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In situ produced ^{10}Be measurements at great depths: implications for production rates by fast muons

R. Braucher [a,*], E.T. Brown [a,b], D.L. Bourlès [a], F. Colin [c]

[a] CEREGE UMR 6635, Plateau de l'Arbois, 13545 Aix en Provence, France

[b] Large Lake Observatory, University of Minnesota, 2205 East 5th Street, Duluth, MN 55812, USA

[c] IRD/UM GECO, CEREGE UMR 6635, Plateau de l'Arbois, 13545 Aix en Provence, France

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Abstract

In situ cosmogenic ^{10}Be values have been used to investigate a Brazilian quartz vein from the surface to a depth of 15 m. At depths greater than 1000 g/cm^2, deep enough for neutron-induced reactions to be insignificant, there is only a slight decrease in ^{10}Be concentration with increasing depth. Our results are consistent with deep production of ^{10}Be by a mechanism, presumably induced by fast muons, with an attenuation length of 5300 ± 950 g/cm^2 and a contribution of 0.65 ± 0.25% to the total surface production. Results are compared with values from the literature and implications of this re-evaluation are discussed.

Keywords: beryllium; cosmogenic; muons; production rate

1. Introduction

Accumulation of in situ produced cosmogenic nuclides allows quantification of denudation or burial of geomorphologic surfaces, as well as dating formation of landforms such as moraines, alluvial fans or terraces. To quantify erosion and exposure time using cosmogenic nuclides, it is necessary to know how their production varies with depth below the Earth's surface. Beryllium-10, one of the most commonly used cosmogenic radionuclides, is produced by three types of mechanisms in exposed surface rock: neutron-induced spallation, slow (or stopping) muon capture, and fast muon-induced reactions. Neutron-induced reactions produce the vast majority of ^{10}Be in surface rocks, but muons penetrate more deeply into the Earth's surface and dominate production at depths greater than several meters. There is a growing body of theoretical and experimental literature examining production of cosmogenic nuclides induced by muons [1–7]. In addition, field studies have demonstrated the potential importance of muon-induced reactions for a range of applications using ^{36}Cl [8–10] in addition to ^{10}Be [11,12].

These studies have led to semi-empirical math-

* Corresponding author. Tel.: +33-4-42-97-15-09; Fax: +33-4-42-97-15-40.

E-mail addresses: braucher@cerege.fr (R. Braucher), ebrown1@d.umn.edu (E.T. Brown), bourles@cerege.fr (D.L. Bourlès), colin@cerege.fr (F. Colin).

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Table 1
^{10}Be sample location and data

Sample	Sample location	Sampling depth (g/cm^2)	^{10}Be (10^4 at/g)	Error (1σ) (10^4 at/g)
BR96-15A	Site 2	0	149.00	43.72
BR96-15B	Site 2	85	107.01	7.42
BR96-15C	Site 2	136	64.30	7.08
BR96-15D	Site 2	300	32.03	3.52
BR96-15E	Site 2	900	8.04	0.923
BR96-13A	Itaipu mine	0	6.61	0.92
BR96-13-5	Itaipu mine	750	3.71	0.43
BR96-13-4	Itaipu mine	1000	2.47	0.35
BR96-13-3	Itaipu mine	1250	4.06	0.47
BR96-13-2	Itaipu mine	1500	3.46	0.48
BR96-13-1	Itaipu mine	1750	2.15	0.51
BR96-13G	Itaipu mine	2125	2.56	0.18
BR96-13F	Itaipu mine	2375	3.14	0.44
BR96-13E	Itaipu mine	2625	2.82	0.39
BR96-13D	Itaipu mine	2875	3.52	0.57
BR96-13C	Itaipu mine	3125	2.10	0.45
BR96-13B	Itaipu mine	3375	2.02	0.25
BR96-13I	Itaipu mine	3750	2.77	0.61

