Chapter 9

ACTIVATION MEASUREMENTS FOR FAST NEUTRONS

Part B. ^{63}Ni Measurements by Accelerator Mass Spectrometry (AMS)

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Introduction

The capability to measure ^{63}Ni in copper samples was developed to provide modern validation data for fast neutrons in Hiroshima and Nagasaki (Shibata 1994; Shibata et al. 1994a; Shibata et al. 1994b; Straume and Marchetti 1994; Marchetti and Straume 1996; Straume et al. 1996; Marchetti et al. 1997; McAninch et al. 1997; Ito et al. 1999; Rühm et al. 2000; Rugel et al. 2000; Knie et al. 2000). Two approaches involving the measurement of ^{63}Ni in bomb-exposed copper samples were independently considered, an accelerator-mass-spectrometry (AMS)-based approach (Straume and Marchetti 1994) and a low-background scintillation counting approach (Shibata 1994). A meeting of Japanese and U.S. scientists was held at the Radiation Effects Research Foundation in Hiroshima on August 2-3, 1994 to coordinate intercomparisons between these two measurement approaches. At this meeting, both approaches were presented, discussed, and agreements made for subsequent intercomparisons of laboratory-irradiated copper samples.

The present approach was to develop AMS-based methods to detect atoms of ^{63}Ni produced by neutrons above about 1 MeV in copper via the reaction, ^{63}Cu(n,p)^{63}Ni. The half-life of ^{63}Ni is 100.1 ± 2.0 y (Tuli 2000) and is sufficiently long to permit detection more than 50 years after the bombings. For the Hiroshima neutron spectrum, 95% of the ^{63}Ni is produced in copper samples by neutrons above about 1.4 MeV at 1 km from the hypocenter. For comparison, 95% of the ^{32}P in sulfur samples was produced by neutrons above about 2.5 MeV.

This effort involved the development of ultra-pure chemical separation methods to extract nickel from the copper samples and, thereby, reduce the isobaric interference of ^{63}Cu, the development of clean (i.e., low-copper) sample holders, the evaluation and development of low

copper AMS components (e.g., ion sources, pumps, etc.), and the use of a gas filled magnet at the Munich AMS facility, which permitted the measurement of large distance samples and of short distance samples high in stable nickel.

AMS was selected as the analytical tool because it is well suited for high sensitivity measurements of long-lived isotopes in small amounts, e.g., where the decay rate of the sample is too low to be effectively measured by radiation counting methods. AMS is an atom counting technique and, therefore, is independent of the decay rate of the sample. Samples for AMS measurements are generally in solid form with typically a few milligrams of mass. The sample is contained in a target holder that is normally a metal cylinder with a small hole to load the sample. The sample is ionized with a cesium sputter source to produce negative ions (atoms and molecules) that go through a low-energy mass spectrometer for single mass selection. Subsequently, the selected negative ions are accelerated towards the positive terminal of a tandem accelerator. As the ions go through a stripper (gas or thin foil), they experience collisions that destroy interfering molecular species and remove electrons from the atoms forming positive ions. The terminal potential accelerates the positive ions out of the tandem into a high-energy spectrometer. In general, the rare isotope is counted in an ionization chamber, which allows isobaric discrimination, while the corresponding stable isotope is measured as a current in a Faraday cup. The actual measurement is a ratio of the rare isotope counts to the current of the stable isotope. Normalization standards are required to properly determine the ratio of rare to stable isotope in the sample. The quantity of stable isotope present in the sample, that may include carrier, has to be known for an absolute calculation of the rare isotope present in the sample.

It should be noted that fast neutron measurements were made by Japanese scientists in Hiroshima within days following the atomic-bomb explosion (Chapter 9, Part A). Those measurements, which detected ^{32}P in sulfur, have provided indispensable data to help validate the calculated yield for the Hiroshima bomb. However, a method was needed that could extend the fast neutron measurements in Hiroshima to the larger distances most relevant to the atomic-bomb survivor locations. In 2001, as part of an evaluation of RERF dosimetry, the U.S. National Academy of Sciences, National Research Council, reviewed the ongoing program to measure ^{63}Ni in copper samples from Hiroshima and Nagasaki and recommended the completion of those measurements (NAS-NRC 2001).

Materials and Methods

Copper Samples

Before ^{63}Ni could be measured in Hiroshima, suitable copper samples had to be collected. Most importantly, there had to be assurance that the samples were of known location at the time of the bombing. Furthermore, the samples would be most useful if they were in line-of-sight with no or minimal intervening shielding. Also, samples should span the range of distances most relevant to the atomic-bomb survivor health studies. The sampling effort recovered a number of line-of-sight copper samples from Hiroshima. Measurements of ^{63}Ni have been completed for bomb-exposed copper samples located at six distances in Hiroshima ranging from 391 m to 1,470 m from the revised DS02 hypocenter (Table 1).

Table 1. Bomb-exposed copper samples from Hiroshima for which ^{63}Ni measurements have been completed using AMS

Sample location & description	DS86 ground range (m)	DS02 ground range (m)	Sample height above ground (m)	Year of construction[a]
Bank of Japan, lightning conductor (line-of-sight)	380	391	18	1936
Soy Sauce Brewery, lightning conductor (inside an iron pipe)	949	964	15.5	1933
Hiroshima City Hall, lightning conductor (inside an iron pipe)	1014	1018	18	1928
Hiroshima University, Elementary School, rain gutter (line-of-sight)	1301	1308	13	1938
Hiroshima University, Radioisotope Building, rain gutter (line-of-sight)	1461	1470	6	1934

[a]Provided by RERF (Dr. Fujita).

As indicated in Table 1, two copper lightning conductors were inside iron pipes. The conductor obtained from the Soy Sauce Brewery building was inside an iron pipe mounted on a concrete chimney, which survived the blast. The iron pipe had 3.5 cm outside diameter and 0.35 cm thick wall. The conductor obtained from the Hiroshima City Hall was inside an iron pipe located on the roof of the building. This pipe had 5.0 cm outside diameter and 0.40 cm thick wall. The conductor was composed of ten copper wires twisted tightly together to form a wire bundle approximately 1 cm in diameter. The other copper samples listed in Table 1 were either lightning conductors or copper rain gutters and were all in line-of-sight without any intervening shielding. Additional shielding details are given in Chapter 9, Part E.

Distant copper samples have been collected from 1,880 m to 7,500 m in Hiroshima and from 2,683 m to 4,428 m Nagasaki (Table 2). These large distance samples provide information on the ^{63}Ni in copper samples not significantly exposed to bomb neutrons. The sample collected from the entrance pillar to the Ohban Shrine was unfortunately composed of alloy materials with only 69.5% Cu. It was not suitable for electrochemical purification and sample preparation for AMS analysis.

In addition to background copper samples from Hiroshima, we have also measured ^{63}Ni in three aliquots from a modern copper wire obtained from a hardware store in Livermore and three aliquots from a copper sample obtained from an old church roof in Germany.

Copper Chemistry

The major isotopic component of natural Cu is ^{63}Cu (69.17%), which is also an isobar interference for ^{63}Ni AMS. The presence of isobaric interferences in the sample matrix largely limits the sensitivity of the AMS method, as they are the dominant machine background. Therefore, the challenge in sample preparation was to extract efficiently ~10^6 atoms of ^{63}Ni from gram amounts of isobar interference, and convert them into a solid form, avoiding Cu contamination. Also, because of the very limited amounts of copper materials available, it was important to develop a method that could use small samples.

