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Cosmic-ray–induced ^{63}Ni —A potential confounder of fast-neutron–induced ^{63}Ni in copper samples from Hiroshima

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Abstract. Recently, the determination of ^{63}Ni in copper samples has been suggested as a means to assess fast-neutron fluences in Hiroshima and Nagasaki. In those samples, ^{63}Ni (half-life: 100.07 years) was produced by MeV neutrons from the A-bomb explosions via the reaction ^{63}Cu(n,p)^{63}Ni. For large distances from the hypocenters, cosmic-ray–induced production of ^{63}Ni might also be important and, therefore, it is calculated here. The effective probability f^* which is required to quantify the cosmic-ray–induced production by stopped muons, was measured, and a value of $(12.6 \pm 1.6)\%$ obtained. The cross-section for the cosmic-ray–induced production by fast muons was measured to be (0.64 ± 0.33) mb, at a muon energy of 100 GeV. To validate the proposed method, cosmic-ray–induced production of ^{32}P in sulfur and of ^{39}Ar in granite was also calculated, and reasonable agreement with literature values was found. Our estimates indicate that as many as $(4\text{-}5) \cdot 10^3$ ^{63}Ni nuclei per gram copper were produced in a sample that was exposed to cosmic radiation in Hiroshima for about 80 years. A similar concentration due to A-bomb neutrons would be expected in Hiroshima at a distance from the hypocenter of about 1900 m.

PACS. 28.20.Fc Neutron absorption – 96.40.Vw Cosmic-ray effects in meteorites and terrestrial matter – 82.80.Ms Mass spectrometry (including SIMS, multiphoton ionization and resonance ionization mass spectrometry, MALDI)

1 Introduction

Recently, the determination of ^{63}Ni in pure-copper samples has been suggested as a means to assess fast-neutron fluences in Hiroshima and Nagasaki [1–3]. In those samples, ^{63}Ni (half-life: 100.07 years [4]) was produced by MeV neutrons originating from the A-bomb explosions via the reaction ^{63}Cu(n,p)^{63}Ni. Some years ago, the Technical University of Munich and the Lawrence Livermore National Laboratory (LLNL) initiated a collaboration to improve methods to detect ^{63}Ni in copper by means of accelerator mass spectrometry (AMS). The feasibility of AMS had already been demonstrated earlier [5,6]. The Munich Tandem Laboratory provides a unique combination of an MP tandem accelerator and an analyzing system, including a gas-filled magnet, for ultrasensitive detection of different long-lived radionuclides such as ^{63}Ni, ^{36}Cl, ^{53}Mn, ^{59}Ni and ^{60}Fe [7,8]. For ^{63}Ni, current background levels correspond to a ratio of $^{63}\mathrm{Ni/Ni} < 2 \cdot 10^{-14}$ and suggest that, with adequate copper samples, the assessment of fast-neutron fluences in Hiroshima and Nagasaki is possible for ground distances from the hypocenters ranging to about 1500 m [9–11].

The low ^{63}Ni activities induced in large-distant copper samples due to neutrons from the A-bomb explosion require, however, a careful discussion of the fraction of ^{63}Ni produced in these samples due to cosmic radiation. In general, 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. It is assumed 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.

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2 Materials and methods

2.1 Experimental procedures to assess production of ^{63}Ni in copper due to muon-induced reactions

2.1.1 Stopped muons

Slow negative muons captured by the Coulomb potential of atoms reach, within about 10^{-11} s, the muonic $1s$ shell [12]. The fraction f_D of the muons in the $1s$ shell, which do not decay via $\mu^- \rightarrow e^- + \nu_\mu + \bar{\nu}_e$ ($\tau = 2.2\,\mu$s), is finally captured by the nucleus and reacts with a proton to form a neutron:

$$\mu^- + {}^A_N Z \rightarrow {}^A_{N+1}(Z-1)^* + \nu_\mu \,. \tag{1}$$

For copper, f_D^{Cu}has a value of 0.928 [13]. The resulting excited nucleus loses its energy by emission of photons, neutrons, protons, etc. The probability that a certain nuclide is produced from the excited nucleus is described by the effective probability f^*. In the case of ^{63}Ni being produced from excited copper nuclei, f^* includes the relevant isotopic abundances for ^{63}Cu and ^{65}Cu and the corresponding branching ratios for the emission of the required particles (*i.e.* emission of no nucleon to produce ^{63}Ni from ^{63}Cu, or emission of two neutrons to produce ^{63}Ni from ^{65}Cu).