Site 2 refers to the unperturbed quartz vein (see text). Rock density is around 2.3 g/cm^3.

ematical descriptions of depth variability of ^{10}Be production. The earliest field measurements suggested that the contribution of muons was small, less than 3% of ^{10}Be production at the Earth's surface [11,12]. However, the profiles examined in these studies were not deep enough to differentiate between stopping and fast muons. Granger and Smith [5] proposed a ^{10}Be production calculation based on measured ^{36}Cl depth variability [9] coupled with production rates of ^{10}Be from neutron-induced (97.5% of surface production) and muon-induced (2% for muon capture and 0.5% for fast muon reactions) reactions [4]. Depth variability of the flux of stopping muons was described as a combination of exponentials with attenuation lengths of 786.6 and 2688 g/cm^2 whereas fast muons were considered to have a simple exponential attenuation length of 4360 g/cm^2. More recently, using results from irradiation experiments with 100 and 190 GeV muons, Heisinger et al. [4] proposed contributions of $\sim$2% in the total ^{10}Be production rate for each type of muon-induced reaction with attenuation lengths of 4320 g/cm^2 for fast muons and 1500 g/cm^2 for negative muons. For depths less than 6000 g/cm^2, this latter value does not greatly differ from the combined exponential of 786.6 and 2688 g/cm^2 presented by Granger and Smith [5]. However, the proportion of production induced by the two types of muon-induced reactions differs between the two studies and has not been directly determined in a natural system. To provide a field-based evaluation of the role of muons for in situ production of ^{10}Be, we collected samples in a depth profile (from the surface to 15 m) along a quartz vein embedded within saprolite.

2. Sample location and description

The city of Cuiaba (15°21′S, 56°03′W) is located on the part of Guapore cratonic block, the southern part of the Amazonian craton, that was cratonized during the early to middle Proterozoic [13]. Regional cover rocks are metasediments and granitoids (500–600 Ma [13]) cut by auriferous or non-auriferous subvertical quartz veins. The regional landscape consists of very gently sloping hills. The present climate is tropical humid.