Table 2. Large distance copper samples available from Hiroshima and Nagasaki

Sample location & description	Distance from hypocenter (m)[a]	Year of construction[b]	Measurement status
HIROSHIMA SAMPLES			
Higashi-Matsubara Branch, Sumitomo Bank (rain gutter)	1880	1921	Completed
Ohban Shrine (entrance pillar)	3392	1930	Not measurable
Kusatsu-Hachiman Shrine (copper roof)	5062	1931	Completed
Kamesaki Shrine (ornament)	7500	~1900	Completed[c]
NAGASAKI SAMPLES			
Shofuku-ji temple (roof ornament)	2683	1703	Not measurable
Minori-en (lightning rod)	3931	1927	One measurement
Nagasaki Branch, Hong Kong-Shanghai Bank (rain gutter)	4187	1896	One measurement
Oura-Tensyudou Church (rain gutter)	4428	1864	One measurement

[a]For these large distance samples, separate distances for the DS02 and DS86 dosimetry systems are not available.
[b]Provided by RERF (Dr. Fujita).
[c]This sample was high in stable Ni (251.9 ppm) and it was only possible to obtain an upper limit value.

Nickel and copper have well separated standard reduction potentials: E^o = +0.337 V for the reduction of Cu^{2+} to Cu^0, and E^o = -0.250 V for the reduction of Ni^{2+} to Ni^0. Therefore, an efficient electrochemical separation of the two metals is achievable. Under ideal conditions, an electrode held at -0.1 V in a solution containing Cu^{2+} and Ni^{2+} will reduce Cu^{2+} to Cu^0 until the concentration of Cu^{2+} decreases to $\sim 2 \times 10^{-15}$ M, while any practical concentration of Ni^{2+} in the same solution will remain unchanged. Thus, an electrochemical method was developed to separate Ni from Cu metal (Marchetti et al. 1997). The method was successfully tested using Cu wires that were irradiated with neutrons from a ^{252}Cf fission source; the results showed ~100% quantitative recovery of the expected ^{63}Ni.

The electrochemical separation yielded the Ni in solution as Ni^{2+}, which had to be converted into a solid form to be loaded into the AMS target holder. Copper is such a ubiquitous element that the addition of reagents and sample manipulation involved in this step is likely to contaminate the sample again with unacceptable levels of Cu. However, Ni forms a unique carbonyl compound, $Ni(CO)_4$, that is a gas at room temperature, while, on the other hand, Cu does not form any volatile carbonyl compound. Based on this, a method was developed to separate Ni in solution as carbonyl gas. The $Ni(CO)_4$ was then transported by a stream of He to a heated target holder where it was thermally decomposed depositing Ni metal directly into the sample cavity (McAninch et al. 1997). This methodology minimized any potential for Cu contamination particularly from reagents.

Initial ^{63}Ni AMS testing indicated high instrumental background due to ambient Cu in the ion source. Dedicated ion source hardware had to be implemented and special Cu-free target holders had to be manufactured from high purity graphite (99.9995%, Johnson Matthey Co.) to reduce

the Cu background to acceptable levels.

The presence of Ni as an impurity in the Cu sample is an important consideration for two reasons: a) AMS measures the ratio of ^{63}Ni to a stable isotope of Ni (^{58}Ni), and b) the contribution to ^{63}Ni from the reaction ^{62}Ni(n,γ)^{63}Ni. The total Ni content of the sample was measured in aliquots of the solution obtained after the electrolytic separation using graphite furnace atomic absorption spectrometry (GFAA). In general, samples required at least ~200 μg of Ni to produce a sample viable for measurement.

Sample Preparation for ^{63}Ni AMS

Each copper sample was polished to eliminate surface oxide, and weighed. Then, it was wrapped with a platinum mesh and connected as the anode in a conventional 3-electrode cell arrangement. The cathode was a cylindrical platinum mesh, the reference electrode was a standard silver chloride electrode (3M NaCl/AgCl/Ag), and the supporting electrolyte was H_2SO_4 1 M.

The electrochemical equipment was set to controlled-current mode and a small initial current (50-100 mA) was circulated through the cell. Subsequently, the current was increased gradually to 0.8-1.0 A. Once the integrated charge corresponded roughly to the amount of Cu of the sample, the cell was disconnected and the anode removed. The remaining copper was rinsed with deionized water, dried, and weighed to determine the amount of Cu dissolved. Then a platinum wire was connected as the anode to finish the electrodeposition of the Cu^{2+} still remaining in solution in controlled-potential mode. The cell was disconnected and the electrodes removed quickly. The electrolyte solution was filtered using a 0.2 μm cellulose acetate disposable filter. The filtrate was taken to a pH>8 by addition of NaOH 4M. The $Ni(OH)_2$ precipitated was filtered using a 0.2-μm cellulose acetate disposable filter. The precipitate was dissolved in 5 mL of 1M HNO_3. The sample solution was placed in a 50-mL three-necked flask and concentrated ammonia was added to pH~8. The solution was purged with a mixture of CO and He, while a ~0.5% fresh solution of sodium borohydride ($NaBH_4$ is a strong reducing agent) was added dropwise. The BH_4^- reduced the Ni^{2+} to Ni^0, which reacted with the CO to form the volatile $Ni(CO)_4$. The carbonyl was condensed in a liquid nitrogen U-trap. After the reaction was completed, CO was cut off and only He flowed through the system. The trap was allowed to warm up to room temperature, and the $Ni(CO)_4$ was carried by the He stream to a glass capillary tip inserted in the sample hole of a graphite target holder that was heated at about 200°C. The carbonyl was thermally decomposed, depositing the Ni metal directly in the target holder.

Calibration ^{63}Ni/Ni ratio standards were prepared using a NIST-traceable ^{63}Ni solution (Du Pont Radiopharmaceutical Division) that was serially diluted using commercial 1,000 μg/mL Ni standard solutions (VWR Scientific). The ^{63}Ni/Ni ratios for the standards ranged from 3×10^{-13} to 1×10^{-8}. About 1 mL of a given ^{63}Ni/Ni ratio solution (~1 mg of total Ni) was processed by the carbonyl method to generate one calibration target. Likewise, blanks were prepared using 1 mL of 1,000 μg/mL Ni standard solutions.

AMS Measurement of ^{63}Ni at Lawrence Livermore National Laboratory (LLNL)

The Center for Accelerator Mass Spectrometry (CAMS) at LLNL has a 10-MV FN tandem. The measuring system consists of a cesium-sputtering source, a low energy 90° spectrometer, a

high energy 90° spectrometer, a 90° rigidity filter, a Wien velocity filter, and a four-anode ionization chamber. For ^{63}Ni measurement, a tandem terminal voltage of 9.0 MV was applied, and carbon foil was used as stripper. The charge state selected was Ni^{10+} that was produced at ~22% efficiency. To reduce instrumental background, a dedicated ionizer made out of high purity molybdenum was used in the ion source. In addition, the installation of a cryopump, replacing a turbomolecular pump, reduced the Cu background by a factor of ~10. Initial test measurements (Table 3) were performed using an X-ray detection system instead of the ionization chamber to measure ^{63}Ni (McAninch et al. 1997). However, all other measurements were made using the ionization chamber. The CAMS instrument was able to reach background levels from ^{63}Cu corresponding to a ^{63}Ni/Ni ratio of ~10^{-12}.