The effective probability f^* was not known for the reactions investigated in this paper and, due to its potential significance for A-bomb survivor dosimetry, was determined experimentally. For this purpose, a high-purity copper target was used which had been irradiated previously with low-energy negative muons at the μE4 beam line of the Paul Scherrer Institute in Villigen, Switzerland. To quantify the number of negative muons stopped in the copper sample, the resulting muonic X-rays were measured in-beam with a coaxial high-purity germanium detector. Detector efficiency was determined by means of standard nuclides such as ^{152}Eu. The required transition probability p_γ to the muonic K-shell was taken from [14]. Finally, for the number of negative muons stopped in the sample, a value of $N_{\mu^-} = (2.32 \pm 0.11) \cdot 10^{10}$ was obtained. After irradiation, a variety of short-lived manganese, iron, cobalt, and nickel radioisotopes were identified off-line in the sample, by means of a lead-shielded coaxial high-purity germanium detector. Details of this experiment which did not include the determination of ^{63}Ni in the copper target, are described in refs. [15] and [16].

The number of ^{63}Ni nuclei produced in the copper sample was determined by means of AMS. The sample was sent to the LLNL, where the stable nickel content was determined by means of graphite furnace atomic absorption spectrometry. Then, the sample (mass: 44.69 g) was chemically prepared using a two-step dedicated procedure which includes a) an electrochemical separation step and b) the formation of volatile nickel carbonyl [5,6]. Both methods allow the effective separation of ^{63}Ni from stable ^{63}Cu which represents a major isorbaric interference in the following AMS measurement. The Munich AMS setup used features, on the low-energy side, a dedicated Cs sputter source for the production of negative ions [10], a 90 ° magnetic dipole, and an 18 ° electrostatic deflector. The negative ions are injected into the 14 MV tandem accelerator. At the terminal the ions pass a carbon foil, electrons are stripped off, and the now positively charged ions are further accelerated. After the tandem accelerator, the following high-energy spectrometer includes a Wien velocity filter, a double focusing analyzing magnet, a time-of-flight detector, a gas-filled magnet, and a Frisch-grid ionization chamber [7,8]. The ^{63}Ni/Ni ratio in a sample of interest is calculated from the ^{63}Ni events in the detector and the stable nickel ions injected into the system, which were measured by means of a Faraday cup (eq. (2)). The relative uncertainty associated with this measurement was assumed to be 10%. For comparison, standards with known ^{63}Ni/Ni ratios are also measured, to determine the transmission through the whole system:

$$\frac{N_{^{63}\mathrm{Ni}}}{N_{\mathrm{Ni}}} = \frac{1.6 \cdot 10^{-10} \cdot Z}{\overline{I} \cdot \overline{T} \cdot t}\,, \tag{2}$$

where

Z: number of ^{63}Ni events in the detector, corrected for background events due to ^{63}Cu,
$\overline{I}$: mean stable nickel current measured at the entrance of the accelerator (nA),
$\overline{T}$: mean transmission through the system during measurement,
t: measuring time (s).

In order to correct for machine background and potential crosstalk, etc., blank samples which do not contain any ^{63}Ni are used. The final uncertainties of the ^{63}Ni/Ni ratios are obtained by standard error propagation. For small signal statistics ($Z < 20$), the approach outlined by [17] is used. The experimental setup and the measurement procedure is described in detail in [9].

The effective probability f^* for the production of ^{63}Ni after μ^- capture in the irradiated copper target was finally calculated as

$$f^* = \frac{N_{^{63}\mathrm{Ni}}}{N_{\mu^-} \cdot f_D^{\mathrm{Cu}}}\,, \tag{3}$$

where

f^*: effective probability for the production of ^{63}Ni in the copper target,
$N_{^{63}\mathrm{Ni}}$: number of produced ^{63}Ni nuclei determined by means of AMS,
N_{μ^-}: number of negative muons stopped in the copper target,
f_D^{Cu}: probability for capture in a copper nucleus.

