Chapter 8

ACTIVATION MEASUREMENTS FOR THERMAL NEUTRONS

Part E. ^{36}Cl Measurements in Germany

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Introduction

The long-lived radioisotope ^{36}Cl (half-life: 301,000 years) was measured in bomb-exposed granite and concrete samples from Hiroshima, and in granite samples not exposed to A-bomb neutrons, by means of accelerator mass spectrometry (AMS). For those samples exposed to A-bomb neutrons, measured ^{36}Cl/Cl ratios as a function of distance to the epicenter were compared with calculated ratios based on the new dosimetry system DS02. Measured and calculated values agree within the experimental uncertainties even at large ground ranges when ^{36}Cl production from both A-bomb neutrons and natural radiation was taken into account (see Chapter 8, Part G). Thus, the so-called Hiroshima discrepancy reported in the literature, i.e. higher measured thermal neutron fluences than predicted by DS86 at larger ground ranges, was not confirmed. Close to the hypocenter measured ratios are somewhat lower than those based on DS02 calculations.

Overview of AMS Measurements at Munich

In the late 1970s and early 1980s, AMS measurement techniques were developed for the detection of long-lived radioisotopes such as ^{10}Be, ^{14}C, and ^{36}Cl (Nelson et al. 1977, Bennett et al. 1977, Raisbeck et al. 1978, Elmore et al. 1979). Short-lived radioisotopes can be easily detected by techniques in which the radiation such as γ-rays after β decay, α-particles or β-particles is counted directly. For long-lived radioisotopes, however, decay counting is difficult due to the very low count rates involved. AMS with its capability to identify those nuclides by mass spectrometry prior to their rare decays offers a solution to this problem.

At the Munich tandem laboratory, an AMS set-up had been installed by end of 1983, which

permitted the measurement of the long-lived radioisotope ^{36}Cl in environmental samples (Kubik et al. 1983, Kubik et al. 1984). It immediately became clear that AMS would also be a versatile tool for the detection of radioisotopes produced by neutrons originating from the A-bombs at Hiroshima and Nagasaki. The need for a reassessment of neutron fluences was evident for two reasons: first, the relatively short half-lives of those radioisotopes that had been used before for this purpose made it more and more difficult to perform additional comtemporary measurements. Second, results of ^{60}Co measurements which had been performed in reinforcing steel bars from concrete buildings in Hiroshima were in apparent disagreement with DS86 neutron fluence calculations (Loewe et al. 1987).

The basic motivation for the Munich efforts was to demonstrate ^{36}Cl to be a fluence monitor for neutrons originating from the A-bomb, and a gravestone was investigated from the Saikoji graveyard at a ground range of 94 m. Initially it was expected that ^{36}Cl could have been produced dominantly in potassium-rich compounds of granite via the fast-neutron reaction ^{39}K(n,α)^{36}Cl. It turned out, however, that the stable chlorine content in granite was not low enough, so that most of the detected ^{36}Cl was produced via the thermal-neutron reaction ^{35}Cl(n,γ)^{36}Cl (Haberstock et al. 1986, Kato et al. 1990). Ratios of stable H/D and ^{18}O/^{16}O were also measured, and concerns that penetrating rain water could have influenced the ^{36}Cl/Cl ratio in granite were not confirmed (Blamart et al. 1992).

Recently, we extended our approach to detect ^{36}Cl in granite to ground ranges where victims could survive while still receiving considerable radiation exposure. A similar approach had already been persued before by Straume and coworkers who detected ^{36}Cl in concrete samples from Hiroshima and Nagasaki at those distances (Straume et al. 1992, 1994). In our study, we concentrated on a ground range of about 1,300 m where deviations between ^{36}Cl, ^{152}Eu, and ^{60}Co measurements on the one hand, and DS86 calculations on the other hand had been reported. When first results of our study were evaluated and did not show a clear discrepancy to DS86 calculations (Huber 1999; Rühm et al. 2000; NAS 2001), it was decided to organize an intercomparison study between different laboratories involved in the ^{36}Cl and ^{152}Eu measurements, which included granite samples exposed to A-bomb neutrons from Hiroshima, and a couple of granite samples which were not exposed to neutrons from the A-bomb.

Here we report on the ^{36}Cl results obtained by means of AMS at the Munich accelerator laboratory. In our study we focused on granite samples, since environmental processes such as weathering are not expected to influence the bomb-induced ^{36}Cl/Cl ratios (Blamart et al. 1992). We also collected a few concrete samples for comparison, since ^{36}Cl levels were reported to be much higher than predicted by DS86 in distant concrete samples at Hiroshima (Straume et al. 1992). The production of ^{36}Cl in the lithosphere by cosmic rays and by neutrons from uranium and thorium decay, sometimes called “natural *in situ* production,” was also studied (see Chapter 8, Part G) and compared with that due to A-bomb neutrons.

Materials and Methods

Location of Samples Exposed to A-Bomb Neutrons

The investigated samples were predominantly granite. The samples were from gravestones and buildings such as the Fukoku building, the “E-building” at Hiroshima University, the Red Cross Hospital, and the Postal Savings Office (PSO). Up to four gravestones per graveyard were

collected to investigate local variations in the bomb-induced $^{36}Cl/Cl$ ratios. The investigated samples also include those that were part of an intercomparison study (see Chapter 8, Part H). These samples were provided by Dr. Hoshi, Hiroshima University.

