Chapter 9

ACTIVATION MEASUREMENTS FOR FAST NEUTRONS

Part D. Evaluation of Cosmic-Ray-Induced ^{63}Ni Background in Copper

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Introduction

As a result of a joint collaboration between the University of Utah, LLNL, the Technical University Munich and the Ludwig Maximilians University Munich, it became possible to determine A-bomb induced ^{63}Ni in pure copper samples from Hiroshima beyond a ground range of 1,000 m (see Chapter 9, Part B). The low ^{63}Ni activities induced in copper samples due to neutrons from the A-bomb explosion at large distance require, however, a careful discussion of the fraction of ^{63}Ni produced in these samples due to cosmic radiation.

In this section, an analysis of the production of ^{63}Ni in copper samples due to cosmic radiation is performed. Production due to neutrons, protons, stopped muons, and photonuclear reactions is discussed. It is obvious from Figure 1 (Pfennig et al. 1995) that a variety of reactions induced by neutrons, protons, muons and photons can contribute to the production of ^{63}Ni in copper. The most important of these processes will be discussed here. Since the cross-sections for the production of ^{63}Ni in copper samples due to fast and stopped muons were not known, they were determined experimentally.

The estimates given in this section are based on the assumption that copper was exposed to cosmic radiation on the Earth's surface for a sufficiently long time for the ^{63}Ni activity to reach saturation. All results are given in terms of ^{63}Ni nuclei per gram copper. Our estimates indicate that as many as $4\text{-}5{\cdot}10^{3}$ ^{63}Ni nuclei per gram copper are present in a copper sample that was exposed to cosmic radiation in Hiroshima for about 80 years. Based on neutron fluences given in this report (see Chapter 3), a similar concentration due to A-bomb neutrons would also be expected in Hiroshima free-in-air and one meter above ground, at a distance from the epicenter of about 1,800 m. An experimental corroboration of our estimates would require, for example,

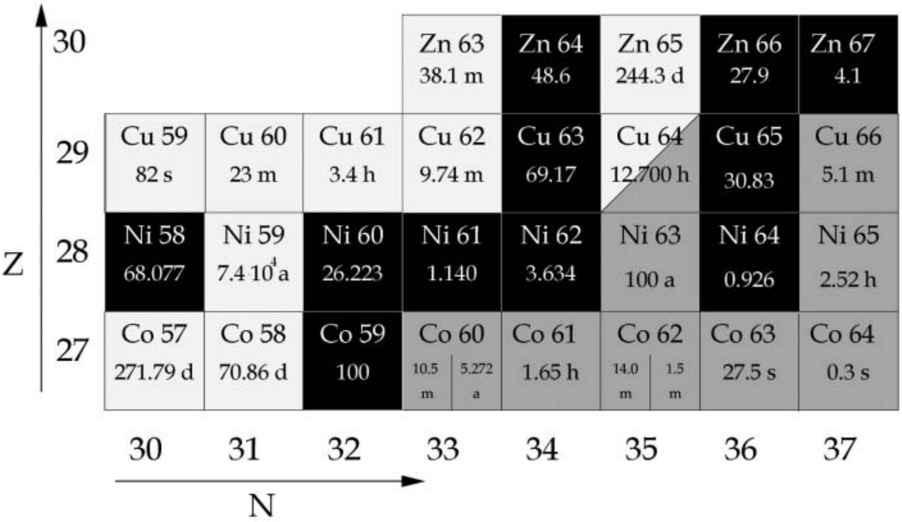


Figure 1. *Part of the Chart of the Nuclides (Pfennig et al. 1995) around* ^{63}*Ni; black: stable nuclide and isotopic abundance; dark gray:* β^-*-decay, and half-life; light gray:* β^+*-decay or electron capture, and half-life.*

measurement of the number of ^{63}Ni nuclei in copper samples exposed to cosmic radiation at different altitudes. Such measurements are presently in progress. At the present stage of investigation we conclude that the ^{63}Ni background measured in distant copper samples from Hiroshima (Chapter 9, Part B) cannot be explained by cosmic-ray induced processes alone.

Materials and Methods

Calculation of ^{63}*Ni Production in Copper due to Neutron- and Proton-Induced Reactions*

Neutron Flux. At mean geomagnetic latitudes and sea level, the neutron flux is about 10^{-2} n/(cm^2 s). In general, it has its origin in the interaction of primary cosmic ray particles with the atmosphere of the Earth and depends on solar activity, altitude, and geomagnetic cut-off. The following estimates are based on the neutron spectrum shown in Figure 2 which was measured from July 26th to August 2nd, 1999, in Hampton, Virginia, USA (geographic latitude: 37.04° N; geographic longitude: 76.35° W; geomagnetic latitude: 48.0° N; geomagnetic cut-off: 2.702 GV), at sea level (Goldhagen et al. 2000; Goldhagen et al. 2002).

