of the buildings to avoid possible shielding effects of neighboring buildings and possibly of fading effects due to outbreak of fire within the buildings. Of the 16 tile samples, 3 (H6-1 to H6-3) were collected from one site, and 4 (H10-1 to H10-4) from another site (Figure 1). In terms of standard errors, variations in the measured exposure values were 5% of the mean for the three H6 tile samples and 6% of the mean for the four H10 tile samples (Table 4).

It should be emphasized that quartz particles extracted from the collected samples showed good reproducibility in emission of TL intensity as is seen from the values of standard errors listed in Table 2 that represent variation of triplicate measurements for each tile (Figure 3). Furthermore, the relation between TL intensity and exposure of ^{60}Co gamma rays was linear for quartz particles extracted from all 16 tiles, as is illustrated in the case of the H9 tile (Figure 4).

However, various uncertainties in regard to the absolute values of estimated gamma-ray exposures exist in the present TL measurements. The contribution of background radiations is one of the major uncertainties. The estimated exposure (Table 2) was as large as 10 to

Table 4. Comparison of Experimental and Theoretical Estimates of Gamma-ray Doses from the Hiroshima A-bomb

Ground Distance (m)	Dose Estimates		A/B
	TL Data[a] (Gy) A	Loewe's Data[b] (Gy) B	
1268	1.44	1.23	1.17
1277	1.49	1.18	1.26
1295	1.45	1.10	1.32
1314	1.13	1.02	1.11
1335	1.02	0.93	1.09
1387	0.87 ± 0.50[c)]	0.75	1.17 ± 0.06[c]
1397	0.79	0.73	1.08
1421	0.71	0.66	1.08
1426	0.63	0.64	0.98
1449	0.73 ± 0.05[d)]	0.58	1.26 ± 0.06[d]
1460	0.48	0.56	0.87
Average			1.15 ± 0.12

[a] From the values in sixth column of Table 3 after multiplying 3.68×10^{-2} $Gy \cdot mC^{-1} \cdot kg$ (see text).
[b] From Loewe[22]
[c] From three tiles (see Table 3).
[d] From four tiles (see Table 3).

27% of the total exposure (Table 3). Quartz grains as large as 50 to 150μm were used for TL measurement after their surface layers were removed by treatment with hydrofluoric acid. Therefore, the contribution of alpha particles to the TL intensity was made negligible.[10,16] Thus, major contributors to the background exposure in the quartz grains were natural gamma rays from outside the tile sample and beta particles from natural radionuclides in the surrounding environment of each quartz particle. The dosimetry of natural gamma and beta rays was carried out by the method routinely used for TL dating (Figure 5), in which $CaSO_4$:Tm phosphors were used instead of quartz grains.[15]

From experience in TL dating of earthenware by measurement of TL intensities induced by natural radiation, the errors in the present estimates of background exposure would be less than ± 10%. In fact, TL dating of roof tiles from a building with negligible A-bomb radiation at 3400 m from the hypocenter gave an estimated age of 67.5 years against the actual age of 71 years,[15] indicating only a 5% error. Furthermore, using a slightly different method, Maruyama et al[17] obtained a value of 0.338 rad y^{-1} for the annual background dose in a tile from Hiroshima University. This value is almost identical to our estimates (Table 2).

Quartz grains extracted from tile samples were calibrated by ^{60}Co gamma-ray irradiation under conditions of secondary electron equilibrium achieved by placing the quartz samples between 4 mm thick Lucite plates. As an attempt to simulate the actual exposure conditions, one of the 2.6 cm thick tile samples was annealed at 500°C for 24 hours and then exposed to ^{60}Co gamma rays. Quartz grains extracted from the front surface layer (2 mm thick) and those from four successive layers (6 mm thick) were subjected to TL measurements. From the results obtained on the TL intensity with depth, it was estimated that reduction in