Special emphasis was placed on the E-building, since it is located at a critical ground range where a large discrepancy between thermal neutron activation measurements and DS86 neutron fluences had been reported in the past. Four samples directly exposed to A-bomb neutrons were taken from granite ditch covers near the north corner of the building (samples d1 - d4 in Figure 1). A fifth sample that was not line-of-sight to the epicenter was also collected from behind the building and used as control (sample d5 in Figure 1). In addition, six line-of-sight cores were taken from the north facade of the building (samples c1 - c6 in Figure 1). Cores 1 to 3 were taken at a height of about 1.2 m above ground and consisted (from outside to inside) of granite (about 10 cm in thickness) followed by mortar (6 cm) and concrete (about 20 cm). Cores 4 to 6 were taken at heights of 5.3 m and 8.04 m, respectively. They consisted (from outside to inside) of tiles (2 cm in thickness), mortar (about 4 cm), and concrete (about 27 cm).

Two concrete samples from the Red Cross Hospital were provided by Dr. Straume, University of Utah. These samples were taken at a height of 20 m, at depths of 5.2 cm and 13.9 cm from the surface of the facade. The samples were also measured at the Lawrence Livermore National Laboratory (LLNL) and at the University of Purdue (see below). Finally, the sample from the Postal Savings Office was taken from a granite stair that was in the line-of-sight to the epicenter. This sample was provided by Dr. Fujita, RERF.

Ground ranges of the investigated samples cover a range from 94 m to 1,591 m. Sample locations, sample types, material and ground ranges are summarized in Table 1. The locations of the investigated samples relative to the hypocenter are shown in Figure 2.

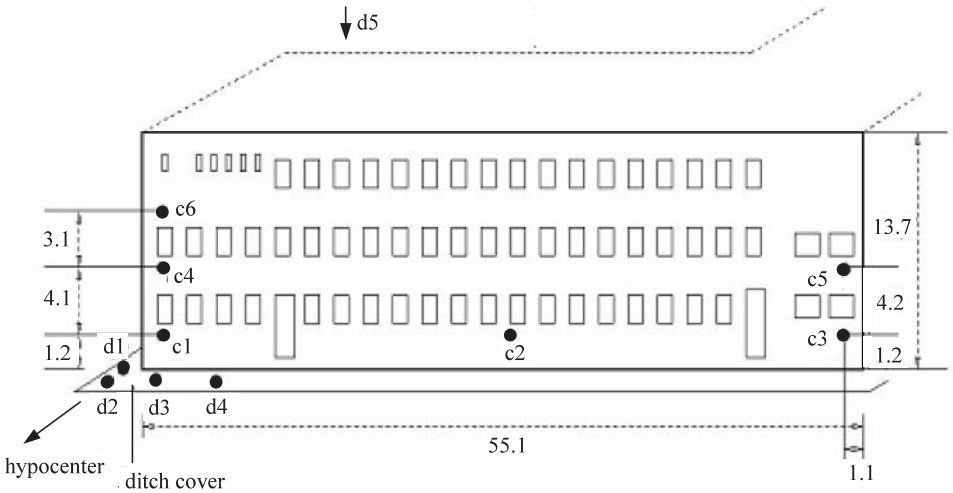

Figure 1. *Location of samples from the building of the Hiroshima Faculty of Sciences ("E-building").*

Table 1. Sample specification, and $^{36}Cl/Cl$ ratios measured in samples exposed to A-bomb neutrons; given ground ranges are based on the new DS02 hypocenter location

Sample	Sample type	Ground range [m]	Material	Height above ground [m]	Depth from the surface [cm]	$^{36}Cl/Cl$ [10^{-13}]
Saikoji 1	Gravestone	94 ± 35	Granite	1.5	9.4 – 11.4	1570 ± 70
Saikoji 2			Granite	1.3	0 – 21	1300 ± 70
Motoyasu Bridge[c]	Railing	134 ± 18	Granite	1	Surface	1670 ± 118
Fukoku 1	Wall	328 ± 20	Mica	6.8	0 – 1	633 ± 64
Fukoku 2			Granite		2.2 – 4.5	266 ± 16
Fukoku 3			Concrete		18	322 ± 36
Shirakami Shrine[c]	Fence	504 ± 18	Granite	1	Surface	188 ± 15
Myochoji[c]	Gravestone	639 ± 29	Granite	1	Surface	114 ± 10
Shinkoji 1	Gravestone	818 ± 39	Granite	1	Surface	19.9 + 3.9 – 3.6
Shinkoji 2	Gravestone		Granite	1	Surface	20.4 + 2.9 – 2.8
Shinkoji 3	Gravestone		Granite	1	Surface	21.5 ± 2.4
Shinkoji 4	Gravestone		Granite	1	Surface	21.2 + 4.2 – 4.1
Old Prefectural Office[c]	Pillar	877 ± 64	Granite	1	Surface	22.1 ± 2.1
Honkeiji[c]	Gravestone	896 ± 29	Granite	1	Surface	17.7 ± 2.0
Enryuji[c]	Stone lantern	925 ± 29	Granite	1	Surface	16.8 ± 2.1
Shingyoji[c]	Gravestone	915 ± 18	Granite	1	Surface	9.9 ± 1.1
Ganjyoji 1	Gravestone	992 ± 44	Granite	1	Surface	5.5 ± 0.6
Ganjyoji 2	Gravestone		Granite	1	Surface	3.2 ± 0.3
City Office pavement[c]	Pavement	1022 ± 18	Granite	0	Surface	3.3 ± 0.4
Tokueiji	Gravestone	1173 ± 34	Granite	1	Surface	2.6 ± 0.4
Kozenji[c]	Stone lantern	1177 ± 29	Granite	1	Surface	2.2 ± 0.4
Hosenji 1	Gravestone	1156 ± 50	Granite	1	Surface	2.8 + 0.7 – 0.6
Hosenji 2	Gravestone		Granite	1	Surface	2.3 ± 0.5
Hosenji 3	Gravestone		Granite	1	Surface	2.2 + 0.7 – 0.6
Jyunkyoji	Gravestone	1235 ± 47	Granite	1	Surface	2.3 ± 0.4