Figure 2 shows a lethargy representation of those data. Such a plot has the advantage that the area under the curve in any energy range corresponds to the neutron fluence in that energy range. Therefore, equal areas represent equal numbers of neutrons. The first peak in Figure 2 corresponds to a Maxwell distribution at thermal energies. The second peak at about 2 MeV originates from neutrons evaporating from highly excited residual nuclei. Finally, the third peak is due to a broad minimum in the corresponding neutron-air reaction cross sections at energies of

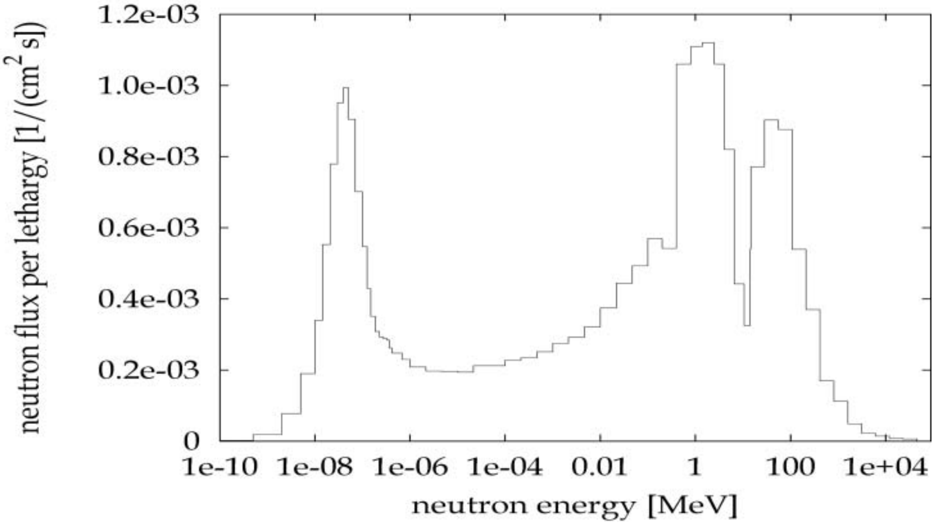


Figure 2. *Neutron spectrum at sea level, measured from July 26th to August 2nd, 1999, in Hampton, Virginia, USA (Goldhagen et al. 2000; Goldhagen et al. 2002).*

about 100-300 MeV (Roesler et al. 1998). The use of the neutron spectrum shown in Figure 2 for other geographic locations and years requires corrections for the 11-year solar cycle, altitude and different geomagnetic latitude.

Solar Cycle. The influence of the solar activity on the neutron flux at sea level is rather small. This is e.g. demonstrated by the data obtained with a neutron monitor located at Kiel, Germany (Figure 3), which consists of a BF_3 proportional counter surrounded by polyethylene and lead. The data represent a measurement program that has continued for more than 40 years. Maxima and minima of the curve (which correspond to minima and maxima in solar activity, respectively) differ by less than 10% from the mean.

Altitude. The interaction of primary cosmic-ray particles with the atmosphere causes an increase of secondary particles such as neutrons, and a decrease of primary particles, with increasing vertical atmospheric depth x towards the Earth's surface. Therefore, the altitude h is a parameter that must be taken into account if one would like to apply the neutron spectrum given in Figure 2 to other locations. Up to an altitude of about 16 km (i.e. above a vertical atmospheric depth x of about 100 g/cm^2), where the cosmic radiation intensity reaches a maximum, the shape of the neutron spectra below 1 GeV does not depend on altitude and can therefore be expressed by equation (1) (Roesler et al. 1998):

$$\frac{d\Phi(E,x)}{dE} = f(x)\cdot\frac{d\Phi(E)}{dE}, \qquad x > 100\, g/cm^2 \tag{1}$$

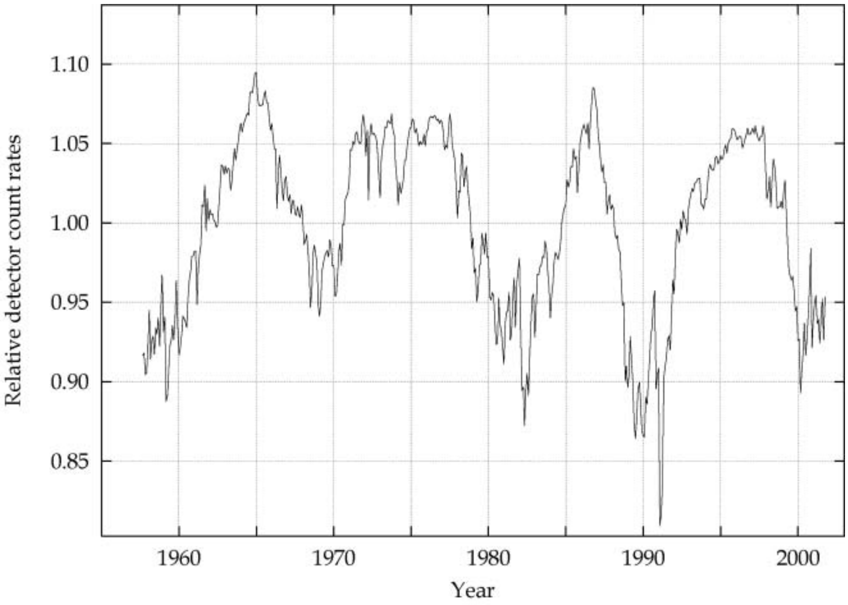


Figure 3. *Relative count rate (normalized to the mean of the count rate) measured by a neutron monitor over the past 40 years in Kiel, Germany (National Geophysics Data Center 2002).*

The function f(x), which describes the dependence of the neutron flux on vertical atmospheric depth, can be expressed by equation (2) (Schraube et al. 1997; Roesler et al. 1998):

$$f(x) = f(0) \cdot e^{-\beta \cdot x}, \qquad \beta = (7.21 \pm 0.01) \cdot 10^{-3} \quad cm^2 / g \tag{2}$$

