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Measurements of ^{60}Co in massive steel samples exposed to the Hiroshima atomic bomb explosion

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Executive Summary

Man's knowledge on the effect of ionising radiation is to a large extent based on follow-ups of Hiroshima victims. It is of utmost importance that the dose they received from neutrons from the A-bomb in 1945 be estimated as accurately as possible. Regular re-assessments of the dose received by Hiroshima victims have been performed ever since 1945. In the assessment from 1986 (Dosimetry System 1986, DS86) there were big discrepancies between model calculations and measurements of ^{60}Co induced in steel samples from distances >1000 m (long range) from the epicentre. The 5.2 y half-life of ^{60}Co means that the massic activity in steel samples from Hiroshima is extremely low nowadays. Therefore it is necessary to measure the ^{60}Co from steel samples in a semideep or deep underground laboratory and to use massive samples in order to reach low detection limits for the massic activity (measured in mBq/kg). This report gives the details of measurements of 14 steel samples from Hiroshima. The sample selection carried out by Hiroshima University was an essential and a very complex task, as it is important to know that the samples in question were present in 1945 and to understand what other structures could possibly have been shadowing the steel. The details of the sample selection process are, however, not dealt with in detail in this report.

The underground gamma-ray spectrometry measurements were carried out as a collaboration within three CELLAR (Collaboration of European Low-level underground LAboRatories) institutes. IRMM was coordinating the work and performing measurements in the underground laboratory HADES. Samples were also measured in the Gran Sasso underground laboratory in Italy and in the UDO laboratory in the ASSE mine, operated by PTB. This report does, however, not include the results from the three samples that were measured by PTB as PTB has been reorganising their low-level radioactivity work and assessing the future of their underground laboratory. As soon as these results are known, this report will be updated and re-printed.

The results of the measurements show that it is no longer possible to assess the activity induced by neutrons from the A-bomb at ground distances greater than about 1000 m (slant range ~ 1200 m) since environmental activation starts to dominate at that distance. The measurements were, however, successful in showing that measurements reported in DS86 on ^{60}Co in steel at long range show too high activity. The new measurements support the computer models of DS86 and DS02 (Dosimetry System 2002) and convey the message that the ^{60}Co dilemma from DS86 is solved. The measurements thus provide greater confidence in the existing dosimetry system.

Table of contents

List of Tables

1. Introduction

Knowledge of the effect of ionising radiations on humans and the resulting dosimetry regulations concerning exposure are mainly based on follow-up of the population exposed to the atomic bomb explosions above Hiroshima and Nagasaki. The Radiation Effects Research Foundation (RERF) is an organisation that started as a collaboration between USA and Japan. It conducts research into the medical effects of radiation on man. The RERF started in 1975 and took over the work started by the US organisation, Atomic Bomb Casualty Commission (ABCC), which was established in 1947. An important task for RERF is to retrospectively determine the effective doses at various locations in Hiroshima. This is a work that has been under continuous development and refinement ever since 1945. In recent years computer models for dose determination have been developed. The first dose re-assessment that was based on computer models was Dosimetry System 86 (DS86) from 1986. It is important that the model calculations are supported by measurements. One way of doing this is to measure the activity of certain radionuclides that were produced by the neutrons from the A-bomb. There were certain disagreements between model calculations and activity measurements after DS86 (Hoshi, 1998; Gold, 1999). Of particular importance for this study was the disagreement in activity of ^{60}Co found in steel samples. The most recent re-assessment of doses is the Dosimetry System 02 (DS02). Many disagreements from DS86 have been resolved but there are still a few that persist and amongst them ^{60}Co in steel. The values in the vicinity of ground zero at short range are in good agreement but at ground distances of 1000 m from ground zero and further, there are still discrepancies. It is important to clarify if these disagreements are due to problems in computer models or due to the technically very challenging measurements of extremely low-levels of ^{60}Co in steel. A previous series of measurements was performed by IRMM and PTB in 2003. The specific activities determined in samples collected at a ground distance shorter than 993 m (city hall) were in agreement with DS02. The small masses of the samples (between 8 and 160 g) permitted, however, only calculation of decision thresholds at longer distances. Those decision thresholds were lower than the specific activity values from earlier measurements. It was thus decided to perform a second series of measurements using larger samples in order to determine ^{60}Co specific activities in the order of 0.1 mBq/kg at the time of measurement and this is the scope of this report. It is outside the scope of this report to study the effect of neutron transport inside the samples albeit this being a vital part when dealing with massive samples like the ones in this study. The neutron transport inside the samples influences the activity calculations since an inhomogeneous activity distribution is expected. This report forms the basis for continued studies and calculations on the effect of inhomogeneous activity distribution and neutron transport in the samples. Furthermore, this report seeks to estimate the effect of the environmental neutrons and if the ^{60}Co activity induced by the A-bomb is still measurable at long distance or swamped by environmental activation.

2. Materials and Methods

The samples were collected at locations in Hiroshima that were carefully selected. The samples must have had a direct view of the A-bomb in 1945. The sampling process is described by Cullings at al. (2003) in the DS02 report. Sixteen "large mass" samples were found suitable and shipped to IRMM by boat in order to minimise the activation by cosmic rays. Fifteen samples were shipped in November 2004 and arrived January 4, 2005. A sixteenth sample (Hiroshima Bank Kanayama-cho branch iron ring on the rooftop) was shipped in May 2006 and arrived at IRMM August 4, 2006.
IRMM made sample preparations and shipped 4 samples further to two laboratories in the CELLAR (Collaboration of European Low-level underground LAboRatories) network. The majority of samples (12) were measured by IRMM using the underground laboratory HADES. One sample was measured by LNGS (Laboratori Nazionali del Gran Sasso) in their underground laboratory, 3 by PTB (Physikalisch-Technische Bundesanstalt) in the ASSE salt mine. However, as PTB was undergoing reorganisation and assessing the future of their underground laboratory, it has not yet been able to deliver the final quantitative results. When these arrive an updated version of this report will be issued.

In order to determine the stable cobalt content, a small piece (~ 1 g) was cut off from each sample for elemental analyses using 2 different methods, ICP-OES (Inductively Coupled Plasma Optical Emission Spectroscopy) and k_0-NAA (k_0-Neutron Activation Analysis),.

When calculating the specific activities it was an important issue to make correct background subtraction as ^{60}Co is present in the background, and the massive steel samples in this project shield (=protect/cover) to some extent the background radiation from the shield (= the lead+copper surrounding the detector). This is also discussed below.

2.1. Overview of samples

Table 1 summarises the data of each sample indicating the mass of the sample used for gamma-ray spectrometry. They are sorted by increasing slant range.

Table 1. List of Hiroshima samples described in this report.

	Sample	Short description	Ground distance (m)	Slant range (m)	Mass as arrived (g)
1	Sumitomo bank	Thick bent bar	260	654	386.48
2	Aioi bridge	5 slabs kept together by a spanner to form a thick piece	300	671	1769.03
3	Fukoku Life assurance	Thick rod bent to form a semi-circle	331	685	192.96
4	Chugoku electricity	Section of a ring with steps	687	912	613.31
5	Kodokan building	Short piece of pipe	720	937	120.78
6	City hall	Tube with enlarged part at one end - thread visible	993	1160	2321.54
7	City hall-2	Solid bar bent 90 deg.	993	1160	798.69
8	Hiroshima bank Kanayamacho	Thick rod bent to form semi-circle - resembles Fukoku sample	1210 or 1203	1351	190.27
9	Hiroshima bank Kanayamacho 6	Thick rod bent to form semi-circle	1222		233.41
10	Kyu-Fuzoku elementary school	Straight tube with white letters	1269	1404	634.44
11	Yokogawa bridge	Seemingly 4 pieces kept together by a spanner -traces of molten metal	1290	1423	2777.36
12	Hiroshima university plate	Thick square plate	1400	1523	523.65
13	Hiroshima university window	Three bars connected by a flat strip	1400	1523	704.69
14	Nisseki "5"	Tube	1480	1597	1127.56
15	Nisseki "1"	Tube slightly bent	1480	1597	806.98
16	Postal Saving Building (Chokin Kyoku)	Very thick rod in T-shape	1591	1700	1667.36

* Nisseki "5" and Nisseki "1" were measured together.
** Hiroshima University plate and window were measured together

2.1.1. Description and preparation of the samples

All samples were brushed with a steel brush and cleaned as described below under each subsection. Some samples were sawed in smaller pieces, which were sometimes flattened with a bench vise in order to obtain a more suitable geometry for measurement. As the procedures were slightly different for each sample, a more detailed description is given for each sample.

- ## 2.1.1.1 Sumitomo bank

This sample consisted of a small thick bent bar with a diameter of 24 mm and a length of 132 mm. The sample was partly covered with a grey-reddish paint. It was first brushed with a steel brush, then immersed in HCl for one hour, rinsed with water, brushed and scraped with a knife. This was followed by 3 cycles of immersion in acetone (for resp. 20 minutes, 10 minutes and 1 hour) and scraping. Then it was immersed in HCl for 40 minutes, rinsed with water and cleaned with tissue. It was subsequently immersed in HNO_3 for 10 minutes and then rinsed with water and alcohol. It was then sent to PTB for measurement (the small pieces for elemental analysis were cut off afterwards).

Figure 1. Picture of the "Sumitomo bank" sample as received

- ## 2.1.1.2 Aioi bridge

This sample consisted of 6 pieces of metal welded together. The sample was showing heavy corrosion and molten metal deposit. The pieces were separated and the small samples for elemental analysis were cut off before cleaning. They were first brushed, then immersed in HCl for 70 minutes, washed with water and paper tissue, brushed, cleaned again with water and paper tissue. They were subsequently immersed in HNO_3 for 30 minutes and rinsed with alcohol. Only the connecting piece and the two outer pieces were used for measurement.

Figure 2. Pictures of the "Aioi bridge" sample as received (first two) and with the pieces separated (last two).

- 2.1.1.3 Fukoku life assurance

This sample consisted of a thick rod, bent to form part of a ring. It was partly covered with paint, most of which was removed by scraping. The sample was then cut in two pieces that were immersed in HCl for 105 minutes in order to remove rust and the last traces of paint. It was then rinsed with water and immersed in HNO_3 for 30 minutes and subsequently washed with water and alcohol before being sent to PTB for measurement.

Figure 3. Pictures of the " Fukoku life assurance" sample as received and after sawing.

