Chapter 9

ACTIVATION MEASUREMENTS FOR FAST NEUTRONS

Part A. Sulfur (^{32}P) Activation

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Introduction

In early September 1945, the Japanese Science Promotion Society, now the Science Council of Japan, mobilized available scientists to conduct a large-scale investigation of all aspects of the Hiroshima and Nagasaki bombings, and in late September 1945, U.S. Army, Navy, and Manhattan Project teams entered Japan and began working with the Japanese scientists. The results of these investigations are summarized in numerous U.S. reports published in the late 1940s and a 1953 book published in Japan entitled *Collection of the Reports on the Investigation of the Atomic Bomb Casualties* (Science Council of Japan 1953).

One item discussed in several of these reports was measurements of the ^{32}P radioactivity in sulfur that was used in porcelain insulators mounted on electric power lines. The radionuclide ^{32}P can be induced in sulfur by neutrons with energies of 1 MeV or more, but the activation is mainly due to neutrons with energies greater than the effective threshold energy of about 3 MeV for the ^{32}S(n,p)^{32}P reaction. The ^{32}P has a half-life of 14.3 days and decays by the emission of very energetic beta particles (E_{max} = 1.71 MeV). Thus, the ^{32}P radioactivity can be measured easily by very simple electronic instruments with a thin aluminum window for transmission of the energetic beta particles to their detection chambers. Because high energy neutrons are required to activate sulfur by the ^{32}S(n,p)^{32}P reaction, the ^{32}P measurements are more easily related to neutron leakage from the bombs than the thermal-neutron activation of other materials such as cobalt, europium, and chlorine (Chapter 8).

The ^{32}P radioactivity measurements at Hiroshima can be traced back to two survey teams. One of the survey teams was from the Physical and Chemical Research Institute (Riken) at Tokyo (Yamasaki and Sugimoto 1953; Yamasaki 1958; Tajima and Oda 1958; Hamada 1983), and the other survey team was from Kyoto University (Arakatsu 1953; Shimizu 1982; Shimizu and Saigusa 1987). The measurements of the Kyoto University survey team were made by pulse counting of individual beta particles from the sulfur samples using a thin-window Geiger-Mueller

(GM) counter (Shimizu and Saigusa 1987). The Riken survey team made ionization measurements of the beta-particle intensity from one-gram samples of sulfur using thin-window Lauritsen electrometers (Hamada 1983).

No equivalent measurements of sulfur activation have been found for Nagasaki. However, the activation of sulfur by fast neutrons and the activation of gold foils by thermal neutrons were measured at a number of early tests in New Mexico, the Pacific (Bikini), and the Nevada Test Site (NTS). Data from several NTS tests of Nagasaki-like weapons are used here to verify the reliability of the newer DS02 calculations for the air-over-ground transport of neutrons at Nagasaki (Chapter 3).

The sulfur activation measurements at Hiroshima have been useful in refining and verifying the neutron source-term calculations for the Hiroshima bomb (Kerr and Pace 1988). For example, the Riken measurements of sulfur activation at Hiroshima were used to demonstrate that a two dimensional calculation of the neutron leakage was required for the Hiroshima bomb in order to accurately account for the blind spot in the neutron leakage through the nose of the device (Kerr 1982a,b; Kerr et al. 1983; Pace and Kerr 1984). The sulfur activation measurements at Hiroshima are discussed and compared in this section with the results of DS86 and DS02 calculations for the air-over-ground transport of neutrons at Hiroshima.

The Riken Survey

Drs. Yamasaki and Sugimoto of the Riken survey team were in Hiroshima from August 30, 1945 to September 6, 1945 to make measurements of soil activation and also to collect human bones and porcelain insulators from electric poles (Hamada 1983). They took these materials back to Tokyo and determined the ^{32}P radioactivity. Thirty porcelain insulators were collected and their location plotted on a small map of Hiroshima, the points of collection were numbered from 1 through 30, and a brief description of each sample location and its estimated ground range was provided in a table of initial counting results for 23 of the 30 sulfur samples. The initial measurements on these 23 samples using two thin-window Lauritsen electrometers with different sensitivities and different backgrounds are shown in Figure 1 (Munch 1946). The sample activity is only twice the instrument background for samples located near the hypocenter, and it approaches instrument background levels at ground ranges of approximately 1,000 m.

All sulfur samples within a ground range of 1,000 m were measured eventually using the electrometer with the highest sensitivity (Hamada 1983, 1984, 1987). The sulfur taken from the insulators was pulverized, measurements were made of the ^{32}P beta particles from 1 g of the sulfur power spread over an area of 35×35 mm, and the electrometer measurements converted to disintegrations per minute (dpm) of ^{32}P per g of S based on a calibration of the instrument using the ^{234m}Pa beta particles from a 1-g powder of uranium oxide (U_3O_8), also spread over an area of 35×35 mm. The results of these measurements are shown in Figure 2, which is taken from Pace and Smith (1946), and Table 1, which is taken from Yamasaki and Sugimoto (1953). Drs. Yamasaki and Sugimoto also showed the sample locations on a small map of Hiroshima in a companion 1953 report dealing with their measurements of the ^{32}P activity in bone samples (Yamasaki et al. 1953). On this map, they used the letters A through K (I was omitted) to designate the collection locations for the sulfur samples and the numbers 1 through 39 to designate the collection locations for the bone samples.

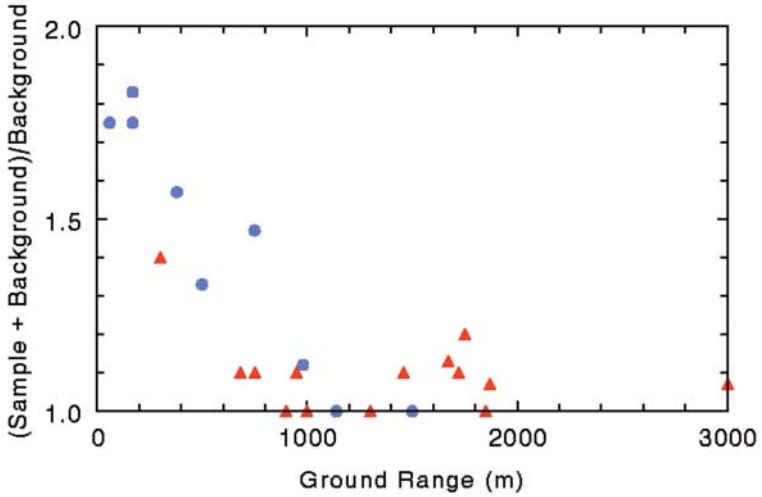

Figure 1. *Initial measurements of sulfur activation in 23 Hiroshima samples by Riken survey team using two thin-window Lauritsen electroscopes with different sensitivities and different backgrounds (Munch 1946).*

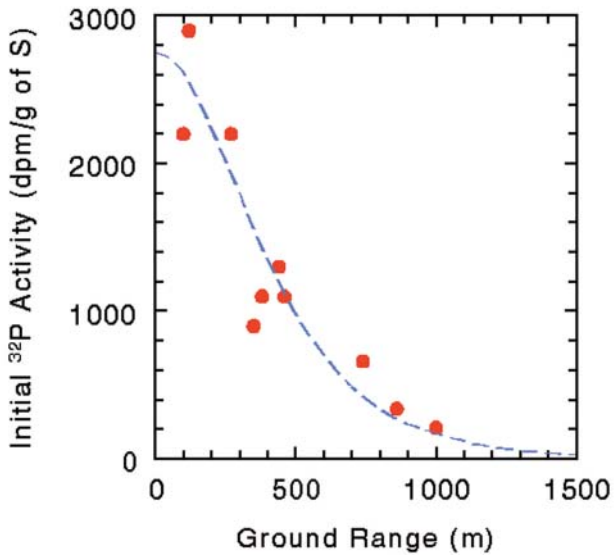

Figure 2. *Measurements of sulfur activation in 10 Hiroshima samples by Riken survey team using a thin-window Lauritsen electroscope that was calibrated with a U_3O_8 beta-particle source (Pace and Smith 1946). The dashed line shows a point source function that was fitted to the sulfur-activation data by Pace and Smith (1946).*

