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Cosmogenic production of ^{7}Be and ^{10}Be in water targets

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Abstract. We have measured ^{10}Be ($t_{1/2}$ = 1.5 x 10^{6} years) and ^{7}Be ($t_{1/2}$ = 53.28 days) concentrations in water targets exposed for 1 to 2 years at Echo Lake, Colorado (elevation = 3246 m) and at La Jolla, California (140 m). Neutron monitor data were used to normalize the measured concentrations in order to calculate production rates equivalent to the cosmic ray flux averaged over four solar cycles (43 years). The ^{7}Be production rates thus obtained correspond to 6.03 ± 0.07 x 10^{-6} atom g^{-1}·O s^{-1} at Echo Lake and 5.06 ± 0.20 x 10^{-7} atom g^{-1}·O s^{-1} at La Jolla. The ^{10}Be production rates correspond to 3.14 ± 0.18 x 10^{-6} atom g^{-1}·O s^{-1} at Echo Lake and 2.68 ± 0.47 x 10^{-7} atom g^{-1}·O s^{-1} at La Jolla. When compared with ^{10}Be production rates determined in ^{10}Be-saturated rocks from the Antarctic and with theoretical calculations based on meteorite and lunar sample data, we find that the million-year average production rate is about 14 -17% greater than the present production rate averaged over the last four solar cycles. Comparison with production rates determined by measuring glacially polished rocks from the Sierra Nevada in California indicates that average production (based on a revised 13,000-year deglaciation age and a geographic latitude correction) is about 11% greater than the average over the last four solar cycles. The measured ^{10}Be/^{7}Be production ratio in oxygen is 0.52 ± 0.03 at Echo Lake and 0.55 ± 0.07 at La Jolla.

Introduction

Knowledge of the precise production rate for each nuclide as a function of depth, geographical location, and altitude is the most important factor for effective application of cosmogenic radionuclides to geochronology. Since the cosmic ray flux at Earth's surface is attenuated by about 3 orders of magnitude below that in space by passage through Earth's atmosphere, major applications of cosmogenic radionuclides to geochronology, with the exception of ^{14}C dating, were not possible until the development of accelerator mass spectrometry (AMS). AMS increased the measurement sensitivity for long-lived radionuclides by several orders of magnitude and, for the first time, made it possible to accurately measure a wide range of radionuclides, even at the very low concentrations at which they are produced by cosmic rays at Earth's surface [*Elmore and Phillips*, 1987; *Finkel and Suter*, 1993].

A variety of experimental studies of naturally exposed targets and of materials exposed in accelerators as well as theoretical calculations have been used to determine these cosmogenic radionuclide production rates. Production rates depend on the cross sections for production of each cosmogenic nuclide from each target element in the irradiated material, on the concentration of target elements, and on the cosmic ray flux in that material. Production from neutrons is especially important since at sea level, energetic neutrons compose about 95% of the total spallogenic production [*Masarik and Reedy*, 1995]. By combining measured or theoretical cross-section data with estimates of the cosmic ray flux, it has been possible to estimate production rates. They can also be measured directly in certain specifically selected geologic samples. However, laboratory neutron cross sections are difficult to measure and the production rates obtained using cross-section data and by measurements of naturally exposed samples are not always consistent and in some cases scatter widely. *Lal and Peters* [1967] and *Lal* [1988] have reviewed nuclide production mechanisms, rates, and applications.

In certain cases, it has been possible to calibrate cosmogenic nuclide production rates by measuring cosmogenic nuclide concentrations in geologic samples with known exposure times and exposure geometries. For mineral samples of simple and fixed composition, such as quartz, this technique has the advantage that precise information about cross sections is not required. The production rates of ^{10}Be ($t_{1/2}$ = 1.5x10^{6} years) and ^{26}Al (7.05x10^{5} years) and the ratio of ^{26}Al/^{10}Be are very important for many terrestrial applications. *Nishiizumi et al.* [1989] calibrated the ^{10}Be and ^{26}Al production rates in quartz by measurements on samples with glacially polished surfaces from several sites in the Sierra Nevada of California. These samples experienced exposures of known duration determined by the time of deglaciation [*Nishiizumi et al.*, 1989]. Although the deglaciation dates were not well established at the time (see below), *Nishiizumi et al.* [1991] confirmed that the production rates determined from these glacially polished surface rocks are identical to the production rates on a million-year timescale derived from concentrations of ^{10}Be and ^{26}Al in rocks from Antarctica which have been exposed long enough to saturate the activities. *Brown et al.* [1991] studied samples from Antarctic moraines which had exposure ages up to about 2 M.y. old and also confirmed that the production rates determined for the 11-kyr-old surfaces studied by *Nishiizumi et al.* [1989] were representative of the average production over the past 2 M.y., within a rather large uncertainty. *Niedermann et al.* [1994] obtained the production rate of ^{21}Ne in quartz using the same Sierra Nevada glacially polished rocks.