Equation (3) is used to convert the vertical atmospheric depth x (in g/cm^2) to altitude h (in km) (Roesler et al. 1998):

$$h = 44.34 - 11.86 \cdot x^{0.19}, \qquad x > 230\ g / cm^2 \tag{3}$$

Geomagnetic Latitude. Different approaches are used to correct for different geomagnetic latitudes. Yokoyama et al. (Yokoyama et al. 1977) published correction factors for the production of cosmogenic radionuclides that are independent of the altitude, but dependent on geomagnetic and geographic latitude (Table 1). They based their values on calculations, and normalized them to a cut-off rigidity of 4.7 GV. For example, the relative production rate for Hampton, USA, (line C in Table 1, geomagnetic latitude 48.0° N) is 1.26 (linear interpolation between 1.23 and 1.39, see Table 1), whereas that for Hiroshima (line E in Table 1, geomagnetic latitude 27.0° N) is 0.55 (linear interpolation between 0.56 and 0.45, see Table 1). Accordingly, in order to convert the neutron spectrum for Hampton to Hiroshima conditions, a factor of 0.55/1.26 = 0.43 is used. Lal, on the other hand, deduced relative correction factors that depend on geomagnetic latitude and altitude (Lal 1991). His values are based on measurements of nuclear disintegration rates in the atmosphere (Table 2). In this work we are using both approaches in order to illustrate the uncertainties involved in the corrections applied for different geomagnetic latitudes.

Proton Flux. High energy protons can contribute to the production of radionuclides via reactions such as, for example, (p,2pn) etc. To estimate the proton energy spectrum at sea level, the proton/neutron ratio given by Lal and Peters as a function of energy was used and folded with the neutron spectrum shown in Figure 2 (Lal and Peters 1967).

Cross Sections. The contribution of high-energy neutrons and protons to the production of any radionuclide is difficult to quantify, since most cross-section libraries such as ENDF or JENDL usually do not supply data above energies of 20 MeV. Therefore, we used the computer code CEM95 (Gudima et al. 1983) to generate the cross sections of the relevant reactions, for energies between 1 MeV and 221 MeV. Cross sections calculated with CEM95 are believed to be accurate within a factor 2, for target nuclei between ^{27}Al and ^{197}Au (Mashnik et al. 1998). At an energy of about 15 MeV, the ENDF/B-VI cross sections matched reasonably well to the corresponding calculated CEM95 cross sections. Therefore, the ENDF/B-VI cross sections were used for energies below 15 MeV, and the CEM95 cross sections for energies above.

Table 1. Calculations of relative production rates (Production) of cosmogenic radionuclides (lower line) as a function of geomagnetic latitude; Table from Yokoyama et al. (1977)

Cut-off riginity	0	1	2	3	4	6	8	10	12	15
A	80	62	56	52	49	43	39	35	30	15
B	-80	-60	-50	-43	-38	-31	-25	-18	-11	0
C	80	58	52	47	44	38	32	28	20	–
D	-80	-64	-58	-53	-49	-41	-34	-28	-14	–
E	80	56	49	44	40	35	31	28	25	17
F	-80	-57	-51	-47	-44	-39	-34	-30	-26	-17
Production	1.49	1.49	1.39	1.23	1.09	0.86	0.70	0.56	0.45	0.37

Note: A: Europe, North Africa; B: South Africa; C: North America; D: South America; E: Asia; F: Australia.

Table 2. Nuclear disintegration rates in the atmosphere for altitudes below 10 km, s ($g^{-1} \cdot y^{-1}$), are fitted to a third-order polynomial in altitude, h (km): $s = a_1 + a_2 \cdot h + a_3 \cdot h^2 + a_4 \cdot h^3$ for different geomagnetic latitudes (Lal 1991)

Geomagnetic latitude	0°	10°	20°	30°	40°	50°	60° - 90°
a_1	330.7	337.9	382.1	469.3	525.6	571.1	563.4
a_2	255.9	252.1	272.1	394.6	505.4	588.1	621.8
a_3	98.43	111.0	132.5	97.76	142.0	170.9	177.3
a_4	20.50	20.73	24.83	47.20	58.87	76.12	78.91

Calculation of ^{63}Ni Production in Copper due to Stopped Muons

High energy protons react with nuclei in the atmosphere and produce e.g. short-lived $\pi^{\pm}$ pions that decay ($\tau = 2.6 \cdot 10^{-8}$ s) to produce muons. At sea level, 44% of the total muon intensity is due to negative muons (Takagi and Tanaka 1968). In matter, negative muons are slowed down and captured by the Coulomb potential of atoms. If the stopping material consists of several elements, the proportion of negative muons captured by one of these elements is described by the chemical compound factor f_C. Within 10^{-11} s, a captured negative muon reaches the muonic 1s shell (Charalambus 1971). The fraction f_D of the muons in the 1s shell that does not decay via $\mu^- \rightarrow e^- + \nu_\mu + \nu_e$ ($\tau = 2.2$ µs) is finally captured by the nucleus and reacts with a proton to form a neutron (equation 4):

$$\mu^- + {}^{A}_{N}Z \rightarrow \nu_\mu + {}^{A}_{N+1}(Z-1)^* \tag{4}$$

The resulting excited nucleus loses its energy by emission of photons, neutrons, protons, etc. The probability that a certain nuclide (e.g. ^{63}Ni) is produced from a certain target element (e.g. copper) is described by the effective probability f^*, which in turn includes the relevant isotopic abundances (e.g. for ^{63}Cu and ^{65}Cu) and the corresponding branching ratios for the emission of the required particles (e.g. emission of no nucleon to produce ^{63}Ni from^{63}Cu, or emission of two neutrons to produce ^{63}Ni from ^{65}Cu).