- 2.1.1.4 Chugoku electricity

This sample consisted of one stepped piece showing some corrosion. After cutting off a sample for elemental analysis, it was cut in three pieces, brushed, immersed in HCl for 100 minutes, rinsed with water, then immersed in HNO_3 for 70 minutes and subsequently washed with water and alcohol before being sent to PTB for measurement.
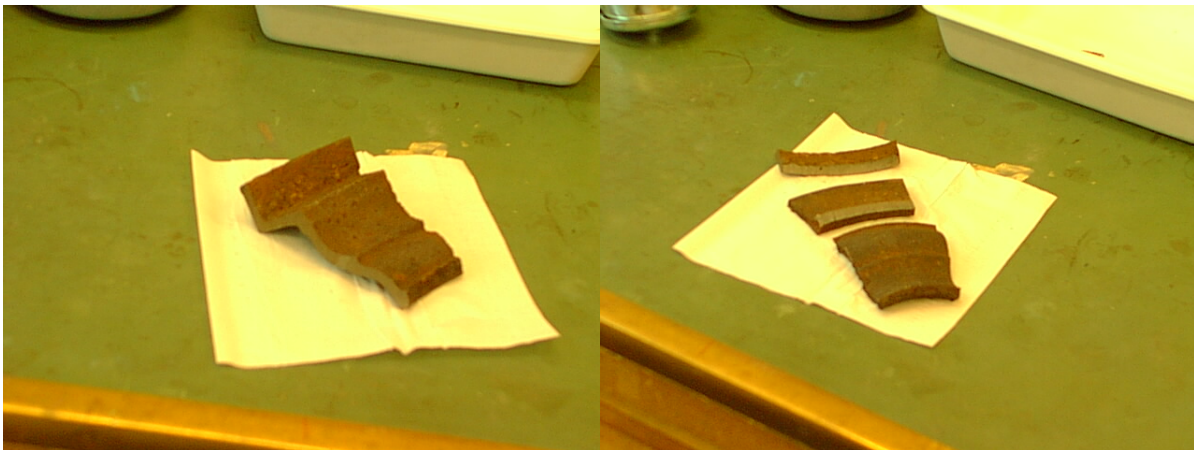
Figure 4. Pictures of the "Chugoku electricity" sample as received and after sawing.

- 2.1.1.5 Kodokan building

This sample consisted of a small piece of tube with a length varying from 21 to 25 mm, an outer diameter of 60 mm and a material thickness of 3 mm. It was first brushed with a steel brush, then immersed in HCl for 10 minutes and subsequently rinsed with water, then immersed in HNO_3 for 5 minutes and subsequently rinsed with water and alcohol. Small pieces were then cut off for elemental analysis, after which it was immersed again in HCl for 40 minutes, rinsed with water, immersed in HNO_3 for 10 minutes and rinsed with alcohol.

Figure 5. Picture of the Kodokan sample as received.

- ### 2.1.1.6 City Hall

This sample consisted of a tube with an enlargement on one side. It was immersed in HCl for 1 hour and then cleaned with water and paper tissue. It was subsequently immersed in HNO_3 for 30 minutes and rinsed with alcohol. The tube was then cut into pieces having a height of around 8 cm that were then cut in two along the vertical axis and then flattened. All those pieces were then again brushed, immersed in HCl, washed with water and paper tissue, immersed in HNO_3 and rinsed with alcohol. Finally, the piece with the enlargement was sawed in two along the vertical axis. Pieces that were screwed together came apart in a total of 6 pieces. Four of them were flattened. All pieces were then cleaned again in HCl and HNO_3. A Marinelli beaker was then cut in order to remove the outer part and the pieces were centred around the inner part (after cleaning it in ultrasonic bath). This was done in order to measure in a geometry as close as possible to a Marinelli geometry. The pieces were wrapped in plastic foil to keep them together.
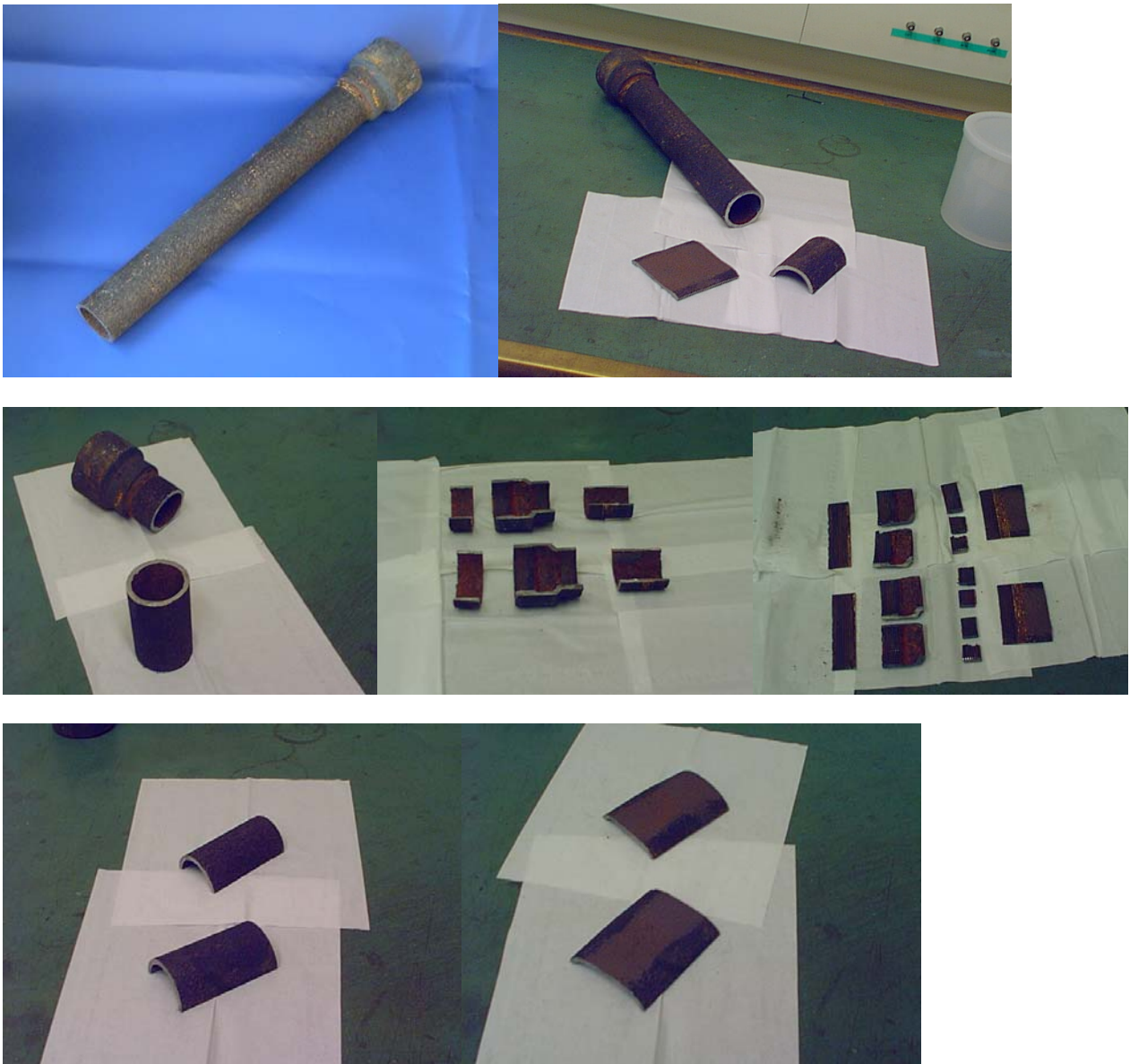
Figure 6. The "City Hall" sample cut in pieces which are cut longitudinally and flattened.

- ### 2.1.1.7 City Hall-2

This sample consisted of a solid bar, bent 90 degrees. It had a length of 418 mm and a diameter of 15 to 18 mm. It was sawed in 13 pieces which were then brushed and scraped. The pieces were then immersed in HCl for 30 minutes and rinsed with water. The 5 pieces with paint left were again scraped, brushed, immersed in HCl for 10 minutes and rinsed with water. All pieces were then immersed in HNO_3 for 20 minutes and rinsed with water. Those pieces were then placed in a standard Teflon container for measurement.

Figure 7. The "City Hall-2" sample as it arrived.

- ### 2.1.1.8 Hiroshima bank Kanayamacho

This sample consisted of a thick rod, bent, forming a part of a ring. It was brushed, immersed in HCl for 1 hour, washed with water and paper tissue, scraped to remove paint and then immersed in HNO_3 for 40 minutes and rinsed with alcohol. This sample was possibly shielded from directly exposure to the A-bomb neutrons.
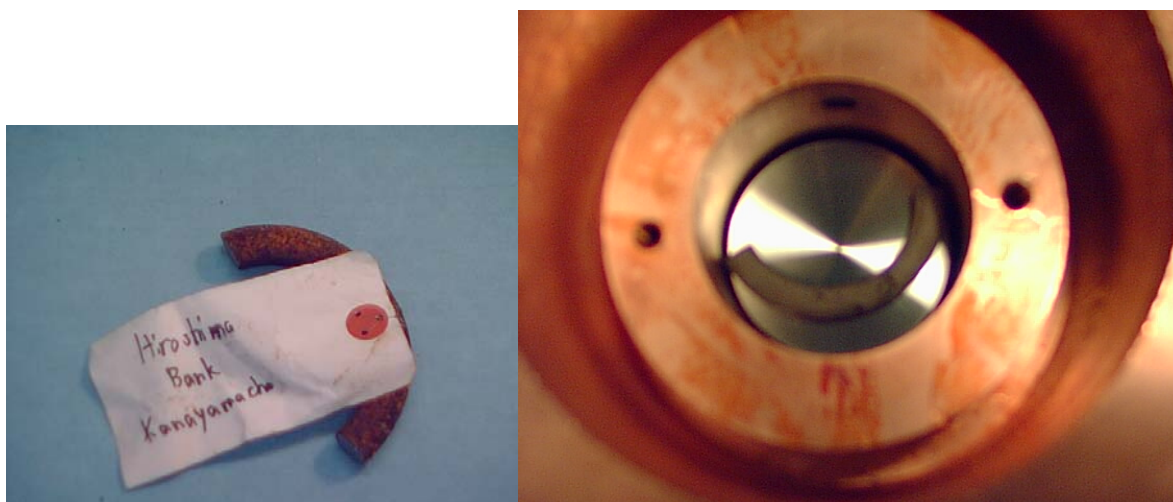
Figure 8. The "Hiroshima bank Kanayamacho" sample as it arrived and on a Germanium detector.

10

- 2.1.1.9 Hiroshima bank Kanayamacho 6

This sample consisted of a thick rod, bent to form part of a ring. It is claimed to have been directly exposed to the bomb while this should not be the case for the other Kanayamacho sample. It was sawed in two pieces that were brushed and scraped, then immersed in HCl for 30 minutes, rinsed with water, immersed in HNO_3 for 20 minutes and rinsed with water and alcohol.

Figure 9. The "Hiroshima bank Kanayamacho-6" sample as it arrived.

- 2.1.1.10 Kyu-Fuzoku elementary school

This sample consisted of a piece of tube with a length of 298 mm, a diameter of 48 mm and a material thickness of 2 mm. Heavy rust deposits were present on the inside of the tube. It was sawed in 8 pieces which were then flattened. During this procedure, most of the rust and paint came off. The pieces were then brushed, immersed in HCl for 20 minutes, rinsed with water, immersed in HNO_3 for 15 minutes and rinsed with water and alcohol.

Figure 10. The " Kyu-Fuzoku elementary school" sample as it arrived, cut in pieces and these pieces cut longitudinally and flattened.

