Chapter 8

ACTIVATION MEASUREMENTS FOR THERMAL NEUTRONS

Part A. Cobalt (^{60}Co) Activation

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Introduction

Studies of cobalt activation are an important part of the history of radiation dosimetry for the Hiroshima and Nagasaki survivors. The first ^{60}Co measurements made by Saito (1961) were found to be important in estimating the radiation doses to early entrants into the hypocenter area of the two cities (Arakawa 1962), and the ^{60}Co activation studies by the Japanese National Institute Radiological Sciences (JNIRS) in the mid-1960s have been important in the estimation of radiation doses for all survivors in the two cities (Hashizume et al. 1967; Hashizume and Maruyama 1975; Hashizume 1983). The large discrepancy found between ^{60}Co measurements and calculations during the DS86 studies (Loewe 1985; Loewe et al. 1987) was a major impetus for subsequent neutron activation measurements and calculations, including the current DS02 reassessment.

While ^{60}Co radioactivity can be induced in stable cobalt (^{59}Co) by neutrons of all energies, it is mainly due to thermal neutrons (Figure 1). There is also a large resonance capture of epithermal neutrons at approximately 100 eV that is important in the activation of some of the more heavily shielded samples. The cross section for activation of cobalt is well known (Figure 2), and it has a relatively simple decay scheme. Cobalt-60 has a half-life of 1925.12 ± 0.46 days or approximately 5.271 years (Unterweger et al. 1992), and it decays over 99.9% of the time by the emission of a beta particle (E_{max}= 318 keV) and two gamma-rays with energies of 1173 and 1333 keV (International Commission on Radiological Protection 1983). The early 1960 studies by Saito (1961) and JNIRS (Hashizume et al. 1967; Kawamura et al. 1967) counted beta particles from the decay of ^{60}Co in their studies, while more recent studies have counted the 1173- and 1333-keV gamma rays emitted during the decay of ^{60}Co (Hoshi and Kato 1987; Kerr et al. 1990;

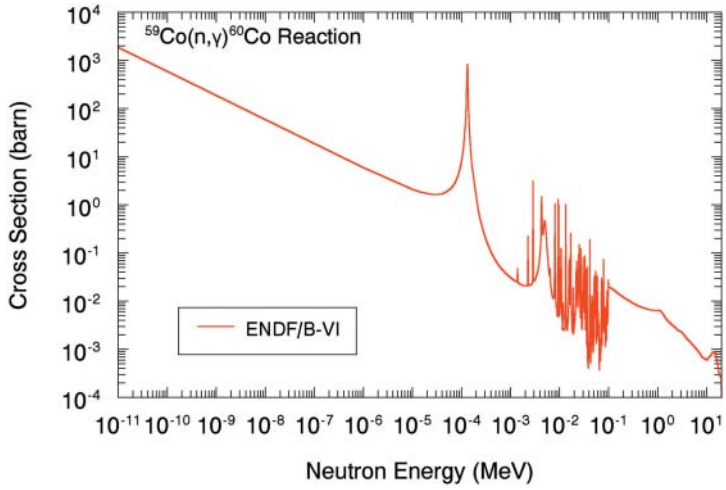

Figure 1. *Energy dependence of the cross sections for the $^{59}Co(n,\gamma)^{60}Co$ reaction (McVane 2001)*

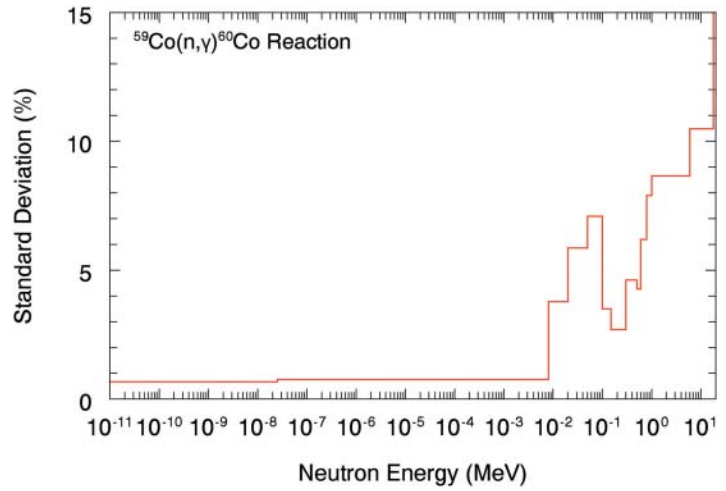

Figure 2. *Energy dependence of the uncertainty in the cross sections for the $^{59}Co(n,\gamma)^{60}Co$ reaction (Remec 2002)*

Kimura et al. 1990; Kimura and Hamada 1993; Komura 2001a; Nakanishi et al. 1987, 1991; Okumura and Shimasaki 1997; Shizuma et al. 1992, 1993, 1997, 1998, 2002).

In Hiroshima, there are now over 104 measurements of cobalt activation in 43 samples taken from 35 different sample sites, and in Nagasaki, there are over 32 measurements of cobalt activation in 24 samples from 24 different sites. The types of samples include rocks, roof tiles, steel plates from bridges, and numerous iron and steel samples from buildings such as steel

reinforcing rods embedded in concrete, pipes, handrails, guttering, and iron rings from the roofs of the buildings. The iron rings were used to secure ropes for purposes of routine building maintenance and for escape from the buildings in emergencies. The widely varying kinds of samples used in the measurements make it difficult to analyze the extensive cobalt activation data from the many different studies.