AMS Measurement of ^{63}Ni at Munich

The Munich AMS facility is a combination of an MP tandem and an analyzing system that includes a 135° gas-filled magnet (GFM) (Knie et al. 1997). The experimental setup to measure ^{63}Ni consists of a dedicated cesium sputter source (Rugel et al. 2000), an injection system with a 90° magnetic dipole, and a subsequent 18° electrostatic deflector. Following the tandem accelerator, the high-energy spectrometer includes a Wien velocity filter, a 90° rigidity filter, a time-of-flight detector, the GFM, and an ionization chamber (Knie et al. 1997, 2000).

The ion source used normally at Munich produced unacceptably high Cu backgrounds. Pure Ni samples resulted in Cu/Ni ratios of 10^{-5} to 10^{-2}, depending on sample, sample holder, source parameters, etc. In order to decrease this background level, a new Cs sputter source was developed. The basic features of this source consist of an open geometry with large pumping units and a water-cooled surface to reduce cross talk and improve vacuum. All technical components were checked for their copper content, and those normally made of copper such as wires, target holders, gaskets, and so forth were replaced with copper-free materials. The sample introduction system was equipped with an air-lock to maintain the ion source vacuum during target changing. This new source reduced copper background by about two orders of magnitude, as determined by the ^{63}Cu/Ni ratio (as low as 10^{-6}) measured in ultrapure nickel samples (Rugel et al. 2000).

Table 3. First ^{63}Ni measurements of test samples

Sample	Lab	Cu Mass (g)	Carrier Ni (μg)	Total Ni (μg)	Measured ^{63}Ni/Cu (10^7 at/g)	Calculated ^{63}Ni/Cu (10^7 at/g)[a]
Unirradiated Cu wire	LLNL	2.92	1093	1102	–9 ± 17[b]	0
LLNL irradiated Cu wire No. 1	LLNL	19.81	1105	1172	54 ± 11	47
LLNL irradiated Cu wire No. 2	LLNL	2.24	1095	1102	57 ± 15	52
LLNL irradiated Cu wire No. 2	LLNL	4.33	1083	1197	117 ± 11	101
LLNL irradiated Cu wire No. 2	LLNL	8.73	1103	1132	213 ± 14	204

[a]ENDF/B-VI cross sections.

[b]Negative mean value due to the subtraction of background. Uncertainties include background errors.

The terminal voltage of the tandem accelerator used for the ^{63}Ni measurements was typically set to 13.5 MV. The charge state selected was Ni^{12+}, which was formed with 14% probability using a carbon foil stripper (thickness 4 $\mu g/cm^2$) at the terminal, resulting in an ion beam energy of 176 MeV. During the measurement series the GFM was moved to a position much closer to the tandem accelerator compared to its position at the beginning of this study, which increased the transmission through the system by a factor of about 2. In addition, a new control system for the terminal voltage improved the quality of the beam in terms of stability and reproducibility.

After passing through the GFM, isobars such as ^{63}Ni and ^{63}Cu showed a different mean charge state due to charge changing collisions with the gas molecules in the magnet and exit the GFM at different positions. This offers the possibility to distinguish between isobars and to improve the detection limits for an AMS measurement significantly. In this context, the use of a large tandem accelerator and the corresponding high particle energies is of great advantage, since multiple scattering in the GFM is reduced and a high gas pressure is possible. This results in improved statistics of charge state fluctuations. Additionally, the higher residual energy and lower energy straggling at the exit of the GFM improves the quality of the additional atomic charge determination, which is performed by energy loss measurements in the ionization chamber.

The final detector is a Frisch-grid ionization chamber. The anode of this ionization chamber is divided into five energy loss sections along the flight direction of the incoming projectiles. The first and third sections of the divided anode are split diagonally to determine the position and the horizontal angle of the incoming particles. To obtain information about the vertical angle, the time difference between the second and fourth energy loss signals is measured. An independent signal for the total residual energy is taken from the Frisch-grid itself. Isobutane was used as detector gas at a pressure of about 40 mbar. By measuring a standard with high ^{63}Ni concentration (^{63}Ni/Ni ~10^{-10}), the detector signals are analyzed and the position of the ^{63}Ni peak in the corresponding spectra defined. Events other than those contributed to ^{63}Ni are rejected by means of software cuts.

For the AMS measurements, the magnetic field of the GFM was chosen in a way that most of the ^{63}Ni could still enter the detector (about 80%), but the ^{63}Cu ions were blocked by an aperture due to their higher mean charge state in the gas and the corresponding smaller bending radius in the magnet. With nitrogen as a gas and a gas pressure of about 7 mbar (corresponding to an energy loss of 1/3 of the total energy), the count rate of the ^{63}Cu interference in the detector was reduced by a factor of up to 3,000. This allows measurements with high ion currents from the ion source at still modest detector count rates.

During our AMS measurements, the combination of a GFM and an ionization chamber resulted in a total isobaric suppression of about 5×10^9. That is, one ^{63}Ni ion can be distinguished from 5×10^9 ^{63}Cu ions entering the gas-filled analyzing system. After the modifications described above, the Munich instrument was able to reach background levels of ^{63}Ni/Ni as low as 2×10^{-14} (Rühm et al. 2000).

Method Validation

The first step was to produce Cu samples with known concentrations of ^{63}Ni that would be good surrogates for Hiroshima Cu samples. One way to achieve this is by irradiating Cu with neutrons from a fission source. Two irradiations of electrical Cu wire were conducted at LLNL

using a ^{252}Cf source. One irradiation lasted 1 day and the other 10 days. The fast neutron fluence was monitored with sulfur detectors by the reaction ^{32}S(n,p)^{32}P and by measuring the activity of ^{32}P using electrometers. The Monte Carlo code MCNP (Briemeister 1997) was used to calculate the yield of ^{63}Ni in the irradiated samples. The neutron spectrum for ^{252}Cf required as input for the MCNP code was obtained from an evaluation made at the National Institute of Standards and Technology (Lamaze and Grundl 1988), and the ENDF/B-VI cross-section library used for the reaction ^{63}Cu(n,p)^{63}Ni was obtained from Rose (1991). The results of the calculations were 2.37×10^7 and 2.33×10^8 ^{63}Ni/g of Cu for the one and ten day irradiations, respectively. The relative uncertainty in the calculation due to neutron fluence and cross-section values was estimated to be about 20%.

Aliquots of the irradiated wires were processed as described above. Once each aliquot was placed in the electrolytic cell, 1 mg of Ni carrier was added (i.e., 1 mL of a commercial standard solution of Ni^{2+} 1,000 μg/mL). Unirradiated wire was also processed as a blank. The Cu wire contained Ni at a concentration of 3.4 ± 0.5 μg/g, as measured by GFAA in three one-gram samples of wire. The Ni contribution of the Cu wire was not significant compared to the carrier, but it was included in the total Ni calculation. For each of the samples, total Ni was determined by spectrophotometry (Marczenko 1976) in an aliquot of the final solution that was used later in the carbonyl generation. The average yield was 94.7 ± 4.3%, which may be considered practically 100%, because of expected loses during transfer, precipitation, and dissolution processes. The AMS measurement of the ratio ^{63}Ni/Ni was used to verify that ^{63}Ni was extracted quantitatively from the Cu wire. The results are presented in Table 3. A weighted least square fit of the measured ^{63}Ni as a function of the calculated ^{63}Ni resulted in a slope of 1.066 ± 0.052 (note that the calculated values have an uncertainty of ~20%), which indicates that the extraction of ^{63}Ni from Cu was 100% efficient within the uncertainties. Because of its nature, the carbonyl procedure is not quantitatively depositing all the Ni into the target holder, and a substantial fraction of the sample is lost. Early estimates indicated that only about 20-30% of the Ni mass ended up in the AMS target holder. Therefore, the 200 μg of Ni required to produce a measurable AMS target are only nominal. It is also important to note that the efficiency of this step should not affect the value of the ratio measured, because it is not expected that Ni would fractionate isotopically during the carbonyl procedure.