Table 1. Continued

Sample	Sample type	Ground range [m]	Material	Height above ground [m]	Depth from the surface [cm]	$^{36}Cl/Cl$ [10^{-13}]
E-Building, d1	Ditch cover	1384 ± 25	Granite	0	0 – 2	2.1 + 0.5 – 0.4
E-Building, d2	Ditch cover	1383 ± 25	Granite	0	0 – 2	1.4 ± 0.2
E-Building, d3	Ditch cover	1384 ± 25	Granite	0	0 – 2	1.1 ± 0.3
E-Building, d4	Ditch cover	1385 ± 25	Granite	0	0 – 2	0.9 ± 0.2
E-Building, d5	Ditch cover	1398 ± 25	Granite	0	0 – 2	0.9 ± 0.3
E-Building, c1	Wall	1384 ± 25	Granite	1.2	1 – 3	1.7 ± 0.4
			Granite		6 – 8	1.2 + 0.3 – 0.2
			Granite		8 – 10	2.0 ± 0.3
			Concrete		24 – 26	1.0 + 0.4 – 0.3
			Concrete		33 – 35	0.7 ± 0.2
E-Building, c2	Wall	1391 ± 25	Granite	1.2	1 – 3	1.2 ± 0.2
E-Building, c3	Wall	1398 ± 25	Granite	1.2	1 – 3	1.0 ± 0.2
E-Building, c4	Wall	1384 ± 25	Concrete	5.3	6 – 8	1.1 ± 0.2
E-Building, c5	Wall	1398 ± 25	Concrete	5.3	6 – 8	0.8 ± 0.2
E-Building, c6	Wall	1384 ± 25	Concrete	8.4	6 – 8	0.6 + 0.2 – 0.1
			Concrete		24 – 26	0.5 ± 0.2
			Concrete		31 – 33	0.9 + 0.3 – 0.2
Red Cross Hospital 1[a]	Wall	1501	Concrete	20	5.2	1.5 + 0.7 – 0.5
Red Cross Hospital 2[a]	Wall	1501	Concrete	20	13.9	1.0 + 0.6 – 0.5
Postal Savings Office[b]	Stair	1591 ± 32	Granite	0	Surface	1.2 ± 0.3
Senngyoji[c]	Gravestone	–	Granite	1	Surface	2.6 + 0.8 – 0.6

[a]Samples from the Red Cross Hospital were provided by Dr. Straume, University of Utah, USA.
[b]Samples from the Postal Savings Office were provided by Dr. Fujita, RERF, Japan.
[c]Samples which are part of the intercomparison excercise (see Chapter 8, Part H); these samples were provided by Dr. Hoshi, Hiroshima University, Japan; uncertainties for the given ground ranges were estimated from Shizuma et al. 1993, for these samples.

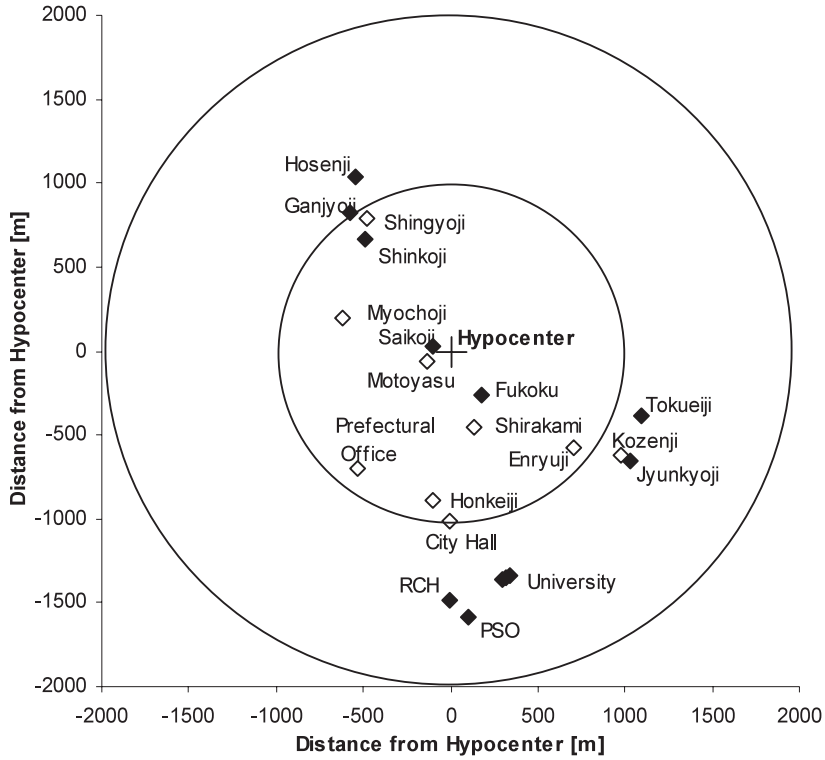

Figure 2. *Location of samples analyzed by the Munich group for the $^{36}Cl/Cl$ ratio. The open symbols are samples that were part of the intercomparison study. The Red Cross Hospital and Postal Savings Office are designated using RCH and PSO, respectively.*

Chemical Processing

Typical sample masses used in the study were about 100 g. The samples were crushed and milled. In some cases, the granite or concrete powder was washed with deionized water to remove any meteoric chloride that might have entered the material after the ^{36}Cl had been formed by A-bomb neutrons. The powder was put into a flask and mixed with diluted nitric acid. No stable chloride carrier was added. The flask was heated and the extracted chlorine distilled into a wash bottle filled with a silver nitrate solution. Silver chloride (AgCl) precipitated in the wash bottle and was separated from the silver nitrate solution by centrifugation. The separated AgCl was dried in an oven at 70° C and finally put into a sample holder.