DS86 Neutron Discrepancy

The primary source of data for the testing of the DS86 calculations for neutrons was the JNIRS measurements of ^{60}Co activation in steel reinforcing bars (rebars) located at depths of 6-10 cm in concrete (Table 1) and the iron rings from the roofs of large buildings (Table 2). The steel rebars and iron rings were collected at locations that were unshielded in the line-of-sight direction toward the bomb (i.e., line-of-sight samples). A major disappointment of the DS86 dose reassessment effort was the poor agreement between the measurements and calculations for ^{60}Co activation (Loewe et al. 1987). At Hiroshima, the measured-to-calculated (M/C) ratios were less than one at slant ranges of less than approximately 750 m (or ground ranges of about 500 m) and greater than one at slant ranges of more than 750 m as shown in Figure 3. This suggested that the neutron doses were overestimated close to the bomb and underestimated at the larger distances of most interest in survivor dosimetry. The steel rebar calculations by Loewe (1985) were later confirmed by Dolatshahi et al. (1987).

Table 1. Summary of data on cobalt activation in steel rebar samples from report by Hashizume et al. (1967)

City and sample location[a]	Ground range (m)[b]	Sample activity (cpm/g of Co)	Decay correction[c]	Initial activity (Bq/mg of Co)[d]
Hiroshima				
Hiroshima Bank	260	2.09 ± 0.17	12.90	3.74 ± 0.305
Sentry Box	640	0.324 ± 0.0204	12.90	0.58 ± 0.0366
Water Trough for Horses	779	0.146 ± 0.0013[e]	12.91	0.262 ± 0.0233
Powder Magazine	1180	0.0124 ± 0.0023	12.93	0.0223 ± 0.0041
Nagasaki				
Nagasaki University Hospital	590	0.140 ± 0.0066	12.90	0.251 ± 0.0118
Municipal Commercial H.S.	1030	0.0127 ± 0.0010	13.47	0.0238 ± 0.0019

[a]Additional information on sample location is provided by Maruyama (1987). The samples were steel reinforcing rods (rebars) in concrete at a depth of 6-10 cm from the concrete surface.
[b]Ground ranges as originally reported by Hashizume et al. (1967).
[c]Derived using Equation 1 and data in Table 1 from Hashizume et al. (1967).
[d]Detection efficiency for cobalt deposited on a planchet at 5 mm from the end of thin-window GM counter used in the measurements was 12% (Hashizume et al. 1967).
[e]The uncertainty for this measurement is reported as 0.89% compared to 4.7 to 18.4% for the other rebar measurements at Hiroshima and Nagasaki. Data from the original counting logs indicate the uncertainty for this value should be approximately 8.9% (i.e., 0.013 cpm/mg of Co).

Table 2. Summary of data on cobalt activation of iron ring samples from roofs of buildings in Hiroshima (Hashizume 1983)

Sample location	Ground range (m)[a]	Sample activity (cpm/mg of Co)	Initial activity (Bq/mg of Co)[b]
Honkawa Primary School	373	2.445	4.38
Fukuromachi Primary School	441	1.840	3.30
Kirin Brewery	650	0.548	0.982
Kodokan Building	727	0.298	0.534
Hiroshima City Hall	997	0.064	0.115

[a]Ground ranges as originally reported by Hashizume (1983).
[b]See Table 4 of Shizuma et al. (1992) and Table 16 of Kerr et al. (1990). The sample activity at time of measurements was converted to an initial activity at time of bombing by the use of data from the rebar measurements of Hashizume et al. (1967). These conversions assumed a counting efficiency of 12% (Hashizume et al. 1967) and a decay correction of 12.9 (Table 1).

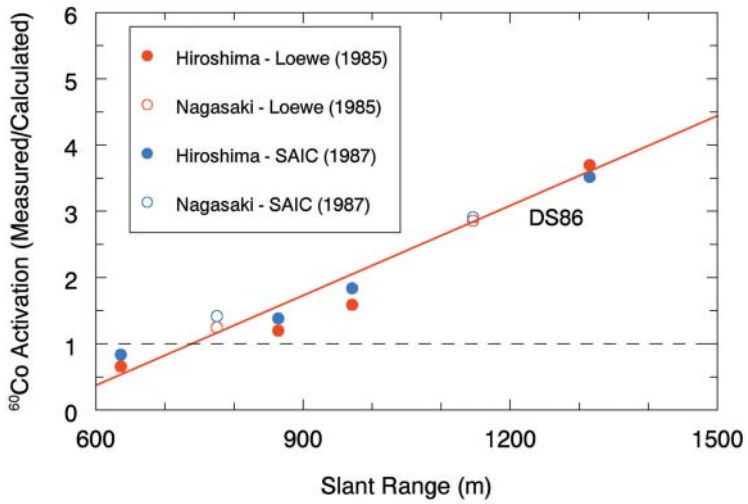

Figure 3. *Measured-to-calculated (M/C) ratios for ^{60}Co activation at Hiroshima and Nagasaki from Loewe (1985) and SAIC (Dolatshahi et al. 1987). A 580-m burst height and 15-kt yield were used in the DS86 calculations for Hiroshima, and a 503-m burst height and 21-kt yield were used in the DS86 calculations for Nagasaki.*