Table 1. Measurements of sulfur (^{32}P) activation at Hiroshima as reported by Yamasaki and Sugimoto (1953)

Sample	Ground range (m)	Initial activity (dpm g^{-1} of S)
A	270	2200
B	120	2900
C	350	900
D	380	1100
E	100	2200
F	460	1100
G	440	1300
H	740	660
J	1000	210
K	860	340

The results of the sulfur activation measurements by the Riken team were reexamined by Dr. Yamasaki (1958) and Dr. Hamada (1983, 1984, 1987). Their reexaminations of the measurements have led to some small changes in the beta-particle intensity of the samples as shown in Tables 2 and 3. Dr. Hamada also added data on duplicate measurements of several samples (Samples D and G), added data on several additional samples (Samples L through N), and provided estimates of the uncertainties in the sulfur samples measurements (one standard deviation). The samples have been designated in Hamada's reports and in this report by a combination of both of their previous designations; that is, Sample A in Yamasaki and Sugimoto (1953) and Sample 4 in Munch (1946) are now designated as Sample A-4, and so forth.

Table 2. Initial ^{32}P activity in sulfur samples at Hiroshima

Sample	Initial activity (dpm g^{-1} of S)	
	Yamasaki and Sugimoto (1953)	Hamada (1987)[a]
A-4	2200	2200 ± 430
B-5	2900	2940 ± 440
C-6	900	880 ± 400
D-12	1100	1140 ± 340
		1620[b] ± 310
E-13	2200	2430 ± 340
F-14	1100	1260 ± 340
G-15	1300	1370 ± 350
		1620 ± 350
H-7	660	630 ± 360
J-8	210	190 ± 290
K-29	340	370[b] ± 350
L-1		174 ± 330
M-3		174 ± 330
N-16		0 ± 360

[a]Also see Table 3.

[b]Corrected values (This work).

Table 3. Summary of sulfur activation data for Hiroshima (Hamada 1987)

Sample	Electrometer reading[a]		^{32}P activity	Decay	Initial activity
	(div/s)	RSD[b]	(Bq g^{-1} of S)	correction[c]	(dpm g^{-1} of S)
Background	0.00124	0.06			
A-4	0.00200	0.19	6.7 ± 1.3	5.5	2200 ± 430
B-5	0.00224	0.14	8.8 ± 1.3	5.6	2940 ± 440
C-6	0.00154	0.45	2.6 ± 1.2	5.6	880 ± 400
D-12	0.00162	0.29	3.3 ± 1.0	5.7	1140 ± 340
	0.00178	0.21	4.7 ± 0.9	5.7	1620[d] ± 310
E-13	0.00205	0.13	7.1 ± 1.0	5.7	2430 ± 340
F-14	0.00166	0.26	3.7 ± 1.0	5.7	1260 ± 340
G-15	0.00167	0.24	3.9 ± 1.0	5.8	1370 ± 350
	0.00175	0.20	4.6 ± 1.0	5.8	1620 ± 350
H-7	0.00144	0.53	1.8 ± 1.0	6.0	630 ± 360
J-8	0.00130	1.58	0.53 ± 0.8	6.1	190 ± 290
K-29	0.00136		1.05[d] ± 1.0[e]	5.8	370[d] ± 350
L-1	0.00130		0.53 ± 1.0[e]	5.5	174 ± 330
M-3	0.00130		0.53 ± 1.0[e]	5.5	174 ± 330
N-16	0.00120		0 ± 1.0[e]		0 ± 360

[a]Detection efficiency of electrometer was 1.14×10^{-4} division (div) per disintegration (dis) of ^{32}P (Hamada 1984,1987).

[b]Relative standard deviation (RSD) of electrometer readings (Table 2 of Hamada 1987).

[c]Correction factors to account for radioactive decay from time of bomb to the time of sample counting (Table 3 of Hamada 1984 and Table 6 of Hamada 1987).

[d]Corrected values (This work).

[e]Based on an early estimate that the standard deviation of the electrometer measurements of the ^{32}P was probably ±1 Bq or less (Table 3 of Hamada 1984 and Table 6 of Hamada 1987).

The calibration value used originally by Drs. Yamasaki and Sugimoto to convert the electrometer readings to ^{32}P activity per gram of sulfur was 1.09×10^{-4} division (div) of the electrometer scale per disintegration (dis) of ^{32}P (Hamada 1984). In 1958, Dr. Yamasaki revised the calibration factor based on newer half-life values for uranium and ^{32}P and on consideration of the difference of beta-ray self-absorption in sources caused by the differences in beta-ray energies of ^{32}P and ^{234m}Pa (E_{avg} = 0.695 MeV and 0.825 MeV, respectively). The revised calibration value for the electometer was 0.97×10^{-4} div/dis (Hamada 1984). However, Dr. Yamasaki introduced a correction factor for beta-ray self-absorption using a parallel beam approximation into his calibration calculations that did not represent the actual counting geometry of the electrometer measurements very well. Because it is difficult to establish the calibration value on a strictly theoretical basis, Dr. Hamada calibrated a similar electrometer using one gram of pure powdered sulfur containing a known amount of ^{32}P and one gram of uranium oxide, both spread over an area of 35 mm by 35 mm (Hamada 1984). The ratio of the electrometer measurements using the two sources in approximately the same counting geometry as the original ^{32}P measurements allowed Dr. Hamada to obtain a better founded calibration value of 1.14×10^{-4} div/dis with an estimated uncertainty of approximately ±2%.

The Riken survey team also used a rather small inaccurate map of Hiroshima to record the sample locations in 1945, and the sample locations were transferred to the more accurate U.S. Army map of Hiroshima in a 1968 study by Dr. Yamasaki and Dr. H. H. Hubbell, Jr. of the Oak Ridge National Laboratory. The recent 1983 report by Dr. Hamada provided map coordinates for all samples except G-15, which was located near the age-old camphor tree on the grounds of the Kokutaji Temple. Hence, the map coordinates for G-15 have been established in this work, and the locations of several other samples have also been reevaluated in this work based on data from Dr. Hubbell's log books, sample locator maps found in Munch (1946) and Yamashita et al. (1953), and information provided previously by Dr. Eizo Tajima (1982) and Mr. Hiroaki Yamada (1984).

The age-old camphor tree was a well-known landmark in Hiroshima (Nagaoka 1960). It was located on the grounds of the Kokutaji Temple close to the Bank of Japan, and the streetcar tracks near the tree were curved and raised to avoid damage to its roots. The coordinates of Sample G-15 taken at a location near this camphor tree have been established to within an estimated accuracy of ±20 m on the U.S. Army map of Hiroshima (Tajima 1982), and the coordinates for Sample K-29 have also been established to a similarly estimated accuracy. Sample K-29 was located near the Hiroshima Infantry Regiment Headquarters, but this site was placed on the U.S. Army map at the former site of the Brigade Headquarters in the Taisho Period (1912-1925). Sample K-29 is now located near the correct site for the Hiroshima Infantry Regiment Headquarters (Tajima 1982; Yamada 1984).