To summarize, the production rate $P_{\mu^-}(d)$ of a certain nuclide by capture of stopped muons as a function of depth d in the lithosphere can be calculated according to equation (5).

$$P_{\mu^-}(d) = \theta_{\mu^-}(d) \cdot f_C \cdot f_D \cdot f^* \tag{5}$$

where

$P_{\mu^-}(d)$: production rate of the radionuclide of interest at depth d in the lithosphere,

θ_{μ^-} : total stopping rate of negative muons,

f_C : chemical compound factor,

f_D : probability for capture in the nucleus,

f^* : effective probability for production of a certain nuclide.

In this paper, a rate of stopped negative muons of 190 μ^- per gram and year is used (Heisinger et al. 2002b) for estimation of the number of any radionuclide produced due to stopped muons at sea level.

The effective probability f^* was not known for the reactions investigated in this paper and, therefore, was determined experimentally. For this purpose, a high-purity copper target was used (99.999%; mass: 45.5 g; thickness: 2.0 mm), which had been irradiated previously with low-energy negative muons at the Paul-Scherrer-Institute in Villigen, Switzerland. The number of negative muons stopped in the sample was determined to be $N_{\mu^-} = (2.32 \pm 0.11) \cdot 10^{10}$ by the muonic X rays measured in-beam with a coaxial high-purity germanium detector. Details of this experiment are described in Heisinger et al. (2002b) and Heisinger (1998). The stable nickel content of the sample was determined at LLNL. There, about 45 g of the sample were chemically prepared. The number of ^{63}Ni nuclei in the sample was measured by means of AMS at Munich (Rühm et al. 2000). Finally, a value of 0.928 was used for f_D^{Cu} (Eckhause et al. 1966), and the effective probability f^* for the production of ^{63}Ni after μ^- capture in the copper target calculated with equation (6):

$$f^{*} = \frac{N_{^{63}Ni}}{N_{\mu^{-}} \cdot f_{D}^{Cu}} \tag{6}$$

where

f^{*} : effective probability for production of ^{63}Ni in the copper target,

$N_{^{63}Ni}$: number of produced ^{63}Ni nuclei determined by means of AMS,

$N_{\mu^{-}}$: number of negative muons stopped in the copper target,

f_{D}^{Cu} : probability for capture in a copper nucleus.

Calculation of ^{63}Ni Production in Copper due to Photonuclear Reactions

In some cases, photonuclear reactions such as (γ,n) or (γ,np) might also contribute to the production of certain radionuclides. For the production of ^{63}Ni via the reaction ^{65}Cu(γ,np)^{63}Ni, the production cross section was taken from a webpage provided by the Moscow State University (Center for Photonuclear Experiments Data 2001), folded with the differential energy spectrum of photons Φ_{phot} at sea level, which decreases with increasing energy (Daniel and Stephens 1974; Ryan 1979), and which was parameterized using equation (7):

$$\Phi_{phot} \quad \left[cm^{2} s\ ster\ MeV\right]^{-1} = \begin{cases} 3.0 \cdot 10^{-3} \cdot E^{-1.1} & E \leq 20 MeV \\ 8.1 \cdot 10^{-2} \cdot E^{-2.2} & 20 MeV < E \leq 1 GeV \\ 2.6 \cdot E^{-2.7} & E > 1 GeV \end{cases} \tag{7}$$

Verification of the Applied Methodology to Calculate ^{63}Ni Production in Copper

By Use of Production due to Fast Muons. Fast muons interact with nuclei via exchange of virtual photons and produce electromagnetic and hadronic showers. The excited residual nuclei lose their energy by the emission of evaporation nucleons or gamma radiation. In this way, fast muons contribute to neutron and proton fluences discussed above. At sea level and high geomagnetic latitudes the production of cosmogenic radionuclides such as ^{10}Be, ^{14}C, ^{26}Al, ^{36}Cl, and ^{53}Mn, in target elements such as oxygen, silicon, sulfur, potassium, calcium and iron, due to the hadronic component of the cosmic radiation was shown to be 40-80 times larger than that due to fast muons (Heisinger et al. 2002a). Therefore, the estimation of the ^{63}Ni production in copper due to fast muons offers an indirect possibility to estimate the ^{63}Ni production in copper due to neutrons and protons.

In general, the production rate $P_{\mu,fast}$ of any radionuclide by fast muons can be described by equation (8) (Heisinger et al. 2002a):

$$P_{\mu,fast} = \sigma_{0} \cdot \beta \cdot \Phi_{\mu,fast}(d) \cdot \overline{E}_{\mu,fast}^{\alpha}(d) \cdot N_{T} \tag{8}$$

where

σ_0 : effective cross section [b],

β : average factor,

$\Phi_{\mu,fast}(d)$: muon flux [1/(cm^2·s)] as a function of depth d in the lithosphere,

$\overline{E}_{\mu,fast}^{\alpha}(d)$: total mean muon energy [GeV],

α : 0.75,

N_T : number of target nuclei [1/g].

At sea level, $\beta \cdot \Phi_{\mu, fast}(0) \cdot \overline{E}^{\alpha}_{\mu, fast}(0)$ is approximately $2.54 \cdot 10^6$ /(cm$^2 \cdot$y) (Heisinger et al. 2002a).