As a check of the JNIRS rebar measurements, it was decided to make newer measurements of ^{60}Co activation in two line-of-sight samples collected previously by the RERF. These samples were a handrail from a smokestack on the Chugoku Electric Company and a section of a steel girder from the top of the Yokogawa Bridge (Kerr et al. 1990; Kimura et al. 1990). The Yokogawa Bridge sample was taken at a somewhat larger ground range than the rebar samples previously reported by JNIRS. Pieces of the two samples were distributed by the RERF to the

Oak Ridge National Laboratory (ORNL) and the Japan Chemical Analysis Center (JCAC) for analysis. The results of both the ORNL measurements (Kerr et al. 1990) and the JCAC measurements (Kimura et al. 1990; Kimura and Hamada 1993) were found to be consistent with the JNIRS measurements of ^{60}Co activation in the iron rings from the roofs of buildings (Table 2). As shown in both Figures 3 and 4, the measured-to-calculated (M/C) ratios were always less than one at smaller slant ranges and always greater than one at the larger slant ranges. Thus, it appeared that there was a systematic uncertainty in DS86 neutron calculations and the neutron doses at a slant range (or ground range) of about 1,500 m were underestimated by a factor of two to five. Later thermal neutron measurements suggested that the calculated neutron doses at Hiroshima were too small by a distance-dependent factor ranging up to as much as ten or more at a ground distance of 1,500 m (Shizuma et al. 1992; Straume et al. 1992; Straume 1994; Shizuma et al. 1998).

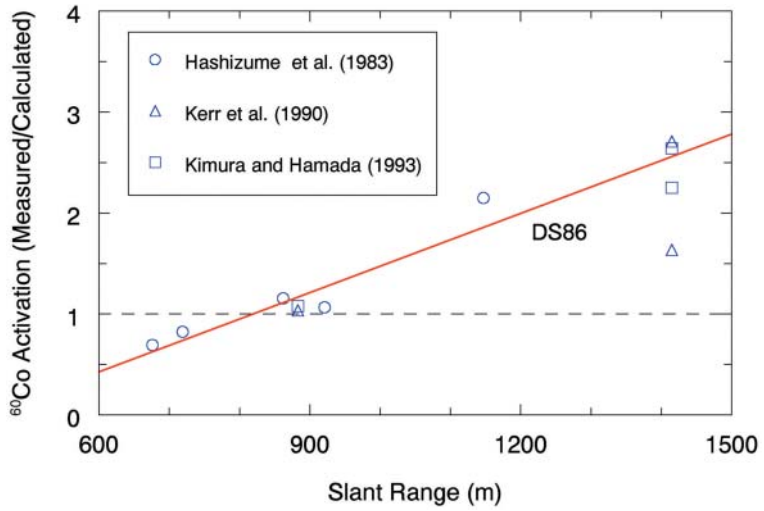

Figure 4. *Measured-to-calculated (M/C) ratios for ^{60}Co activation at Hiroshima from Kimura and Hamada (1993), Kerr et al. (1990), and Hashizume (1983). A 580-m burst height and 15-kt yield were used in the DS86 calculations for Hiroshima.*

Measurements Database for Cobalt Activation

As part of the DS02 reassessment, the RERF established a measurements database for both Hiroshima and Nagasaki (Chapter 6). The ^{60}Co information in this database was obtained from the literature, papers presented at meetings and workshops, and information provided by the measurers. The ^{60}Co data from the different studies are summarized in this section, and the ground ranges of the various ^{60}Co samples are updated using new map evaluation tools developed during the DS02 reassessment effort (Chapter 5).

Hashizume et al. (1967), Hashizume (1983)

The samples used in these JNIRS studies were collected in 1964 by personnel of the Field Investigation of the Atomic Bomb Casualty Commission (ABCC), now the Radiation Effects Research Foundation (RERF), and the specific ^{60}Co radioactivity in samples was measured by JNIRS in 1965. A limited number of these measurements, confined to those in steel rebars embedded in concrete at depths of 6-10 cm, were reported in 1967 in ABCC Technical Report TR 6-67 and in *Health Physics* (Hashizume et al. 1967). In 1983, an additional set of the original 1965 measurements, but of iron rings from the roofs of buildings, was reported in a dosimetry workshop held at RERF (Hashizume 1983). Finally, the rebar measurements first reported in 1967 were reported again in the DS86 Final Report by Loewe et al. (1987) and by Maruyama and Kawamura (1987). The latter publication by Maruyama and Kawamura provides some additional data on the locations of the rebar samples at Hiroshima.

Our reanalysis of the published JNIRS counting data is presented in Table 3. The original detailed JNIRS counting logs from 1965 were obtained and provided the basis for the reanalysis presented here. The original JNIRS counting logs were used: (1) to check the previously published values for the steel-rebar samples because it was noticed that the uncertainty of one of the measurements was extremely small compared to those of the other rebar-sample measurements (Table 1); (2) to derive better specific activity values for the ^{60}Co measurements of the iron ring samples (Table 2); and (3) to use the JNIRS counting logs along with other available information to make better estimates of the ground ranges and their uncertainties for both the steel-rebar and iron-ring samples (Table 3). The other available information for estimating the ground ranges of the various samples were as follows: (1) internal RERF reports prepared in

Table 3. Summary of current reassessment studies of data on cobalt activation from 1965 JNIRS studies by Hashizume et al. (1967) and Hashizume (1983)