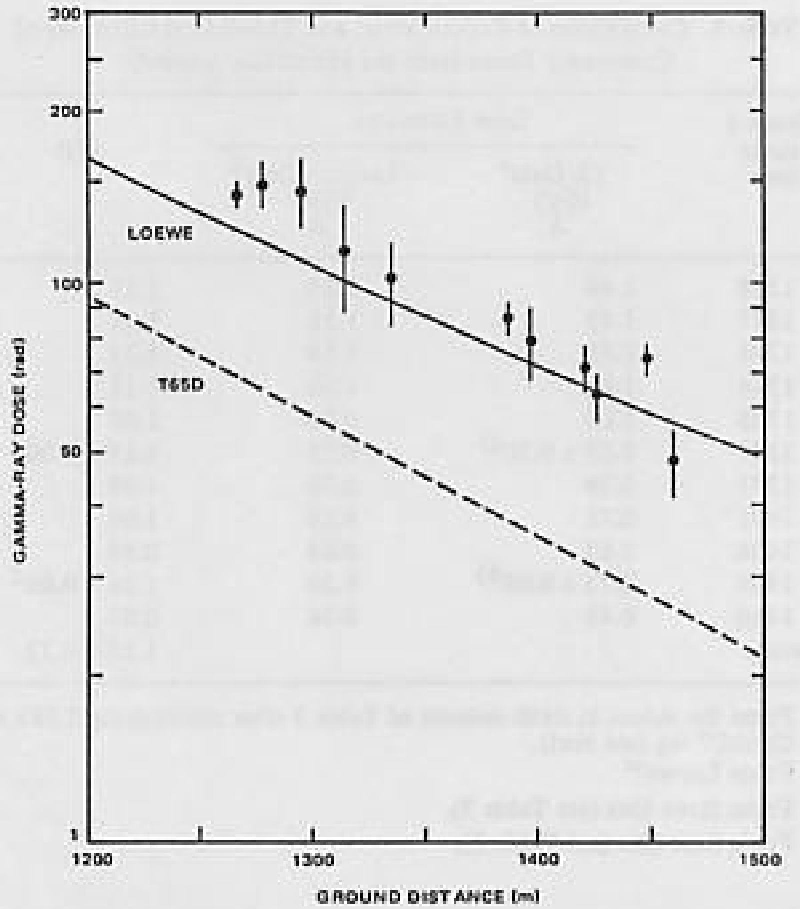

Figure 6. Measured values versus ground distance from hypocenter. Each value was obtained using one tile sample except those at 1387 m (three tiles) and 1449 m (four tiles)

the average TL intensity due to self-shielding is 1.05 for the 2.6 cm thick tile and 1.02 for the 2.0 cm thick tile.[15] These factors may be used as correction factors for filling the gap between the conditions for calibration irradiation and the actual conditions of exposure to A-bomb radiations. However, the tile samples had been exposed to A-bomb radiation while placed on the concrete blocks of the buildings. Therefore, the actual buildup effects of A-bomb gamma rays in the tiles could have been larger than those in the tiles exposed to ^{60}Co gamma rays without backscatter of concrete blocks. Furthermore, the intensity of gamma rays from the Hiroshima A-bomb was highly directional with most coming from the direction parallel to the line connecting the buildings, from which the samples were taken, with the hypocenter.[18] Thus, the major fraction of the A-bomb gamma rays must have impinged at large slant angles on the tiles on the floor of the roof of the Hiroshima University building. These problems should be studied in more detail.

Any contributions of fast neutrons from the A-bomb to TL intensities of tile samples seems negligible as judged from the work of Haskell et al.[19] Dependence of TL reponse of quartz particles on the energy of gamma rays should also be taken into consideration. The TL response of quartz to gamma rays at 40 and 100 keV are 3 to 4 and 1.2 to 2 times as large as that to ^{60}Co gamma rays, respectively.[17,20,21] Fortunately, the fluences of such low-energy gamma photons in A-bomb radiation were much smaller than those of photons of 1 MeV or higher energy.[18] Nevertheless, one needs to know the magnitude of the contribution of low-energy photons to TL intensities.

In spite of the various uncertainties discussed above, it is interesting to note that the present estimates of 23.7 ± 1.3 mC kg^{-1} (92 ± 5 R) from three tiles at ground distance 1387 m and 19.9 ± 1.3 mC kg^{-1} (77 ± 5 R) from four tiles at ground distance 1449 m are close to the computed estimates, 75 and 58 rad in tissue, respectively, that are based on the report by Loewe[22] (Table 4), when we consider that 0.258 mC kg^{-1} (1 R) of ^{60}Co gamma rays is about 0.95 rad in tissue (i.e., 3.68×10^{-2} Gy kg mC^{-1}).[23] The measured values are shown in Figure 6 compared with the computed estimates of Loewe[22] and the T65D estimates. The ratios of the present estimates to the corresponding computed doses by Loewe[22] are in the range of 0.83 to 1.32 with a mean value of 1.15 ± 0.12 (Figure 6). On the other hand, the T65D values[4] at ground distances 1387 and 1449 m are lower than our exposure values by a factor of about 2.5.

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