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Production rate measurements for ^{3}He [*Cerling and Craig*, 1994; *Kurz et al.*, 1990; *Poreda and Cerling*, 1992], ^{14}C [*Jull et al.*, 1994], and ^{36}Cl [*Phillips et al.*, 1990; *Stone et al.*, 1994; *Zreda et al.*, 1991] have been carried out in similar studies. These nuclides are produced in terrestrial material by cosmic ray neutrons and muons. However, the neutron cross sections for these cosmogenic nuclides are not well known because appropriate neutron beams are difficult to obtain. Only a few experimental results are available. Recently, data have been obtained by high-energy neutron and muon bombardments of SiO_2 targets at Los Alamos Clinton P. Anderson Meson Physics Facility (LAMPF) [*Reedy et al.*, 1994]. *Imamura et al.* [1990] have also measured high-energy neutron cross sections for production of ^{14}C and ^{26}Al.

Although the production rates of ^{10}Be, ^{26}Al, and ^{21}Ne from SiO_2 have been determined from measurements of glacially polished rocks and from saturated Antarctic rocks, measurements under more controlled conditions are desirable in order to better define these important parameters. The major target element for the production of ^{10}Be is oxygen and that for ^{26}Al is silicon. Therefore in 1988 we started two experiments to measure production rates directly by exposing water and pure Si metal at high altitude and at sea level. Similar experiments have been performed previously for the following short-lived nuclides: ^{7}Be, ^{22}Na, ^{24}Na, ^{28}Mg, ^{32}P, ^{33}P, ^{35}S, and ^{60}Co [*Bhandari et al.*, 1969; *Lal et al.*, 1960; *Mabuchi et al.*, 1971; *Nakamura et al.*, 1972; *Rama and Honda*, 1961; *Takagi and Tanaka*, 1969; *Yokoyama et al.*, 1977]. In order to compare our results with earlier experiments and because cosmogenic ^{7}Be is a useful radionuclide in studies of atmospheric circulation, we included ^{7}Be in our study. We present here measurements of ^{7}Be and ^{10}Be production rates from water (oxygen) by cosmic rays. The silicon data will be reported separately.

Experimental Procedures

Since the expected production rate of ^{10}Be from water at sea level is quite low, only about 10 atom g^{-1}-H_2O yr^{-1}, it is necessary to expose several hundred kilograms of water for an extended time in order to obtain a measurable amount of ^{10}Be. In this work, water was exposed to cosmic rays at high elevation and at sea level for 5 years. Water beds were selected as containers for the target water because of their durability and low cost, good exposure geometry and because they can be heated to avoid freezing during winter. In order to ensure that cosmic ray produced ^{10}Be and ^{7}Be became isotopically mixed with added ^{9}Be carrier and to avoid any hot atom effects that might occur in solid ice, the acidified water target was kept liquid during the whole exposure period. One water bed was exposed at the University of Denver's High Altitude Laboratory at Echo Lake, Colorado. This location was selected to take advantage of existing laboratory facilities and in order to facilitate comparison with a similar experiment performed 36 years ago at this site [*Lal et al.*, 1960]. The water bed was set up in an unheated room covered by a total thickness of 5 ± 1 g cm^{-2} of wood roof, ceiling, and fiber glass insulation. Snow cover on the roof was monitored throughout the course of the experiment, and the measured thicknesses were used as described later in making shielding corrections. A second water bed was exposed at La Jolla, California. It was placed on top of the roof of our laboratory building, Mayer Hall at University of California, San Diego (UCSD). This water bed was covered by a plastic roof of less than 0.3 g cm^{-2} thickness to minimize damage by rain and direct sunlight. There were no additional shielding corrections for this sample. Exposure conditions for both samples are summarized in Table 1.