City and sample location	Material	Ground range (m)	Sample height (m)	Initial activity (Bq/mg of Co)[a]
Hiroshima				
Hiroshima Bank	Rebar	257 ± 17	20	3.74 ± 0.305
Honkawa Primary School	Iron ring	408 ± 17	12	4.80 ± 0.79
Fukuromachi Primary School	Iron ring	468 ± 26	12	3.62 ± 0.10
Sentry Box	Rebar	683 ± 23	2.5	0.581 ± 0.037
Kirin Beer Hall	Iron ring	690 ± 18	14	1.08 ± 0.05
Kodokan Building	Iron ring	748 ± 17	15	0.586 ± 0.020
Watering Trough for Horses	Rebar	779 ± 53	0.6	0.262 ± 0.023
Hiroshima City Hall	Handrail	998 ± 17	15	0.126 ± 0.012
Powder Magazine	Rebar	1178 ± 81	4.3	0.0223 ± 0.0041
Nagasaki				
Nagasaki University Hospital	Rebar	613		0.251 ± 0.0118
Municipal Commercial H.S.	Rebar	1042		0.0238 ± 0.0019

[a]The iron-ring samples and the handrail sample from City Hall are about 10% greater than the values given in Table 2 because the assumed decay correction of 12.9 in Table 2 was too small for these sample measurements.

1981 by Hiroaki Yamada and 1985 by Tadaaki Watanabe; (2) a 1985 letter to J. V. Pace III from G. D. Kerr with various attachments including drawings and other excerpted materials from the U.S. Strategic Bombing Survey Report (1947) and other sources (Kerr 1985); and (3) the discussion in the DS86 Final Report by Maruyama and Kawamura (1987).

In the JNIRS measurements, the cobalt was chemically separated from 50-g samples of the steel rebars and iron rings as discussed by Hashizume et al. (1967) and Kawamura et al. (1967). The radioactivity of the ^{60}Co in the chemically separated Co from a steel or iron sample was determined by electroplating the Co onto a planchet, and the beta particles emitted during the radioactive decay of the ^{60}Co were counted with a thin-window GM tube. The GM tube was surrounded by a large scintillation counter and the two were operated in coincidence to minimize the background in the GM tube from cosmic rays (Hashizume et al. 1967). The background count rate in the GM tube was determined to be 0.069 ± 0.002 cpm (or approximately 1 count every 15 minutes). The method of determining the amount of stable Co in each chemically separated sample is well documented by Kawamura (1967). The uncertainty estimate is based on the standard deviation of five repeated measurements in an aliquot of the sample prior to electroplating, using an absorptiometric method with o-nitroresorcin monomethyl ether in the solution, and the calculated amount of Co in the solution to be used for electroplating was based on the concentration so determined and the volume of the remaining solution. The amount of Co on a planchet and its standard deviation, and the counts per minute (cpm) of ^{60}Co and its standard deviation were recorded in the log. The original JNIRS counting log was used to check the cpm/mg values and their uncertainties, which were estimated by combining the uncertainties for the cpm and mg values in quadrature. An error found in the calculated uncertainty for the rebar sample from the watering trough for horses was corrected in this analysis (see footnote e to Table 1).

Prior to the publication of the DS86 Final Report, there was a series of communications with RERF in 1981 and 1985, involving requests by JNIRS to recalculate the ground distances for the rebar samples. Although these requests in the 1980s were motivated by considerations of suggested changes in the location of the hypocenter at Hiroshima, they revealed some previous inconsistencies in information about sample locations for the rebars and resulted in reports being prepared by Hiroaki Yamada in 1981 and Tadaaki Watanabe in 1985. The ground distances based on these reports with the exception of the Hiroshima Bank are accompanied by unusually large uncertainty because: (1) the samples were collected in the 1960s from the military areas in Motomachi that were heavily damaged by the bomb; (2) the sample locations were not very well documented at the time of sample collection and did not involve large distinct structures that were well represented on the U.S. Army map; and (3) the areas of sample collection were extensively redeveloped by the time of the new city map in 1979. The sample locations of the iron rings were reasonably well documented for all of the samples and the specific locations of the iron rings from the tops of buildings were well defined for the Honkawa Primary School, Kodokan Building, and Hiroshima City Hall (Kerr 1985). The specific location of the rebar sample from the Hiroshima Bank was also well defined in the report by Maruyama and Kawamura (1987).

Kerr et al. (1990)

Steel samples from the Yokogawa Bridge located at ground range of 1,283 m and the Chugoku Electric Company located at a ground range of 692 m were used in 1988-1989 studies by Kerr et al. (1990). The steel sample from the Chugoku Electric Company was a flat piece of steel cut

from a stand at the top of the smokestack when the building was demolished in 1985, and the steel samples from the Yokogawa Bridge consisted of two steel plates that were cut from a section of the top arch on the east side of the bridge when it was demolished in 1983. The top arch was cut into two sections as shown in Figure 5. The section facing west and away from the hypocenter was discarded, and the section facing east and toward the hypocenter was cut into six pieces. Plates A and B of Piece 3 were used in the JCAC studies by Kimura et al. (1990) and Kimura and Hamada (1993), and Plates C and D of Piece 4 were used in the ORNL studies by Kerr et al. (1990). Plates C and D were located approximately 14 m above water in the river at the time of bombing and 10 m above ground, and the steel plate from the stand on the smokestack of the Chugoku Electric Company was located about 25 m above ground (Kerr et al. 1990).