After the initial test at LLNL, Cu was sent to NIST to be irradiated with a calibrated ^{252}Cf source. NIST irradiated coils of Cu wire with the following fluences: a) 3.89×10^{13}, b) 4.02×10^{13}, and c) 4.03×10^{14} n/cm^2. In addition, Cu metal irradiated in Japan using a ^{252}Cf source was provided to us by T. Shibata for method intercomparison purposes. This sample had a concentration of ^{63}Ni of 0.0096 ± 0.0009 Bq/g or $(4.36 \pm 0.41) \times 10^7$ atoms/g. Aliquots of these samples were processed with the addition of Ni carrier and measured at LLNL. Results of these measurements are presented in Table 4. One sample is listed as irradiated by Munich and corresponds to a piece of Cu irradiated with cold neutrons at the Garching Research Reactor close to Munich. For the NIST irradiations, the amount of ^{63}Ni/g of Cu was calculated using MCNP to estimate the neutron fluence as a function of energy, which was then folded with the cross section for the reaction ^{63}Cu(n,p)^{63}Ni. As noted in Table 4, two values are presented for each sample corresponding to the libraries ENDF/B-VI and JENDL-3 (Shibata et al. 1990). The production of ^{63}Ni using JENDL-3 is about 25% higher than that obtained using ENDF/B-VI. The ^{63}Ni expected in the sample irradiated in Japan was calculated using the experimental activity and a half-life of 100.1 y for ^{63}Ni. There was no calculation performed for the expected

Table 4. ^{63}Ni measurements of irradiated test samples

Sample	ID	AMS facility[a]	Cu mass (g)	Carrier Ni (μg)	Total Ni (μg)	Measured ^{63}Ni/Ni (10^{-12} at/at)	Measured ^{63}Ni/Cu (10^{6} at/g)	Calculated ^{63}Ni/Cu (10^{6} at/g)
MUNICH irradiated Cu	H568	LLNL	9.08	390.3	392 ± 22	23.8 ± 0.8	10.5 ± 1.0	na
MUNICH irradiated Cu	H577	MUNICH	9.08	390.3	392 ± 22	25 ± 4	11.1 ± 1.8	na
Shibata irradiated Cu	H456	LLNL	2.79	193.5	207 ± 10	65 ± 22	49 ± 17	43.6
NIST irradiated Cu wire No. 1	H571	LLNL	11.51	194.5	229 ± 16	129 ± 9	26 ± 3	16.8[b]–21.0[c]
NIST irradiated Cu wire No. 5	H455	LLNL	11.55	193.1	228 ± 10	115 ± 37	23 ± 8	17.4[b]–21.8[c]
NIST irradiated Cu wire No. 5	H464	LLNL	11.73	195.0	230 ± 10	111 ± 3	22 ± 1	17.4[b]–21.8[c]
NIST irradiated Cu wire No. 5	H641	MUNICH	6.68	1203	1302	10.2 ± 1.0	20.4 ± 2.1	17.4[b]–21.8[c]
NIST irradiated Cu wire No. 6	H465	LLNL	10.53	190.3	222 ± 10	940 ± 21	203 ± 12	174[b]–218[c]
NIST irradiated Cu wire No. 6	H572	LLNL	5.46	196.5	229 ± 15	604 ± 35	260 ± 24	174[b]–218[c]

[a]The chemistry and sample preparation for all samples were performed at Livermore.
[b]ENDF/B-VI cross sections.
[c]JENDL-3 cross sections.

amount of ^{63}Ni for the sample irradiated with cold neutrons. However, it was included as a quality control sample to test consistency between the AMS results from both laboratories.

Total Ni Content in Hiroshima Cu Samples

The concentration of Ni impurity in Cu samples had to be known accurately to quantify the contribution from the ^{62}Ni(n,γ) reaction to ^{63}Ni, and to allow the determination of the absolute concentration of ^{63}Ni in Cu from the measured ^{63}Ni/Ni ratio. In addition, it was necessary to know the mass of Cu sample needed to obtain a minimum of about 200 μg of Ni required to prepare a viable AMS target. The addition of Ni carrier was not desirable for samples at large distances from the hypocenter, because such addition could bring the ^{63}Ni/Ni ratio below the detection limit. In principle, an ideal Cu sample would not have any Ni impurity. Thus, for a given amount of carrier, a measurable ^{63}Ni/Ni ratio could be obtained just by processing enough Cu material. However, in practice, the sensitivity is limited by the intrinsic content of Ni in the sample that fixed the highest possible limit to the ^{63}Ni/Ni ratio.

The analytical method of choice to measure the Ni^{2+} concentration in solution after the electrochemical separation was GFAA. A few samples were also analyzed by inductively coupled plasma atomic emission spectrometry. The first samples received from Hiroshima were expected to contain enough ^{63}Ni that Ni carrier was added to the electrolysis solution.

Occasionally, significant differences in the Ni content between aliquots of Cu from the same sample were observed. To detect analytical errors, sample solutions were submitted as blind duplicates for analysis. In addition, an independent private laboratory analyzed some of the sample aliquots for quality control, which were also submitted as blind duplicates (Galbraith Laboratories, Tennessee). One possibility for variations in Ni content is that these represent inhomogeneities in the Ni distribution within the Cu sample. Another possibility is that they represent Ni contamination from an external source. The samples were prepared very carefully, and all the materials were acid washed and copiously rinsed with deionized water between preparations. Therefore, contamination seems very unlikely. It is noteworthy that the copper sample with the largest inter-aliquot differences in Ni content was the lightning conductor from the Hiroshima City Hall. That sample was composed of a bundle of about ten copper wires. Typically, one or two wires would be used to make a sample for AMS measurement. Hence, each wire could potentially be from a different copper source and then bundled together to form the lightning conductor cable.

To deal with the potential intra-sample variability of Ni concentration, it was decided that the Ni content used to obtain ^{63}Ni from the ^{63}Ni/Ni ratio would be based on the Ni content in each aliquot evaluated. That is, no average sample concentration would be used. It was the correct approach if the samples were indeed inhomogeneous. On the other hand, this approach intrinsically assumed that if there was an external source of Ni contamination, it would not likely contain ^{63}Ni. Therefore, the effect of external contamination would be to dilute the ^{63}Ni/Ni ratio, but the ^{63}Ni content would be correct insofar as the total Ni concentration was correct. The stable nickel analyses were done as blind replicates. Note that some of the resulting solutions were subsequently combined to produce one target for AMS measurement of ^{63}Ni. This was done when the total amount of Ni in the solution was less than the desired 200 μg necessary to produce a viable AMS target. Some solutions contained enough Ni to produce more than one AMS target. These are solutions originating from Cu samples with high Ni content.

Measurement Uncertainties

All ^{63}Ni AMS measurements were performed "blind" (i.e., without the measurement laboratory knowing the Ni content of the sample until after the measurement had been completed), preventing any potential measurer-induced bias.

The uncertainty associated with the mass of the sample aliquot is very small (less than 0.1%) and is primarily dependent on the accuracy of the scale used to weigh the sample. However, the uncertainty in the Cu fraction of the sample is larger, on the order of 1%. This is due to uncertainties in the Cu measurements of the samples.