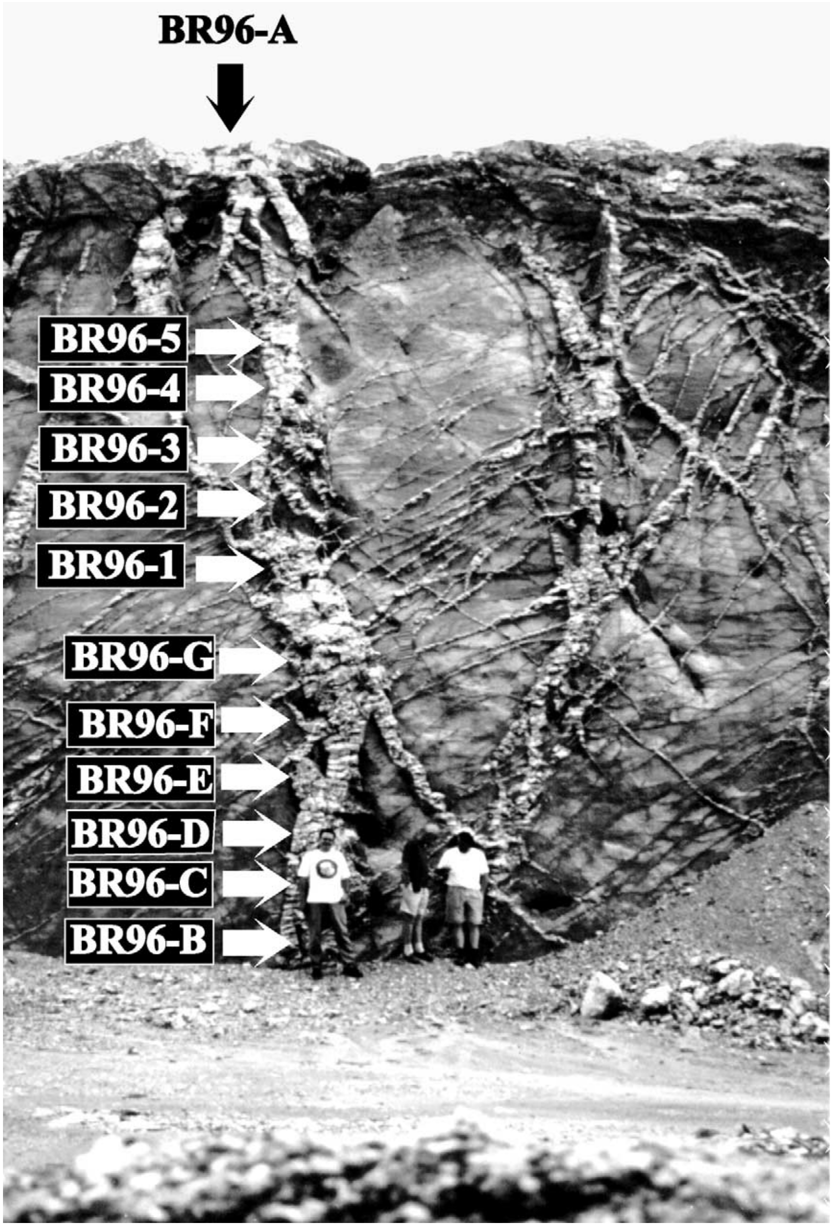

Fig. 1. Photo from the Itaipu quartz vein and sample location.

Two quartz veins were sampled. One is located at the Itaipu mine ($\sim$6 km northeast of the city) where a profile of 12 quartz samples was collected between the surface and a depth of 15 m (Table 1 and Fig. 1). Because the soil surface at this site appears to have been disturbed by mining activities, we also collected a five-point profile (to depths up to 5 m; Table 1) from an unperturbed quartz vein (Site 2) located 3 km north of the mine.

3. Analytical procedures

Quartz was isolated from crushed and sieved samples by dissolving all other minerals with mixtures of HCl and H_2SiF_6. Atmospheric ^{10}Be was then eliminated by successive HF sequential dissolutions [14]. This purified quartz was dissolved in Suprapur HF and the resulting solution was spiked with 0.3 mg of ^{9}Be carrier [14]. Beryllium was separated from these solutions by successive solvent extractions and precipitations [15,16]. All ^{10}Be measurements were performed with the AMS Tandétron facility at Gif-sur-Yvette, France [17,18]. ^{10}Be uncertainties were calculated by propagating a conservative estimate of 3% instrumental uncertainty with 1σ uncertainties associated with counting statistics, blank correction [17] and 15% in production rate estimates [19–21]. All ^{10}Be concentrations were calculated using the NIST standard with its certified ^{10}Be/^{9}Be of 2.68×10^{-11}.

Uranium and thorium were determined by ICP-MS at the LMTG (Toulouse) using aliquots of dilute nitric acid solutions containing the dissolved purified quartz.

4. Results

Data from Site 2 show a simple exponential decrease with depth in ^{10}Be concentrations (black triangles in Fig. 2). The erosion of 2.2 ± 0.3 m/Myr deduced from the profile ^{10}Be concentration is in agreement with a previous study in this area [22]. In contrast, the ^{10}Be surface concentration from the Itaipu mine is far lower, consistent with field observations that mining activities may have removed significant quantities of material. To estimate this soil loss, the profile of the mine samples was matched to that from the unperturbed quartz vein, yielding a depth correction of about 4 m (open triangles in Fig. 2).

At depths greater than 1000 g/cm^2, deep enough for neutron-induced reactions to be insignificant, there is only a slight decrease in ^{10}Be concentration with increasing depth. Low U and Th contents (Table 2) indicate that radiogenic ^{10}Be production is negligible, producing a steady-state concentration of only 21 at/g in sandstones [23]. It thus cannot be invoked as the cause of the relatively high ^{10}Be contents, suggesting that the deep production is indeed cosmogenic.