- 2.1.1.11 Yokogawa bridge

Figure 11. The " Yokogawa bridge" sample as arrived and disassembled,

This sample consisted of four pieces of metal welded together. It was showing heavy corrosion as well as traces of molten metal. It had a length of 298 mm and a maximum height of 68 mm. The long flat pieces were sawed into 10 pieces in total while the connecting piece was left alone. The pieces were then cleaned individually by brushing, immersions in HCl (2 to 4 immersions for a total time of 100 to 190 minutes) followed by rinsing with water and brushing and a final immersion in HNO_3 for 15 to 35 minutes in order to remove rust and traces of molten metal. Subsamples for analysis were taken from the connecting piece. A Marinelli beaker was then cut in order to remove the outer part and the pieces were centred around the inner part (after cleaning it in an ultrasonic bath) in order to measure in a geometry as close as possible to a Marinelli geometry. The pieces were wrapped in a radiopure plastic foil to keep them together.

Figure 12. The " Yokogawa bridge" with its pieces cut and cleaned, the inner part of a Marinelli beaker and the sample arranged around it and fixed with plastic foil for measurement.

- 2.1.1.12 Hiroshima University plate

This sample consisted of one thick plate having a width of 69 mm and a length of 82 mm. Its thickness was between 12 and 17 mm. It was partly covered with paint. It was first brushed, then immersed in HCl for 55 minutes, rinsed in water, brushed again and subsequently immersed in HNO_3 for 30 minutes and rinsed with water and alcohol. This sample was then prepared for measurement together with the university window sample (see section 2.1.1.14 hereunder).

Figure 13. The " Hiroshima University plate" sample as it arrived.

- 2.1.1.13 Hiroshima University window

This sample consisted of three bars connected by a flat strip. It was partly covered by paint. The pieces were separated and a small sample was taken for analysis. The strip was scraped and brushed and then sawed in two pieces. Those were then immersed in HCl for 65 minutes, rinsed with water, scraped again and then immersed in HNO_3 for 40 minutes and rinsed with water. As paint was still present this procedure was done a second time. The bars were cut into small pieces and immersed in HCl for 30 minutes. This was followed by scraping and the bars were then again immersed in HCl for 50 minutes and rinsed with water. The paint left was removed by scraping and the sample was then immersed in HNO_3 for 20 minutes and rinsed with water and alcohol. A Marinelli beaker was then cut in order to remove the outer part and the pieces were centred around the inner part (after cleaning it in ultrasonic bath) in order to measure in a geometry as close as possible to a Marinelli geometry. The plate sample described in 2.1.1.13 was placed on top of the Marinelli while the pieces from the window sample were placed on the side around the inner wall of the beaker. The pieces were wrapped in radiopure plastic foil to keep them together.
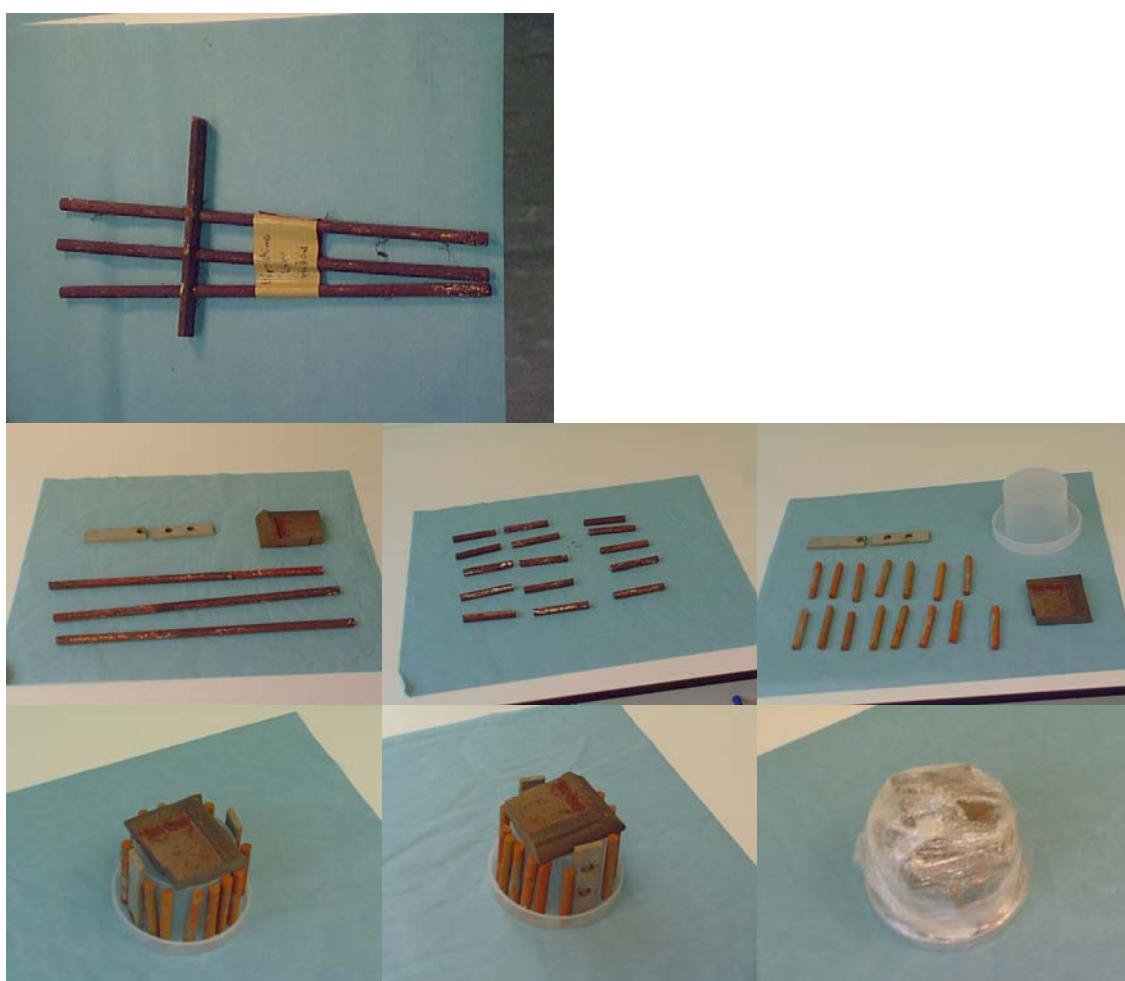
Figure 14. The " Hiroshima University window" sample as it arrived, cut in pieces and arranged around the inner part of a Marinelli beaker together with the University plate and fixed with plastic foil for measurement.

- 2.1.1.14 Nisseki "1"

This sample consisted of a slightly bent tube with a length of 225 mm, a diameter of 47 mm and a material thickness of 3 mm. The tube was cut into six pieces. Five of them were flattened until they fitted into a Marinelli beaker along the side wall. The sixth piece was completely flattened to fit on the

upper part of the beaker. The pieces were then brushed and scraped, immersed in HCl for 30 minutes, rinsed with water, scraped and brushed again to remove traces of paint, immersed in HNO_3 for 15 minutes, rinsed with water and alcohol.
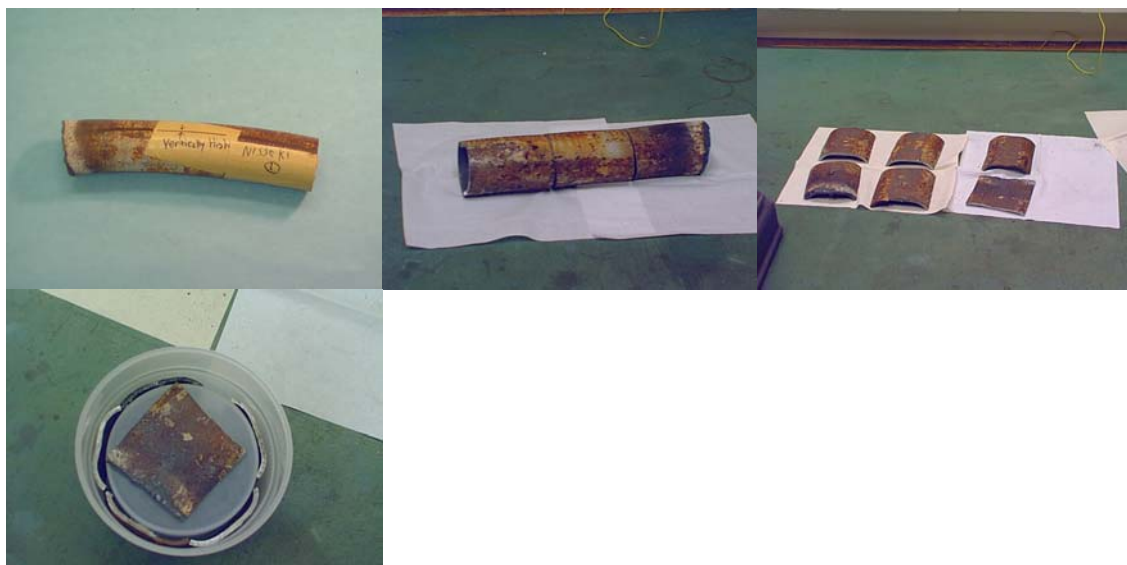
Figure 15. The "Nisseki 1" sample as it arrived, cut in pieces, these pieces cut longitudinally and partly flattened to fit in a Marinelli beaker.

- 2.1.1.15 Nisseki "5"

This sample consisted of a straight tube with a length of 280 mm, a diameter of 49 mm and a material thickness of 3 mm. The tube was cut into eight pieces that were then flattened. They were then brushed, immersed in HCl for 30 minutes, rinsed with water, brushed and scraped to remove traces of paint, immersed in HNO_3 for 15 minutes and rinsed with water and alcohol. They were then placed for measurement on the top of the Marinelli beaker on which the pieces of Nisseki "1" had already been placed.

Figure 16. The "Nisseki 5" sample as it arrived, cut in pieces and these pieces cut longitudinally and flattened.

- 2.1.1.16 Postal Saving Building (Chokin Kyoku)

This sample consisted of a very thick rod (diameter of 50 mm), in T-shape, composed of 3 pieces: the rod itself, a smaller piece and a pin connecting them. Those pieces were partly covered with paint. The sample was first brushed using a steel brush, then immersed in HCl for 75 minutes, then scraped and brushed, subsequently immersed again in HCl for 55 minutes and a second time for 80 minutes with brushing between both immersions. It was then cleaned for 15 minutes in an ultrasonic bath and then immersed in HNO_3 for 30 minutes. Then the sample was drilled at the level of the pin to separate the two main pieces. Those were then immersed in HCl for 70 minutes, brushed, immersed again in

HCl for 15 minutes, brushed, immersed in HNO_3 for 15 minutes and rinsed with water and alcohol. Subsamples for elemental analysis were taken from the smallest of both pieces. The biggest piece was then sawed in two through the central pin. The two resulting pieces were again sawed in two. The pieces were then again immersed in HCl for 20 minutes, cleaned with water and paper tissue, immersed in HNO_3 for 40 minutes and then rinsed with water and alcohol.

Figure 17. The "Postal Saving Building" sample separated in its components, these when cut in pieces and cleaned and the connecting pin cut longitudinally

2.2. Detectors and measurements

2.2.1. Detectors

Five detectors were used for this study, three at IRMM (named Ge-3, Ge-4 and Ge-5), one at PTB (named PTB-ULB) and one at LNGS (named GeCris). Their main characteristics are summarized in Table 2. Note that the results from the measurements on PTB-ULB are not included in this report.