The dependence of the cross section on energy can be parameterized as follows:

$$\sigma(E) = \sigma_0 \cdot E^{\alpha} \tag{9}$$

Experimental data on σ(E) were not available for the production of ^{63}Ni in copper by fast muons. The relevant cross section was therefore determined experimentally. For this purpose, a high-purity copper target (mass: about 288 g; thickness: 5 mm; area: 64 cm^2) that had been exposed previously to 100 GeV positive muon beams at the NA54 experimental setup at CERN was investigated (Heisinger et al. 2002a). The target had been irradiated by $1.533 \cdot 10^{12}$ muons behind about 3 m of concrete blocks to generate muon-induced showers. The lateral extent of the showers had been measured with a laterally movable scintillator and with Fe, Ni and Cu monitor disks. The induced radioactivities of those disks were determined off-line by gamma spectroscopy. These profiles were used to calculate the shower fluxes seen by the copper target. Details of this experiment are given in (Heisinger 1998; Heisinger et al. 2002a). The determination of the stable nickel content of the copper sample and its chemical preparation was performed at LLNL. The number of ^{63}Ni nuclei in the samples was measured by means of AMS at Munich (Rühm et al. 2000). With the measured cross-section we estimated the production of ^{63}Ni in copper due to fast muons (equation 8) at sea level, scaled it by factors 40-80 (see above) to calculate the production due to hadrons, and compared the result to that obtained directly from the neutron and proton fluences discussed above.

By Use of Data Available for Other Radionuclides. The methodology discussed in the previous sections was also used to estimate the production of ^{32}P in sulfur and of ^{39}Ar in granite by cosmic radiation, and the results were compared with data from the literature. Both radionuclides are of interest in the reconstruction of fast neutron spectra in Hiroshima and Nagasaki. ^{32}P was measured in Hiroshima immediately after the explosion in porcelain insulators that contained sulfur as an adhesive (Arakatsu 1953; Sugimoto 1953; Yamasaki and Sugimoto 1953). ^{39}Ar has recently been proposed as an additional monitor for fast neutrons originating from the A-bombs (Rühm et al. 1998).

Results and Discussion

Production of ^{32}P in Sulfur

Mabuchi et al. (1971) measured ^{32}P in sulfur exposed to cosmic radiation at altitudes from sea level up to 3,720 m at Mt. Fuji, Japan (geomagnetic latitude: 25° N). At sea level they obtained a total production rate of 0.18 ^{32}P per minute and kilogram sulfur and estimated that 0.04 ^{32}P (per min·kg sulfur) was due to muons and 0.14 ^{32}P (per min·kg sulfur) due to neutrons. At 3,720 m above sea level they obtained a total value of 1.83 ^{32}P (per min·kg sulfur).

The basis for our estimate of the production of ^{32}P in sulfur is the neutron spectrum shown in Figure 2. The cross section for the reaction ^{32}S(n,p)^{32}P was taken from ENDF/B-VI. For energies above 14.5 MeV, the cross section was calculated by means of the CEM95 code. The same procedure was used for some other relevant reactions such as ^{33}S(n,pn)^{32}P, ^{34}S(n,p2n)^{32}P and

^{36}S(n,p4n)^{32}P (see Figure 4). Correction factors for the different geomagnetic latitudes (48.1° N (Hampton) compared to 25° N (Mt. Fuji)) obtained are 0.36 (approach a, using Table 1) and 0.76 (approach b, using Table 2), respectively. Finally, an additional correction factor of 1.03 was used to account for different solar activity in 1970 and in 1999, when the ^{32}P measurements and the neutron spectrum measurements were made, respectively.

Due to the high isotopic abundance of ^{32}S, the (n,p) reaction is dominant for the production of ^{32}P in sulfur. Other reactions such as ^{33}S(n,pn)^{32}P, ^{34}S(n,p2n)^{32}P and ^{36}S(n,p4n)^{32}P are of minor importance. For sea level, the neutron-induced production rates of 0.18 and 0.39 ^{32}P 1/(min·kg sulfur) deduced here are somewhat higher than the measured value (see Table 3). For an altitude of 3,720 m, the resulting production rates are 2.9 and 4.9 ^{32}P 1/(min·kg sulfur), respectively. Interestingly in their paper, Mabuchi et al. already noticed that their experimental data were a factor 2 lower compared to calculated values, which has also been observed here.

In order to estimate the production of ^{32}P in sulfur due to stopped muons, equation 5 is applied. A stopping rate of negative muons at sea level of 190 μ^- per gram and year is used (Heisinger et al. 2002b). The chemical compound factor f_C is equal to 1, since sulfur is an element, and the probability f_D for capture in sulfur is 0.747 (Eckhause et al. 1966). Concerning the effective probability f^* for the production of ^{32}P, no experimental data are available. However, data of Heisinger, who measured f^* for a variety of (n,p) reactions, would indicate values between 10% und 25% (Heisinger 1998). Therefore, we used a mean value of 17%. With these numbers and equation 5, a value of 0.05 ^{32}P nuclei per minute and kilogram sulfur is estimated, which is in agreement with the measured value (0.04 ± 0.02) of ^{32}P nuclei per minute and kilogram sulfur given by Mabuchi et al. (1971) (Table 3).

As for Hiroshima and Nagasaki, it is evident from Table 3 that production of ^{32}P by cosmic rays is negligible compared to that by A-bomb neutrons at relevant ground ranges (see Chapter 9, Part A).