The inside surfaces of the new water beds were soaked with dilute HCl and rinsed several times with deionized water and Milli-Q water. The target water, 488.2 kg of acidified (0.01 N HCl) Milli-Q water, was mixed with 3.1 mg of Be carrier, prepared from beryl. The beryl was collected from a deep mine and contains $6 \pm 2 \times 10^{-15}$ $^{10}Be/Be$. This solution was passed through a 100-mL cation exchange column (Dowex AG50W-X8) to remove the 3.1 mg of Be, which was extracted from the column and used as a blank (EL-blank-1). The acidified water was then transported to Echo Lake and pumped into the water bed, along with 1.5 mg of Be carrier. A similar procedure was followed at the Mayer Hall site, except that a larger amount of water target solution was used (850.9 kg). A blank (MH-blank-1) was also prepared from this solution. These Be blanks can place an upper limit on the ^{7}Be and ^{10}Be blank levels since they were directly extracted from the Milli-Q water used and therefore include any ^{10}Be and ^{7}Be which were contained in the Milli-Q water. The Milli-Q water which was exposed in the water beds was further purified by first passing

Table 1. Exposure Conditions of Water Targets

	Echo Lake, Colorado	Mayer Hall, UCSD, La Jolla, California
Elevation, m	3246	140
Longitude, °W	105.60	117.21
Geographic latitude, °N	39.66	32.88
Geomagnetic latitude, deg	48.7	40.1
Atmospheric thickness, g cm^{-2}	693	1016
H_2O exposed, kg	489	850
Thickness of water bed, cm	18.8	21.8
First exposure	320 days, Oct. 27, 1988 to Sept. 11, 1989	268 days, Jan. 23 to Oct. 18, 1989
Second exposure	795 days, Sept. 12, 1989 to Nov. 16, 1991	748 days, Oct, 20, 1989 to Nov. 7, 1991
Third exposure	634 days, Nov. 17, 1991 to Aug. 12, 1993	602 days, Nov. 11, 1991 to July 5, 1993

UCSD is University of California, San Diego.

it through a 100-mL cation exchange column. After an exposure period of 1 or 2 years, Be in each water bed was extracted by passing the solution through a 100-mL cation exchange column. The elution speed was approximately 20 L h^{-1}. At each extraction the Be was recovered from the 100-mL column with 1 N HCl and purified with a 5-mL anion exchange column followed by a 5-mL cation exchange column. For the Echo Lake sample the 100-mL cation exchange extraction was performed at Echo Lake and the Be fraction was transferred to La Jolla for further purification. After the Be was extracted, the water was pumped back into the original water bed and 1.5 mg of Be carrier was added. After the second extraction of the Mayer Hall water sample, the solution was transferred to the original water bed along with Be carrier and immediately passed through a 100-mL cation exchange column again before the third exposure. This Be was extracted from the column and used as a blank (MH-blank-2). We used atomic absorption spectrometry to determine Be carrier yields. It was necessary to trace the chemical yields of Be through the various separation and purification steps for two reasons. First, we determined the overall yield of Be in the counting sample. Knowledge of the counting sample yield was necessary for calculation of the ^{7}Be concentration in the water bed. The values in Table 2 give the overall extraction efficiencies for the ^{7}Be counting samples. The Be that is missing from the counting samples resides in the various fractions produced during the chemical purification. It did not remain in the water beds. We could ascertain that 100% of the added Be carrier was recovered from the water exposed in the water beds by summing the Be yields in all the chemical fractions. Loss of Be carrier to the walls of the water bed or contribution of Be carrier to subsequent exposure could therefore be excluded.

Each purified Be fraction was transferred into a 5-mm-diameter quartz tube and the γ ray from ^{7}Be ($t_{1/2}$ = 53.28 days) decay was measured. After the γ ray counting, a small aliquot was taken for chemical yield determination. The main fraction was ignited to BeO, and ^{10}Be was measured by AMS. The ^{7}Be measurements were performed using a well-type Ge detector with a NaI(Tl) anticoincidence background suppression system at UCSD. The background of the system was 0.0186 ± 0.0013 cpm (477.6 ± 3.0 keV), and the absolute efficiency for the ^{7}Be γ ray photo peak (477.6 ± 3.0 keV) was 22.8%. Since the γ ray branching ratio of ^{7}Be is 0.1035, the over-all efficiency for ^{7}Be was 2.36%. The ^{7}Be counting for the third extraction was performed using a well-type Ge detector at Lawrence Livermore National Laboratory (LLNL), which had a background of 0.0261 ± 0.0044 (477.6 ± 1.5 keV) and an absolute efficiency for the ^{7}Be γ ray photo peak (477.6 ± 1.5 keV) of 16.1%. The uncertainties of ^{7}Be quoted in the tables do not include absolute uncertainties in the counting standards. The ^{10}Be measurements for the first and second extractions and the blanks were performed at the University of Pennsylvania AMS facility, and those for the third extraction were performed at the LLNL AMS facility [*Davis et al.*, 1990; *Klein et al.*, 1982].