The uncertainties associated with the mass of nickel carrier added to the sample are principally dependent on the accuracy of weighting 100-1,000 μL aliquots from a stock solution, which is in the 0.1% range. The uncertainties associated with the concentration of nickel in the copper are based on our GFAA measurements for each sample after dissolution of the copper sample in preparation for making the AMS sample target.

The uncertainties listed for the ^{63}Ni/Ni ratios include AMS statistical and systematic uncertainties. Systematic uncertainties included variation in AMS normalization (due to changes in the transmission of the machine), and uncertainty in the subtraction of machine background ^{63}Cu events (estimated from measurements of blanks). The uncertainties for the ^{63}Ni/Ni ratios measured in Munich were obtained according to the approach outlined by Feldman and Cousins (1998) for small signal statistics.

After calculating the means and deviations for each measured sample aliquot, the weighted means and weighted deviations were obtained for each sample location (note that multiple measurements were made at each sample location). The weighting used 1/variance and resulted in the number of atoms ^{63}Ni/g Cu at the time of measurement for each sample location.

The next step was to subtract ^{63}Ni background. This employed the results from the copper samples from the Sumitomo Bank (1,880 m) and the Kusatsu Hachiman Shrine (5,062 m). The ^{63}Ni/g Cu in the background copper samples have a weighted mean of 7.3 $(+2.4, -1.9) \times 10^4$.

Finally, this mean background value was subtracted from the line-of-sight sample results and then corrected for radioactive decay since 1945 using the half-life of 100.1 years for ^{63}Ni. The uncertainties were propagated in quadrature.

Sample-Specific Calculations

The calculations of the activation of ^{63}Ni in copper samples were performed using the Monte Carlo Adjoint Shielding Code System (MASH) (Johnson 1999). This is the same methodology used for both DS86 and DS02. The code is ideal for situations in which a shielded detector point is exposed to a large field of incident neutrons or gamma rays. Neutrons are started at the detector point and run backwards in space and time to exit a closed surface that surrounds the sample and shield. A further description of this code is in Chapter 9, Part E.

Geometry Model. The shielding around the point detector is modeled using a combination of primitive bodies, like boxes and ellipsoids. For each of the copper samples, geometric models consisting of about ten primitive bodies are constructed. The important aspects of the geometry are the detector point and building height, the incident angle, and any shielding between the detector point and the direction of the main source. For thermal neutron activation, and to a lesser

extent for fast neutron activation, structures not in the line-of-sight must be included when neutron scattering is important.

Materials. The building and sample materials can play a role in the calculation. The materials chosen for the nickel calculations are given in Chapter 9, Part E. For the Bank of Japan, a large variety of materials were used to determine the range of responses.

Transport Cross Sections. The neutrons were transported through the materials using ENDF/B-VI cross sections in the DABL69 group format (Ingersoll et al. 1989).

Fluences. The unperturbed air-over-ground neutron fluences are obtained from discrete ordinate calculations of both the prompt and delayed neutron sources. The fluences are discussed in Chapter 3.

Responses. The $^{63}Cu(n,p)^{63}Ni$ and $^{62}Ni(n,\gamma)^{63}Ni$ neutron responses are derived from ENDF/B-VI cross sections. They were converted from fine multipoint data to the DABL69 group structure by collapsing with representative spectra at Hiroshima 1 m above ground. There are other cross-section sets for the ^{63}Cu reaction that differ somewhat from ENDF/B-VI and are discussed in Chapter 9, Part E.

Coupling. Each neutron that reaches the closed surface is multiplied by the neutron fluence according to its energy and angle. That represents the activation at the detector point. The contributions from all the neutrons are averaged.

Results. The results from the DS02 sample-specific modeling calculations are listed in Table 5. The transmission factor includes both the effects of shielding and height of the sample from the 1-m above ground reference point. Notice that the fast transmission factor tends to be close to unity. That is because the copper samples were all in direct line-of-sight with no or minimal shielding. The thermal transmission factor is affected mainly by the height of the samples, which takes them away from the main thermal neutron source, i.e., the ground.

Uncertainties. It is estimated that the largest random uncertainties are transmission factors (~8%) and sample distances (~5%). The largest systematic uncertainty is from the activation cross section (~10%). It is estimated that the total random uncertainty associated with the calculation for each copper sample derived from the DS02 free-field fluence is about 10%. In addition, there is a 10% systematic uncertainty equally affecting all sample calculations. It is also noted that, due to a positive Q-value for the $^{63}Cu(n,p)^{63}Ni$ reaction, the cross section might not be zero at thermal energies, although measurements to date have suggested that it is small. Based on preliminary information, this could result in an additional systematic uncertainty of less than 10%. Future work to improve the cross section for $^{63}Cu(n,p)^{63}Ni$ could reduce somewhat the present systematic uncertainties associated with the Hiroshima sample calculations. For the present report, we have used what we consider a best-estimate SD for the sample-specific calculations of ±15%.

Table 5. Results for DS02 sample-specific modeling calculations

Location	Ground range (m)	Ni in Cu sample (μg/g)[a]	Transmission factor		^{63}Cu(n,p)[b] (fast)	^{62}Ni(n,γ)[b] (thermal)	Total[b] (fast + thermal)
			(fast)	(thermal)			
Bank of Japan	391	207	0.826	0.723	5.03E + 06	1.80E + 06	6.83E + 06
Soy Sauce Brewery	964	258	1.000	0.600	3.79E + 05	3.02E + 04	4.09E + 05
City Hall	1018	10	1.004	0.597	2.48E + 05	7.93E + 02	2.49E + 05
Elementary School	1308	16	0.820	0.793	4.49E + 04	1.77E + 02	4.51E + 04
Hiroshima U. E-Bldg.							
#1 (line-of-sight)	1385	25	1.046	0.904	3.68E + 04	1.76E + 02	3.70E + 04
#2 (line-of-sight)	1388	25	1.050	0.891	3.64E + 04	1.71E + 02	3.66E + 04
#4 (shielded)	1469	25	0.221	0.692	4.84E + 03	7.30E + 01	4.92E + 03
Radioisotope Bldg.	1470	10	1.067	0.812	2.38E + 04	3.28E + 01	2.38E + 04

[a]Total Ni concentration in Cu samples used to calculate ^{62}Ni(n,γ)^{63}Ni. These concentrations are approximations of the actual measured values, which varied somewhat between aliquots.

[b]Calculations of atoms ^{63}Ni/g Cu are for August 6, 1945 (at time of the bomb). The transmission factors are for the samples *in situ* and thus include both shielding and height. The uncertainties (SD) in the calculations are estimated to be about ±15%.

Results and Discussion

The results of the ^{63}Ni measurements and of total nickel concentration for line-of-sight samples from Hiroshima are presented in Table 6. For these samples, multiple measurements were made during independent beam times, and the results are weight averaged to provide ^{63}Ni/g Cu values. These ^{63}Ni/g Cu results are without background subtraction and without accounting for decay since 1945. Results for those samples that have been measured only once (i.e., not yet confirmed by additional independent measurements) are not included in Table 6.

The measured results after background subtraction and decay correction are listed in Table 7 along with sample-specific calculations based on either DS02 or DS86. The DS02 calculations were done using distances based on the revised hypocenter, 600-m source height, and 16-kt yield. The DS86 calculations were done using sample distances from the DS86 hypocenter and DS86 calculated air-over-ground fluences based on a 580-m source height and 15-kt yield (Roesch 1987). A background of 7.3 (+2.4, -1.9) $\times 10^4$ was subtracted from the line-of-sight measurements. The sample-specific calculations include both the ^{63}Cu(n,p)^{63}Ni fast neutron reaction and the ^{62}Ni(n,γ)^{63}Ni thermal neutron reaction (i.e., total ^{63}Ni produced in the copper samples by the bomb neutrons). The nickel concentrations listed in Table 6 for each sample were used in these calculations.