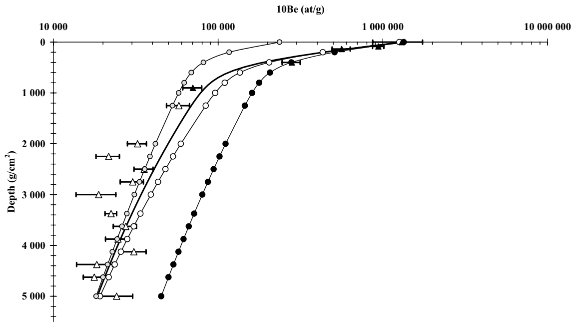

Fig. 2. Depth evolution of ^{10}Be concentrations for the Itaipu mine quartz vein (open triangles). Depths have been adjusted for soil loss during mining operations; for comparison, we include surface data (black triangles) from an unperturbed quartz vein located near the mine site. The solid line represents theoretical ^{10}Be concentrations obtained using a fast muon attenuation length of 5300 ± 950 g/cm^2 with a contribution of 0.65 ± 0.25% and an erosion rate of 2.2 m/Myr. Black and gray dotted lines represent the expected steady-state erosion depth profiles using Heisinger production parameters for erosion rates of 2.2 m/Myr and 16 m/Myr respectively. Open circles show the expected steady-state erosion depth profiles using the local erosion rate of 2.2 m/Myr and all the calculated Heisinger parameters apart from the fast muon contribution which has to be adjusted at 0.65%.

5. Discussion

Our results are not compatible with the production parameters proposed by either Granger or Heisinger. In particular, the measured ^{10}Be concentrations are substantially lower at depth than predicted by either calculation. There are three potential explanations for this discrepancy.

(a) The mine work induced removal of a substantial layer of soil and saprolite. To make our data fit with the depth variability expected from Heisinger's production calculations, this removal would have to be ca. 20 m. This is incompatible with field observations and information from mine operators.

(b) The physical parameters, in particular the proportions of total production attributed to

Table 2
Uranium and thorium concentrations

Sample	Th (ppb)	U (ppb)
BR96-13I	2.96	0.71
BR96-13D	1.72	0.53
BR96-13E	6.93	1.16
BR96-13G	0.48	0.19
BR96-13-3	7.71	2.64

each reaction, reported by Heisinger et al. are based on laboratory experiments for discrete muon energies and thus may not be directly applicable to the broader energy spectrum present in the natural environment.

To model our data set, we use the equation:

$$C(x;t) = \frac{P_o p_n}{\frac{\varepsilon}{\Lambda_n}+\lambda} e^{\left(-\frac{x}{\Lambda_n}\right)} \left[1 - e^{-t\left(\frac{\varepsilon}{\Lambda_n}+\lambda\right)}\right] + \frac{P_o p_{\mu s}}{\frac{\varepsilon}{\Lambda_{\mu s}}+\lambda} e^{\left(-\frac{x}{\Lambda_{\mu s}}\right)} \left[1 - e^{-t\left(\frac{\varepsilon}{\Lambda_{\mu s}}+\lambda\right)}\right] + \frac{P_o p_{\mu f}}{\frac{\varepsilon}{\Lambda_{\mu f}}+\lambda} e^{\left(-\frac{x}{\Lambda_{\mu f}}\right)} \left[1 - e^{-t\left(\frac{\varepsilon}{\Lambda_{\mu f}}+\lambda\right)}\right] + C_o e^{(-\lambda t)} \quad (1)$$

where Λ_n, $\Lambda_{\mu s}$, and $\Lambda_{\mu f}$ are effective apparent attenuation lengths (g/cm^2) for neutrons, slow muons, and fast muons, respectively, p_n, $p_{\mu s}$ and $p_{\mu f}$ the relative contributions of the three reactions to total ^{10}Be production ($p_n+p_{\mu s}+p_{\mu f}=100\%$), and C_o the number of atoms present at the initiation of exposure.