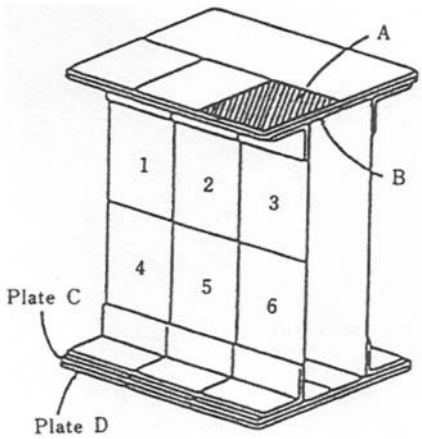

Figure 5. *Schematic of the steel sample from the arch of the Yokogawa Bridge, showing the location of Plates A and B of Piece 3 and Plates C and D from Piece 4 (Kimura and Hamada 1993). The total sample was approximately 75 cm in length, 70 cm in height, and 45 cm in width.*

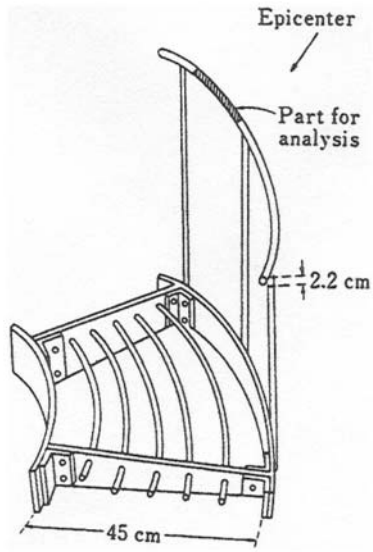

Figure 6. *Schematic of stand from the smokestack to the Chugoku Electric Company showing the location of the steel rod sample used in the cobalt actions studies by Kimura et al. (1990). The front steel plate at the bottom of the stand to the smokestack was used in the cobalt activation studies by Kerr et al. (1990).*

Analyses were made to determine the elemental composition for all major pieces of the steel samples used in the ORNL studies. The primary technique for obtaining data on a sample was inductively coupled plasma-atomic emission spectrometry (ICP-AES). Other techniques were used as required to obtain data on a few additional elements or to verify the data on a specific element, particularly cobalt. These other techniques included flame-excited atomic absorption spectrometry (AAS), neutron activation analysis (NAA), and inductively coupled plasma-mass spectrometry (ICP-MS). The NAA measurements were made at the National Institute of Standards and Technology (NIST), the ICP-MS measurements were made at the Pacific Northwest Laboratories (PNL), and the AAS and ICP-AES measurements were made at ORNL.

The initial measurements of the ^{60}Co activity in the steel samples were made using so-called "moon rock" counters at PNL. These PNL counters consisted of pairs of 15,000-cm^3 NaI(Tl) detectors that were operated in coincidence and also shielded electronically by the use of very large plastic phosphor anticoincidence detectors (Wogman and Brodzinski 1973). Such a system is well suited to counting of ^{60}Co, which emits two gamma rays that can be detected simultaneously in the two NaI(Tl) detectors. In addition, pulses that occur in one (or both) of the NaI(Tl) detectors at the same time as a pulse in the plastic phosphor anticoincidence shields can also be ignored. It is possible, therefore, to achieve both a very low background and a high counting efficiency for ^{60}Co. The plates from the bridge and smokestack were counted on two different PNL "moon rock" counters designated as Systems 8 and 9. To establish the background on the two different systems, measurements were also made using other steel samples: one from a light steel-frame building erected in the early 1940s at the Aluminum Company of America at Alcoa, Tennessee, and another from an old rail track at the Homestake Mine, a deep underground mine in South Dakota. The background rates were approximately one count every 15 minutes on Detector System 8 and one count every 28 minutes on Detector System 9. Each of the steel samples used in the PNL measurements measured $8 \times 8 \times 0.5$ inches ($20.3 \times 20.3 \times 1.27$ cm) and weighed approximately 9 pounds (4 kg).

Initially, it was planned to chemically separate the cobalt from all of the samples and then to measure the specific activity of the separated cobalt from each sample as had been done previously by JNIRS (Kawamura et al. 1967). It was found, however, that the ^{60}Co in the samples could be determined with very good counting statistics by nondestructive measurements at PNL. To convert the PNL counting rate data to disintegration rate data for the samples, it was necessary to make a specific activity measurement using only one of the samples counted at PNL. Thus, the steel plate from the smokestack of the Chugoku Electric Company with the highest ^{60}Co activity was used to minimize the effort involved in making a reliable determination of the specific activity in the sample.

The cobalt was separated by dissolving a 1-kg piece of the sample from the Chugoku Electric Company in HCl, oxidizing the iron to Fe^{+3}, absorbing the Fe^{+3} and Co^{+2} on a Dowex-1 resin at 9M HCl, and eluting the Co^{+2} from the Dowex-1 with 4M HCl. The Dowex-1 was pretreated with 9M HCl before the process was begun, and the Fe^{+2} was oxidized to Fe^{+3} by adding nitric acid to the HCl solution containing the dissolved sample. The excess nitric acid was eliminated by distillation, and the HCl solution was adjusted carefully to 9M before the Fe^{+3} and Co^{+2} were absorbed on the Dowex-1. After the Co^{+2} was eluted from the Dowex-1, the Fe^{+3} was removed with 0.5M HCl, and the process was repeated. It was possible to process only 17 g of the dissolved steel per day in an ion exchange column containing 500 g of the Dowex-1 resin. Thus, five columns were used, and a total of eight days were required to separate the cobalt from 425 g

of dissolved steel.