The measured ^{63}Ni/g Cu results without background subtraction or decay corrections are plotted in Figure 1. It is observed that the ^{63}Ni activation in the copper samples decreases rapidly from 391 m to about 1,500 m from the hypocenter and then levels off beyond 1,500 m and appears to remain essentially constant between 1,880 m and 5,062 m.

The weighted means and deviations with background subtraction and corrected to 1945 are plotted in Figure 2. For comparison, we have also plotted the sample-specific calculations using the DS02 dosimetry system. The calculations are for total atoms of ^{63}Ni/g Cu produced in the sample by the Hiroshima bomb neutrons, i.e., including both the fast neutron reaction ^{63}Cu(n,p)^{63}Ni and the thermal reaction ^{62}Ni(n,γ)^{63}Ni. Note that this is the correct comparison, since the measurements also include ^{63}Ni production from both reactions (Table 5). As indicated above, the calculations used the measured nickel concentration in each sample as input to obtain the production of ^{63}Ni from the (n,γ) reaction. It is observed in Figure 2 that DS02 agrees well with the ^{63}Ni measurements. The mean from the Bank of Japan measurements appears suggestively lower than the DS02 calculation but is not significantly different at the 95% confidence level.

Comparisons between our ^{63}Ni measurements in Hiroshima and DS86 calculations are seen in Figure 3. As with DS02, good agreement is observed between 900 m and 1,500 m from the hypocenter. However, in this case, the disagreement between measurements and calculations is significant at the Bank of Japan (380 m, DS86 ground distance). At this distance, the measurement is 36% lower than the DS86 calculation (M/C ratios listed in Table 7).

We have also compared the mean of our ^{63}Ni measurements at 391 m ground range with the mean of six ^{32}P measurements ranging from 364 m to 445 m (Chapter 9, Part A, Table 8). For DS02, the mean measured-to-calculated ratio is 0.85 ± 0.08 for our three ^{63}Ni measurements and 1.07 ± 0.22 for the six ^{32}P measurements. Neither of these M/C ratios is significantly different from unity. If the estimated uncertainty associated with the sample-specific calculations (15% for ^{63}Ni and 7% for ^{32}P) is included in the M/C ratios, then the standard deviations become ±0.15 for ^{63}Ni and ±0.24 for ^{32}P. It is concluded that the result obtained for the Bank of Japan copper sample is consistent with both the DS02 calculations and the ^{32}P measurements at that distance.

Table 6. Completed ^{63}Ni measurements in line-of-sight copper samples from Hiroshima

Sample location[a]	Sample ID	AMS facility[b]	Cu Mass (g)	Carrier Ni (μg)	Ni in Cu (μg/g)	Measured ^{63}Ni/Ni	-SD	+SD	^{63}Ni/g Cu measured[c]
Bank of Japan	H769	MUNICH	9.52	0	212.8 ± 14.5	2.05×10^{-12}	2.0×10^{-13}	2.1×10^{-13}	$(4.0 \pm 0.4) \times 10^{6}$
(391 m)	H823	MUNICH	3.37	0	199.1 ± 7.5	2.7×10^{-12}	9×10^{-13}	7×10^{-13}	
	H843	MUNICH	5.94	0	207.8 ± 1.2	1.62×10^{-12}	2.4×10^{-13}	2.4×10^{-13}	
Soy Sauce Brewery	H672+H673	MUNICH	12.31	0	258.3 ± 4.8	1.3×10^{-13}	0.9×10^{-13}	1.0×10^{-13}	$(4.4 \pm 1.4) \times 10^{5}$
(964 m)	H770	MUNICH	12.31	0	258.3 ± 4.8	1.8×10^{-13}	0.6×10^{-13}	0.6×10^{-13}	
City Hall	H461	LLNL	15.59	194.0	19.7 ± 11.9	2.2×10^{-12}	1.1×10^{-12}	1.1×10^{-12}	$(2.65 \pm 0.27) \times 10^{5}$
(1018 m)	H462	LLNL	20.75	193.7	18.6 ± 9.5	2.2×10^{-12}	3.0×10^{-13}	3.0×10^{-13}	
	H563	LLNL	36.89	192.8	4.1 ± 0.7	2.9×10^{-12}	3.3×10^{-13}	3.3×10^{-13}	
	H582	MUNICH	36.89	192.8	4.1 ± 0.7	1.9×10^{-12}	6×10^{-13}	6×10^{-13}	
	H583	MUNICH	29.63	192.9	9.1 ± 2.5	1.5×10^{-12}	4×10^{-13}	4×10^{-13}	
	H640	MUNICH	39.78	193.6	6.9 ± 2.1	3.1×10^{-12}	4×10^{-13}	4×10^{-13}	
Elementary School	H593[d]	MUNICH	24.16	103.5	16.1 ± 3.2	6.3×10^{-13}	1.1×10^{-13}	1.1×10^{-13}	$(1.10 \pm 0.14) \times 10^{5}$
(1308 m)	H638	MUNICH	13.71	104.2	18.4 ± 1.7	4.2×10^{-13}	1.1×10^{-13}	1.1×10^{-13}	
	H639	MUNICH	24.72	105.9	13.8 ± 0.4	5.5×10^{-13}	1.0×10^{-13}	1.0×10^{-13}	
Radioisotope Bldg.	H637	MUNICH	29.75	103.9	8.8 ± 0.2	5.5×10^{-13}	2.5×10^{-13}	2.5×10^{-13}	$(1.03 \pm 0.17) \times 10^{5}$
(1470 m)	H767	MUNICH	24.41	0	9.3 ± 0.8	1.09×10^{-12}	2.5×10^{-13}	2.5×10^{-13}	
	H671	MUNICH	26.92	0	11.1 ± 0.8	1.30×10^{-12}	2.8×10^{-13}	3.4×10^{-13}	

[a]Distance from DS02 hypocenter to sample.
[b]The chemistry and sample preparation for all samples were performed at Livermore.
[c]Not corrected for background and decay since 1945.
[d]Total Ni was not measured in aliquot H593. It is taken as the mean of the two other aliquots from the same sample.

Table 7. Completed ^{63}Ni measurements corrected for background and decay and compared with calculated ^{63}Ni in Hiroshima[a]

Sample location	DS02 ground range (m)	DS86 ground range (m)	Measured-bkg ^{63}Ni/g Cu ($\times 10^4$)			DS02 ($\times 10^4$) calculation (*in situ*)	DS86 ($\times 10^4$) calculation (*in situ*)	M/C DS02	M/C DS86
			Mean	-SD	+SD				
Bank of Japan	391	380	580	50	50	683	904	0.85 ± 0.15	0.64 ± 0.11
Soy Sauce Brewery	964	949	54	20	20	40.9	50.3	1.32 ± 0.54	1.07 ± 0.44
City Hall	1018	1014	28	5	5	24.9	30.3	1.12 ± 0.26	0.92 ± 0.22
Elementary School	1308	1301	5.4	3.4	4.1	4.5	5.6	1.20 ± 0.85	0.96 ± 0.68
Radioisotope Bldg.	1470	1461	4.5	3.8	4.5	2.4	3.0	1.88 ± 1.72	1.50 ± 1.38

[a]All ^{63}Ni/g Cu values are at time of bombing. Background of 7.3 (+2.4, -1.9) $\times 10^4$ ^{63}Ni/g Cu (Table 8) was subtracted from the measurements and then corrected for decay since 1945. Calculations are sample-specific *in situ* and include both the ^{63}Cu(n,p)^{63}Ni and ^{62}Ni(n,γ)^{63}Ni reactions. The uncertainties associated with the M/C ratios were estimated by combining the measurement and calculation uncertainties in quadrature. The SDs for the *in situ* calculations are ±15% of the means.