Another problem is that the locations of Samples D-12 and F-14 were switched in the 1983 report by Hamada. This is verified easily by comparing the maps in Munch (1946) and Yamasaki et al. (1953). The coordinates for Sample F-14 have now been assigned to Sample D-12, and Sample D-12 taken near the First Army Hospital, known more accurately as the Hiroshima Military Hospital, Motomachi Branch (Yamada 1982), is now located north of the hypocenter, as it should be. A more reliable coordinate was determined for Sample F-14 taken near the Fukuromachi Elementary School, and Sample F-14 is now located south of the hypocenter, as it should be.

Finally, the coordinates of the locations of the sulfur activation measurements on the U.S. Army map of Hiroshima were relocated on the new city map of Hiroshima, and adjustments were made in the locations of several sulfur sample measurements because of localized distortions in the older U.S. Army map of Hiroshima (Chapter 5). The locations of the sulfur activation measurements most affected by these localized distortions on the older U.S. Army map were found to the west of the hypocenter at Sakan-cho and the West Branch Office of the Telephone Company (Table 4). The locations of the all of the sulfur activation measurements and the DS02 hypocenter are shown in Figure 3 as coordinates on the new city map for Hiroshima. The north-south coordinates are negative on the new city map for Hiroshima and decrease in magnitude toward the north, and east-west coordinates are positive and increase in magnitude toward the east (Figure 3). The north-south and east-west coordinates of the DS02 hypocenter on the new city map of Hiroshima are -178.395 and 26.721, respectively (Chapter 1).

Table 4. Revised location coordinates for sulfur activation measurements at Hiroshima by the Physical and Chemical Research Institute (Riken) at Tokyo

Sample	General description of sample location[a]	U.S. Army Map		New City Map	
		Sample coordinates[b]	Ground range[c]	Sample coordinates	Ground range[d]
A-4	Kamiya-cho, main traffic intersection	1261.77 × 744.61	291 m	-178.330 × 27.008	294 m
B-5	Gokoku Shrine, torii or gate to shrine	1261.89 × 744.31	168 m	-178.228 × 26.726	167 m
C-6	Aioibashi, west end of bridge	1261.965 × 744.00	360 m	-178.167 × 26.437	364 m
D-12	Motomachi Branch, Hiroshima Army Hospital	1261.95 × 744.665	402 m	-178.164 × 27.052	404 m
E-13	Motoyasubashi, east end of bridge	1261.66 × 744.21	91 m	-178.441 × 26.641	92 m
F-14	Fukuromachi Elementary School	1261.95 × 744.62	440 m	-178.714 × 27.031	445 m
G-15	Kokutaiji Temple, age-old camphor tree	1261.28 × 744.45	414 m	-178.782 × 26.877	417 m
H-7	Sakan-cho	1262.06 × 743.585	728 m	-178.084 × 26.018	769 m
J-8	Telephone Company, West Branch Office	1262.185 × 743.275	1032 m	-177.984 × 25.758	1047 m
K-29	Hiroshima Infantry Regiment Headquarters	1262.21 × 744.91	724 m	-177.921 × 27.270	725 m
L-1	Yanagibashi, west end of bridge	1261.35 × 745.67	1296 m	-178.695 × 27.982	1296 m
M-3	Fukuya Department Store, west side	1261.60 × 745.05	695 m	-178.475 × 27.421	705 m
N-16	Hiroshima City Hall	1260.64 × 744.23	978 m	-179.373 × 26.695	978 m

[a]I.V. Munch, U.S. Army, Translated Japanese Language Documents, Translation M-7 (March 15, 1946).

[b]Previously unpublished data of G. D. Kerr.

[c]Based on DS86 hypocenter coordinates at 1261.707 × 744.298 on the U.S. Army map of Hiroshima.

[d]Based on DS02 hypocenter coordinates at -178.395 × 26.721 on the new city map of Hiroshima.

The Kyoto University Survey

Survey teams from Kyoto University made three trips to Hiroshima within a few weeks after the bombing (Arakatsu 1953). The second survey team consisted of ten members and was lead by Dr. Shimizu. They spent August 13 and 14 in Hiroshima collecting samples of sulfur from porcelain insulators, bone fragments, iron, and soil (Shimizu 1982). The various samples were counted at Kyoto University using a Duraluminum G-M counter with a tube having a 12-mm diameter, 0.1-mm wall thickness, and 40-mm effective length. The tube was filled with air at a pressure of 9.0 cm of mercury and ethyl-alcohol vapor at a pressure of 1.5 cm of mercury. The natural background of the counter was about 18 counts per minute (cpm). The sulfur samples taken from the porcelain insulators were spread uniformly in paper boats of 3 cm by 2 cm and placed 4 mm beneath the counter tube. The detection efficiencies of the G-M counter for the sulfur samples were later estimated from Monte Carlo calculations, and the observed count rates

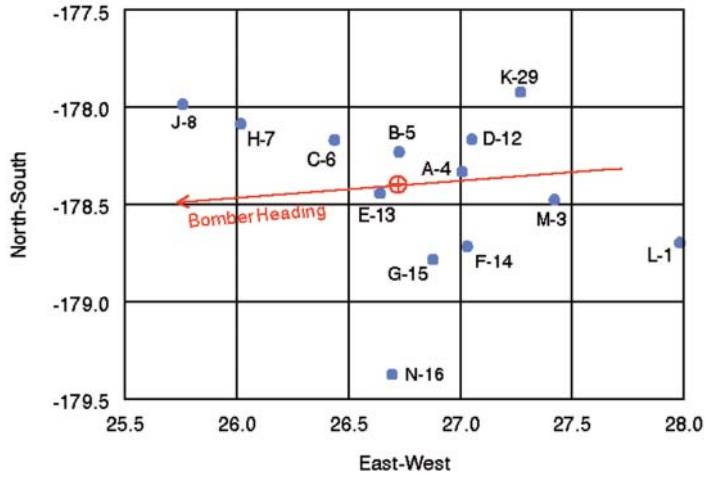

Figure 3. *Location of Riken survey measurements of sulfur activation and DS02 hypocenter shown as coordinates on the new city map of Hiroshima. The locations of the sulfur activation samples are shown as solid blue dots, and the DS02 hypocenter is designated by the cross inside the open red circle. The north-south coordinates are negative on the new city map of Hiroshima and decrease in magnitude toward the north, and the east-west coordinates are positive and increase in magnitude toward the east.*

(cpm) were converted to disintegration rates (dpm) as shown in Table 5 (Shimizu and Saigusa 1987). One significant problem with these sulfur-activation data is that the sample locations were documented only on a rather small inaccurate map of Hiroshima, and there are large uncertainties involved in estimating the sample locations on the U.S. Army map of Hiroshima (see footnotes to Table 5).

Table 5. Sulfur activation at Hiroshima from measurements made by Kyoto University

Sample number	Ground range (m)		Sample mass (g)	Count rate (cpm)	Detection efficiency[c]	Initial activity (dpm g^{-1} of S)[d]
	Old[a]	New[b]				
407	250	550	1.5	35	4.54%	840
411	350	780	2.2	33	3.27%	741
518	800	980	2.6	23	2.80%	518

[a]Arakatsu (1953) and Shimizu (1982).