Results

The concentrations of ^{7}Be in the six extractions are shown in Table 2. EL-1 indicates the first extraction of the Echo Lake sample, and MH-1 indicates the first extraction of the Mayer Hall sample at UCSD. The decay of ^{7}Be activity was corrected to the middle of the extraction time, t = 0, from the middle of the counting period. The ^{7}Be counting time was between 5800 min and 24,800 min. The activity of ^{7}Be in EL-blank-1 was found to be 0.028 ± 0.013 Bq $sample^{-1}$ after correcting for chemical yield or 0.38 ± 0.18 atom ^{7}Be g^{-1} H_2O. The ^{7}Be contamination level in the exposed water bed is actually much lower than this value since the Milli-Q water was first passed through a cation column with Be carrier before being put into the water bed. Because of this purification of the target water and because the observed ^{7}Be activity in the blank was less than 0.02 % of observed ^{7}Be in EL-1 and less than 0.5 % of that in MH-1, after the correction for decay during the exposure period, we can consider ^{7}Be contamination in the exposed water beds to be negligible. The ^{7}Be in MH-blank-1 and MH-blank-2 were not measured.

The observed ^{7}Be activity was converted to the ^{7}Be production rate using undersaturation and shielding corrections summed over the exposure time. For the Echo Lake samples it was necessary to correct for shielding by snow on the roof. The correction f was calculated daily and summed as follows.

$$f = \sum_{t=0}^{T} [e^{-d_t / \Lambda} \cdot e^{-\lambda t} \cdot (1 - e^{-\lambda \Delta t})] \quad (1)$$

where t is the time in days before the end of exposure, T is total exposure in days, λ is the ^{7}Be decay constant, Λ is the $1/e$ attenuation length of cosmic ray interaction (165 g cm^{-2}), d_t is the snow cover thickness on day t assuming a snow density of 0.25 ± 0.15 g cm^{-3} [*Paterson*, 1969], and Δt is the summation interval (1 day). The combined corrections for shielding by the snow and roof mass and for the undersaturation of ^{7}Be are shown in Table 2 for each sample. The errors

Table 2. Beryllium 7 in Water Beds

	Measured ^{7}Be, Bq at t=0	Chemical Yield, %	Beryllium 7 Concentration, 10^{-6} Bq g^{-1} H_2O	Shielding and Exposure Time Correction	Beryllium 7 Production Rate, atom g^{-1} H_2O y^{-1}
EL-1	1.942 ± 0.018	94.0	4.233 ± 0.074	0.9473 ± 0.0047	141.0 ± 2.6
EL-2	1.820 ± 0.011	90.3	4.122 ± 0.067	0.9510 ± 0.0111	136.8 ± 2.7
EL-3	2.198 ± 0.037	92.8	4.834 ± 0.110	0.9473 ± 0.0131	161.1 ± 4.3
MH-1	0.2968 ± 0.0053	94.4	0.3695 ± 0.0086	0.9694	12.03 ± 0.28
MH-2	0.2803 ± 0.0038	93.8	0.3530 ± 0.0072	0.9999	11.14 ± 0.23
MH-3	0.2998 ± 0.0165	87.3	0.406 ± 0.023	0.9996	12.80 ± 0.73
EL-blank-1	0.019 ± 0.009	69.6	0.057 ± 0.027		

EL is Echo Lake, and MH is Mayer Hall. See text for site descriptions.

quoted in Table 2 are the quadratically added 1σ counting statistics and a 1.5% chemical yield uncertainty but do not include the uncertainty in the ^{7}Be standard (≤ ± 5%). The extractions of the Echo Lake samples were performed near the beginning of the snow season, so that the roof was not covered by snow during that part of the exposure which dominates the measured ^{7}Be concentration, i.e., near the end of the exposure.