AMS Measurement

The AgCl samples were put into a high current cesium sputter source to produce negative Cl^- ions. The ions were accelerated by a HVEC MP Tandem (terminal voltage about 10 MV) to an energy of 2.5 MeV per nucleon and the Cl^{8+} ions produced in the terminal foil (probability 30%)

were selected by the 90° analyzing magnet. In a second stripper foil, the Cl^{8+} ions were stripped to charge state 14^+ (probability 33%) and accelerated by a RF Interdigital H-type structure to an energy of 4.3 MeV per nucleon. In a third foil, they were finally stripped completely (probability 7%), analyzed for the charge state 17^+ and detected in a Bragg-type ionization chamber. The overall transmission from the entrance of the Tandem accelerator to the detector was about 1×10^{-3}. With $^{37}Cl^-$ currents of about 2 μA at the entrance of the Tandem accelerator, the detection limit defined as one event per hour in the detector corresponds to a $^{36}Cl/Cl$ ratio of approximately 5×10^{-15}. No interfering events from other isotopes were observed. Cross talk in the ion source was $<10^{-3}$. Details of the set-up are given in Kubik et al. (1983) and Haberstock et al. (1986).

Quality Assurance

To quantify any potential cross contamination during sample preparation and in the ion source blank samples made of NaCl were also prepared, before and after the preparation of a set of granite samples. Before and after a granite sample was measured by means of AMS, a standard with known $^{36}Cl/Cl$ ratio was also measured for normalization. Every granite sample was measured several times during the beam time. A couple of months later, these measurements were repeated independently, and a weighted mean for the $^{36}Cl/Cl$ calculated.

Determination of Natural In Situ Production of ^{36}Cl

In the lithosphere, ^{36}Cl is produced by different mechanisms, such as reactions induced by the nucleonic and muonic components of cosmic rays or by neutrons originating from the decay of uranium and thorium. Factors affecting the natural production of ^{36}Cl include the chemical composition of a sample, its original depth in the lithosphere, and the local erosion rate. Accordingly, the natural $^{36}Cl/Cl$ ratio is expected to be different for every individual mineral sample.

In Chapter 8, Part G, we present a method for estimating $^{36}Cl/Cl$ ratios in granite, based on the chemical composition and assumptions on sample depths and local erosion rates. We have applied this method to granite samples from Japanese quarries that were not exposed to any A-bomb neutrons. Reasonable agreement was found between the calculated $^{36}Cl/Cl$ ratios and those measured in the samples by means of AMS. With this method, we also calculated natural *in situ* $^{36}Cl/Cl$ ratios for the exposed granite samples and subtracted the result from the measured $^{36}Cl/Cl$ ratios, to obtain net $^{36}Cl/Cl$ ratios for A-bomb neutrons alone.

Results and Discussion

Discussion of Uncorrected $^{36}Cl/Cl$ Ratios

The $^{36}Cl/Cl$ ratios measured in granite and concrete samples exposed to A-bomb neutrons are given in Table 1. Measured ratios are in the range from 5×10^{-14} to 1.6×10^{-10}.

Saikoji Gravestone. In the past, a ^{36}Cl profile was measured in a gravestone from the Saikoji graveyard, along the central vertical axis of the upper granite block. A slight maximum in the $^{36}Cl/Cl$ ratios was found at a depth of about 10 cm (see Figure 3).

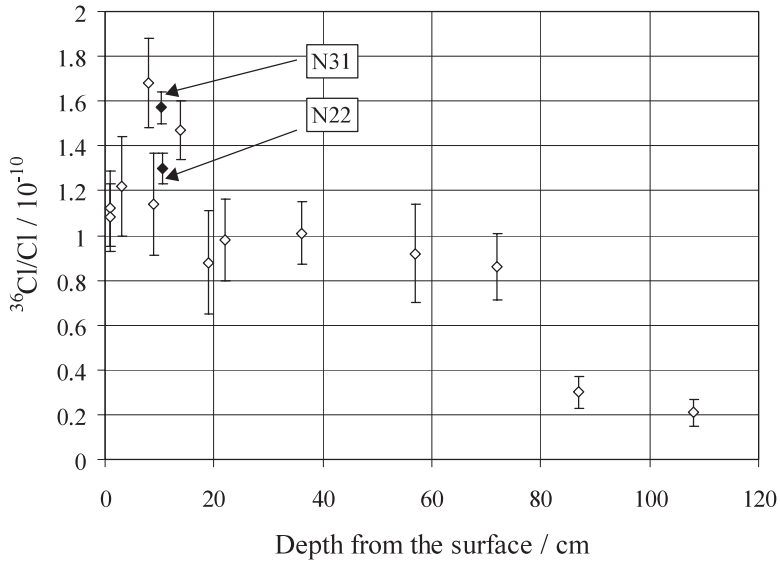

Figure 3. *Depth profile of ^{36}Cl/Cl along central vertical axis of gravestone from Saikoji graveyard. Open symbols refer to data from Haberstock et al. (1986) and Kato et al. (1990), while black symbols refer to data from this work. Sample N31 was taken from an almost central vertical core at a depth of 9.4 to 11.4 cm, while sample N22 was from a 21-cm long horizontal core 29 cm below the top surface.*

In the present work two samples were measured: one was from an almost central vertical core from a depth from 9.4 to 11.4 cm (sample N31), another was from a 21-cm long horizontal core taken 29 cm below the top surface (sample N22). The result obtained here for sample N31 (see Figure 3) agrees with that found by Haberstock and co-workers, but shows much smaller uncertainties. The smaller uncertainties in the current study arise from the chemical procedure that did not—in contrast to the procedure used by Haberstock and co-workers—require addition of a stable chloride carrier. The result for sample N22 (see Figure 3) is slightly lower, since it was obtained from a core that was processed as a whole, and represents, therefore, a mean value for the first 21 cm from the top surface of the block.