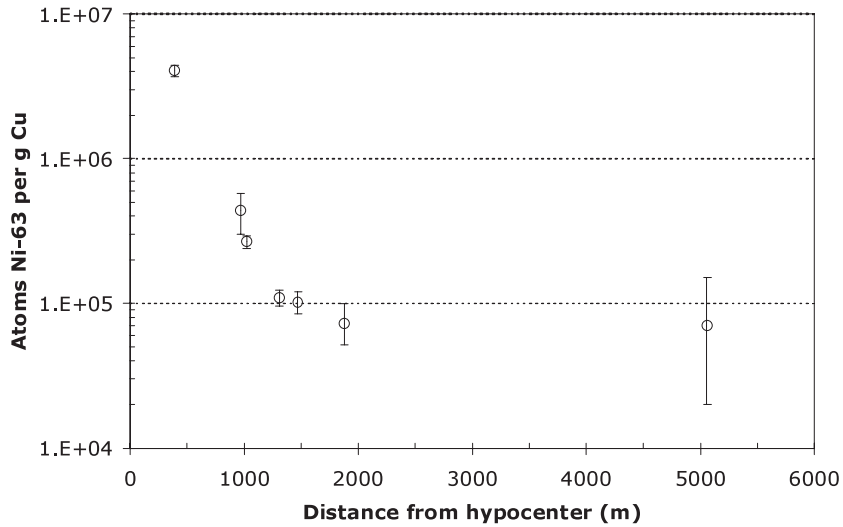

Figure 1. *Means and standard deviations of measured ^{63}Ni in line-of-sight copper samples from Hiroshima. Atoms ^{63}Ni/g Cu are at time of measurement without background subtraction or correction for decay since 1945.*

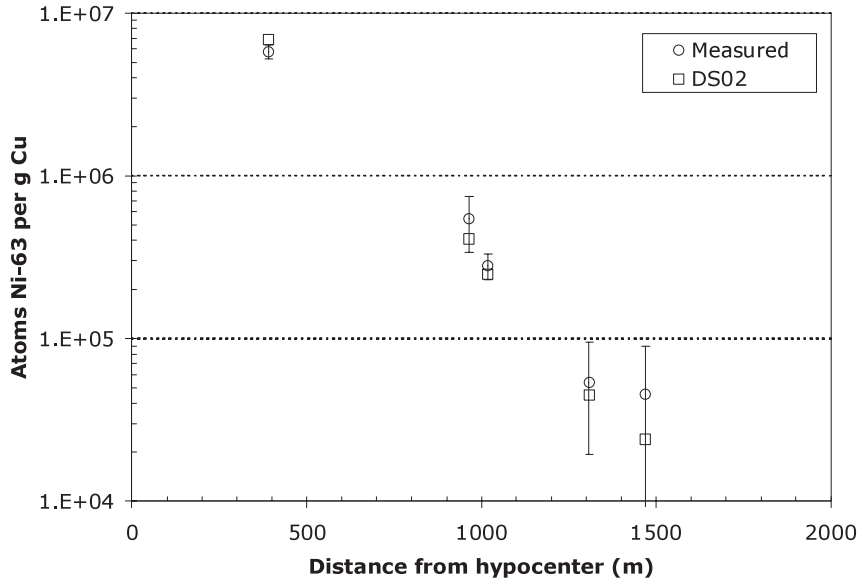

Figure 2. *The open circles are the measurement means after subtracting a background of 7.3×10^4 atoms ^{63}Ni/g Cu and correcting for decay since August 6, 1945. A comparison is made with DS02 sample-specific calculations (open squares).*

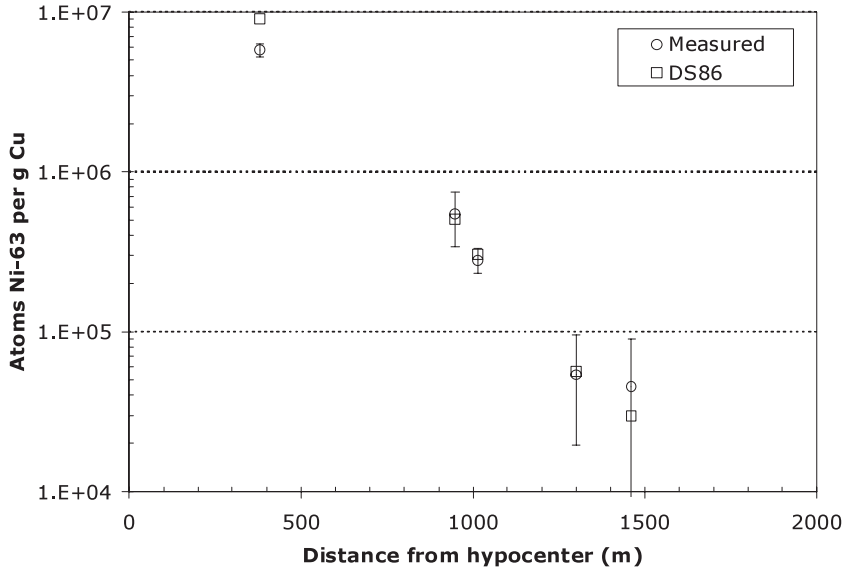

Figure 3. *The open circles are the measurement data after subtracting a background of 7.3 × 10^4 atoms ^63Ni/g Cu and correcting for decay since August 6, 1945. A comparison is made with DS86 sample-specific calculations (open squares).*

As seen in Table 6, the Bank of Japan copper sample had about 200 μg Ni/g Cu. Based on sample-specific calculations (Table 5), the high concentration of nickel at this close distance results in the thermal neutrons producing 26% of the total ^{63}Ni via the ^{62}Ni(n,γ)^{63}Ni reaction. Although this reaction was included in the DS02 calculations, we believe it does have somewhat more uncertainty than the samples, where only the fast neutron component is significant. The fast neutron fluence near the hypocenter may be validated more accurately by the measurement of ^{63}Ni in Cu samples collected from the Hiroshima A-bomb Dome at about 160-m ground range. Such samples have been collected, shown to have relatively low Ni content, and are now in the process of being measured using AMS.

The apparent constant "background" beyond about 1,800 m is not yet fully understood. It could potentially originate from a combination of sources, including sample processing, the AMS machine, and in-situ production of ^{63}Ni due to cosmic rays. It is significant to note that the contribution from cosmic rays to the ^{63}Ni in the copper samples is estimated to be only about 1×10^4 atoms of ^{63}Ni/g Cu at saturation (Chapter 9, Part D). This is substantially lower than the atoms of ^{63}Ni/g Cu actually measured in our background samples, which should not be at saturation based on our estimates of the duration the samples have been exposed to cosmic rays. This difference is currently being evaluated.

Completed measurements of copper samples obtained from large distances in Hiroshima, i.e., at distances where the bomb neutrons are insignificant, are listed in Table 8. Each of these background results are based on at least two independent measurements (i.e., separate processing of sample aliquots and AMS runs) and therefore meet our criterion for a reliable measurement.