2.1.2 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, proton and photon fluences discussed below. Therefore, data on the production of ^{63}Ni in copper due to fast muons from cosmic radiation can be used to test the calculated production of ^{63}Ni in copper due to neutrons and protons from cosmic radiation.

Since the production cross-section of ^{63}Ni in copper due to fast muons was not known, it was determined experimentally. For this purpose, a high-purity copper target (target mass: 288.22 g; target area: $64\,\text{cm}^2$), which had been exposed previously to 100 GeV positive muon beams at the NA54 experimental setup at CERN, was investigated. As described in detail in [16,18], the muon fluxes were measured using a hodoscope consisting of a scintillation counter and an ionization chamber. The copper target was irradiated by muons behind about 3 m of concrete blocks to generate muon-induced showers, and the lateral extent of the showers was measured with a movable scintillator. Iron, nickel and copper monitor disks were also used, in which the produced radionuclides were determined off-line by γ-spectroscopy. From the resulting beam profiles, the shower fluxes seen by the copper sample were deduced. As a result, it was calculated that the copper target had been irradiated by $1.84 \cdot 10^{11}$ muons.

In the copper target, a variety of short-lived radionuclides such as manganese, iron, and cobalt isotopes, were produced and had already been measured by means of off-line γ-spectroscopy. However, no attempt had been made to detect ^{63}Ni in this sample. Therefore, the stable nickel content of the copper sample was determined and its chemical preparation was performed (see sect. 2.1.1). The number of ^{63}Ni nuclei in the samples was again measured by means of AMS at Munich in an analogous way as described in sect. 2.1.1.

2.2 Theoretical procedures to calculate cosmic-ray–induced ^{63}Ni production in copper

2.2.1 Production due to neutrons and protons

Neutron flux

At mean geomagnetic latitudes and sea level, the total neutron flux is about $10^{-2}\,\text{cm}^{-2}\text{s}^{-1}$. The following estimates are based on a neutron spectrum 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 [19,20]. In the lethargy representation this neutron spectrum exhibits three peaks. The first peak 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 about 100–300 MeV [21].

The use of this neutron spectrum for other geographic locations and years requires corrections for the 11-year solar cycle, altitude and different geomagnetic latitude. The influence of the solar activity on the neutron flux at sea level is rather small. This has been demonstrated by means of the so-called neutron monitors during measurement programs which have continued for more than 40 years. In this study, any correction for the solar cycle is based on the neutron monitor data given in [22]. If the neutron spectrum used was applied to locations which were not at sea level, altitude corrections were applied following [21,23]. Different approaches are used to correct for different geomagnetic latitudes. Yokoyama *et al.* [24] published correction factors for the production of cosmogenic radionuclides which are independent of the altitude, but dependent on geomagnetic and geographic latitude. They based their values on calculations, and normalized them to a cut-off rigidity of 4.7 GV. Lal, on the other hand, deduced relative correction factors which depend on geomagnetic latitude and altitude [25]. His values are based on measurements of nuclear disintegration rates in the atmosphere. 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 [26] and folded with the neutron spectrum from [19,20].

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 [27], which is based on the Cascade Exciton Model [27,28], 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 [29]. At an energy of about 15 MeV, the ENDF/B-VI cross-sections [30] matched reasonably well with 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.

2.2.2 Production due to stopped muons

Negative muons produced by the interaction of high-energy protons with nuclei in the atmosphere are slowed down in matter, 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}. A fraction f_D of the muons in the 1*s* shell is finally captured by the nucleus and reacts with a proton to form a neutron (eq. (1)). The resulting excited nucleus loses its energy by emission of photons, neutrons, protons, etc., and finally forms, with the effective probability f^*, a certain nuclide (see sect. 2.1.1).

Table 1. AMS results obtained for the copper targets irradiated by fast muons at the CERN facility, and by stopped muons at the PSI facility; for details see text.