[b]Shimizu and Saigusa (1987). The new ground ranges are based on the DS86 hypocenter and the error in the new ground ranges is estimated to be within ± 100 m.

[c]Estimated by Shimizu and Saigusa by Monte Carlo calculations that included self-absorption of beta particles in sulfur and in the thin aluminum window of the G-M counter.

[d]Correction for decay from time of bombing to time of counting was 0.619. The uncertainty in the initial activities is estimated to be ±15% or less.

Shielding of Sulfur by Porcelain Insulators

The first measurements of the shielding of the sulfur by a porcelain insulator were made by Drs. Tajima and Oda (1958). They used fast neutrons generated through Be-D reaction at the Riken cyclotron and D-D reaction by a Crockroft-Walton accelerator at Rikkyo (St. Paul's) University in Tokyo. The insulators were arranged as shown in Figure 4, irradiated, and the ^{32}P radioactivity in the sulfur was measured with a thin-window GM counter. If A was the measured activation of the sulfur outside the insulators, and B and C were the measured activation of the sulfur inside the insulators, then the transmission coefficient for lateral irradiation was B/A = 0.77, and for irradiation from the top, C/A = 0.52 (Tajima and Oda 1958). Thus, the shielding for lateral irradiation was 23%, and the shielding for irradiation from the top was 48%.

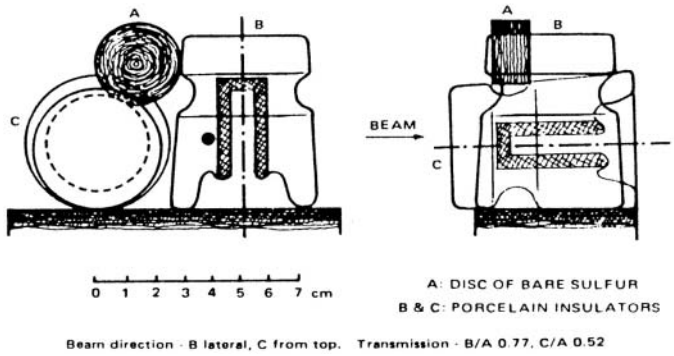

Figure 4. *Arrangement of porcelain insulators and sulfur samples for shielding studies (Tajima and Oda 1958).*

Several previous calculations of the shielding of sulfur by the porcelain insulators have been reported in the literature (Loewe 1983; Kerr et al. 1983; Pace and Kerr 1984; Gritzner and Woolson 1987). These calculations all used the dimensions of the insulators from Tajima and Oda (1958) and the density and composition for the porcelain insulators from Sugimoto (1953). Dr. Sugimoto gave the density as 2.6 g cm^{-3} and the composition as $Al_2O_2 + 2SiO_2 + 2H_2O$. It was assumed typically that the water was removed by the firing process during the manufacture of the porcelain insulators, and the composition used for the shielding calculations was 0.714 SiO_2 + 0.286 Al_2O_2 (Loewe 1983). The calculated shielding of sulfur was typically about 15% for irradiation of an insulator from all directions (Gritzner and Woolson 1987) with a range from about 10% for lateral irradiation of the insulator to about 30% for irradiation from the top of the insulator (Loewe 1983). The calculations suggested significantly less shielding of sulfur by the porcelain insulators than the measurements of Drs. Tajima and Oda (1958).

A porcelain insulator of the same type used in the early Japanese studies was obtained from an old warehouse in Hiroshima by Dr. Tajima and sent to the Oak Ridge National Laboratory (ORNL) in 1985 for further studies. The results of these ORNL studies have not been reported previously in the literature. Measurements of the density and composition of the insulator are

summarized and compared in Table 6 with values that were used previously (Sugimoto 1953; Loewe 1983), and the purity of the sulfur was investigated (Table 7) and found to be very pure, as in the studies of Hamada (1987). The shielding of sulfur by the insulator was also determined using neutrons from the Health Physics Research Reactor (HPRR), a small unshielded and unmoderated fast-burst reactor (Auxier 1965). The insulators were exposed at 3 m from the center of the reactor core and 1.5 m above the concrete floor of the HPRR facility. Three small sulfur pellets were irradiated outside the insulator, three were irradiated inside the insulator, and the insulator was irradiated from the top (Figure 4). The shielding of sulfur for the unshielded spectrum of neutrons from the HPRR was about 4% (Swaja 1985a), and the shielding for sulfur using a 13-cm steel-shielded spectrum of neutrons from the HPRR was about 7% (Swaja 1985b). The unshielded and steel-shielded neutron spectra at 3.0 m from the center of the reactor core and 1.5 m above the concrete floor of the HPRR facility are provided and discussed by Poston et al. (1974) and the International Atomic Energy Agency (IAEA 1978, 1990).

Table 6. Chemical composition for porcelain insulators from Hiroshima

Element	Percent by mass	
	Sugimoto (1953)[a]	This study[b]
O	46.57	48.63
Al	26.18	11.65
Si	27.25	31.88
K		6.39
Fe		1.45
Density	2.6 g cm^{-3}	2.4 g cm^{-3}

[a]Also see Loewe (1983).
[b]Previously unpublished data of G. D. Kerr.

Table 7. Sulfur sample impurities[a]

Chemical element	Relative mass (μg g^{-1} of S)
Zn	1400
Fe	365
Al	100
La	100
Mg	40
Ca	37
K	35
Na	22
Hg	15

[a]Previously unpublished data of G. D. Kerr.

The shielding of sulfur by a porcelain insulator was calculated in these studies using the MCNP4C computer code (Briesmeister 2000), the ENDF/B-VI cross-section library (Hendricks et al. 1994), and the densities and compositions for porcelain insulators from the ORNL studies in Table 6. The insulators were modeled using concentric spheres. The sulfur was represented by a spherical shell with an inner radius of 0.5 cm and an outer radius of 1.0 cm, and the porcelain was represented by a spherical shell with an inner radius of 1.0 cm and outer radius of 2.5 cm. The porcelain insulator was irradiated with a parallel beam of neutrons to obtain results that were applicable to both the irradiations at the HPRR and the irradiations at Hiroshima by neutrons from all directions. The shielding calculations for both HPRR irradiations gave a value of approximately 5%, based on the spectral data for the unmoderated and steel-shielded HPRR neutrons in IAEA TRS No. 318 (International Atomic Energy Agency 1990), and the shielding calculations for Hiroshima irradiations gave a value of approximately 15%, based on the air-transport spectrum of neutrons at a ground range of 250 m and a height above ground of 6 m (20 feet) at Hiroshima (Chapter 3).

The value of 15% calculated in this study for the shielding of sulfur by the porcelain insulators at Hiroshima is in good agreement with that calculated in several earlier studies (Loewe 1983; Kerr et al. 1983; Pace and Kerr 1984; Gritzner and Woolson 1987), and the value of 5% calculated in this study for the shielding of sulfur by the porcelain insulators exposed at the HPRR is in good agreement with experimental measurements (Swaja 1985a,b). It is not possible to make calculations for the shielding measurements of Tajima and Oda (1958), because the spectra of the accelerator-produced fast neutrons used in their studies is not known. The energies of the accelerator-produced neutrons would be mainly in the resonance region of the $^{32}S(n,p)^{32}P$ reaction below 5 MeV (Figure 5), which could account for the much higher shielding values observed in the studies of Tajima and Oda (1958). For example, a down scattering collision of a neutron in the porcelain insulator can significantly alter its activation cross section for sulfur inside the porcelain insulator compared to its activation cross section for sulfur exposed free-in-air.