The final ^{60}Co measurement was made at ORNL using a high-resolution Ge detector with a volume of 85 cm^3 and a well of 4 cm^3 for the highly efficient counting of small samples. To use the well, the detector was calibrated with the mixed radiation sources in an Amersham Standard Solution that was diluted very carefully to 3 cm^3. The separated Co product was then reduced by distillation to the same volume (i.e., 3 cm^3) and counted for 10.3 days to obtain the disintegration rate of ^{60}Co to within one standard deviation of ±5%. The elemental cobalt in the condensed Co sample was measured easily and reliably by both AAS and ICP-AES methods. From these data, it was determined that the specific activity was 1.49×10^{-3} Bq per mg of Co when the data were normalized arbitrarily to 6 August 1988. All counting data were arbitrarily normalized to 6 August 1988, because the measurements at ORNL and PNL were carried out over a period of several months. It was also possible using the above data on the smokestack sample to establish the counter efficiency for the PNL measurements and the specific activity of the cobalt in the C- and D-Plates from Piece 4 of the bridge sample. The counter efficiency was (3.99 ± 0.24)% or about one count per 25 disintegrations in the 9-pound (4-kg) plates, and the specific activities were $(9.03 \pm 1.00) \times 10^{-6}$ Bq per mg of Co for the C-Plate and $(1.97 \pm 1.00) \times 10^{-5}$ Bq per mg of Co for the D-Plate on 6 August 1988. A factor of 285 was used to correct these specific activities back to the time of bombing on 6 August 1945 (Table 4). These results are found to be in excellent agreement with the reported results of an independent Japanese study at the JCAC (Kimura et al. 1990; Kimura and Hamada 1993).

Table 4. Summary of data on cobalt activation in bulk steel samples from structures at Hiroshima (Kerr et al. 1990)

Sample location and description	Ground range (m)	Sample height (m)	Initial activity (Bq/mg of Co)
Chugoku Electric Company: Handrail from top of smokestack	690	25	0.423 ± 0.025
Yokogawa Bridge: Plate-4D from Top of arch on east side of bridge	1285	10	0.00560 ± 0.00050
Yokogawa Bridge: Plate-4C from Top of arch on east side of bridge	1285	10	0.00257 ± 0.00025

Kimura et al. (1990), Kimura and Hamada (1993)

As noted above, steel samples from the Yokogawa Bridge (Figure 5) and Chugoku Electric Company (Figure 6) were also analyzed in the independent studies by the JCAC (Kimura et al. 1990). They performed radiochemical separation of the ^{60}Co from the steel using a zinc-oxide method and an ion exchange method. The steel samples were etched, dissolved in 6M HCl, and HNO_3 was added to oxidize Fe^{+2} to Fe^{+3}. A mixture of ZnO and water was then added to the solution and $Fe(OH)_3$ was precipitated. After centrifugation, the pH of the supernatant was raised to 14 by adding NaOH. The precipitate was passed through a glass filter, washed, and dissolved in HCl. For further purification of Co, the residue was dissolved in 8M HCl and the solution passed through an anion exchange column. The eluate containing the Co was transferred to a small vessel and evaporated to dryness to prepare the sample for gamma-ray spectrometry. The

amount of stable Co in an enriched sample was determined by neutron activation. Only the sample from the Chugoku Electric Company and Plate A from Piece 3 of the Yokogawa Bridge were measured in the late 1980 studies by Kimura et al. (1990), but early 1990 studies by Kimura and Hamada (1993) also determined the specific activity of the ^{60}Co in Plate B of Piece 3 from the Yokogawa Bridge (Figure 5) and in three additional steel plates from the A-Bomb Dome (Table 5).

Table 5. Summary of data on cobalt activation in bulk steel samples from structures at Hiroshima (Kimura et al. 1990; Hamada 1991; Kimura et al. 1993)

Sample location and description	Ground range (m)	Sample height (m)	Initial activity (Bq/mg of Co)
A-Bomb Dome: Steel support-S16 for the arch of dome at base of dome on the east side of building	147	21	8.82 ± 0.67 (16A2) 9.35 ± 0.63 (16B2) 8.73 ± 1.23 (16C2)
Chugoku Electric Company: Handrail from top of smokestack	690	25	0.440 ± 0.063
Yokogawa Bridge: Plate-3A from Top of arch on east side of bridge	1285	10.7	0.00515 ± 0.00080
Yokogawa Bridge: Plate-3B from Top of arch on east side of bridge	1285	10.7	0.00353 ± 0.00100

Shizuma et al. (1992, 1993, 1997, 1998, 2002)

An extensive set of data on cobalt activation in Hiroshima and Nagasaki is provided by the Hiroshima University studies by Shizuma et al. (1992, 1993, 1997, 1998, 2002). Shizuma et al. (1992, 1993) collected six mineral samples located at ground ranges of less than 250 m at Hiroshima. The six samples consisted of three roof tiles, one wall tile, and two granite samples (Table 6). Because the cobalt content of mineral samples is quite small compared to steel samples (i.e., about 20 µg/g versus about 200 µg/g in steel), only the specific activity of ^{60}Co could be determined in samples close to the hypocenter. The specific activity in the six mineral samples was determined by direct gamma-ray spectrometry without chemical separation of the cobalt from the mineral samples.