	$\overline{I} \cdot \overline{T} \cdot t$ (nA s)	$Z_{^{63}\mathrm{Ni}}$	Z_{BG}	^{63}Ni/Ni
Stopped muons (PSI)	7750	152	3.7	$(3.1 \pm 0.3) \cdot 10^{-12}$
Fast muons (CERN)	4560	19	8.0	$(3.9 \pm 2.0) \cdot 10^{-13}$

To summarize, the production rate P_{μ^-} of a certain nuclide by capture of stopped muons at sea level can be calculated according to

$$P_{\mu^-} = \theta_{\mu^-} \cdot f_{\mathrm{C}} \cdot f_D \cdot f^* , \tag{4}$$

where

P_{μ^-}: production rate of the radionuclide of interest at sea level,
θ_{μ^-}: total stopping rate of negative muons at sea level,
f_{C}: chemical compound factor,
f_D: probability for capture in the nucleus,
f^*: effective probability for the production of a certain nuclide.

In this paper, a rate of stopped negative muons of 190 μ^- per gram and year is used [15], for the estimation of the number of any radionuclide produced due to stopped muons at sea level. The dependence of the muon flux on geomagnetic latitude and altitude is small (see, *e.g.*, [16]) and has therefore been neglected in this paper. For the production of ^{63}Ni in copper, f_{C} was taken to be 1 since copper is an element. For copper, f_D^{Cu}has a value of 0.928 [13], and f^* was determined in this work.

2.2.3 Production due to photons

To estimate 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 [31], and folded with the differential energy spectrum of photons Φ_{phot} at sea level [32,33].

2.2.4 Verification of the applied methodology to calculate cosmic-ray–induced ^{63}Ni production in copper

By use of data available for other radionuclides

The methodology described above 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 [34–36]. ^{39}Ar has recently been proposed as an additional monitor for fast neutrons originating from the A-bombs [37].

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 the neutron, proton and photon 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 obtained in samples irradiated with fast muons [15]. 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.

With the experiment described in sect. 2.1.2, we measured the cross-section for the production of ^{63}Ni in copper due to fast muons with an energy of 100 GeV. Following [18], it is assumed that the mean energy of fast muons from cosmic radiation at sea level is 7.6 GeV, and that the energy dependence of the corresponding cross-section can be described by $\sigma(E) = \sigma_0 \cdot E^{0.75}$. With this information, the production of ^{63}Ni in copper due to fast muons at sea level can be calculated. A detailed discussion is given in [18]. The resulting value is scaled by factors 40–80 (see above) to calculate the production due to hadrons, and the result is compared to that obtained directly from the neutron and proton fluences discussed above.

3 Results and discussion

3.1 Cosmic-ray–induced production of ^{63}Ni in copper

3.1.1 Production due to stopped muons

For the copper target which had been irradiated with stopped muons at the PSI facility, Switzerland, a stable-nickel content of 1.9 ± 0.2 ppm was determined. The total sample was electrochemically dissolved, and from the resulting solution of 849.3 g a fraction of 50.0 g further processed. To this fraction 5.00 mg of stable-nickel carrier was added, prior to the carbonyl chemistry. For the resulting sample, a value of ^{63}Ni/Ni $= (3.1 \pm 0.3) \cdot 10^{-12}$ was measured, by means of the Munich AMS setup (table 1). This corresponds to a number of $(2.7 \pm 0.3) \cdot 10^9$ ^{63}Ni nuclei in the total copper sample produced by stopped muons. Based on eq. (3), a value of (12.6 ± 1.6)% is finally obtained, for f^*. Together with eq. (4), a chemical compound factor f_{C} of 1 (since copper is an element), and the probability f_D for capture in copper of 0.928 [13], we deduce a saturation concentration of 3200 ^{63}Ni nuclei

Table 2. Saturation concentrations of ^{63}Ni per gram copper, due to neutrons, protons, stopped muons, and photons from cosmic radiation (for Hampton, 48.1 °N geomagnetic latitude, 1999, sea level).