The measurements indicate a background in the 7×10^4 ^{63}Ni/g Cu range. Additional measurements of background samples are underway (Table 2) and should strengthen the background data and perhaps also provide a better understanding of the sources of ^{63}Ni observed in the measurements. Our preliminary results of the large distance copper samples from Nagasaki appear to be consistent with the background observed for Hiroshima. That is, single measurements have been made of copper from the Minori-en (3,931 m), the Nagasaki Branch of the Hong Kong-Shanghai Bank (4,187 m), and the Oura-Tensvudou Church (4,428 m). The weighted mean ± SD of these preliminary measurements is 1.2 (+0.4, -0.6) $\times 10^5$ atoms ^{63}Ni/g Cu, which does not differ significantly from the background measured in Hiroshima copper samples.

Single measurements have also been made of three Cu rain gutters from the Hiroshima University "E-Building." Two of the Cu samples were in line-of-sight with the bomb (1,385 and 1,388 m ground range) and a third sample (1,469 m ground range) was from the shielded side of the building. The rain gutters appeared identical and presumably made of the same or similar copper. The measured mean for the two line-of-sight samples was 8.3 (+1.9, -1.5) $\times 10^4$. The result for the shielded sample was 4.1 (+2.2, -1.7) $\times 10^4$. If we assume that the shielded sample represents the background for these rain gutters plus a small contribution from the bomb neutrons (Table 5), then the measurements would translate into a bomb-induced ^{63}Ni concentration of 5.6 (+3.2, -2.5) $\times 10^4$ atoms/g Cu on August 6, 1945. Although preliminary, these results are consistent with our ^{63}Ni measurements made at similar distances in Hiroshima. Also, if we compare these preliminary measurements with the sample-specific DS02 calculations in Table 5, the estimated M/C ratio would be 1.5 (+0.9, -0.7), which does not differ significantly from unity.
We have measured other background copper samples as well. A modern copper wire from a hardware store in Livermore, Ca. was measured, and copper from a church roof in Bavaria, Germany was also measured (Rugel et al. 2004). The copper wire obtained from the hardware store is most likely from recently produced copper, while the church roof is known to have been exposed to cosmic rays directly for more than 80 years. These data are listed in Table 9. The results suggest that ^{63}Ni/g Cu in the old church roof may be higher than the modern hardware store wire. However, due to the inter-sample variations observed in the limited number of

Table 8. Results of completed background ^{63}Ni measurements in copper samples from Hiroshima[a]

Sample location & description	Distance from hypocenter (m)	Ni in Cu (μg/g)	^{63}Ni/g Cu	-SD	+SD
Higashi-Matsubara Branch, Sumitomo Bank (rain gutter)	1880	21.3 ± 3.9	7.3×10^4	2.1×10^4	2.6×10^4
Kusatsu-Hachiman Shrine (copper roof)	5062	129.8 ± 10.0	7×10^4	5×10^4	8×10^4
Kamesaki Shrine (ornament)	7500	251.9 ± 19.6	$<9.3 \times 10^4$		

[a]The weighted average of the results for Sumitomo Bank and Kusatsu Hachiman Shrine was used for background subtraction in this report. The Kamesaki Shrine sample had high stable Ni content and could only provide an upper limit estimate. It was therefore not included in the weighted average for background subtraction. It should be noted that these buildings were constructed about the same time as those from which the line-of-sight samples were obtained and thus should have similar cosmic-ray exposure histories (see Tables 1 and 2).

Table 9. Results of background ^{63}Ni measurements in other copper samples[a]

Sample location & description	Sample ID	Height above sea level (m)	Cosmic-ray exposure duration (y)	Ni in Cu (μg/g)	^{63}Ni/g Cu	-SD	+SD
Copper wire from hardware store (Livermore, Ca.)	H579	75	<5[b]	5 ± 1	2.4×10^4	1.7×10^4	4.2×10^4
Copper wire from hardware store (Livermore, Ca.)	H670	75	<5	1.2 ± 0.2	$<3 \times 10^4$		
Copper wire from hardware store (Livermore, Ca.)	H820	75	<5	1 ± 0.2	3.3×10^4	2.3×10^4	4.1×10^4
Thannhausen church roof (Germany)	H674	550	127[c]	44.8 ± 2.0	$<16 \times 10^4$		
Thannhausen church roof (Germany)	H771	550	127	41.0 ± 3.3	14×10^4	6×10^4	7×10^4
Thannhausen church roof (Germany)	H772	550	127	41.0 ± 3.3	12×10^4	4×10^4	4×10^4

[a]The mean and SD for the hardware store wire is $(2.4 \pm 1.5) \times 10^4$ atoms ^{63}Ni/g Cu and for the Thannhausen church roof is $(10 \pm 3) \times 10^4$ atoms ^{63}Ni/g Cu.
[b]We assume this wire has been exposed to cosmic-ray neutrons for less than 5 years.
[c]During the first 80 years the sample was on the surface of the church spire, afterwards it was stored in the workshop of a blacksmith.

background samples measured to date, we cannot conclude that there is a detectable correlation with cosmic-ray exposure duration.

Finally, the magnitude and variations observed in the background have little effect on the results within 1,100 m from the hypocenter, but can substantially affect our bomb-induced ^{63}Ni/g Cu estimates beyond about 1,300 m.

Conclusions

The AMS-measurements of ^{63}Ni in copper samples obtained from different distances in Hiroshima represent the first detection of fast neutrons after more than 50 years, and the first reliable bomb-induced fast neutron measurements beyond 700 m from the hypocenter.

A major significance of these ^{63}Ni results is that they provide, for the first time, fast neutron measurements at the distances most relevant to atomic-bomb survivor locations (900-1,500 m).

The measurement of ^{63}Ni in copper samples using AMS provides a substantial improvement in fast neutron detection compared with the ^{32}P measurements made in 1945. For example, the measurement-to-background ratio at about 400 m from the hypocenter is about 55 for ^{63}Ni and only about 2 for the ^{32}P measurements. Also, the ^{63}Ni measurements reach background levels at about 1,800 m from the hypocenter compared with only about 700 m for ^{32}P.

At distances of ~1,800 m from the hypocenter, out to at least 5,000 meters, the measurements appear to level off at a value on the order of 7×10^4 atoms of ^{63}Ni/g of Cu, suggesting a background of approximately that magnitude.

When this background is subtracted and the resulting data corrected to 1945, the measured ^{63}Ni in copper samples from Hiroshima are in good agreement with DS02 sample-specific calculations. Comparisons with DS86 calculations also show good agreement except at the Bank of Japan. The difference at that distance is significant when compared with DS86 calculations.

At present, calculations of cosmic-ray induced ^{63}Ni in copper (Chapter 9, Part D) have not accounted for the higher background measured in Cu samples. It is possible that the ^{63}Ni background measured in copper samples may come primarily from sources such as the sample chemistry, the sample holders, and the AMS machine. This should be investigated further.

Finally, it is unfortunate that bomb-induced ^{63}Ni could not have been measured in Nagasaki. Three line-of-sight samples obtained a few hundred meters from the Nagasaki hypocenter were made available to us for AMS analysis. However, we determined that they were composed of an alloy with ~4,500 ppm Ni, which is much too high for useful fast neutron information from ^{63}Ni. If suitable line-of-sight Cu samples could be made available from Nagasaki, fast neutron measurements would be possible using the approach described here.

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