Shizuma et al. (1997, 1998) also reported on measurements made for seven steel samples that were collected at ground ranges out to approximately 1,700 m and consisted of plates, pipes, and ladders from the roof of large buildings. Measurements were also reported for a control sample that was collected at a ground range of 4,578 m and consisted of thin steel plates measuring 25 cm × 25 cm × 1.5 mm. Small chips machined from the samples were dissolved in HCl so that the Co could be chemically separated from the steel. A small amount of HNO_3 and water were added to the solution to oxidize Fe^{+2} to Fe^{+3}. Isopropyl ether was then used to remove major ions in a solvent extraction method similar to that used by Kawamura (1967). The solution was then heated to reduce the volume. For further purification of the Co, an anion exchange process was applied to separate Co from the Ni, Cu, and residual Fe ions. The Co concentration in the enriched samples was determined by the Kawasaki Steel Techno-Research Company, using atomic absorption spectrometry.

Table 6. Summary of data on cobalt activation in sample materials from structures at Hiroshima (Shizuma et al. 1992, 1993, 1996, 1998)

Sample location	Material	Ground range (m)	Sample height (m)	Initial activity (Bq/mg of Co)	Reference
Shima Surgical Hospital	WT[a]	15	1	12.6 ± 1.3	b
Hiroshima Post Office	RT	36	10	10.9 ± 1.4	b
Saikou-Ji Temple	RT	82	10	11.2 ± 2.0	b
A-Bomb Dome	P	147	21	10.0 ± 1.0	c
	P	147	21	8.70 ± 0.47	d
	P	149	21	9.96 ± 0.55	e
Nippon Life Insurance Co.	G	160	1	7.7 ± 0.5	b
Monument of Victory	G	230	0	6.5 ± 0.5	b
Yasuda Life Insurance Co.	WT	253	1	7.4 ± 1.0	b
Kirin Beer Hall	L	685	18	0.364 ± 0.02	e
Kodakan Building	PP	722	14	0.304 ± 0.03	e
Hiroshima City Hall	PP	1018	18	0.105 ± 0.01	e
Animal Building, HU[f]	PP	1428	5	Not detected	g
Seifu ryo Dormitory, HU[f]	PP	1438	5	Not detected	g
Red Cross Hospital	PP	1485	29	0.022 ± 0.008	e
	L	1488	29	0.033 ± 0.010	e
	L	1491	29	Not detected	g
Postal Savings Bureau	PP	1601	22	Not detected	g
Hiroshima Bank of Credit	L	1676	18	0.016 ± 0.007	e
Army Food Warehouse	P	4576	2	Not detected	e and g

[a]RT: roof tile; P: steel plate; G: granite; WT: wall tile; L: steel ladder; PP: steel pipe.
[b]See Table 4 of Shizuma et al. (1993).
[c]See Table 1 of Shizuma et al. (1992).
[d]See Table 1 of Shizuma et al. (1997).
[e]See Table 1 of Shizuma et al. (1998).
[f]HU: Hiroshima University
[g]See Endo (2001).

Five additional steel samples were also collected at ground ranges out to approximately 1,000 m at Nagasaki (Shizuma et al. 2002). Three samples were steel rebars embedded at a depth of 1.5-4.5 cm in concrete, one sample was a steel handrail from the roof of a building, and one sample was a steel rail on the ground surface. Chemical separation of the Co in these samples was performed in the same way as the Hiroshima samples, and gamma-ray measurements were carried out with a well-type high-resolution Ge detector. The results of the Hiroshima ^{60}Co measurements are summarized in Table 6, and the Nagasaki ^{60}Co measurements are summarized in Table 7.

Table 7. Summary of data on cobalt activation in sample materials from structures at Nagasaki (Shizuma et al. 2002)

Sample location	Material	Ground range (m)	Sample height (m)	Sample depth (cm)	Initial activity (Bq/mg of Co)
Takatani house	Rebar[a]	290	1	3.3	1.03 ± 0.09
Shiroyama School	Rebar[a]	543	10	1.5	0.26 ± 0.02
Nagasaki University Hospital	Hand rail	655	20	0	0.10 ± 0.02
Motoki Bridge	Rebar[a]	743	-1.5	4.5	0.070 ± 0.012
Mitsubishi Steel	Steel rail	943	0	0	0.033 ± 0.006

[a]Steel reinforcing bars in concrete.

Endo (2001)

Endo (2001) addressed the problem of determining net or "peak" counts vs. "background" counts at the very low count rates from ^{60}Co in distant samples. As specific examples, he used five samples that had previously been classified as "ND" or "not detectable" for ^{60}Co in the studies by Shizuma et al. (1997, 1998). The data on these five samples have been added to Table 6 for the sake of completeness. The five samples were classified as ND-samples for ^{60}Co based on the application of a trapezoid method for obtaining net or "peak" vs. "background" counts from the high-resolution data obtained in gamma-ray spectrometry. Endo (2001) fitted a Gaussian distribution to the counting data in the region of a ^{60}Co gamma-ray peak and then estimated the "peak" counts using the fitted Gaussian distribution. Of the five ND-samples, the fitted Gaussian distributions gave negative or "not detectable" results in three cases, but positive or "detectable" results in the other two cases (Table 8). Thus, the assessment of "peak" vs. "background" counts in the distant samples is dependent to some extent on the method used in the spectral analysis (Tables 6 and 8).

Table 8. Summary of data from a reanalysis of cobalt activation in steel samples from Hiroshima by Endo (2001)[a]

Sample location	Material	Ground range (m)	Initial activity (Bq/mg of Co)
Animal Building, HU[b]	Handrail	1428	0.054 ± 0.031
Seifu ryo Dormitory, HU[b]	Steel pipe	1438	-0.031 ± 0.058
Red Cross Hospital	Handrail	1491	-0.0016 ± 0.050
Postal Savings Bureau	Handrail	1601	0.038 ± 0.017
Army Food Warehouse	Steel plate	4576	-0.002 ± 0.116

[a]These samples were designated as not-detected samples for ^{60}Co previous studies by Shizuma et al. (1997, 1998).
[b]HU: Hiroshima University.