Fukoku Building. The ^{36}Cl/Cl ratios obtained for the samples from the Fukoku Building can be compared to results already obtained for other radioisotopes in samples from this building. It turns out that effective thermal neutron fluences deduced from the ^{36}Cl/Cl ratios of the surface samples (Table 1) agree within the uncertainties with those deduced from ^{152}Eu (Kato 2002; Shizuma et al. 1993) for other granite samples from the facade. Other results reported of ^{152}Eu (Nakanishi et al. 1987) and of ^{60}Co (Kato 2002), however, are about a factor 2 to 3 higher. The reason for the different results is not yet understood.

Granite Samples from Intermediate Distances. Results obtained from different gravestones located within the same graveyard show remarkably consistent results (see, for example, those

for the 4 gravestones from the Shinkoji graveyard in Table 1). This suggests that detailed knowledge on shielding and location of the investigated gravestones within the graveyard is not necessary. Results obtained on gravestones from similar ground distances, but different locations, also do not differ greatly. For example, ^{36}Cl/Cl ratios measured for the Hosenji gravestones (ground range of 1,156 ± 50 m to the north of the hypocenter) and those for the gravestones from Tokueiji, Kozenji, and Jyunkyoji (ground ranges of 1,173 ± 34 m, 1,177 ± 29 m, 1,235 ± 47 m, respectively, to the south east of the hypocenter) are very similar. The ^{36}Cl/Cl ratios obtained on granite samples from similar but smaller ground ranges also agree within a factor 2 (Old Prefectural Office at a ground range of 877 ± 64 m to the southwest, Honkeiji at a ground range of 896 ± 29 m to the south, Enryuji at a ground range of 925 ± 29 m to the southeast, and Shingyoji at a ground range of 915 ± 18 m to the north). Thus, a potential asymmetry in thermal neutron activation, if there is any, is not very large (see also Shizuma et al. 1993).

E-Building. The ^{36}Cl/Cl ratios measured in the four line-of-sight ditch covers from the E-building (d1 - d4) are of the order of 1×10^{-13}, and the weighted average is $(1.2 \pm 0.1) \times 10^{-13}$. The result obtained for the shielded ditch cover (d5) is not significantly different from the results for the line-of-sight ditch covers. This is due to diffusive thermal-neutron propagation and to the fact that natural *in situ* production is dominant for these samples.

Results obtained from the facade of the E-building support the above observation. For example, the weighted average for the granite surface samples of cores 1 through 3 is $(1.1 \pm 0.1) \times 10^{-13}$, in perfect agreement to that for the line-of-sight ditch covers. The surface samples from the concrete cores (numbers 4 through 6) were covered by 6 cm of tiles and mortar and gave a weighted average of $(0.8 \pm 0.1) \times 10^{-13}$. This compares to a value of $(1.2 + 0.3 - 0.2) \times 10^{-13}$ obtained at a depth from 6 to 8 cm in the granite core 1. Thus, a significant difference between the ^{36}Cl/Cl ratios measured in concrete and granite samples from similar locations and depths was not found at large ground ranges. It should be noted that the results obtained for the near-surface samples are confirmed by the few measurements performed at greater depths (cores 1 and 6 in Table 1).

Red Cross Hospital and Postal Savings Office. Aliquots of the two samples from the Red Cross Hospital had been measured before at LLNL and at Purdue University. Our results agree within the uncertainties with those results [sample 1: $(2.29 \pm 0.21) \times 10^{-13}$ obtained at LLNL, and $(2.12 \pm 0.32) \times 10^{-13}$ obtained at Purdue; sample 2: $(1.38 \pm 0.18) \times 10^{-13}$ obtained at Purdue] (see Chapter 8, Part D). Thus, consistent results were obtained at several different AMS laboratories. Another systematic intercomparison between AMS laboratories at LLNL, Tsukuba, and Munich is described below (Chapter 8, Part H). The result obtained on the sample from the Postal Savings Office is also of the order of 10^{-13}.

Comparison with DS02 Free-in-Air Activation without Sample-Specific Calculation of ^{36}Cl In Situ Production

Figure 4 shows the experimental results obtained for near-surface samples in terms of $(^{36}\text{Cl/Cl})_{\text{exp}}$ ratios, as a function of slant range. For comparison, calculated $(^{36}\text{Cl/Cl})_{\text{calc}}$ ratios are also shown, which include the contribution from A-bomb neutrons, $(^{36}\text{Cl/Cl})_{\text{DS02}}$, and the contribution from cosmic radiation and from neutrons produced by U and Th decay, $(^{36}\text{Cl/Cl})_{\text{in situ}}$.

The $(^{36}Cl/Cl)_{DS02}$ was calculated by folding DS02 free-in-air thermal neutron fluences with the corresponding ENDF/B-VI (n,γ) production cross section, and ratios of $(^{36}Cl/Cl)_{in\ situ}$ equal to 0.6×10^{-13} and 1.0×10^{-13} measured in unexposed Giin-type granite samples were added as a typical background contribution by natural *in situ* production (see Chapter 8, Part G). Samples from the Giin quarry were used here for this purpose, since it is believed that this type of granite is representative of the granite from the E-building.

It is evident from Figure 4 that close to the hypocenter, measured $(^{36}Cl/Cl)_{exp}$ ratios are significantly lower than the calculated ratios. At large ground ranges, the experimental results are higher than those calculated, if only production due to A-bomb neutrons is included. If the upper and lower limits for $^{36}Cl/Cl$ ratios measured in the Giin quarry are added to the DS02 calculations (dotted lines in Figure 4), the agreement between calculation and measurement is excellent.