Reaction	Isotope	Isotopic abundance (%)	Production (^{63}Ni/g Cu)
(n,p)	^{63}Cu	69.16	4410
(n,p2n)	^{65}Cu	30.83	1720
(p,2pn)	^{65}Cu	30.83	160
(Stopped muons)	^{65}Cu, ^{63}Cu	100	3200
(Photons)	^{65}Cu	30.83	230
Total			**9720**

Table 3. Estimated production rates of ^{32}P (min · kg sulfur)$^{-1}$ produced by neutrons and muons from cosmic radiation (Asia, 25 °N geomagnetic latitude, 1970) and compared to experimental data [38]; (a) latitude correction according to [24] and altitude correction according to [21,23]; (b) latitude and altitude corrections according to [25].

		Estimate (a)	Estimate (b)	Measurement [38]
Sea level	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

per gram copper produced by stopped muons at sea level (table 2).

3.1.2 Production due to neutrons and protons

The reactions investigated include ^{63}Cu(n,p)^{63}Ni, ^{65}Cu(n,p2n)^{63}Ni, and ^{65}Cu(p,2pn)^{63}Ni, respectively. As expected, the production branch via the ^{63}Cu(n,p)^{63}Ni reaction is the most important (table 2). The ^{63}Ni production on nickel and zinc, which can be found in copper as impurities, is negligible, unless their concentrations are orders of magnitudes above typical values. In total, about 6300 ^{63}Ni nuclei are produced per gram copper at saturation, for Hampton, USA.

3.1.3 Production due to photons

The energy threshold for the ^{65}Cu(γ,np)^{63}Ni 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 [31]. Folding the photon spectrum and the production cross-section results in a saturation concentration at sea level of 230 ^{63}Ni nuclei per gram copper (table 2).

3.2 Test of the method

3.2.1 Production of ^{32}P in sulfur

Mabuchi *et al.* measured ^{32}P in sulfur exposed to cosmic radiation at altitudes from sea level up to 3720 m at Mt. Fuji, Japan (geomagnetic latitude: 25 °N) [38]. Their results are shown in table 3.

For comparison, we have estimated the production of ^{32}P in sulfur including the reactions ^{32}S(n,p)^{32}P, ^{33}S(n,pn)^{32}P, ^{34}S(n,p2n)^{32}P and ^{36}S(n,p4n)^{32}P based on the method proposed here. Correction factors for the different geomagnetic latitudes (48.1 °N (Hampton) compared to 25 °N (Mt. Fuji)) obtained are 0.36 (estimate (a)) and 0.76 (estimate (b)), respectively. A correction factor of 1.03 was used to account for the 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. For sea level and an altitude of 3720 m, the neutron-induced production rates deduced here are somewhat higher than the measured value (see table 3). 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.

As for the production due to stopped muons, the chemical compound factor f_C was taken to be 1 since sulfur is an element, and a value for f_D of 0.747 was used [13]. Concerning the effective probability f^* for the production of ^{32}P, no experimental data were available. However, data of Heisinger who measured f^* for a variety of (n,p) reactions would indicate values between 10% und 25% [16]. Therefore, we used a mean value of 17%. With these numbers and eq. (2), a value of 0.05 ^{32}P (min · kg sulfur)$^{-1}$ is estimated which is in agreement with the measured (0.04 ± 0.02) ^{32}P (min · kg sulfur)$^{-1}$ [38] (table 3).

3.2.2 Production of ^{39}Ar in granite

Yokoyama *et al.* calculated the production of ^{39}Ar in granite by cosmic radiation [24], at an altitude of 3840 m

Table 4. Estimated production rates of ^{39}Ar (min · kg granite)$^{-1}$ produced by neutrons, protons, and muons from cosmic radiation (Europe, 47.7 °N geomagnetic latitude, 3840 m above sea level) compared to literature values [24]; (a) latitude correction according to [24] and altitude correction according to [21,23]; (b) latitude and altitude corrections according to [25].

	Estimate (a)	Estimate (b)	Estimate [24]
Hadrons	0.20	0.23	0.4
Stopped muons	0.0015	0.0015	< 0.04

(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% (table 4).

In our study we assumed that the production of ^{39}Ar in granite is mainly due to spallation reactions on potassium and calcium, and the reactions ^{39}K(n,p)^{39}Ar, ^{41}K(n,p2n)^{39}Ar, ^{41}K(p,2pn)^{39}Ar and ^{40}Ca(n,2p)^{39}Ar were considered. Table 4 summarizes our results compared to those given by Yokoyama *et al.*, for granite containing 3.5% K_2O and 0.5% CaO.