For depths greater than 3000 g/cm^2, production by neutrons and negative muons becomes negligible and Eq. 1 becomes:

$$C(x;t) = \frac{P_o p_{\mu f}}{\frac{\varepsilon}{\Lambda_{\mu f}}+\lambda} e^{\left(-\frac{x}{\Lambda_{\mu f}}\right)} \left[1 - e^{-t\left(\frac{\varepsilon}{\Lambda_{\mu f}}+\lambda\right)}\right] + C_o e^{(-\lambda t)} \quad (2)$$

If inherited ^{10}Be is negligible and if steady state is achieved, Eq. 2 can be rewritten as:

$$\ln[C(x;t)] = -\frac{x}{\Lambda_f} + \ln\left(\frac{P_o p_{\mu f}}{\frac{e}{\Lambda_{\mu f}}+\lambda}\right) \quad (3)$$

which is a straight line:

$$y = Ax + B \text{ with } A = \frac{-1}{\Lambda_{\mu f}} \text{ and } B = \ln\left(\frac{P_o p_{\mu f}}{\frac{e}{\Lambda_{\mu f}}+\lambda}\right)$$

A weighted least squares fit of experimental data from the Itaipu mine yields an apparent attenuation length of 5300 ± 950 g/cm^2 and a fast muon contribution of about 0.65 ± 0.25% (Fig. 2). This attenuation length is comparable to the value for fast muons reported by Heisinger [6,7] (4320 g/cm^2), but the contribution by fast muons appears to be a factor of 3–4 lower than the 2% proposed by Heisinger.

The theoretical ^{10}Be concentration–depth profile obtained using the attenuation lengths proposed by Heisinger and an erosion rate of 2.2 m/Myr does not fit the experimental data for the samples below 300 g/cm^2 (Fig. 2). To fit these experimental data using the calculated Heisinger production parameters, the mean erosion rate must be forced to 13 ± 3 m/Myr. This is incompatible with the ^{10}Be concentrations measured in the samples collected from 900 g/cm^2 to the surface (Fig. 2). However, the experimental data presented in this paper, including the upper part of the profile which constrains the local erosion rate at 2.2 m/Myr, may be fitted using all the calculated Heisinger parameters except the fast muon contribution which has to be adjusted at 0.65%. This experimental study thus points out that if the Heisinger calculated attenuation lengths are comparable to those obtained from measured ^{10}Be concentrations at Cuiaba, the fast muon contribution to the total in situ produced ^{10}Be production rate has to be lowered to 0.65 ± 0.25%.

Samples in the zone where production by stopping muons is expected to be most significant (400–3000 g/cm^2) would have allowed accurate evaluation of the contribution of stopping muons. Unfortunately the sampling density in the upper part of our profile as well as the perturbation of the surface by mining activity make evaluation problematic. Nevertheless, using the attenuation lengths proposed by Heisinger coupled with the preceding value of the percentage of surface production by fast muons, the contribution of stopping muons can be graphically bracketed between 0.6% and 1.8%, with a best value of 1.2%. This is somewhat lower than the 2% reported elsewhere.

(c) The mean erosion rate over the past 10 kyr is substantially lower than the average on 100 kyr timescales. An erosion rate of 2.2 m/Myr fits only the surface data. For deep samples, the best fit is obtained for an erosion rate of 30 m/Myr. Such a drastic change in erosion rate is not probable on stable cratons, however, this result highlights a potential use of coupled deep and surface ^{10}Be analyses for evaluation of erosion rates on a range of timescales.