It should be noted, however, that the $^{36}Cl/Cl$ ratio expected from cosmic radiation and neutrons produced by U and Th decay generally depends on the chemical composition of the corresponding sample, its original depth in the lithosphere, and local erosion rate, and it is expected to be different for each sample investigated. For concrete, which is comprised of different components, the origin of the components must be known in order to perform

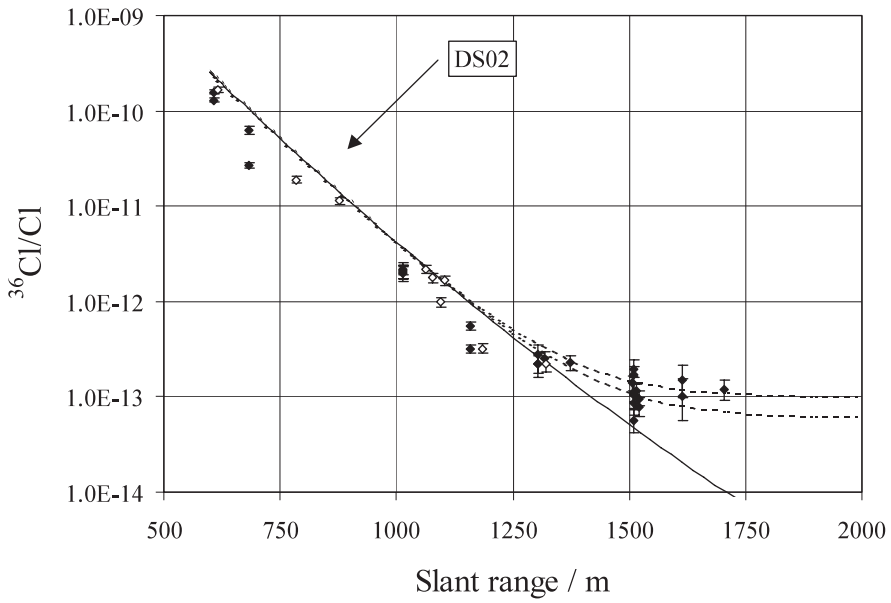

Figure 4. *Measured and calculated ($^{36}Cl/Cl$) ratios for near-surface granite and concrete samples. The open symbols are for samples that were part of the intercomparison exercise (see Chapter 8, Part G), the solid line is the DS02-based free-in-air (n,γ) calculation for a height of burst of 600 m and yield of 16 kt, and the dotted lines were obtained by adding the upper and lower limits for in situ production in measured samples from the Giin quarry (0.6×10^{-13} and 1.0×10^{-13}, respectively) (Chapter 8, Part G).*

calculations of the ^{36}Cl signal induced by natural *in situ* production. Therefore, the natural $(^{36}Cl/Cl)_{in\ situ}$ ratio from the Giin sample might not be representative of that for the other granite samples or the few measured concrete samples. A more detailed discussion of $(^{36}Cl/Cl)_{in\ situ}$ ratios from cosmic radiation and neutrons produced by U and Th decay is provided in Chapter 8, Part G. The results are shown in Table 2.

It can also be inferred from Figure 4 that for the production of ^{36}Cl in granite samples from Hiroshima, cosmic radiation and neutrons from U and Th decay become important at slant ranges beyond about 1,300 m.

Finally, it should be emphasized that most of the samples investigated were taken from environments rich in granite. As is pointed out elsewhere (e.g., Chapter 10, Part C), these environments tend to decrease thermal neutron fluence above ground, compared with DS02 free-in-air fluences and, accordingly, the calculated free-in-air activations shown in Figure 4 are believed to be somewhat too high for some of the investigated samples (see below).

Comparison with DS02 Free-in-Air Activation with Sample-Specific Calculation of ^{36}Cl In Situ Production

The *in situ* production of any radionuclide in the lithosphere depends on several parameters such as the chemical composition of the sample, the depth where the sample was initially located in the lithosphere, and local erosion rates. Therefore, those parameters must be known specifically for each of the investigated samples.

Most of the distant granite samples were analyzed for their elemental composition by the XRAL Company, and a local erosion rate of $\varepsilon = 100\ \mu m/a$ is assumed (see Chapter 8, Part G). Information on the probable depths of mined granite stones was obtained from detailed interviews of miners of granite and owners of granite quarries. These interviews indicated that granite taken from quarries on Kurahashi Island (Giin-type and Odachi-type) and on Oshima island (Iyo-type) were generally taken from 5-15 m depths, before the Second World War. Most of the exposed granite samples are expected to be made of these three granite types. The only exception is the gravestone from the Saikoji graveyard, which is made of granite from a quarry on Shikoku Island (Aji-type) and which had been taken from a probable depth of 10-15 m (Oda 2001; Nakano 2001; Tanimura 2001).

Table 2 summarizes calculated $(^{36}Cl/Cl)_{in\ situ}$ ratios based on the method described in Chapter 8, Part G for some of the granite samples exposed to A-bomb neutrons. Emphasis was placed on those samples that were located at slant ranges beyond about 1,000 m. It was assumed that the samples were taken from depths of 10 ± 5 m (see above), and the uncertainty in the depth was included as a systematic error.

In Figure 5, the differences between measured and calculated natural $^{36}Cl/Cl$ ratios, $(^{36}Cl/Cl)_{exp} - (^{36}Cl/Cl)_{in\ situ}$, are compared to DS02 free-in-air ratios $(^{36}Cl/Cl)_{DS02}$, as a function of slant range. The results obtained on samples closer than about 1,000 m were not corrected for *in situ* background, since for those samples the A-bomb induced ^{36}Cl signal is much larger than the signal induced by cosmic radiation and by neutrons from U and Th decay, and a background correction is not necessary. For the E-building, only one sample was analyzed for its elemental composition. This sample was assumed to be representative of all granite samples investigated from the E-building.