The above sulfur-activation calculations were all based on the ENDF/B-VI cross sections for the $^{32}S(n,p)^{32}P$ reaction. Because of the large uncertainties of the cross sections for sulfur activation in the resonance region below 5 MeV (Figures 5 and 6), the activation of sulfur inside a porcelain insulator at Hiroshima was also calculated using the cross sections for the $^{32}S(n,p)^{32}P$ reaction from the Japanese Evaluated Nuclear Data Library (JENDL Version 3.3) and the International Reactor Dosimetry File (IRDF-90 Version 2). The IRDF-90 data shown in Figures 5 and 6 were obtained from Kocherov and McLaughlin (1993), the American Society for Testing and Materials (1999), and Remec (2002). The results of the calculations for sulfur exposed inside a porcelain insulator at a ground range of 250 m and a height of 6 m above ground at Hiroshima are shown in Figure 7. In spite of the large uncertainties in the cross sections at energies below 5 MeV, the cumulative sulfur activation shows only minor differences when plotted as a function of the neutron energy. The total sulfur activation at Hiroshima predicted by the ENDF/B-VI and IRDF-90 cross sections agree to within 1%, while the JENDL 3.3 data predict about 7% more sulfur activation at Hiroshima than the ENDF/B-VI cross sections for the $^{32}S(n,p)^{32}P$ reaction.

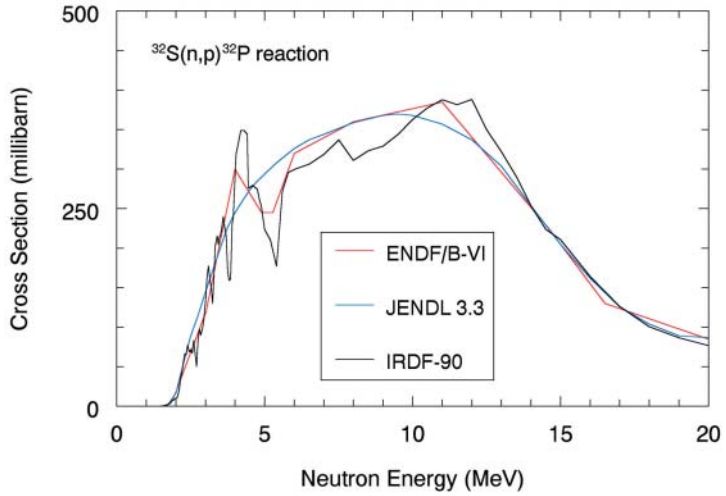

Figure 5. *Comparison of cross sections for $^{32}S(n,p)^{32}P$ reaction from various nuclear data libraries.*

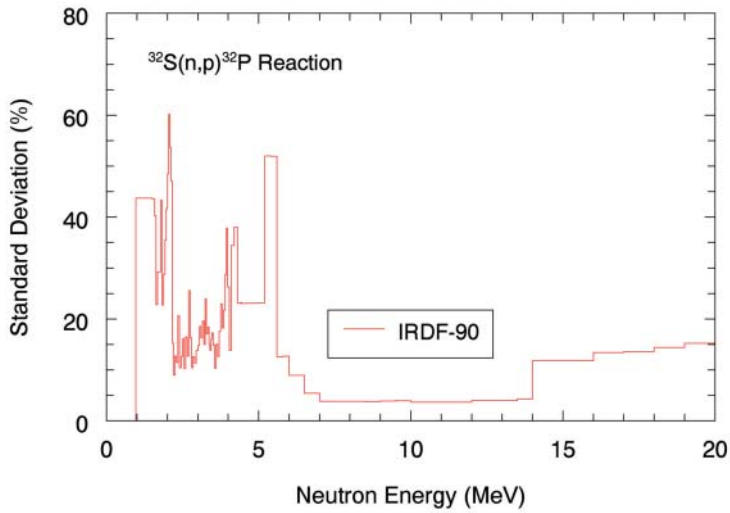

Figure 6. *Energy-dependence of uncertainty in the cross sections for the $^{32}S(n,p)^{32}P$ reaction (American Society for Testing and Materials 1999; Remec 2002).*

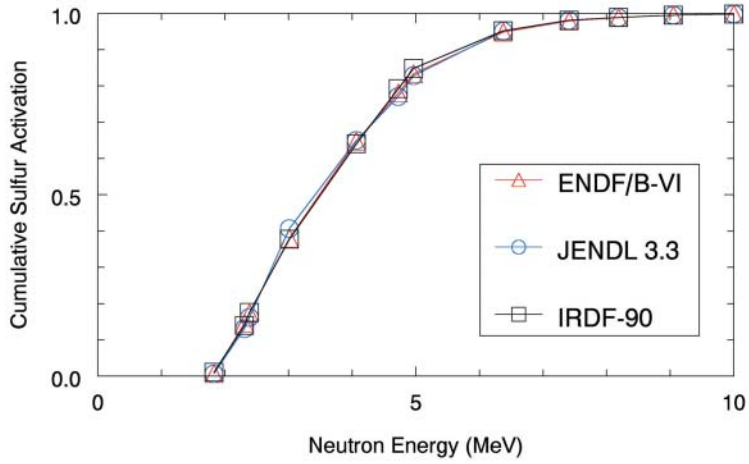

Figure 7. *Energy dependence of cumulative ^{32}P activation of sulfur inside a porcelain insulator at a ground distance of 250 m and a height of 6 m above ground at Hiroshima.*

Comparison of DS86 and DS02 Calculations for Sulfur Activation at Hiroshima

The results of DS02 calculations for sulfur activation at Hiroshima are compared in Figure 8 with the measured values at Hiroshima and the results of former DS86 calculations for sulfur activation at Hiroshima (Kerr et al. 1987). The DS86 and DS02 calculations shown in Figure 8 are symmetric about the hypocenter because they were calculated for an untilted bomb (i.e., the nose of the bomb was pointed directly downward toward the hypocenter). It was also assumed in these calculations that the porcelain insulators containing the sulfur were located on electric poles at a height of about 6 m (20 feet) above ground. In the DS86 calculations, the bomb yield and burst height were taken to be 15 kt and 580 m, respectively, and in the DS02 calculations, the bomb yield and burst height were taken to be 16 kt and 600 m, respectively. The burst height was raised to 600 m in the DS02 studies to obtain better agreement with all neutron-activation measurements, including the new ^{63}Ni fast-neutron data, and the TLD measurements for gamma rays at Hiroshima. While some of the decrease noted in the DS02 calculations of sulfur activation at Hiroshima can be attributed to changes in the burst height, it is also due to a number of other factors as discussed below.

At the time of explosion, the Hiroshima bomb was tilted about 15° with respect to the vertical, and the nose of the bomb was pointed toward a point on the ground about 160 m from the hypocenter. The neutron output from the bomb was also not uniform in angle but had a distribution where the output in the direction of the nose was quite suppressed (Chapter 2). Hence, the activation of sulfur was not symmetric about the hypocenter of the Hiroshima bomb, but varied with both ground distance and direction with respect to the bomber heading or the flight path of the bomber (Figure 3 and Chapter 1). The effects of bomb tilt on the calculated activation in sulfur samples that were measured by the Riken survey group are summarized in Table 8, and the sulfur activation ratios that were calculated for a bomb tilted at 15° and an untilted bomb are shown in Figure 9. The solid line in Figure 9 was calculated using the bomb tilt

equation for sulfur in Chapter 2, the triangles are the ratios calculated for the sulfur activation samples using this equation and the ground range for each sample along the bomber path from Table 8, and the circles are the ratios calculated in a previous study by Pace and Kerr (1984). Hence, the effect of bomb tilt on sulfur activation is considered to be reasonably well established at Hiroshima (Figure 9). A comparison between the sulfur-activation measurements of the Riken survey team and the sulfur-activation calculations for both an untilted bomb and a tilted bomb are shown in Figure 10. The solid line in Figure 10 is a least-squares fit to the calculated sulfur-activation values for an untilted bomb in Table 8.