Table 2. Data of the HPGe-detectors used for the measurements

	Ge-3	Ge-4	Ge-5	PTB-ULB	GeCris
Bkg cr[a] at 1173 keV (d[-1])	0.31(7)	0.53(10)	0.58(9)	0.68(15)	0.36(11)
Bkg cr[a] at 1332 keV (d[-1])	0.20(5)	0.41(9)	0.46(10)	0.61(14)	0.28(9)
Relative efficiency	60%	106%	50%	95%	120%
Volume (cm[3])	251	412	150	357	468
Crystal configuration	coaxial	coaxial	planar	coaxial	coaxial
Diameter (cm)	6.4	8.0	8.0	7.7	8.4
Depth of laboratory (m)	225	225	225	925	1400
Overburden (m w.e.)	500	500	500	2100	3800

[a]: counting rate of the peak at given energy in the background spectrum

2.2.2. Measurements of samples

The duration of a measurement was determined by the activity of the sample. A sample with relatively high activity was given shorter measurements time than one with relatively low activity. The activity in the samples depended both on the slant range and the mass of the sample. Long measurements were generally performed as several shorter ones since other measurements like quality control, background measurements with the same shield configuration or measurements of other samples being done between two measurements of the sample. Table 3 hereunder gives the list of all the measurements for each sample.

Table 3. Overview of the γ-ray measurements

Sample	Detector	Live time (d)	Start date	End date	Geometry
Aioi bridge	Ge-5	43.6	14-06-2005	28-07-2005	3 pieces directly on endcap
Kodokan building	Ge-3	41.5	22-02-2005	05-04-2005	Directly on endcap
City hall	Ge-3	40.4	11-05-2005	21-06-2005	1 piece directly on endcap
	Ge-5	41.6	16-08-2005	27-09-2005	Marinelli
City hall-2	Ge-5	15.6	06-12-2006	22-12-2006	In Teflon container (directly on endcap)
	Ge-5	13.7	09-01-2007	23-01-2007	
Hiroshima bank Kanayamacho	Ge-4	42.6	21-06-2005	03-08-2005	Directly on endcap
Hiroshima bank Kanayamacho 6	Ge-3	34.6	16-01-2007	20-02-2007	Directly on endcap
Kyu-Fuzoku elementary school	sandwich	46 (data not usable)	12-01-2007	27-02-2007	Pieces piled on detector
	Ge-4	27.4	11-08-2009	08-09-2009	Pieces piled on detector
	Ge-4	19.5	21-10-2009	10-11-2009	Pieces piled on detector
Yokogawa bridge	Ge-5	8.9	30-03-2005	08-04-2005	Pieces piled on detector
	Ge-5	21.7	19-04-2005	11-05-2005	
	Ge-5	46.6	24-03-2006	10-05-2006	Marinelli
	Ge-5	13.6	16-05-2006	30-05-2006	Marinelli
	Ge-5	14.9	07-06-2006	22-06-2006	Marinelli
	Ge-4	38.5	26-08-2005	04-10-2005	Marinelli
	Ge-4	65.4	21-11-2005	26-01-2006	Marinelli
Hiroshima university	Ge-3	102.1	03-11-2005	14-02-2006	Marinelli
	Ge-3	46.6	24-03-2006	10-05-2006	Marinelli
	Ge-3	6.8	17-05-2006	24-05-2006	Marinelli
	Ge-3	8.1	14-06-2006	22-06-2006	Marinelli
Nisseki	Ge-4	2.9	23-01-2007	26-01-2007	Marinelli
	Ge-4	28.4	05-02-2007	06-03-2007	Marinelli
	Ge-4	29.4	13-03-2007	12-04-2007	Marinelli
Postal Saving Building	GeCris	26.6	30-01-2006	27-02-2006	As shown on picture
	GeCris	37.3	16-06-2006	25-07-2006	

After control of each individual spectrum to check for the absence of energy shifting and other problems that can occur, spectra of the different measurements were added and Table 4 hereunder indicates the live time of the spectra sums used for the determination of the activities.

Table 4. Overview of the net live time for each sample.

Sample	detector	Live time (d)
Aioi bridge	Ge-5	43.60
Kodokan building	Ge-3	41.48
City hall	Ge-5	41.62
City hall-2	Ge-5	29.28
Hiroshima bank Kanayamacho	Ge-4	42.58
Hiroshima bank Kanayamacho 6	Ge-3	34.55
Kyu-Fuzoku elementary school	Ge-4	46.88
Yokogawa bridge	Ge-4	106.03
Yokogawa bridge	Ge-5	75.08
Hiroshima university	Ge-3	163.54
Nisseki	Ge-4	60.69
Postal Saving Building	GeCris	63.93

The spectra were not analysed using automatic peak-fitting routines. Three regions of interest were determined for each peak, one encompassing the peak itself, one at lower energy and one at higher energy (those last two being used to determine the continuum). All counts within a region of interest are taken into account.

2.2.3. Measurements of background and mercury samples in HADES

In order to lower the background in HADES, and in particular the contribution of radon daughters to it, pieces of OFHC copper have been specifically designed to fill the space around the detector and the sample. As the samples are generally placed in standard Teflon containers, those copper pieces have been designed for this specific geometry. In order to measure bigger samples, it is necessary to remove some of the copper pieces. Consequently, several shielding configurations are possible for each detector and background measurements were performed for each of the shielding configurations used during this study. Table 5 shows the list of background measurements used for this study.

Table 5. List of background measurements in HADES for the long range samples.

Detector	Configuration	Live time (d)	Counting Rate (d^{-1})	
			1173 keV	**1332 keV**
Ge-3	All Cu in	97.29	0.37	0.32
Ge-4	F out	23.76	0.72	0.45
Ge-4	E and F out	41.99	0.30	0.43
Ge-4	All Cu in	44.51	0.82	0.29
Ge-5	2 Cu out	77.49	0.46	0.43
Ge-5	3 Cu out	46.44	0.05	0.25
Ge-5	All Cu in	52.64	0.38	0.39

As the samples measured were made of steel and had a mass up to 2.7 kg (in a Marinelli geometry), the influence of the sample on the detection of background events can not be neglected. Consequently, mercury samples were also measured (in a Marinelli geometry) in order to be able to assess the influence the sample has on the detection of background counts. Mercury was chosen because of its very high attenuation coefficient, its high radiopurity and the ease with which it can be poured into containers of different shapes.

After selecting mercury batches (based on their radiopurity), two samples were prepared: one for the detectors Ge-4 and Ge-5 and the other one for the detector Ge-3 as this detector has a different outer diameter. The results were presented at the ICRM (International Committee for Radionuclide Metrology) conference in 2007 (Hult et al., 2008).

2.2.4. Subtraction of background to the peak counts

Background counts in the energy region interesting for this study are due to the presence of ^{60}Co, mainly in the copper shield and inside (or in) a detector's endcap. The contribution from ^{60}Co present inside (or in) an endcap was considered constant whatever the sample placed outside is. This assumption was made because the probability of having a photon emitted inside the endcap and scattered in the sample depositing an energy of 1173 or 1332 keV in the crystal is very small. The contribution of ^{60}Co present in the copper shield was determined using background and mercury samples measurements. It was considered that the decay of ^{60}Co was similar for each measurement and efficiencies for detecting decays occurring in the shield in the presence of air (background), a mercury sample or a steel sample were calculated by Monte-Carlo simulations. Using the measurements of background and mercury allowed determining the background counting rate to subtract it from the sample counting rate. The equation used to determine the net number of counts is shown in chapter 2.4.1.

2.3. Efficiency calculation models and programs

All the efficiency calculations were performed with Monte-Carlo simulations using EGS4. A MORTRAN code implementing a cylindrical model, UCCYSL was systematically used (Nelson et al., 1985). In this model we incorporated the source, the detector with its mounting included in some detail and the surrounding shield. Models of the detectors have been elaborated based on the manufacturers' data sheets and radiographs. Standards consisting of point sources and volume sources have been used to adjust the deadlayer thicknesses. For radionuclides having multiple gamma-rays in cascade (including ^{60}Co), the complete decay scheme is introduced in the code so that proper correction for summing-in or summing-out is applied. For ^{60}Co, the weak angular correlation was neglected. Fig.18 shows as example the model used for the measurement of mercury on the detector Ge-4.

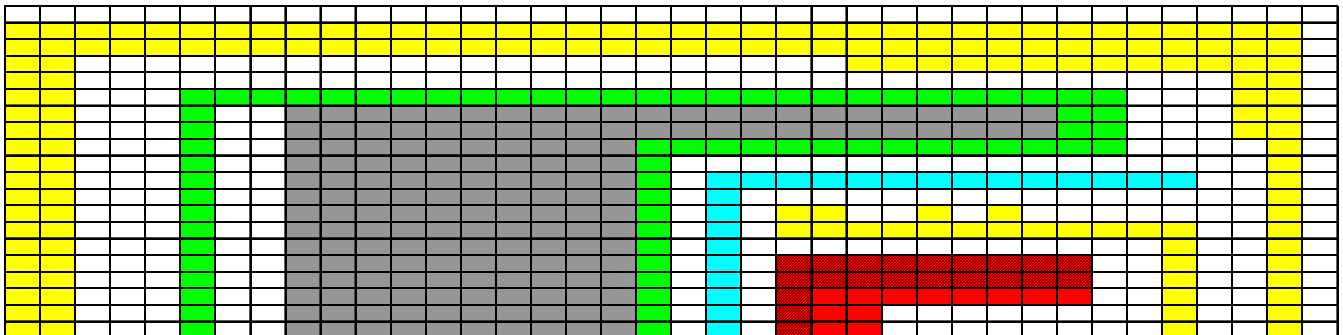**Figure 18.** Model of Ge-4 with mercury sample. Yellow represents Cu, green represents low density polyethylene, grey represents Hg, turquoise represents Al, red represents the active part of the Ge crystal and the red hashed part is the Ge deadlayer.

In EGS4, the geometry is defined by planes and radii, thus resulting in ring-shaped regions. The models representing Ge-3 (resp. Ge-4 and Ge-5) have 1723 regions (resp. 761 and 749).

2.4. Activities and decision thresholds

All calculations in this report assume a homogeneous activity distribution, which is correct only if there was a constant neutron flux inside the samples. Because of the relatively thick samples, this assumption is not entirely correct but is used here as it is considered outside the scope of this study to estimate the inhomogeneous activity. It should, however, be possible in a follow-up study to assess the inhomogeneous activity since track was kept on the position each sample was placed on a HPGe-detector. By knowing the neutron distribution inside the sample it should be possible to divide the samples into slices in the computer model and obtain activities for the different slices.