Table 2. Calculated $(^{36}Cl/Cl)_{in\ situ}$ ratios for natural *in situ* production in distant granite samples

Sample	$(^{36}Cl/Cl)_{in\ situ}$ [a]	$(^{36}Cl/Cl)_{exp} - (^{36}Cl/Cl)_{in\ situ}$ [b]
Ganjyoji 1	$(6.8 \pm 0.5) \times 10^{-14}$	$(4.9 \pm 0.6) \times 10^{-13}$
Ganjyoji 2	$(6.5 \pm 0.6) \times 10^{-14}$	$(2.6 \pm 0.3) \times 10^{-13}$
City Office pavement[c]	$(7.0 \pm 0.9) \times 10^{-14}$	$(2.5 \pm 0.4) \times 10^{-13}$
Tokueiji	$(9.1 \pm 0.4) \times 10^{-14}$	$(1.7 \pm 0.4) \times 10^{-13}$
Kozenji[a]	$(6.0 \pm 0.9) \times 10^{-14}$	$(1.6 \pm 0.4) \times 10^{-13}$
Hosenji 1	$(6.5 \pm 0.7) \times 10^{-14}$	$(2.2 + 0.7 - 0.6) \times 10^{-13}$
Hosenji 2	$(5.5 \pm 0.5) \times 10^{-14}$	$(1.7 \pm 0.5) \times 10^{-13}$
Hosenji 3	$(4.1 \pm 0.1) \times 10^{-14}$	$(1.8 + 0.7 - 0.6) \times 10^{-13}$
Jyunkyoji	$(6.7 \pm 0.6) \times 10^{-14}$	$(1.6 \pm 0.4) \times 10^{-13}$
E-building c1	$(11.5 \pm 0.5) \times 10^{-14}$	$(5.5 + 3.6 - 3.5) \times 10^{-14}$
E-building c2	$(11.5 \pm 0.5) \times 10^{-14}$	$(0.02 + 2.2 - 2.0) \times 10^{-14}$
E-building c3	$(11.5 \pm 0.5) \times 10^{-14}$	$(-1.9 + 1.9 - 1.8) \times 10^{-14}$
E-building d1	$(9.3 \pm 0.8) \times 10^{-14}$	$(1.0 + 0.5 - 0.3) \times 10^{-13}$
E-building d2	$(9.3 \pm 0.8) \times 10^{-14}$	$(4.7 \pm 2.4) \times 10^{-14}$
E-building d3	$(9.3 \pm 0.8) \times 10^{-14}$	$(1.3 + 3.4 - 3.0) \times 10^{-14}$
E-building d4	$(9.3 \pm 0.8) \times 10^{-14}$	$(-7.9 \pm 21.5) \times 10^{-15}$
Postal Savings Office	$(9.5 \pm 0.7) \times 10^{-14}$	$(2.3 \pm 3.0) \times 10^{-14}$

[a]Calculated by method described in Chapter 8, Part G.

[b]Measured ratio (exp) (Table 1) corrected for natural ratio (*in situ*).

[c]A chlorine concentration of 50 ppm was assumed for the samples from City Office pavement and Kozenji due to a lack of more specific data (see Chapter 8, Part G on natural *in situ* production).

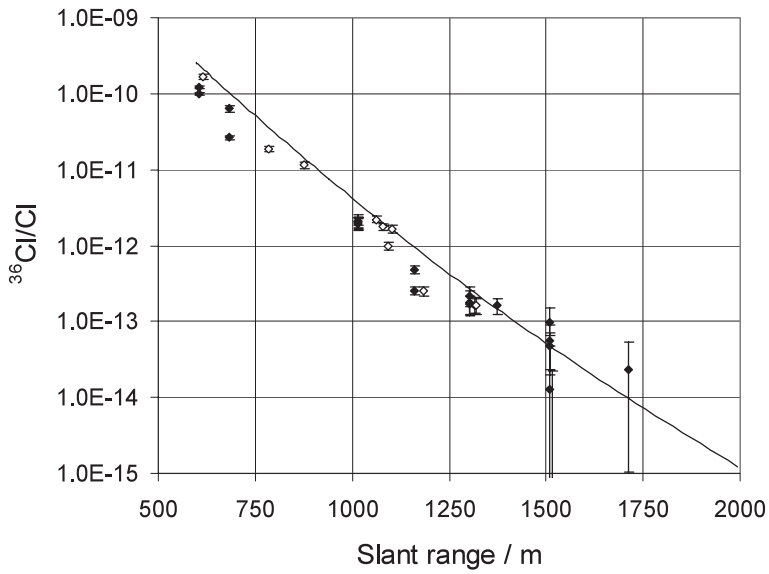

Figure 5. *Measured $^{36}Cl/Cl$ ratios for near-surface samples of granite after subtraction of calculated natural in situ ratios (see Table 2). The open symbols are data for intercomparison samples (see Chapter 8, Part H). The results obtained for the Saikoji gravestone at about 10 cm depth were divided by a factor 1.3 to account for the smaller values measured in the past near the surface (see Figure 3).*

Correction for Local Environments

For most of the samples, detailed DS02 calculations were performed by SAIC, and the resulting $^{36}Cl/Cl$ ratios are shown in Table 3 (see Chapter 8, Part J).

In Figure 6, the ratios for $((^{36}Cl/Cl)_{exp} - (^{36}Cl/Cl)_{in\ situ}) / (^{36}Cl/Cl)_{DS02}$ are plotted. It is seen by comparing Figures 5 and 6 that the experimental and calculated ratios at slant ranges beyond 800 m from the epicenter agree within the uncertainty of the experimental measurements after the subtraction of the *in situ* production. There is no evidence for a discrepancy between measurements and DS02 calculations at large slant ranges.