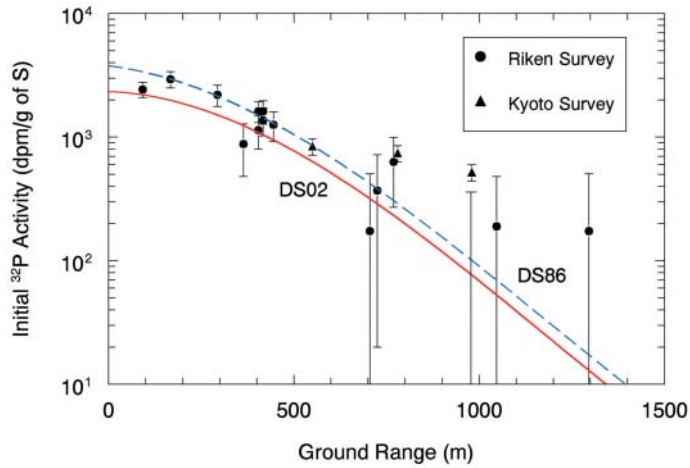

Figure 8. *Comparison of the DS02 and DS86 calculations for sulfur activation with measurements by the Riken and Kyoto survey teams.*

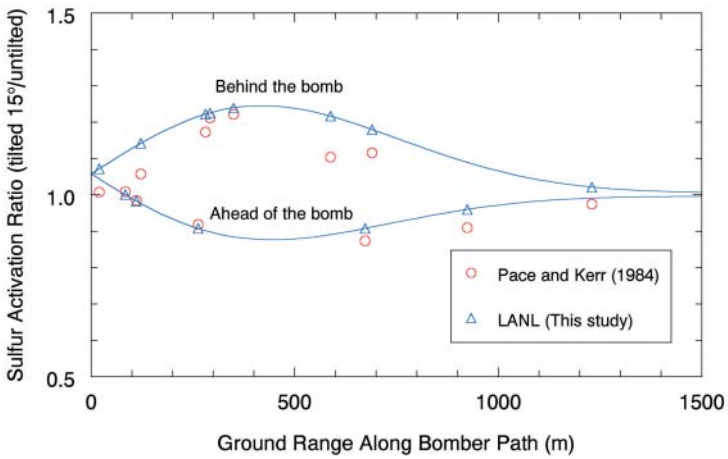

Figure 9. *Calculated ratios for sulfur activation from the Hiroshima bomb tilted at 15° to that of an untilted Hiroshima bomb. The circles and triangles in the figure are the ratios calculated for the sulfur samples measured by the Riken survey team (see Table 8).*

Table 8. Summary of calculated ^{32}P activities for sulfur activation samples that were measured by Riken survey team

Sample number	Measured activity (dpm g^{-1} of S)	Ground range (m)	Ground range along bomber path (m)	Activity ratio with and without bomb tilt	Calculated ^{32}P activity (dpm g^{-1} of S)	
					No bomb tilt[a]	With bomb tilt[b]
A-4	2200 ± 430	294	-292	1.225	1452	1778
B-5	2940 ± 440	167	-20	1.072	2040	2186
C-6	880 ± 400	364	263	0.908	1230	1117
D-12	1140 ± 340	404	-350	1.239	1089	1349
	1620 ± 310					
E-13	2430 ± 340	92	84	1.001	2258	2260
F-14	1260 ± 340	445	-281	1.222	953	1165
G-15	1370 ± 350	417	-122	1.142	1043	1191
	1620 ± 350					
H-7	630 ± 360	769	673	0.908	237	233
J-8	190 ± 290	1047	924	0.960	51.5	49.5
K-29	370 ± 350	725	-588	1.217	291	354
L-1	174 ± 330	1296	-1230	1.021	12.9	13.2
M-3	174 ± 330	705	-690	1.180	315	372
N-16	0 ± 360	978	111	0.983	76.0	74.7

[a]Calculated value for sulfur activation at 6 m in air over wet ground corrected for shielding by porcelain insulator and for scattering from a concrete or dry ground interface.

[b]Calculated value for sulfur activation with no bomb tilt in Column 6 multiplied by activity ratio with and without bomb tilt in Column 5 of table.

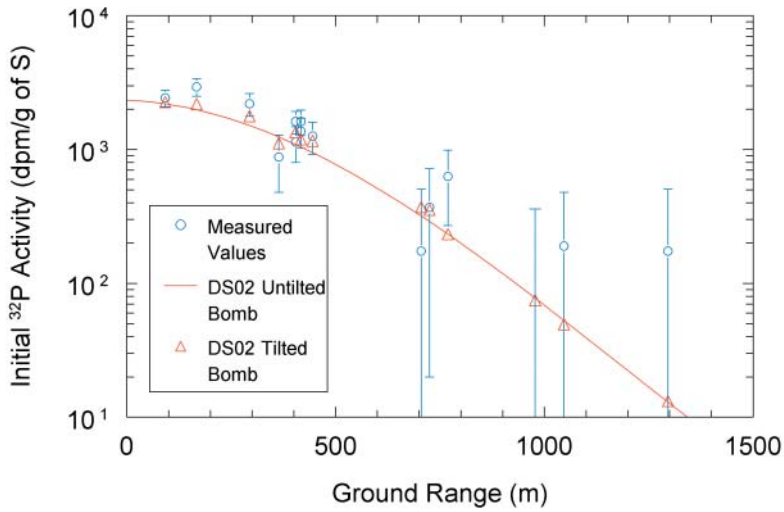

Figure 10. *Comparison between the sulfur-activation measurements of the Riken survey team and the DS02 sulfur-activation calculations for both an untilted bomb and a tilted bomb at Hiroshima. The solid line in the figure is a least-squares fit to the calculated sulfur-activation values for an untilted bomb in Table 8.*

The sulfur activation measurements of the Riken survey team can be used to make an estimate of the energy yield for the Hiroshima bomb (Table 9). To do this, each measured value for sulfur activation (and its standard deviation) are divided by the calculated value for a tilted bomb (Table 8) and then multiplied by the energy yield of 16 kt used in the DS02 calculations for the Hiroshima bomb. It was found that an energy yield of 18 ± 2 kt would provide the best agreement between the DS02 calculations and the sulfur-activation measurements by the Riken survey team (Table 9). This yield estimate was calculated as a weighted mean in which the reciprocal of the square of the standard deviations of the yield estimates from each of the sulfur activation measurements were used as the weighting factor (Beers 1957). The energy yield predicted by these same measurements and the DS86 calculations for sulfur activation at Hiroshima was 13 ± 1 kt (Malik et al. 1987). The sulfur-activation measurements by the Kyoto survey team were not used in estimating the energy yield of the Hiroshima bomb because of the large uncertainties in the ground distances at which the sulfur samples were collected (Table 5).