2.4.1. Activity calculations

The activity was calculated by applying the following formula to each peak:

$$A = \frac{\lambda C e^{\lambda t_d}}{\varepsilon (1 - e^{-\lambda t_m})}$$

where A is the activity at the reference date, λ the decay constant, C the background corrected net number of counts in the peak in question, t_d the decay time, ε the absolute efficiency per decay and t_m the measurement time. In case a signal above decision threshold was determined for both peaks, the weighted mean of the activity from the two peaks was calculated (adding the correlated part separately). The uncertainties are expressed as combined standard uncertainties, following the Guide to the Expression of Uncertainty in measurement. They are given in brackets after the relevant number and their numerical value refers to the corresponding last digit(s) of the quoted result. In order to correctly subtract the background, C has been calculated according to the following equation

$$C = N_c - t_m \left[\left(B_{Hg} - \frac{B_{Hg} - B_{air}}{\varepsilon_{Hg}^B - \varepsilon_{air}^B} \varepsilon_{Hg}^B \right) + \left(\frac{B_{Hg} - B_{air}}{\varepsilon_{Hg}^B - \varepsilon_{air}^B} \right) \varepsilon_{sam}^B \right]$$

where N_c is the net number of counts in the peak considered in the spectrum from the measurement of the sample, B_{Hg} and B_{air} are the net count-rates in the same peak in the mercury and the background spectra respectively. ε_{Hg}^B, ε_{air}^B and ε_{sam}^B are the calculated efficiencies (using EGS4) for detecting, in the germanium detector, decays occurring in the outer copper shield with the presence of respectively mercury, air (background measurement) and sample.

2.4.2. Determination of decision thresholds

Decision thresholds were determined according to the ISO 11929-3[1] standard using a significance level of 0.05 and 0.10 and all measured activities were compared to the decision threshold.

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[1] However, the equation used for the calculations was Eq.5 of the DIN 25482-5 standard as Eq.11 of the ISO 11929-3 standard is not correct.

2.5. Elemental analysis

It was important for this study to perform elemental analysis in order to know the cobalt content of each sample as final results given are expressed in activity of ^{60}Co per mass unit of cobalt. Subsamples were sent to BAM (Bundesanstalt für Materialforschung und –prüfung) for analysis by ICP-OES and to SCK•CEN (Studiecentrum voor Kernenergie•Centre d'étude de l'énergie nucléaire) for analysis using k_0-NAA (Etxebarria et al., 1996).

At BAM the samples were dissolved in HCl/HNO_3. the solution was then heated in H_3PO_4 and H_2SO_4 until fuming. The salts were dissolved in HCl. Finally, cobalt was determined in the sample solution with ICP-OES (λ=228.616 nm).

Samples sent to SCK•CEN were first cleaned for a few seconds in a solution containing HCl, HF and detergent, rinsed and then cleaned in an ultrasonic bath. The samples were then put in plastic bags together with neutron flux monitors. Cobalt, iron and chromium were determined (via the reactions $^{59}Co(n,\gamma)^{60}Co$, $^{58}Fe(n,\gamma)^{59}Fe$ and $^{50}Cr(n,\gamma)^{51}Cr$) after a seven-hour irradiation performed in the 700 kW reactor BR1.

Table 6. The cobalt content from ICP-OES and k_0-NAA as well as the evaluated final results.

Sample	ICP-OES: Mass fraction of Co (mg/kg)	k_0-NAA: Mass fraction of Co (mg/kg)	Rel. Diff. (%) (BAM-SCK)/SCK	final results (Mean value) (mg/kg)	Final Rel uncer. (%)
Sumitomo bank	149 (6)	139 (4)	7.2	144 (7)	5
Aioi bridge	110 (5)	102 (3)	7.8	106 (6)	5
Fukoku Life assurance	96 (5)	90 (2)	6.7	93 (4)	5
Chugoku electricity	24 (1)	22.4 (6)	7.1	23.2 (11)	5
Kodokan building	151 (4)	139 (4)	8.6	145 (8)	6
City hall	162 (6)	154 (4)	5.2	158 (6)	4
City hall-2	182 (5)	173 (4)	5.2	178 (6)	4
Hiroshima bank Kanayamacho	127 (6)	103 (3)	23.3	115 (17)	15
Kanayamacho-6	76 (6)			76 (6)	8
Kyu-Fuzoku elementary school	149 (7)	135 (4)	10.4	142 (10)	7
Yokogawa bridge	196 (9)	194 (5)	1.0	195 (7)	3
Hiroshima university plate	127 (6)	112 (3)	13.4	120 (11)	9
Hiroshima university window	279 (9)	256 (6)	9.0	268 (16)	6
Nisseki "5"	146 (3)	136 (3)	7.4	141 (7)	5
Nisseki "1"	104 (4)	97 (3)	7.2	101 (5)	5
Postal Saving Building	186 (9)	177 (5)	5.1	182 (6)	4
Reference sample (ref. value: 810 mg/kg)	820 (40)	750 (40)	9.2	-	-

3. Results

In this report, the following definitions are used:
- Massic activity: activity of ^{60}Co per unit mass of (steel) sample
- Activity concentration: activity of ^{60}Co per unit volume
- Specific activity: activity of ^{60}Co per unit mass of cobalt (in the steel sample)

Figures 19 to 25 show gamma-ray spectra of samples and the associated background and mercury measurements. All spectra have been re-binned to increase visibility by reducing the statistical fluctuation and the Y-axis systematically represents a normalised counting rate (in counts per day and per keV). Figures 19 and 20 show complete gamma-ray spectra of the Nisseki sample (respectively with a linear and a logarithmic scale as regards the Y-axis) whereas figures 21 to 25 show only the part of the gamma-ray spectra covering the peaks from ^{60}Co (at 1173 and 1332 keV) and thus cover the energy range from 1100 to 1400 keV. Those spectra are sorted out by increasing slant range and are shown using a linear-linear scale.
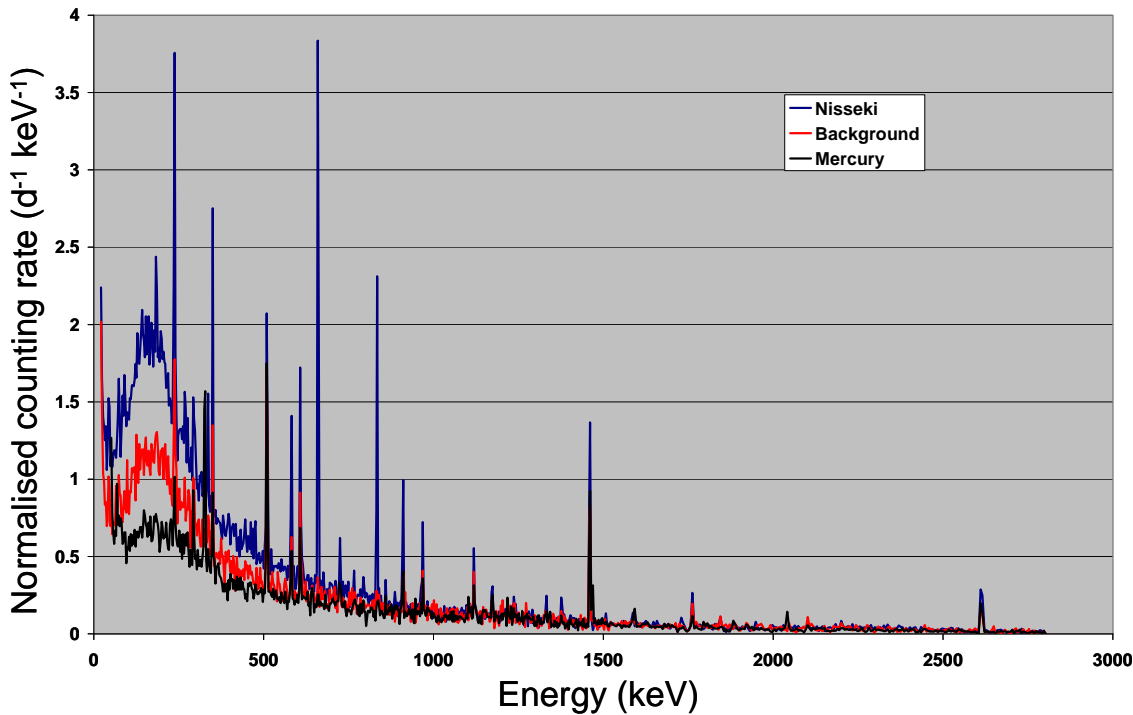

Figure 19. Gamma-ray spectrum (linear-linear scale) showing the counting rates of the sample Nisseki (measured on Ge-4) as well as background and mercury with the same shield configuration.
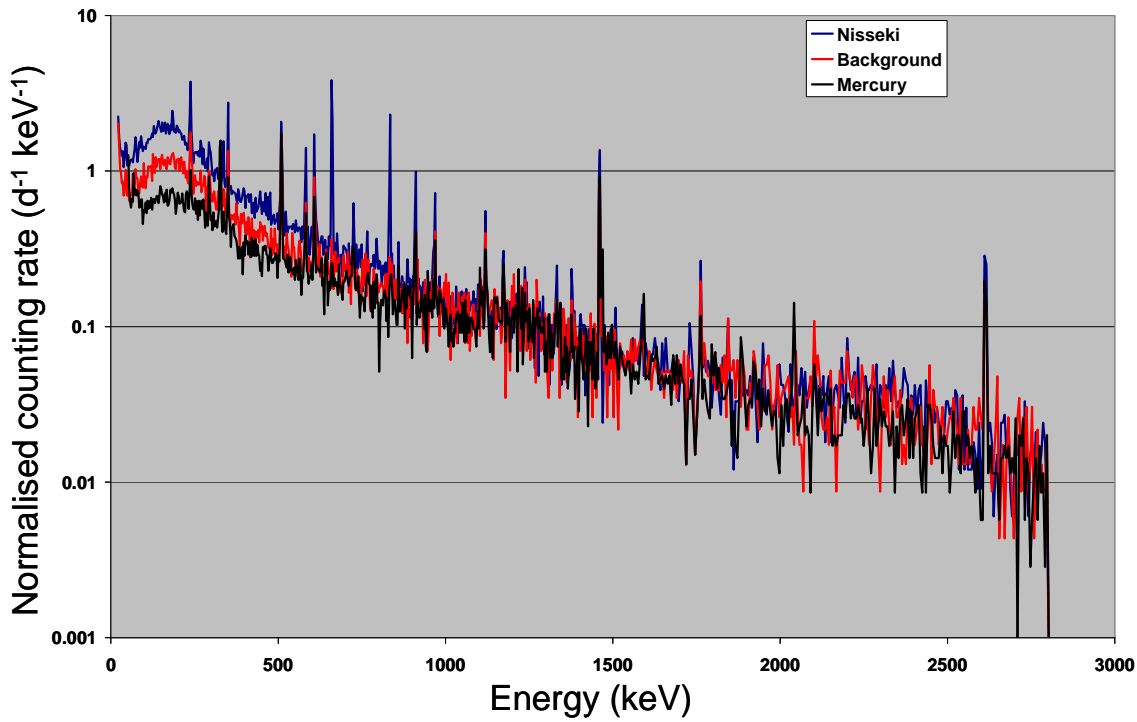
Figure 20. Gamma-ray spectrum (linear-log scale) showing the counting rates of the sample Nisseki (measured on Ge-4) as well as background and mercury with the same shield configuration.
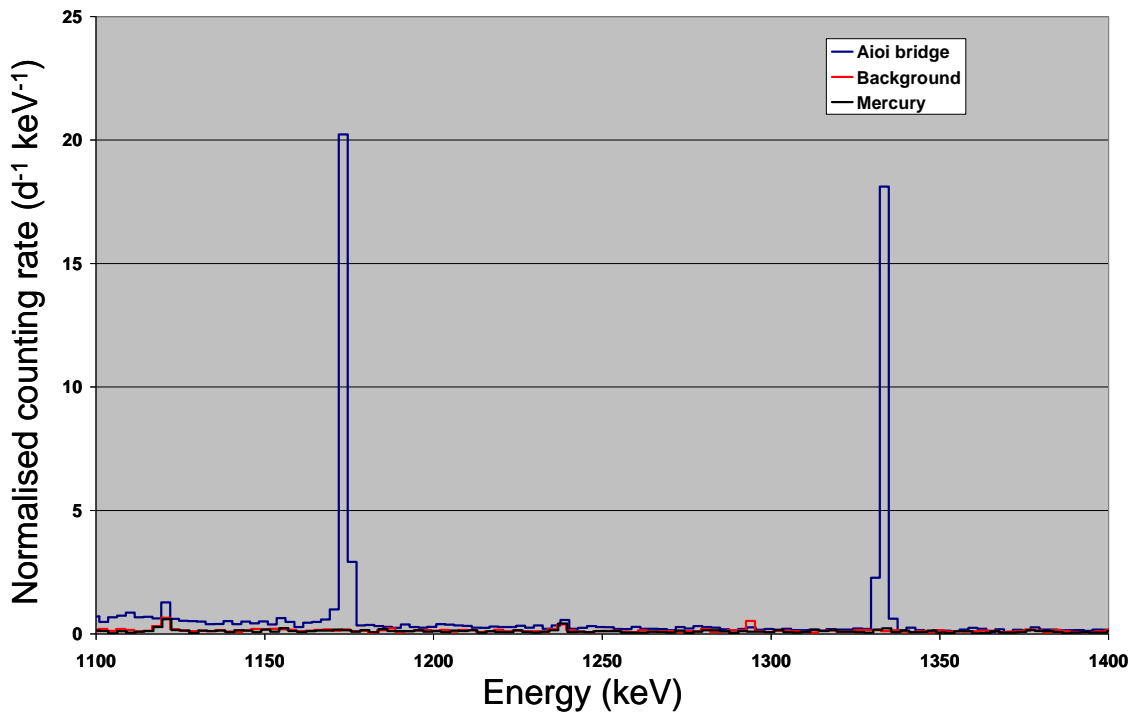