Within an eroding surface, more time is required for ^{10}Be to reach steady state at depth than at the surface. This is a result of the greater depth penetration of muons compared to neutrons (Fig. 3). This means that the muon component is less sensitive to short timescale perturbations and may be used to quantify the mean long-term erosion rate in contrast to the shorter-term erosion rate reflected by neutrons. The muon-produced component retains a longer-term 'memory' of erosion rates. Fig. 4 shows a surface eroded at

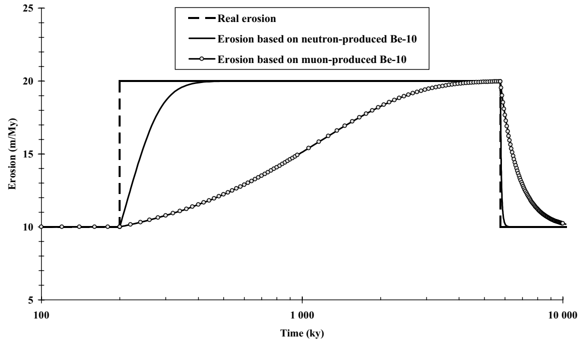

Fig. 4. Evolution of erosion with time. The dashed line represents the real erosion, black and gray dotted lines refer to the erosion rates based on neutron- and muon-produced ^{10}Be respectively. When erosion shifts from 10 to 20 m/Myr and then back to 10 m/Myr, steady-state conditions are reached after ~500 kyr for neutrons and ~5 Myr for muons.

10 m/Myr for a period long enough to have the steady-state conditions for both neutrons and muons and then the erosion rate shifts quickly to 20 m/Myr. It takes around 500 kyr to have the steady-state conditions for the neutrons and 10 times more for the muons. Then, if the erosion rate decrease to 10 m/Myr the steady-state condi-

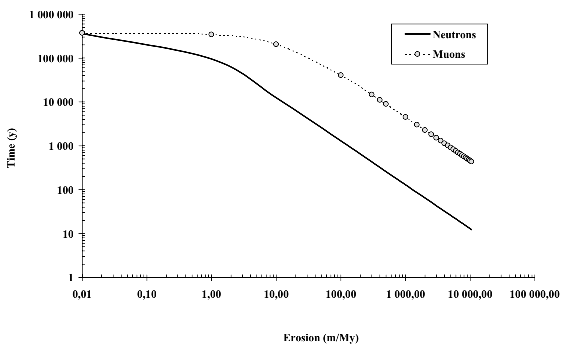

Fig. 3. Time versus erosion. For a given erosion rate, the figure shows the time necessary to reach 95% of the steady-state ^{10}Be concentration for neutrons (solid line) and muons (dotted line). The ^{10}Be fraction produced by muons needs more time to reach its steady-state maximum as the muon attenuation length is much higher than the neutron attenuation length (see text).

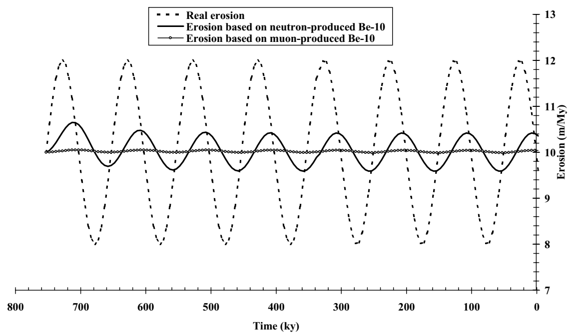

Fig. 5. Evolution of real erosion rate (dashed line) with time considering a sine modulation with an amplitude of 2 m/Myr, a 100 kyr frequency around a mean erosion rate of 10 m/Myr. The black line shows the erosion rates based on neutron-produced ^{10}Be. The gray dotted line shows the erosion rates based on muon-produced ^{10}Be. Considering the analytical uncertainties, both erosions are equal; a time lag of 16 kyr is observed between the real erosion maximum and the erosion rates based on neutron-produced ^{10}Be maximum (erosion rate based on muon-produced ^{10}Be is supposed to be constant).