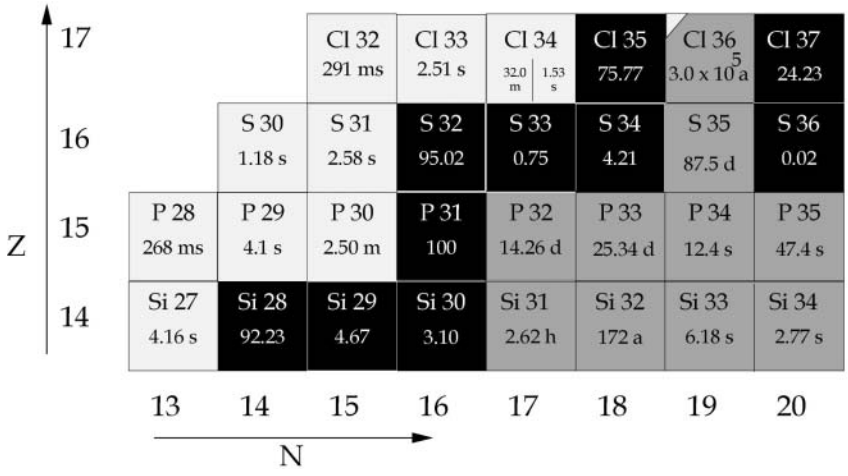


Figure 4. *Part of the Chart of the Nuclides (Pfennig et al. 1995) around* ^{32}*S; black: stable nuclide and isotopic abundance; dark gray:* β^- *decay, and half-life; light gray:* β^+*-decay or electron capture, and half-life.*

Table 3. Estimated production rates of ^{32}P [1/(min · kg sulfur)] produced by neutrons and muons from cosmic radiation (Asia, 25° N geomagnetic latitude, 1970) and compared to experimental data (Mabuchi et al. 1971)

		Estimate[a]	Estimate[b]	Measurement
sea level	^{32}S(n,p)^{32}P	1.8×10^{-1}	3.8×10^{-1}	
	^{33}S(n,pn)^{32}P	9.3×10^{-4}	2.0×10^{-3}	
	^{34}S(n,p2n)^{32}P	3.0×10^{-3}	6.4×10^{-3}	
	^{36}S(n,p4n)^{32}P	7.8×10^{-6}	1.7×10^{-5}	
	Sum hadrons	0.18	0.39	0.14 ± 0.04
	Stopped muons	0.05	0.05	0.04 ± 0.02
3720 m	Hadrons + muons	2.9	4.9	1.83 ± 0.08

[a] Latitude correction according to Table 1 and altitude correction according to equation 2.

[b] Latitude and altitude correction according to Table 2.

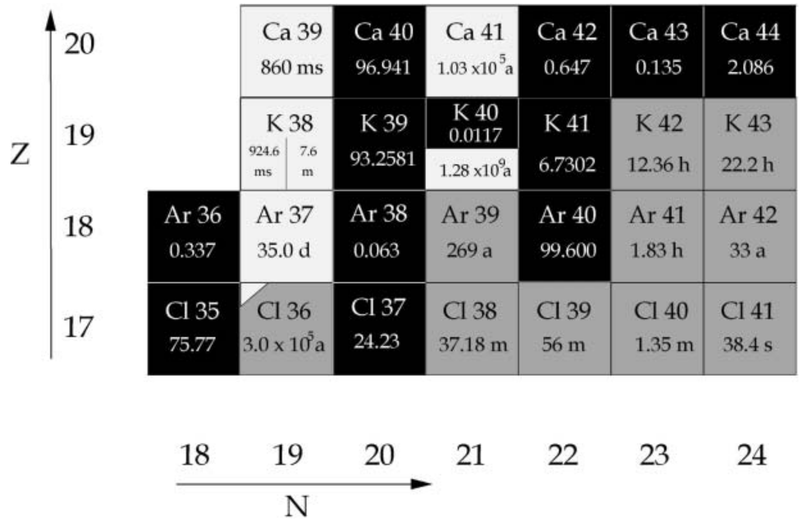
Production of ^{39}Ar in Granite

Yokoyama et al. (1977) investigated the production of radionuclides by cosmic radiation. For ^{39}Ar in granite, they calculated a production rate of 0.40 ^{39}Ar nuclei per minute and kilogram granite at a height of 3,840 m above sea level (Europe, geomagnetic latitude 47.4° N). According to those authors, the contribution of muons to the production of ^{39}Ar in granite is less than 10%.

In our study we assumed that the production of ^{39}Ar in granite is mainly due to spallation reactions on potassium and calcium (Figure 5). The cross section for the ^{39}K(n,p)^{39}Ar reaction was taken from ENDF/B-VI for energies below 14.5 MeV, and calculated with the CEM95 code for energies above 14.5 MeV. Other cross sections for reactions such as ^{41}K(n,p2n)^{39}Ar, ^{41}K(p,2pn)^{39}Ar and ^{40}Ca(n,2p)^{39}Ar were also calculated using the CEM95 code. We used the neutron spectrum at sea level shown in Figure 2, and in the case of the proton-induced reaction on ^{41}K, the proton/neutron ratio given by Lal and Peters (1967). Correction for geomagnetic latitude and altitude was performed according to Table 1 and equation (2) (approach a), or to Table 2 (approach b). Table 4 summarizes our results compared to those given by Yokoyama et al., for granite containing 3.5% K_2O and 0.5% CaO. The resulting values for the production of ^{39}Ar in granite due to neutrons and protons are 0.20 and 0.23 ^{39}Ar nuclei per minute and kilogram granite, respectively, depending on the different latitude and altitude corrections used (Table 4).