The change in the estimated bomb yield from 13 kt based on the DS86 calculations for sulfur activation to 18 kt based on the DS02 calculations for sulfur activation can be explained as follows. Changing from the earlier ENDF/B-IV cross sections used for sulfur activation in the DS86 studies to the ENDF/B-VI cross sections used in the newer DS02 studies caused the sulfur activation to decrease by -10% (and the bomb yield to increase by +10%). Changing from the older coarse-angle (max. 25.8°) bins to the finer-angle (max. 5.3°) bins in the newer LANL

Table 9. Energy yield of Hiroshima bomb derived from sulfur activation data

Sample	Yield ± σ (kt)	Sample weight $(1/\sigma^2)$[a]
A-4	19.8 ± 3.9	6.679E-2[b]
B-5	21.5 ± 3.2	9.642E-2
C-6	12.6 ± 5.7	3.046E-2
D-12	13.5 ± 4.0	3.075E-2
	19.2 ± 3.7	3.699E-2
E-13	17.2 ± 2.4	1.726E-1
F-14	17.3 ± 4.7	4.586E-2
G-15	18.4 ± 4.7	2.262E-2
	21.8 ± 4.7	2.262E-2
H-7	43.3 ± 24.7	1.636E-3
J-8	61.4 ± 93.7	1.138E-4
K-29	16.7 ± 15.8	3.996E-3
L-1	211 ± 400	6.250E-6
M-3	7.5 ± 14.2	4.964E-3
N-16	0.0 ± 77.1	1.682E-4
Weighted mean and standard deviation (σ) = 18 ± 2 kt		

[a]If a sample was measured twice, only one half of the weight of each of the two sample measurements was used in calculating the weighted mean for the yield of the bomb and its standard deviation.

[b]Read as 6.679×10^{-2} or 0.06679.

source calculations for the neutron leakage through the nose of the Hiroshima bomb caused the DS02 sulfur activation in the important sulfur samples at the smaller ground ranges to decrease by -10% (and the bomb yield to increase by +10%). Changing from the earlier ENDF/B-IV cross sections used in the air transport calculations in the DS86 studies to the ENDF/B-VI cross sections used in the air transport calculations in the DS02 studies caused the sulfur activation to decrease by -10% (and the bomb yield to increase by +10%). Changing from the DS86 burst height of 580 m to the DS02 burst height of 600 m caused the sulfur activation to decrease by -15% (or the bomb yield to increase by +15%), and changing the surface under the sulfur samples on the electric poles from wet ground in the DS86 studies to concrete (or dry ground) in the DS02 studies caused the sulfur activation to increase by +10% (and the yield to decrease by -10%). In summary, the change in bomb yield between the DS86 and DS02 studies is equal to 13 kt $\times$ ($1.10 \times 1.10 \times 1.10 \times 1.15 \times 0.90$) or 18 kt.

Discussion and Conclusions

The measurements of the ^{32}P radioactivity in the Hiroshima sulfur samples were evaluated and reported in the DS86 studies. In the current reassessment, the locations of the sulfur samples have been updated and corrected if they were in error. The measurement results for sulfur samples collected at ground ranges of less than 500 m (or slant ranges of less than 800 m) are now as well-established as the available data permit. The bomb yield based on the updated and corrected sulfur activation data of the current assessment is calculated to be 18 ± 2 kt. This yield estimate (and the sulfur activation at Hiroshima) are found to be in substantial agreement with the theoretical bomb yield estimates of 15 to 18 kt (Chapter 2), and highly consistent with the 16 kt selected as the most likely yield of the Hiroshima bomb in the DS02 studies after careful consideration of all of the relevant bomb yield data (Chapter 1).

References

American Society for Testing and Materials (ASTM). *Standard Test Method for Measuring Reaction Rates and Fast-Neutron Fluences by Radioactivation of Sulfur-32*. West Conshohocken, Pennsylvania: American Society for Testing and Materials; E 265-98; 1999.

Arakatsu, B. “Report on Survey of Radioactivity in Hiroshima Several Days After the Atomic Bomb Explosion.” In: *Collection of Investigative Reports on Atomic Bomb Casualties*, Vol. 1, pp. 5-10. Tokyo, Japan: Japanese Science Promotion Society; 1953.

Auxier, J. A. “The Health Physics Research Reactor.” *Health Phys*. 11: 89-93; 1965.

Beers, Y. *Theory of Error*. Reading, Massachusetts: Addison-Wesley Publishing Co.: 1957.

Briesmeister, J. F.; ed. *MCNP—A General Monte Carlo N-Particle Transport Code, Version 4C*. Los Alamos, New Mexico: Los Alamos National Laboratory; LA-23709-M; April 2000.

Gritzner, M. L.; Woolson, W. A. “Sulfur Activation at Hiroshima.” In: *US-Japan Joint Reassessment of Atomic Bomb Radiation Dosimetry in Hiroshima and Nagasaki, Final Report, Vol. 2*, pp. 283-294 (Roesch, W. C.; ed.). Hiroshima, Japan: Radiation Effects Research Foundation; 1987.

Hamada, T. “Measurement of ^{32}P Activity Induced in Sulfur in Hiroshima.” In: *First US-Japan Joint Workshop for Reassessment of Atomic Bomb Radiation Dosimetry in Hiroshima and Nagasaki*, pp. 45-56

(Thompson, D. J.; ed.). Hiroshima, Japan: Radiation Effects Research Foundation; 1983.

Hamada, T. "^{32}P Activity Induced in Sulfur in Hiroshima: Reevaluation of Data by Yamasaki and Sugimoto." In: *Second US-Japan Joint Workshop for Reassessment of Atomic Bomb Radiation Dosimetry in Hiroshima and Nagasaki*, pp. 52-55 (Kato, H.; et al.; eds.). Hiroshima, Japan: Radiation Effects Research Foundation; 1984.

Hamada, T. "Measurements of ^{32}P in Sulfur." In: *US-Japan Joint Reassessment of Atomic Bomb Radiation Dosimetry in Hiroshima and Nagasaki, Final Report, Vol. 2*, pp. 272-279 (Roesch, W. C.; ed.). Hiroshima, Japan: Radiation Effects Research Foundation; 1987.

Hendricks, J. S.; Frankle, S. C.; Court, J. D. *ENDF/B-VI Data for MCNP*. Los Alamos, New Mexico: Los Alamos National Laboratory; LA-12891; 1994.

International Atomic Energy Agency. *Compendium of Neutron Spectra in Criticality Accident Dosimetry*. Vienna, Austria: International Atomic Energy Agency; TRS No. 180; 1978.

International Atomic Energy Agency. *Compendium of Neutron Spectra and Detector Responses for Radiation Protection Purpose*. Vienna, Austria: International Atomic Energy Agency; TRS No. 318; 1990.

Kerr, G. D. "Findings of a Recent Oak Ridge National Laboratory Review of Dosimetry for the Japanese Atom-bomb Survivors." In: *Reevaluations of Dosimetric Factors: Hiroshima and Nagasaki*, pp. 52-97 (Bond, V. P.; Thiessen, J. W.; eds.). Oak Ridge, Tennessee: U.S. Department of Energy; DOE Symposium Series 55; CONF-810928; 1982a.

Kerr, G. D. "Review of Dosimetry for the Atomic Bomb Survivors." *Nucl. Safety* 23(5): 563-571; 1982b.

Kerr, G. D.; Pace, J. V., III; Scott, W. H. "Tissue Kerma vs Distance Relationships for Initial Nuclear Radiation from the Atomic Bombs: Hiroshima and Nagasaki." In: *First US-Japan Joint Workshop on Reassessment of Atomic Bomb Radiation Dosimetry in Hiroshima and Nagasaki*, pp. 57-103 (Thompson, D. J.; ed.). Hiroshima, Japan: Radiation Effects Research Foundation; 1983.