Figure 21. Part of the gamma-ray spectrum covering the peaks from ^{60}Co for the Aioi Bridge sample (slant range 671 m, measured on Ge-5). Background and mercury spectra shown have been measured with the same shield configuration.
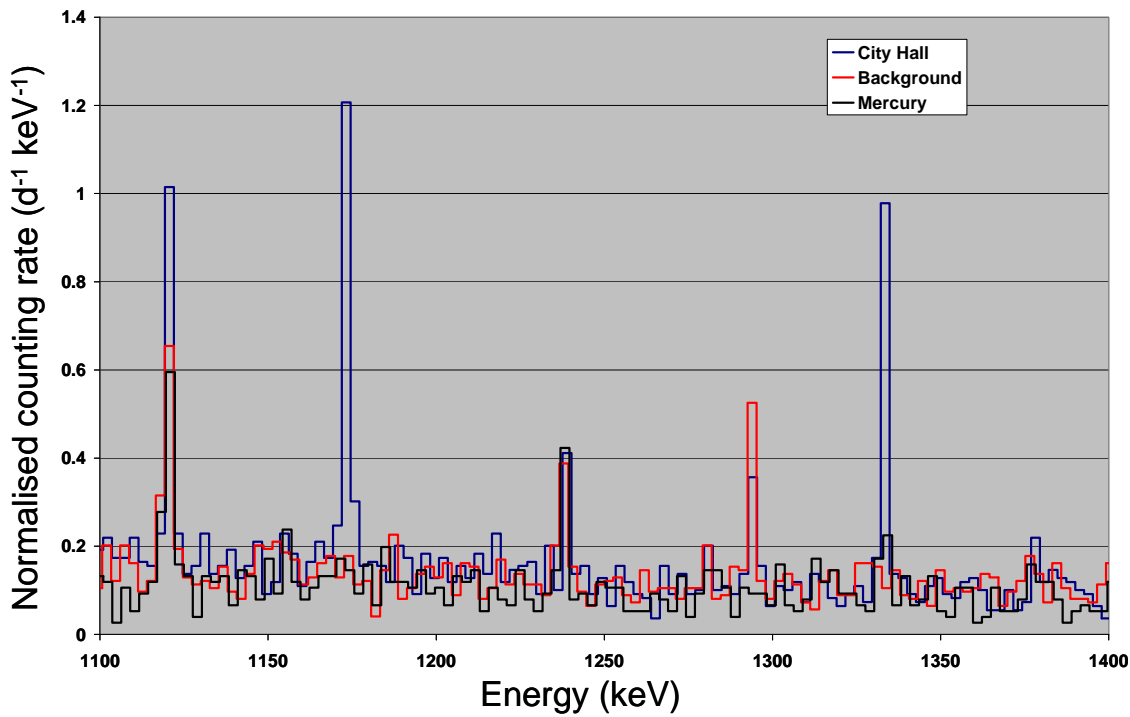
Figure 22. Part of the gamma-ray spectrum covering the peaks from ^{60}Co for the City Hall sample (slant range 1160 m, measured on Ge-5). Background and mercury spectra shown have been measured with the same shield configuration.
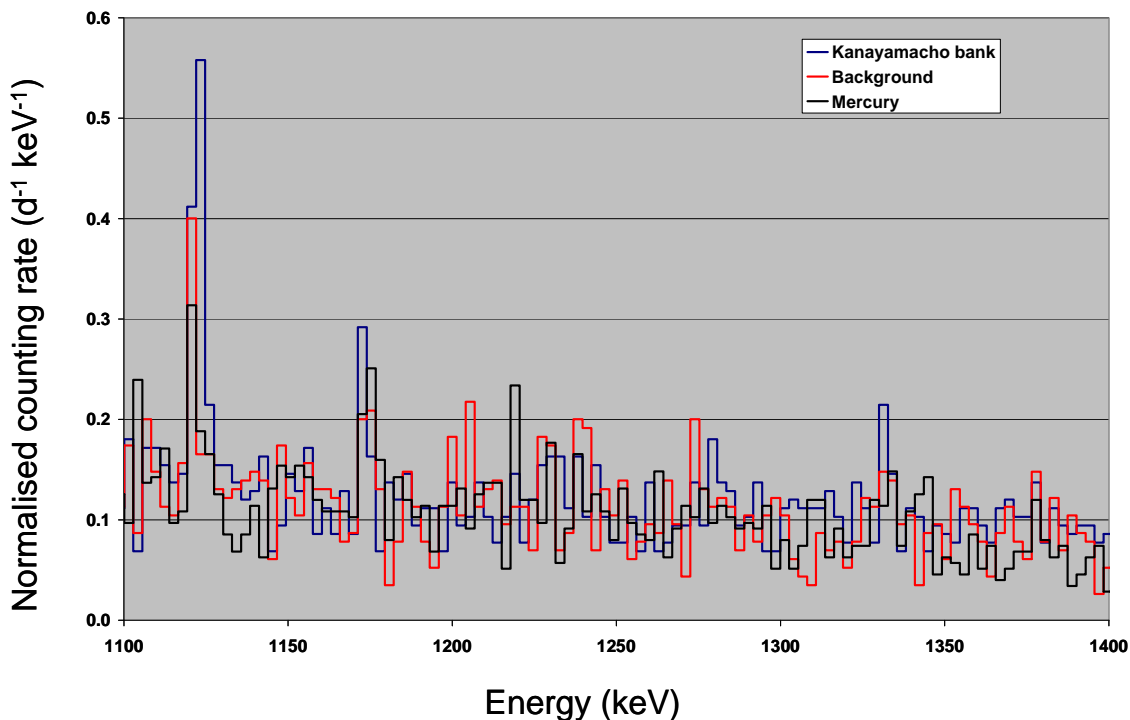

Figure 23. Part of the gamma-ray spectrum covering the peaks from ^{60}Co for the Kanayamacho bank sample (slant range 1351 m, measured on Ge-4). Background and mercury spectra shown have been measured with the same shield configuration.

Figure 24. Part of the gamma-ray spectrum covering the peaks from ^{60}Co for the Yokogawa sample (slant range 1423 m, measured on Ge-4). Background and mercury spectra shown have been measured with the same shield configuration.

Figure 25. Part of the gamma-ray spectrum covering the peaks from ^{60}Co for the Nisseki sample (slant range 1597 m, measured on Ge-4). Background and mercury spectra shown have been measured with the same shield configuration.
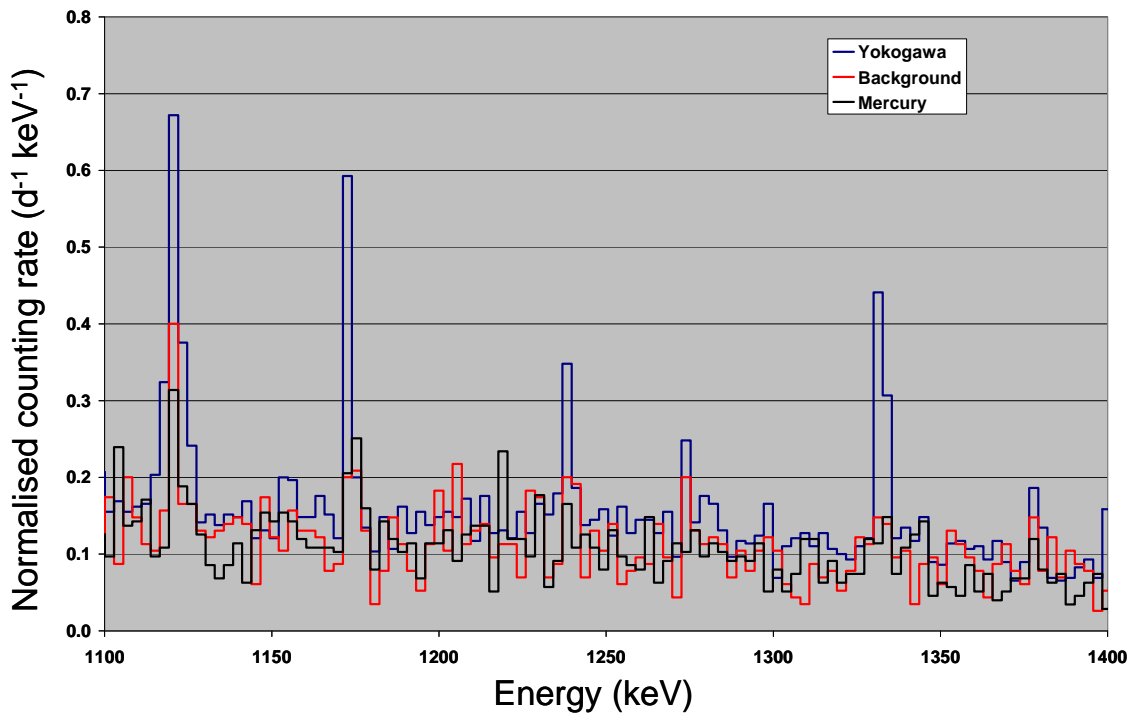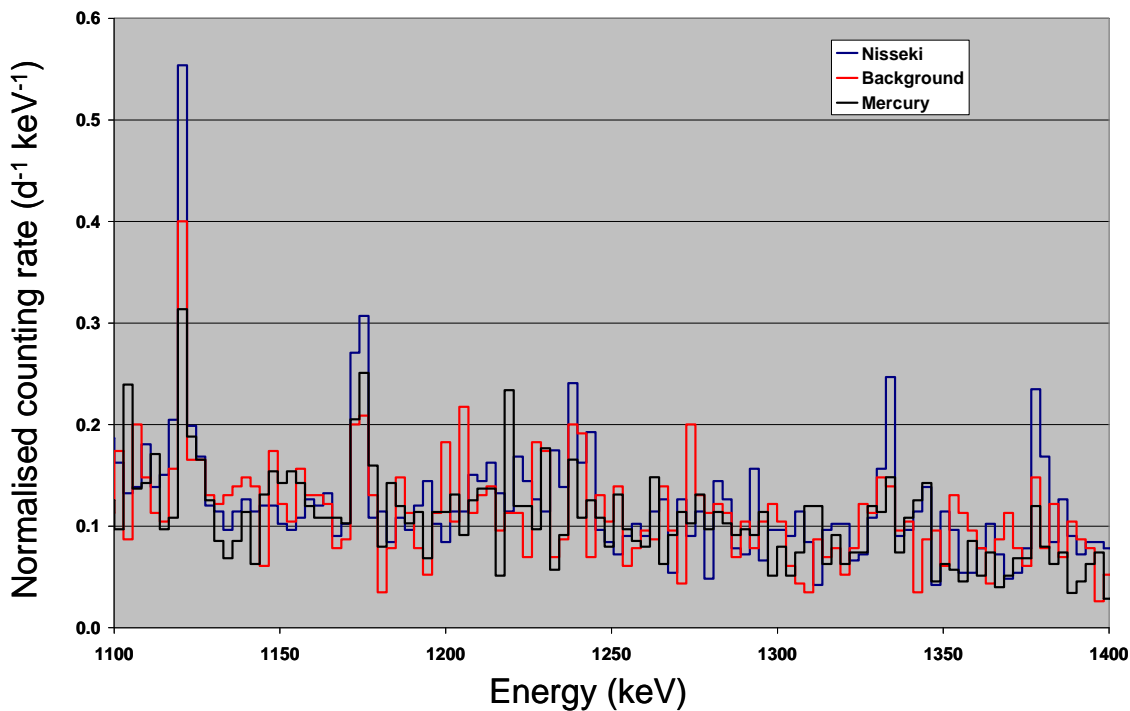
Table 7. Summary of activity at the start of the measurement