For the production of ^{39}Ar due to stopped muons, the chemical compound factor f_C is required, which describes the probability that a stopped muon is captured by potassium. Again, no experimental data are available. A rough estimate for f_C for element n in a matrix that is comprised of i elements can be calculated according to equation (10), where a_i is the concentration of element i in the matrix, and Z_i the corresponding atomic number (Fermi and Teller 1947).

$$f_C = \frac{(a_n \cdot Z_n)}{\sum_i (a_i \cdot Z_i)} \qquad (10)$$



	18	19	20	21	22	23	24
20		Ca 39 860 ms	Ca 40 96.941	Ca 41 1.03×10^5 a	Ca 42 0.647	Ca 43 0.135	Ca 44 2.086
19		K 38 924.6 ms 7.6 m	K 39 93.2581	K 40 0.0117 1.28×10^9 a	K 41 6.7302	K 42 12.36 h	K 43 22.2 h
18	Ar 36 0.337	Ar 37 35.0 d	Ar 38 0.063	Ar 39 269 a	Ar 40 99.600	Ar 41 1.83 h	Ar 42 33 a
17	Cl 35 75.77	Cl 36 3.0×10^5 a	Cl 37 24.23	Cl 38 37.18 m	Cl 39 56 m	Cl 40 1.35 m	Cl 41 38.4 s

Figure 5. *Part of the Chart of the Nuclides (Pfennig et al. 1995) around ^{39}Ar; black: stable nuclide and isotopic abundance; dark gray: β^--decay, and half-life; light gray: β^+-decay or electron capture, and half-life.*

With the chemical composition of granite given in (Yokoyama et al. 1977), a value of 0.03 for f_C is obtained. The corresponding factor f_D is 0.812 (Eckhause et al. 1966), and f^* was assumed to be 17% (see above). Together with the stopping rate of negative muons of 190 μ^- per gram and year, a production rate of $1.5 \cdot 10^{-3}$ ^{39}Ar nuclei per minute and kg granite is obtained. This is well below the production rate due to neutrons and protons, and in agreement with the statement of Yokoyama and coworkers that the contribution of muons to the production of ^{39}Ar in granite is less than 10% (Yokoyama et al. 1977) (Table 4).

As for Hiroshima and Nagasaki, a production rate of $(5\text{-}10) \times 10^{-3}$ ^{39}Ar nuclei per kg granite and minute is estimated for granite samples such as gravestones exposed to cosmic radiation at the surface of the Earth for about 80 years. Similar acitivities should have been induced in Hiroshima by A-bomb neutrons at ground ranges of about 2,000 m.

To summarize, the values for the production of ^{32}P in sulfur and ^{39}Ar in granite obtained here are in reasonable agreement with those found in the literature. From this finding, we deduce that the neutron, proton, and muon fluxes used in this work, as well as the cross sections calculated with the CEM95 code, and the assumptions on the correction for different geomagnetic latitude and altitude are a useful basis to estimate the production of radionuclides in certain target materials. In particular, this might indicate that the method developed in this paper works also reasonably well to estimate the production of ^{63}Ni in copper due to cosmic radiation. It should be emphasized, however, that some essential experimental data on cross sections etc. are still missing, and that the accuracy of our estimates for the production of ^{63}Ni in copper is not expected to be better than about a factor of 2.

Table 4. Estimated production rates of ^{39}Ar [1/(min · kg granite)] produced by neutrons and muons from cosmic radiation (Europe, 47.7° N geomagnetic latitude, 3,840 m above sea level) compared to literature values (Yokoyama et al. 1977)

	Estimate[a]	Estimate[b]	Yokoyama et al. 1977
^{39}K(n,p)^{39}Ar	1.9×10^{-1}	2.3×10^{-1}	
^{41}K(n,p2n)^{39}Ar	3.7×10^{-3}	4.3×10^{-3}	
^{41}K(p,2pn)^{39}Ar	1.1×10^{-3}	1.4×10^{-3}	
^{40}Ca(n,2p)^{39}Ar	6.7×10^{-4}	7.9×10^{-4}	
Sum hadrons	0.20	0.23	0.4
Stopped muons	0.0015	0.0015	< 0.04

[a] Latitude correction according to Table 1 and altitude correction according to equation 2.

[b] Latitude and altitude correction according to Table 2.

Production of ^{63}Ni in Copper

Production due to Neutrons and Protons. Cross sections of the reactions ^{63}Cu(n,p)^{63}Ni, ^{65}Cu(n,p2n)^{63}Ni, and ^{65}Cu(p,2pn)^{63}Ni, respectively, were generated using the CEM95 code, for energies between 1 MeV and 221 MeV. However, for energies below about 15 MeV, the corresponding ENDF/B-VI cross-section data were used. The production of ^{63}Ni on stable nickel and zinc, which are found in copper as impurities at the ppm level, via the ^{62}Ni(n,γ)^{63}Ni, ^{64}Ni(n,2n)^{63}Ni and ^{66}Zn(n,α)^{63}Ni reactions were calculated using ENDF/B-VI cross sections.

Table 5 lists all relevant reactions together with the corresponding contributions to the production of ^{63}Ni in copper. All results are given as saturation values in terms of ^{63}Ni nuclei per gram copper, for Hampton, USA (48.1° geomagnetic latitude, 1999, sea level). As expected, the production branch via the ^{63}Cu(n,p)^{63}Ni reaction is the most important. The ^{63}Ni production due to impurities such as nickel and zinc are negligible if their concentrations are not much above the assumed values. In total, about 6,300 ^{63}Ni nuclei are produced per gram copper at saturation, due to neutrons and protons from cosmic radiation.