Komura (2001a)

Several Hiroshima and Nagasaki samples from the studies by Shizuma et al. (1997, 1998) were re-measured at the underground Low Level Radioactivity Laboratory of Kanazawa University (Komura 2001a). The re-measured samples consisted of six samples from Hiroshima (one close-in sample from the A-Bomb Dome at a ground range of 144 m and five distant samples at ground ranges of 1,429 m or more) and four samples at Nagasaki (samples located at ground ranges from 540 m to 935 m). The results of the Kanazawa University measurements are summarized in Table 9 (Komura 2001a). The backgrounds of the underground Ge detectors at Kanazawa University's counting facility were approximately an order of magnitude lower than the background of the above-ground Ge detector used in the studies by Shizuma et al. (1997, 1998, 2002), but the benefit of a lower background was partially offset by the intervening radioactive decay of the ^{60}Co in the previously measured samples (Komura 2001a). The Hiroshima sample from the A-Bomb Dome demonstrates the general reliability of the ^{60}Co measurements for close-in samples, and the more distant Hiroshima samples provide a further illustration of the difficulty in making precise measurements as the ^{60}Co activity in the samples approaches background levels (Tables 6, 7, 8 and 9).

Table 9. Summary of data from re-measure of cobalt activation in sample materials from structures at Hiroshima and Nagasaki (Komura 2001a)[a]

City and sample location	Material	Ground range (m)	Sample height (m)	Sample depth (cm)	Initial activity (Bq/mg of Co)
Hiroshima					
A-Bomb Dome	Plate	147	21	Surface	7.80 ± 0.30
Animal Building, HU[b]	Handrail	1428	5	Surface	-0.058 ± 0.054
Seifu ryo Dormitory, HU[b]	Steel pipe	1438	5	Surface	0.006 ± 0.086
Red Cross Hospital	Ladder	1488	29	Surface	0.11 ± 0.04
Red Cross Hospital	Handrail	1491	29	Surface	0.037 ± 0.089
Postal Savings Bureau	Handrail	1601	22	Surface	0.034 ± 0.064
Nagasaki					
Shiroyama School	Rebar[c]	543	10	1.5	0.27 ± 0.04
Nagasaki University Hospital	Handrail	655	20	Surface	0.019 ± 0.055
Motoki Bridge	Rebar[c]	743	-1.5	4.5	0.056 ± 0.064
Mitsubishi Steel	Steel rail	943	Surface	Surface	0.005 ± 0.012

[a]Samples previously measured by Shizuma et al. (1997, 1998).
[b]HU: Hiroshima University.
[c]Steel reinforcing bars in concrete.

Okumura and Shimasaki (1997)

Because of the widespread damage in the hypocenter area at Nagasaki and the extensive redevelopment of this damaged area since 1945, it is now difficult to obtain steel samples at ground ranges less than approximately 500 m (Tables 2, 3 and 7). However, Okumura and Shimasaki (1996) have provided important close-in measurements of ^{60}Co in rocks used for shore protection along the Shimonokawa River and Urakami River, which run near the hypocenter, and in garden rocks. These measurements were possible because many of the rocks used for shore protection at Nagasaki are andesite with a high native Co content. The samples measured for ^{60}Co were fragments sliced at a depth of 0-2 cm from the surface of rocks that were found at a ground range of 561 m or less. A high-resolution Ge detector was used to analyze the gamma-ray spectra from the rock samples, and the Co content of the measured rock samples was determined at Kyoto University by neutron activation analysis. The results of these important close-in ^{60}Co measurements at Nagasaki are summarized in Table 10 (Okumura and Shimasaki 1996).

Table 10. Summary of data on cobalt activation of rocks used to protect shore along the Shimonokawa and Urakami Rivers at Nagasaki (Okumura and Shimasaki 1996)[a]

Sample	Ground range (m)[b]	Initial activity (Bq/mg of Co)	Sample	Ground range (m)[b]	Initial activity (Bq/mg of Co)
1	18	7.27 ± 0.73	13	118	5.30 ± 1.23
2	39	6.70 ± 0.67	14	249	3.94 ± 2.60
3	63	5.65 ± 0.57	15	307	2.63 ± 1.06
4	82	8.43 ± 0.84	16	330	3.25 ± 1.59
5	82	7.12 ± 0.71	17	330	2.08 ± 1.02
6	82	5.40 ± 0.54	18	343	3.05 ± 1.04
7	92	3.05 ± 2.06	19	347	4.38 ± 0.81
8	93	10.0 ± 1.73	20	353	3.19 ± 1.43
9	93	6.28 ± 1.29	21	460	1.34 ± 1.09
10	93	5.30 ± 1.09	22	472	2.32 ± 1.34
11	93	4.48 ± 0.92	23	561	0.786 ± 0.401
12	96	8.43 ± 1.74			

[a]Values read from a plot of data in Figure 1 of Okumura and Shimasaki (1996). Samples were taken at a depth of 0-2 cm from surface of rocks that were unshielded in a line-of-sight direction toward the burst point of the bomb.

[b]Ground ranges as originally shown in Figure 1 of Okumura and Shimasaki (1996). These ground ranges were not reevaluated in this study.

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