Kerr, G. D.; Pace, J. V., III; Mendelsohn, E.; Loewe, W. E.; Kaul, D. C.; Dolatshahi, F.; Egbert, S. D.; Grintzer, M.; Scott, W. H., Jr.; Marcum, J.; Kosako, T.; Kanda, K. "Transport of Initial Radiation in Air Over Ground." In: *US-Japan Joint Reassessment of Atomic Bomb Radiation Dosimetry in Hiroshima and Nagasaki, Final Report, Vol. 1*, pp. 66-142 (Roesch, W. C.; ed.). Hiroshima, Japan: Radiation Effects Research Foundation; 1987.

Kerr, G. D.; Pace, J. V., III. "Sulfur Activation in Hiroshima." In: *New Dosimetry at Hiroshima and Nagasaki and Its Implications for Risk Estimates*, pp. 99-106. Bethesda, Maryland: NCRP Publications; Proceedings 9; 1988.

Kocherov, N. P.; McLaughlin, P. K. *The International Reactor Dosimetry File (IRDF-90 Version 2)*. Vienna, Austria: International Atomic Energy Agency; Nuclear Data Services; IAEA-NDS-141 Rev. 2; 1993.

Loewe, W. E. "Calculation and Interpretation of In Situ Measurements of Initial Radiations at Hiroshima and Nagasaki." In: *First US-Japan Joint Workshop on Reassessment of Atomic Bomb Radiation Dosimetry in Hiroshima and Nagasaki*, pp. 138-155 (Thompson, D. J.; ed.). Hiroshima, Japan: Radiation Effects Research Foundation; 1983.

Malik, J.; Tajima, E.; Binninger, G.; Kaul, D. C.; Kerr, G. D. "Yields of the Bombs." In: *US-Japan Joint*

Reassessment of Atomic Bomb Radiation Dosimetry in Hiroshima and Nagasaki, Final Report, Vol. 1, pp. 26-36 (Roesch, W. C.; ed.). Hiroshima, Japan: Radiation Effects Research Foundation; 1987.

Munch, I. V. *Re-investigation of the Hiroshima Disaster, Translation M-7*. U.S. Army: Letter to Capt. W. C. Youngs: Translated Japanese Language Documents; March 5, 1946.

Nagaoka, S. *Hiroshima Under Atomic Bomb Attack*. Hiroshima, Japan: Peace Memorial Museum; circa 1960.

Pace, N.; Smith, R. E. *Measurement of the Residual Radiation at the Hiroshima and Nagasaki Atomic-Bomb Sites*. Bethesda, Maryland: Naval Medical Research Institute; NMRI-160A; April 16, 1946 (also Hiroshima, Japan: Radiation Effects Research Foundation; TR 26-59; 1959).

Pace, J. V., III; Kerr, G. D. "Sulfur Activation in Electric Pole Insulators in Hiroshima." In: *Second US-Japan Joint Workshop for Reassessment of Atomic Bomb Radiation Dosimetry in Hiroshima and Nagasaki*, pp. 56-58 (Kato, H.; et al.; eds.). Hiroshima, Japan: Radiation Effects Research Foundation; 1984.

Poston, J. W.; Knight, J. R.; Whitesides, G. E. "Calculation of the HPRR Neutron Spectrum for Simulated Nuclear Accident Conditions." *Health Phys.* 26: 217-221; 1974.

Remec, I. *Cross Sections*. Oak Ridge, Tennessee: Oak Ridge National Laboratory; E-mail to G. D. Kerr; November 11, 2002.

Science Council of Japan. *Collection of the Reports on the Investigation of the Atomic Bomb Casualties*. Tokyo, Japan: Japanese Science Promotion Society; 1953.

Shimizu, S. "Historical Sketch of the Scientific Field Survey in Hiroshima Several Days After the Atomic Bomb Explosion." *Bull. Inst. Chem. Res. Kyoto University* 60: 34-54; 1982.

Shimizu, S.; Saigusa, T. "Estimation of ^{60}P Induced in Sulfur in Utility-Pole Insulators." In: *US-Japan Joint Reassessment of Atomic Bomb Radiation Dosimetry in Hiroshima and Nagasaki, Final Report, Vol. 2*, pp. 266-268 (Roesch, W. C.; ed.). Hiroshima, Japan: Radiation Effects Research Foundation; 1987.

Sugimoto, A. "Determination of the Number of Fast Neutron Particles Emitted at the Time of the Hiroshima A-Bomb Explosion." In: *Collection of Investigative Reports on Atomic Bomb Casualties*, Vol. 1, pp. 19-20. Tokyo, Japan: Japanese Science Promotion Society; 1953.

Swaja, R. E. *Attenuation of Porcelain Insulator*. Oak Ridge, Tennessee: Oak Ridge National Laboratory; Memorandum to G. D. Kerr; July 22, 1985a.

Swaja, R. E. *Attenuation of Fast Neutrons by Electrical Insulator for the HPRR Steel-shielded Spectrum*. Oak Ridge, Tennessee: Oak Ridge National Laboratory; Memorandum to G. D. Kerr; November 4, 1985b.

Tajima, E. *Locations of Sulfur Samples at Hiroshima*. Tokyo, Japan: Nuclear Safety Commission; Letter to G. D. Kerr; December 1, 1982.

Tajima, E.; Oda, N. "Estimation of Radiation Dose due to Bombing Attack." In: *Annual Report of Cooperative Research*, Radiation Section 33, p. 122. Tokyo, Japan: Japanese Ministry of Education: 1958 [also Oak Ridge, Tennessee; Oak Ridge National Laboratory; Translation OLS-82-125; 1982, and *US-Japan Joint Reassessment of Atomic Bomb Radiation Dosimetry in Hiroshima and Nagasaki, Final Report, Vol. 2*, pp. 269-271 (Roesch, W. C.; ed.). Hiroshima, Japan: Radiation Effects Research Foundation; 1987].

Yamada, H. *Locations of Hiroshima Infantry Regiment Headquarters and Hiroshima Military Hospitals*. Hiroshima, Japan: Radiation Effects Research Foundation; Letter to G. D. Kerr; July 12, 1984.

Yamasaki, F. “Estimation of Radiation Dose to the Victims of Hiroshima and Nagasaki.” In: *Annual Report of Cooperative Research*, Radiation Section 33, p. 121. Tokyo, Japan: Japanese Ministry of Education: 1958 [also Oak Ridge, Tennessee: Oak Ridge National Laboratory; Translation OLS-82-125; 1982].

Yamasaki, F.; Sugimoto, A. “Radioactive ^{32}P in Sulfur in Hiroshima.” In: *Collection of Investigative Reports on Atomic Bomb Casualties*, Vol. 1, pp. 18-19. Tokyo, Japan: Japanese Science Promotion Society; 1953 [also *US-Japan Joint Reassessment of Atomic Bomb Radiation Dosimetry in Hiroshima and Nagasaki, Final Report, Vol. 2*, pp. 246-247 (Roesch, W. C.; ed.). Hiroshima, Japan: Radiation Effects Research Foundation; 1987].

Yamasaki, F.; Sugimoto, A.; Kimura, K. “Radioactive ^{32}P Found in Human Bone in Hiroshima.” In: *Collection of Investigative Reports on Atomic Bomb Casualties*, Vol. 1, pp. 16-18. Tokyo, Japan: Japanese Science Promotion Society; 1953