In order to estimate the production of ^{63}Ni at other geomagnetic latitude locations, data from Table 5 must be corrected using values from Tables 1 or 2. Corrections for different altitudes can be obtained using equation (2). The influence of the solar cycle on the production of ^{63}Ni is small, but can be estimated from Figure 3.

As already noted, the production of ^{63}Ni in copper due to the hadronic component of cosmic radiation might also be estimated from data on fast muons. By means of AMS, we have measured the ^{63}Ni /Ni ratio produced in the copper target, which had been irradiated by 100 GeV muons (see above), and obtained a value of $(3.9 \pm 2.0)\cdot 10^{-13}$. This corresponds to a number of $5.1\cdot 10^{6}$ ^{63}Ni nuclei in the target. Thus, a cross section of (0.64 ± 0.33) mb at 100 GeV is obtained, which in turn corresponds to a σ_0 of 20 μb (equation 9). Finally, a number of (70 ± 35) ^{63}Ni nuclei per gram copper is obtained in saturation, for the production due to fast muons at sea level and high geomagnetic latitudes. Consequently, multiplication of this number by factors 40 to 80 (see above) provides estimated 2,800-5,600 ^{63}Ni nuclei per gram copper to be produced in saturation, due to the hadronic component of cosmic radiation, again at sea level and high geomagnetic latitudes. For comparison, the value obtained for Hampton, USA, of 6,300 ^{63}Ni nuclei per gram copper (Table 5) (at a geomagnetic latitude of 48.1°) corresponds, for those high geomagnetic latitudes, to values of 6,200 or 7,200 ^{63}Ni nuclei per gram copper, if corrections factors given in Tables 1 or 2, respectively, are used.

Table 5. Saturation concentrations of ^{63}Ni per gram copper, due to neutrons and protons from cosmic radiation (for Hampton, 48.1° N geomagnetic latitude, 1999, sea level)

Reaction	Isotope	Element impurity [ppm]	Isotopic abundance [%]	Production [^{63}Ni/g Cu]
(n,p)	^{63}Cu	–	69.16	4410
(n,γ)	^{62}Ni	38	3.59	1.6
(n,2n)	^{64}Ni	38	0.91	0.0017
(n,α)	^{66}Zn	5	27.9	0.0009
(n,p2n)	^{65}Cu	–	30.83	1720
(p,2pn)	^{65}Cu	–	30.83	164

Production due to Stopped Muons. As already indicated, no experimental data on the effective probability f^* for the production of ^{63}Ni in copper were available. Therefore, we measured, by means of AMS, the ^{63}Ni/Ni ratio in the copper target, which had been irradiated at the PSI facility, Switzerland, by stopped muons (see above), and a value of $^{63}\mathrm{Ni/Ni} = (3.1 \pm 0.3)\cdot 10^{-12}$ was obtained. This corresponds to a value for the effective probability f^* of (12.6 ± 1.6)%.

In order to estimate the production of ^{63}Ni in copper due to stopped muons at sea level, a stopping rate for negative muons of 190 μ^- per gram and year is used (Heisinger et al. 2002b). The dependence of the muon flux on geomagnetic latitude and altitude is small (e.g. Heisinger 1998) and has therefore been neglected in this paper. The chemical compound factor f_C is equal to 1, since copper is an element, and the probability f_D for capture in copper is 0.928 (Eckhause et al. 1966). Using these numbers and equation (5), we deduce a saturation concentration of 3,200 ^{63}Ni nuclei per gram copper produced by stopped muons at sea level. It turns out that, compared to the production due to neutrons and protons (Table 5), the production due to stopped muons must not be ignored.

Production due to Photons. It is obvious from Figure 1 that ^{63}Ni can also be produced in copper via the ^{65}Cu(γ,np)^{63}Ni reaction. The energy threshold for this reaction is 17.1 MeV. The evaluated energy dependence of the cross section is gaussian with a maximum of 10.6 mb at an energy of 21.2 MeV (Center for Photonuclear Experiments Data 2001). Folding in the photon spectrum (equation 7) and the production cross section results in a saturation concentration at sea level of 230 ^{63}Ni nuclei per gram copper.

Conclusions

We estimate that, for Hampton, USA, at sea level, as many as about $1 \cdot 10^4$ ^{63}Ni nuclei per gram copper (6,300 ^{63}Ni nuclei per gram copper due to the hadronic component of cosmic radiation, 3,200 ^{63}Ni nuclei per gram copper due to stopped muons, and about 200 ^{63}Ni nuclei per gram copper due to photonuclear reactions) are produced, if saturation is achieved.

If it is assumed that a piece of copper in Hiroshima was exposed to cosmic radiation for about 80 years (which is an age typical for potential rain gutters and lightning rods from Hiroshima, see Chapter 9, Part B), a concentration of $4\text{-}5 \cdot 10^3$ ^{63}Ni nuclei per gram copper can be expected. Based on neutron fluences given in Chapter 3 of this report, a similar concentration due to A-bomb neutrons would be expected, free-in-air and 1 m above ground, at a distance from the epicenter of about 1,800 m.

An experimental corroboration of our estimates would require, for example, measurement of the number of ^{63}Ni nuclei in copper samples exposed to cosmic radiation at different altitudes above sea level. Such work is presently in progress.

At the present stage of investigation we conclude that the ^{63}Ni background measured in distant copper samples from Hiroshima (Chapter 9, Part B) cannot be explained by cosmic-ray induced processes alone.

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