For the production of ^{39}Ar on potassium due to stopped muons, the chemical compound factor f_C was not yet determined experimentally, and had therefore to be estimated by means of eq. (5), below, for element n (potassium) in a matrix (granite) which is comprised of i elements, where a_i is the concentration of element i in the matrix, and Z_i the corresponding atomic number [39]:

$$f_C = \frac{(a_n \cdot Z_n)}{\sum_i (a_i \cdot Z_i)} . \qquad (5)$$

With the chemical composition of granite given in [24], a value of 0.03 for f_C is obtained. The corresponding factor f_D is 0.812 [13], and f^* was assumed to be 17% (see above). Finally, a production rate of $1.5 \cdot 10^{-3}$ ^{39}Ar (min · kg granite)$^{-1}$ is obtained, in agreement with the estimate of Yokoyama and coworkers [24] (table 4).

3.2.3 Production of ^{63}Ni due to fast muons

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 with 100 GeV muons (see sect. 2.1.2), and obtained a value of $(3.9 \pm 2.0) \cdot 10^{-13}$ (table 1). In this target, the content of stable nickel was determined to be 4.4 ± 0.4 ppm. Thus, a concentration of $(1.7 \pm 0.9) \cdot 10^4$ ^{63}Ni nuclei per gram copper was obtained which corresponds to a number of $(5.0 \pm 2.6) \cdot 10^6$ ^{63}Ni nuclei in the target. Thus, a cross-section of (0.64 ± 0.34) mb at 100 GeV could be calculated. Finally, we obtain in saturation, following a procedure given by [33] a number of (70 ± 35) ^{63}Ni nuclei per gram copper, for the production due to fast muons at sea level and high geomagnetic latitudes. Consequently, multiplication of this number by factors 40 to 80 [15] provides estimated 2800–5600 ^{63}Ni nuclei per gram copper to be produced in saturation, due to hadrons from cosmic radiation, again at sea level and high geomagnetic latitudes. For comparison, the value obtained, for example, for the hadronic component at Hampton, USA, of 6300 ^{63}Ni nuclei per gram copper (table 2) (at a geomagnetic latitude of 48.1 °) corresponds, for those high geomagnetic latitudes, to values of 6200 or 7200 ^{63}Ni nuclei per gram copper.

4 Conclusions

We estimate that, for Hampton, USA, at sea level, as many as about $1 \cdot 10^4$ ^{63}Ni nuclei per gram copper (6300 ^{63}Ni nuclei per gram copper due to the hadronic component of cosmic radiation, 3200 ^{63}Ni nuclei per gram copper due to stopped muons, and about 200 ^{63}Ni nuclei per gram copper due to photons) are produced, if saturation is achieved.

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 (tables 3 and 4). 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 are reliable, and that 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 kept in mind, however, that some experimental data on cross-sections are still missing, and therefore have to be calculated by means of the CEM95 code. As suggested by the results obtained on the production of ^{32}P and ^{39}Ar, we believe that the accuracy of our estimates for the production of ^{63}Ni in copper should not be expected to be better than about a factor of 2. This judgement is also supported by our estimate of the production of ^{63}Ni in copper based on fast muons from cosmic radiation.

For a piece of copper in Hiroshima exposed to cosmic radiation for about 80 years (which is typical for potential rain gutters and lightning rods from Hiroshima [40]), a concentration of 4-5 $\cdot 10^3$ ^{63}Ni nuclei per gram copper can be expected. A similar concentration due to A-bomb neutrons would be expected, free-in-air and 1 m above ground, at a distance from the hypocenter of about 1900 m [41].

An experimental corroboration of our estimates on cosmic-ray–induced ^{63}Ni in copper would require, for example, the measurement of the ^{63}Ni concentration in copper samples exposed to cosmic radiation at different altitudes. For example, the ^{63}Ni production is expected to be larger by about a factor 8 at an altitude of 3100 m, compared to sea level. The search for suitable copper samples is presently under way.

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