sample	Slant range (m)	Mass of sample measured (g)	Activity at start of measurement (mBq)	Specific activity (mBq/g[Co])	Measured by
Aioi bridge	671	674.23	58 (3)	810(30)	IRMM
Kodokan building	937	116.00	2.02 (25)	120(11)	IRMM
City hall	1160	1763.24	3.5 (5)	12.5(12)	IRMM
City hall-2	1160	756.60	0.5 (3)?	3.8(24)	IRMM
Hiroshima bank Kanayamacho	1351	181.8	<0.1	<5	IRMM
Hiroshima bank Kanayamacho 6	1351	220.6	<0.21	<13	IRMM
Kyu-Fuzoku elementary school	1404	535.52	<0.11	<1.5	IRMM
Yokogawa bridge	1423	2697.02	0.62 (14)	1.18(18)	IRMM
Hiroshima university	1523	1176.60	<0.1	<0.4	IRMM
Nisseki "1" and "5"	1597	1825.37	0.23(18)?	1.0(8)	IRMM
Postal Saving Building	1700	1628.61	0.18 (11)	0.6(3)	LNGS

Table 8. Summary of activity at reference date

sample	Slant range (m)	Mass of sample measured (g)	Activity at reference date (Bq)	Specific activity at reference date (Bq/g[Co])	Measured by
Aioi bridge	671	674.23	152 (6)	2130 (800)	IRMM
Kodokan building	937	116.00	5.1 (5)	300(30)	IRMM
City hall	1160	1763.24	9.3 (9)	33 (3)	IRMM
City hall-2	1160	756.60	1.6(10)?	12(8)?	IRMM
Hiroshima bank Kanayamacho	1351	181.8	<0.26	<12	IRMM
Hiroshima bank Kanayamacho 6	1351	220.6	<0.7	<40	IRMM
Kyu-Fuzoku elementary school	1404	535.52	<0.5	<6.6	IRMM
Yokogawa bridge	1423	2697.02	1.7 (3)	3.2 (5)	IRMM
Hiroshima university	1523	1176.60	<0.27	<1.1	IRMM
Nisseki "1" and "5"	1597	1825.37	0.7(6)?	3.3(25)?	IRMM
Postal Saving Building	1700	1628.61	0.5 (3)	1.8 (11)	LNGS

Tables 9 and 10 show the activity of each sample at the start of measurement and at reference date, respectively. Decision thresholds using a significance level of 0.05 and 0.10 are also indicated in order to obtain a better understanding of the significance of the results. Activities at the start of measurement are given only to indicate the sensitivity reached during the measurements and to assess the impact of environmental activation. The reference date is 1945-08-05 23:15 UTC (1945-08-06 08:15 local time).

For the long range samples (further than City Hall), the main contribution to the uncertainties are due to the counting statistics from measurements of the sample and the background (measurements with and without mercury). Together their contribution to the combined standard uncertainty is 95% to 99%. However, for two samples (Kodokan and City Hall) their contribution varies between 83% and 88% as the contribution from the efficiency is between 12% and 17%. For the samples closer to epicentre than City Hall, the counting statistics is better and the dominating uncertainty is due to the efficiency calculation. The relative uncertainty for the efficiency for these samples is in the order of 10%.

Table 9. Activities at the start of measurement together with associated decision thresholds.

Sample	Activity at start of measurement (mBq)	Decision threshold (mBq)		Measured by
		α=0.05	α=0.10	
Aioi bridge	58 (3)	0.34	0.26	IRMM
Kodokan building	2.02 (25)	0.16	0.12	IRMM
City hall	3.5 (5)	0.31	0.24	IRMM
City hall-2[a]	0.5 (3)	0.4	0.3	IRMM
Hiroshima bank Kanayamacho	0.01 (10)	0.10	0.08	IRMM
Hiroshima bank Kanayamacho 6	< 0	0.21	0.17	IRMM
Kyu-Fuzoku elementary school	< 0	0.11	0.09	IRMM
Yokogawa bridge	0.62 (14)	0.13	0.12	IRMM
Hiroshima university[a]	0.10 (9)	0.10	0.08	IRMM
Nisseki "1" and "5"[b]	0.23 (18)	0.17	0.13	IRMM
Postal Saving Building	0.18 (11)	0.19	0.15	LNGS

[a]: using only 1332 keV gamma-ray as 1173 keV gamma-ray gives result below decision threshold
[b]: using only 1173 keV gamma-ray as 1332 keV gamma-ray gives result below decision threshold

Table 10. Activities at reference date (1945-08-05 23:15 UTC) together with associated decision thresholds

Sample	Activity at reference date (Bq)	Decision threshold (Bq)		Measured by
		α=0.05	α=0.10	
Aioi bridge	152 (6)	0.9	0.7	IRMM
Kodokan building	5.1 (5)	0.4	0.3	IRMM
City hall	9.3 (9)	0.8	0.6	IRMM
City hall-2[a]	1.6 (10)	1.3	1.0	IRMM
Hiroshima bank Kanayamacho	0.03 (21)	0.26	0.20	IRMM
Hiroshima bank Kanayamacho 6	< 0	0.7	0.5	IRMM
Kyu-Fuzoku elementary school	< 0	0.50	0.39	IRMM
Yokogawa bridge	1.7 (3)	0.4	0.3	IRMM
Hiroshima university[a]	0.27 (24)	0.27	0.21	IRMM
Nisseki "1" and "5"[b]	0.7 (6)	0.6	0.4	IRMM
Postal Saving Building	0.5 (3)	0.56	0.44	LNGS

[a]: using only 1332 keV gamma-ray as 1173 keV gamma-ray gives result below decision threshold
[b]: using only 1173 keV gamma-ray as 1332 keV gamma-ray gives result below decision threshold

More interesting is the specific activity, i.e the activity of ^{60}Co per mass unit of total cobalt, (also given in Table 8) as this is more or less directly proportional to the neutron fluence in the area where the sample was located. Specific activities of the samples together with decision thresholds at reference date are given in Table 11.

Table 11. Specific activities at reference date together with associated decision thresholds.

sample	Specific activity at reference date (Bq/g[Co])	Decision threshold (Bq/g[Co])		Measured by
		α=0.05	α=0.10	
Aioi bridge	2130 (800)	12	10	IRMM
Kodokan building	300 (30)	23	18	IRMM
City hall	33 (3)	3	2.3	IRMM
City hall-2[a]	12 (8)	10	8	IRMM
Hiroshima bank Kanayamacho	1 (9)	12	10	IRMM
Hiroshima bank Kanayamacho 6	< 0	40	30	IRMM
Kyu-Fuzoku elementary school	< 0	6.6	5.1	IRMM
Yokogawa bridge	3.2 (5)	0.6	0.5	IRMM
Hiroshima university[a]	1.1 (10)	1.1	0.9	IRMM
Nisseki "1" and "5"[b]	3.3 (25)	2.5	1.9	IRMM
Postal Saving Building	1.8 (11)	1.9	1.5	LNGS

[a]: using only 1332 keV gamma-ray as 1173 keV gamma-ray gives result below decision threshold
[b]: using only 1173 keV gamma-ray as 1332 keV gamma-ray gives result below decision threshold

3.1. Other activation products

In the gamma spectra from the measurements there are also peaks at 811 keV (^{58}Co) and 835 keV (^{54}Mn). As the half-lives of these radionuclides are relatively short (71 d and 312 d, respectively) the peaks from these radionuclides can possibly be used to estimated the activation due to environmental neutrons and cosmic rays. Table 12 gives the activity of these radionuclides in the samples at the time when a sample was taken underground. Note that also in these calculations a homogeneous activity distribution was assumed.

Table 12. Massic activity (i.e. activity per kg of sample mass) for ^{58}Co and ^{54}Mn. Decision thresholds calculated using α=0.05.