Concrete Samples

Determination of the natural *in situ* background present in the investigated concrete samples was not possible with the method presented in Chapter 8, Part G, because information on the origin of the concrete compounds was not available. Instead, a different approach was used. The weighted means of the 4 deep concrete samples (core c1: depth 24-26 cm and 33-35 cm; core c6: depth 24-26 cm and 31-33 cm; see Table 1) and of the 3 "surface" concrete samples (samples c4, c5 and c6 in Table 1; 6-8 cm depth) are $^{36}Cl/Cl_{mean,\ depth} = (7.3 \pm 1.1) \times 10^{-14}$ and $^{36}Cl/Cl_{mean,\ surface} = (7.6 \pm 1.1) \times 10^{-14}$, respectively. Both include contribution from A-bomb neutrons and from natural in situ production. DS02 calculations performed for core c6 suggest a ratio between deep and surface ^{36}Cl activation due to A-bomb neutrons of k = 0.3. If it is assumed that the m/c values obtained for the 4 deep samples and the 3 "surface" samples are identical, and that all 7 concrete samples are characterized by the same ^{36}Cl signal due to natural *in situ* production, the natural *in situ* background $^{36}Cl/Cl_{background}$ can be deduced from the following equation:

$$^{36}Cl/Cl_{background} = (^{36}Cl/Cl_{mean,\ depth} - k \times {}^{36}Cl/Cl_{mean,\ surface}) / (1-k).$$

The resulting M/C values are 0.1 ± 0.4 for the 3 surface samples, and 0.1 ± 1.3 for the 4 deeper lying samples. As already deduced for the granite samples, there is no evidence for a major discrepancy between measurements and DS02 calculations at large slant ranges.

Conclusions

$^{36}Cl/Cl$ ratios were determined in granite samples exposed to A-bomb neutrons in Hiroshima and in reference granite samples not exposed to A-bomb neutrons. For distant granite samples, *in situ* production of ^{36}Cl by cosmic rays and by neutrons originating from the decay of U and Th was also calculated (see Chapter 8, Part G). Within the experimental uncertainties, no significant discrepancy was found between measured $^{36}Cl/Cl$ ratios and those based on DS02 calculations at ground ranges greater than 800 m, if the natural concentration of ^{36}Cl in the granite samples was taken into account. For smaller ground ranges, evidence was found that experimental fluences derived from ^{36}Cl are lower than those based on DS02 calculations.

Table 3. Calculated ^{36}Cl/Cl ratios for granite samples based on DS02 methodology[a]

Sample	$(^{36}Cl/Cl)_{DS02}$	Sample	$(^{36}Cl/Cl)_{DS02}$
Saikoji	1.61×10^{-10}	Hosenji 2	2.27×10^{-13}
Motoyasu Bridge	1.81×10^{-10}	Hosenji 3	2.27×10^{-13}
Shirakami	3.02×10^{-11}	Jyunkyoji	1.25×10^{-13}
Myochoji	1.17×10^{-11}	E-building c1	4.47×10^{-14}
Shinkoji	3.06×10^{-12}	E-building c4	4.87×10^{-14}
Old Prefectural Office	1.95×10^{-12}	E-building c6	4.90×10^{-14}
Honkeiji	1.68×10^{-12}	E-building c2	4.28×10^{-14}
Enryuji	1.34×10^{-12}	E-building c3	4.02×10^{-14}
Shingyoji	1.45×10^{-12}	E-building c5	4.43×10^{-14}
Ganjyoji 1	8.00×10^{-13}	E-building d1	4.49×10^{-14}
Ganjyoji 2	8.00×10^{-13}	E-building d2	4.50×10^{-14}
City Office pavement	6.33×10^{-13}	E-building d3	4.55×10^{-14}
Tokueji	2.00×10^{-13}	E-building d4	4.47×10^{-14}
Kozenji	1.94×10^{-13}	Postal Sav. Of.	1.04×10^{-14}
Hosenji 1	2.27×10^{-13}		

[a]Calculations are for a depth of 1 cm from the surface and include the production from both the ^{35}Cl(n,γ) and ^{39}K(n,α) reactions. The K/Cl ratios are based on the elemental composition given in Chapter 8, Part G or on an average value of 300 for the intercomparison samples.

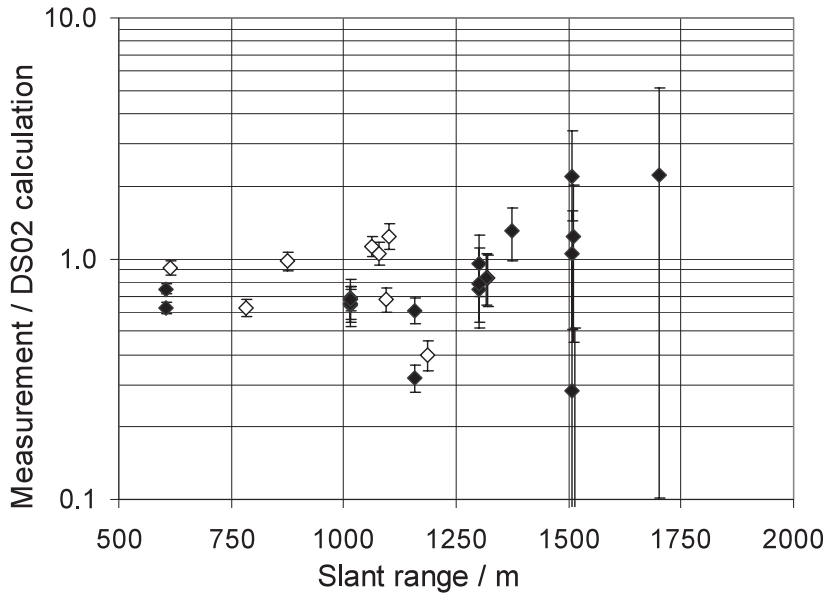

Figure 6. *Comparison of $((^{36}Cl/Cl)_{exp} - (^{36}Cl/Cl)_{in\ situ}) / (^{36}Cl/Cl)_{DS02}$ ratios as function of slant range. The results from the Fukoku building are not included due to the lack of specific DS02 calculation. The open symbols are for data from intercomparison samples (see Chapter 8, Part H).*

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