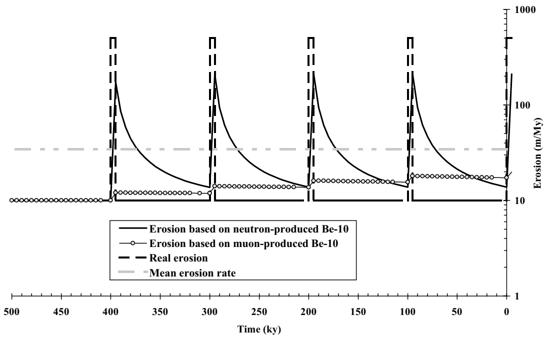

Fig. 6. Evolution of real erosion rate (dashed line) with time considering an erosion rate of 10 m/Myr during 95 kyr followed by pulses of 500 m/Myr during 5 kyr every 100 kyr. The black line represents the erosion rates estimated using the neutron contribution to the total in situ produced ^{10}Be. The gray dotted line represents the erosion rates estimated using the muon contribution to the total in situ produced ^{10}Be. The gray dashed line represents the mean erosion rate ($\sim$34 m/Myr). Erosion rates based on ^{10}Be production by neutrons are more sensitive to the erosion changes. Erosion rates based on ^{10}Be production by muons evolved slowly to the mean erosion rate.

tions are recovered after approximately the same range of time. To illustrate this approach in a more realistic way, we are now considering a surface eroded in two environmental settings: (a) a sine-type erosion evolution around the value of 10 m/Myr with a 100 kyr Milankovitch frequency and an amplitude of 2 m/Myr (Fig. 5) and (b) constant erosion rate (around 10 m/Myr) with high erosion pulses (up to 500 m/Myr) for short period ($\sim$5 kyr) (Fig. 6). In case a (for areas that are not glaciated), erosion rate based on neutron-produced ^{10}Be evolves between 10.5 and 9.5 m/Myr and there is a time lag of $\sim$16 kyr between the real erosion maximum and the maximum erosion rate based on neutron-produced ^{10}Be; erosion rates based on muon-produced ^{10}Be are not affected by the erosion change. In case b (for glaciated areas), neutrons are more sensitive than muons. In this case, erosion rate based on the muon-produced ^{10}Be reflects the long-term erosion rate and tends slowly to the mean erosion rate ($\sim$34 m/Myr).

This different behavior can help to determine if the studied surface has reached steady state. If the erosion rate based on muon-produced ^{10}Be equals or is not too far from the erosion rate based on neutron-produced ^{10}Be, the surface is near the steady state for both muons and neutrons and the measured erosion rate is near the real erosion rate. If the erosion rate based on muon-produced ^{10}Be is very different from the erosion based on neutron-produced ^{10}Be, the surface is not at steady state for at least one production mechanism. In this case, the erosion rate had changed and the erosion rate based on muon-produced ^{10}Be reflects the erosion rate before the change.

6. Conclusion

A depth profile of in situ produced ^{10}Be in vein quartz from the Itaipu mine (Cuiaba, Brazil) yields quantitative information on cosmonuclide production mechanisms. The apparent attenuation rate of fast muons deduced from the ^{10}Be concentration–depth evolution is about 5300 ± 950 g/cm^2. This value is comparable to values ($\sim$4300 g/cm^2) based on laboratory experiments for discrete energy. Linked to the muon's attenuation length, the discussion of the percentage of ^{10}Be produced by muons is not closed. Our data indicate that fast muons contribute 0.65 ± 0.25% of total production whereas stopping muons may contribute 1.2 ± 0.6%. The total ^{10}Be production by muons of $\sim$2% is consistent with previously reported values of 1–3% [12] and 1.5 ± 0.5% [22] determined in rock profiles that were too shallow to allow separate evaluation of the two muon-induced reactions. However, our results are somewhat lower than the values of 2% for each mechanism suggested by Heisinger.

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