Sample	Slant range [m]	Mass [kg]	Activity at the start of measurement [mBq]		Massic activity at the start of measurement [mBq/kg]	
			^{58}Co	^{54}Mn	^{58}Co	^{54}Mn
Aioi bridge	671	0.6742	<0.6	1.3(3)	<0.9	1.9(5)
Kodokan	937	0.1160	<0.18	<0.16	<1.5	<1.3
City Hall	1160	1.7632	<0.5	2.3(4)	<0.27	1.31(22)
City Hall 2	1160	0.7566	<0.4	1.7(3)	<0.6	2.2(4)
Bank	1351	0.1818	0.47(9)	0.41(7)	2.6(5)	2.3(4)
Bank-6	1370	0.2206	0.27(12)	0.54(13)	1.2(6)	2.5(6)
Kyu-Fuzoku	1404	0.5355	<0.11	0.93(13)	<0.21	1.74(25)
Yokogawa	1423	2.6970	0.23(8)	4.3(5)	0.08(3)	1.60(17)
University	1523	1.1766	<0.18	2.4(3)	<0.16	2.02(22)
Nisseki	1597	1.8254	<0.20	3.6(4)	<0.11	1.96(22)
Postal Saving Building	1700	1.628		1.2(2)		0.75(12)

4. Activation of environmental neutrons

The main factor limiting the possibility of using ^{60}Co in steel as a monitor for retrospective neutron exposure, is the level of the activation due to environmental neutrons and cosmic rays. According to Shizuma (Shizuma et al., 2002), without self-shielding the specific saturation activity of ^{60}Co in steel is about 3 mBq/g(Co) (=3·10^{-6} Bq/mg(Co)), which is shown as a red line in Figure 26. This corresponds to a specific activity of 0.009 Bq/mg(Co) at the reference date in 1945. According to model calculations in DS02 this specific activity was obtained at a slant range of about 1400 m. As discussed earlier the self-shielding results in lower activation in massive samples, which could indicate the activities measured for the long range samples in this study presumable to a large extent is still due to A-bomb activation. Nevertheless it is not possible anymore to determine the exact activity of ^{60}Co induced by the A-bomb in steel samples at slant ranges exceeding about 1400 m. It is, however, possible to refute earlier claims of higher specific activities of ^{60}Co at these distances.
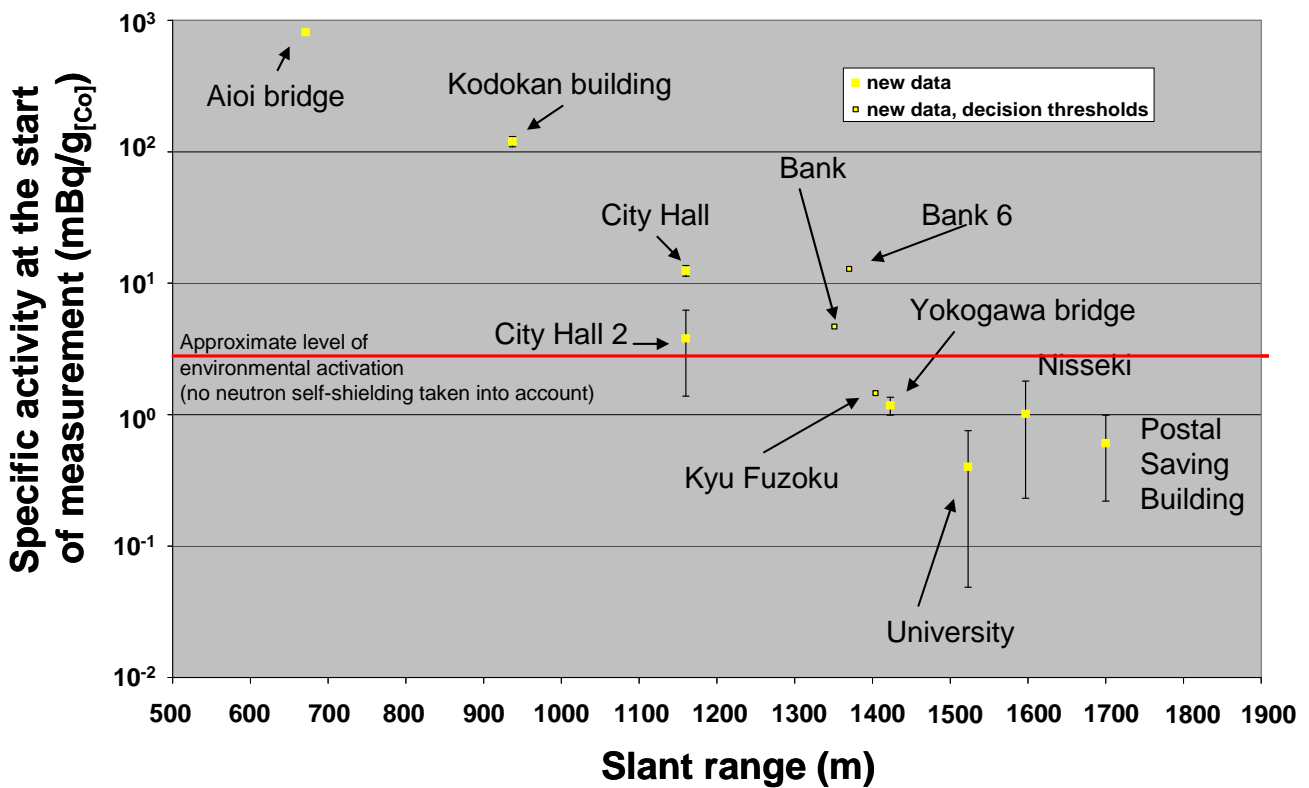

Figure 26. Specific ^{60}Co activity at the start of measurement.

5. Discussion

As already mentioned, it is important to compare the experimental values with model calculations that are the basis of the international regulations concerning doses. Fig. 27 shows two curves from the DS86 and DS02 models, together with the activity results reported here. It shows also the previous results obtained after chemical pre-concentration of cobalt (Shizuma et al., 1998) and the results of the first IRMM-PTB study (Hult et al., 2004).
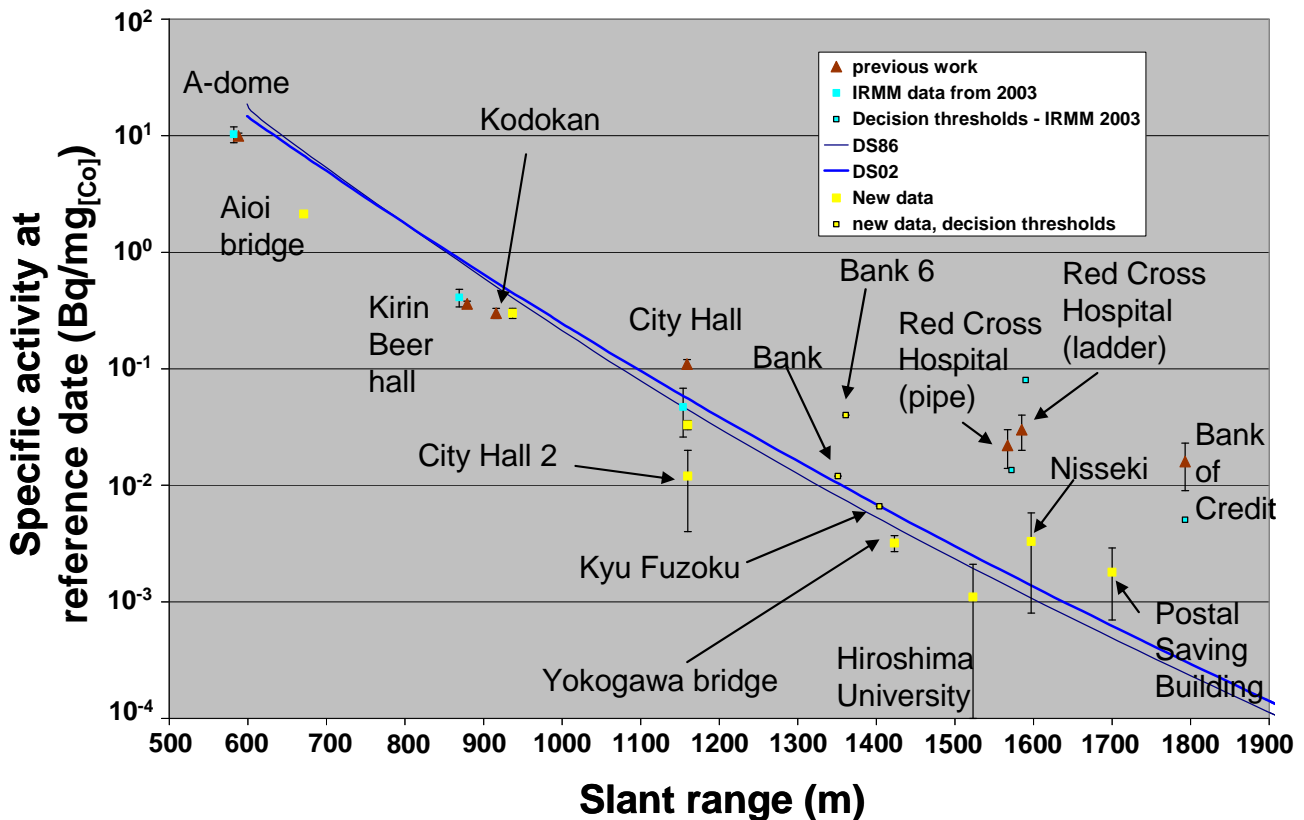

Figure 27: Specific ^{60}Co activity (i.e per mg of Co) at the reference date in 1945 as a function of the slant range from the epicentre.

It should be noted that these results do not take into account corrections for neutrons self-absorption in the samples, which is probably the main reason why the (relatively) thick samples appear in this graph with specific activities below the DS86 and DS02 curves. This is particularly pronounced for the "Aioi bridge" sample (dimensions 108x84x45 mm).
"City Hall 2" is also quite low compared to theoretical value although it is not so thick (diameter 18 mm). This sample was measured for 29 days and the number of counts in the 1173 keV and 1332 keV gamma-rays peaks was around 20 whereas according to the theoretical curves, it should have been around 70. Since the first City Hall sample has a higher specific activity one could speculate that the "City Hall 2" sample was somewhat shielded from the explosion.

For some of the samples, only decision thresholds are given. This is the case for the two Kanayamacho bank samples. "Kanayamacho bank-6" is claimed to have been directly exposed to the bomb while this was not considered certain for the other bank sample. Those two samples were bars with a diameter of 15 mm.

For the University sample, the number of counts is at the level of the (5%) decision threshold and about 60% of what it should be according to the theoretical curves although this sample was also not

amongst the thickest ones. It should be noted that in order to obtain "enough" mass, two different samples from the university were measured together and impact of this might be difficult to assess in case the two samples were activated differently. The low values that were obtained can even so be a useful indication of the level of induced activity.

As discussed in Chapter 4, the effect of environmental neutrons is detrimental at slant ranges above 1400 m. Nevertheless It is possible to refute earlier measurements yielding specific ^{60}Co activities 10 to 100 times higher than DS86 and DS02 predictions at these distances. These measurements should therefore increase confidence in the existing dosimetry model (DS02).

6. Conclusions

It is important for the confidence in the model calculations presented in the DS02 to quantify ^{60}Co in steel at slant ranges greater than 1000 m. This report presents the lowest specific activities of ^{60}Co ever measured in steel samples from Hiroshima. The main limit in the measurements presented here were from ^{60}Co induced by environmental neutrons and cosmic rays. Although the influence of environmental activation is significant at slant ranges greater than 1400 m, these extremely low-level measurements should serve the purpose of creating greater confidence in the DS02 model calculations.

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Abstract
In order to help solving discrepancies in retrospective Hiroshima dosimetry, sixteen steel samples from Hiroshima were measured in the underground laboratory HADES. The aim was to measure the specific activity of ^{60}Co. Although it was found that activities at long range (> 1300 m slant range) were mainly cosmogenically induced, the activities were so low that former measurement that were 10 to 100 times higher than predicted by computer model calculations in DS86 and DS02 could be refuted. As a consequence, the confidence in the existing dose estimates has become stronger.

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