The results of the ^{10}Be measurements in the six samples are shown in Table 3, along with three blanks, EL-blank, MH-blank-1, and MH-blank-2. After correcting for ^{10}Be background due to ^{10}B, the measured ratios were normalized to an ICN Chemical and Radioisotope Division (ICN) ^{10}Be standard which was diluted at La Jolla by one of the authors (K. Nishiizumi) [*Nishiizumi et al.*, 1984]. The errors of the ^{10}Be measurements are quoted as 1σ AMS measurement error and do not include the error in the ^{10}Be standard. We used the same ^{10}Be standard for both AMS measurements at the University of Pennsylvania and LLNL. This standard was calibrated by β counting, and the ^{10}Be/^{9}Be ratio was calculated assuming a ^{10}Be half-life of 1.5 M.y. This standard has been used by us for ^{10}Be AMS measurements in all terrestrial and extraterrestrial samples since 1983 at both the University of Pennsylvania and LLNL [*Nishiizumi et al.*, 1984]. The ^{10}Be concentration in the carrier blank, 6 ± 2 x 10^{-15} ^{10}Be/Be, was then subtracted to calculate the net measured ^{10}Be/Be ratios shown in Table 3. The ^{10}Be concentrations in EL-blank and MH-blank-1 were higher than that in MH-blank-2 because the former blanks were obtained from untreated Milli-Q water. Since a part of the Milli-Q water was passed through a cation column as a practice experiment before extracting the EL-blank, the ^{10}Be concentration in EL-blank is lower than in MH-blank-1. The ^{10}Be concentration in MH-blank-2, 1.10 ± 0.28 atom ^{10}Be $g^{-1}\cdot H_2O$, was adopted as the chemistry blank and was subtracted from the measured net values. This work indicates that our laboratory Milli-Q water contains about 9 atom ^{10}Be g^{-1}, which is negligible for most of our experiments. Shielding corrections, including snow cover and roof thickness, were applied to calculate the ^{10}Be production rate for the Echo Lake samples in a manner similar to that described above for ^{7}Be.

Discussion

Deriving Solar Cycle Average Production Rates

Within the heliosphere of the solar system, which includes the orbit of Earth, the energy spectrum and flux of galactic cosmic rays are modulated by solar magnetic fields, which act to reduce the flux of the low-energy component below levels outside the solar system. As a result of this modulation, the cosmic ray flux at Earth's orbit is anticorrelated with indices of solar activity, such as sunspot number. By the time it reaches Earth's surface, the cosmic ray flux has been further altered by having passed through the atmosphere. This low-energy (below tens of GeV), secondary cosmic ray flux has been well determined at various locations on Earth using an array of neutron monitors. Neutron monitor data from the Climax Neutron Monitor Station, Colorado, operated by the University of Chicago, are available for a time period extending back to 1953 [*Simpson*, 1958]. The station is located at 39°N, 106°W and 3400 m elevation, very close to Echo Lake. The neutron monitor counting rates are in excellent anticorrelation with sunspot number. Furthermore, it is expected that the production rates of ^{10}Be and ^{7}Be are proportional to the count rates of the neutron monitors.

The water bed exposure intervals occurred during a period of solar maximum, at which time the galactic cosmic ray flux on Earth was at a minimum. In order to calculate long-term average production rates of ^{7}Be and ^{10}Be, the observed production rates were normalized to the neutron monitor data in the following manner. The average neutron monitor counting rate at the Climax station between September 1954 and March 1987, which corresponds to three solar cycles, was 395,300 counts h^{-1}. The normalized ^{7}Be production rate P_n is calculated as follows

$$P_n = P_m \frac{N_{av}(1-e^{-\lambda T})}{\sum_{t=0}^{T} N_t \cdot e^{-\lambda t}(1-e^{-\lambda \Delta t})} \quad (2)$$

where P_m is the measured ^{7}Be production rate, t is the time in days before the end of the exposure period, T is the total exposure period (days), λ is the ^{7}Be decay constant, Δt is the summation interval (1 day), N_{av} is the average hourly neutron monitor count rate during the three solar cycles described above, and N_t is the hourly neutron count rate during day t. The normalized ^{10}Be production rates were calculated in a similar manner. The overall average neutron monitor counting rate at Climax station was 393,600 counts h^{-1} (January 1953 to April 1996; 43.3 years), which covers an interval slightly shorter than four solar cycles. This value is only 0.4% lower than the full three solar cycle average (1954-1987). Addition of neutron monitor data for 1 more year will complete coverage of four solar cycles and will slightly increase the average counting rate because it is now near solar minimum (high galactic cosmic ray (GCR) flux). We are confident that

Table 3. Beryllium 10 in Water Beds

	Measured ^{10}Be/Be, 10^{-15}	Measured ^{10}Be, atom $g^{-1}\cdot H_2O$	Blank Corrected ^{10}Be, atom $g^{-1}\cdot H_2O$	Shielding Correction	Beryllium 10 Production Rate, atom $g^{-1}\cdot H_2O$ y^{-1}
EL-1	320 ± 12	67.4 ± 2.4	66.3 ± 2.5	0.941 ± 0.017	80.5 ± 3.3
EL-2	654 ± 16	141.3 ± 3.4	140.2 ± 3.5	0.936 ± 0.020	68.8 ± 2.2
EL-3	665 ± 14	129.3 ± 2.8	128.2 ± 2.8	0.913 ± 0.033	80.9 ± 3.4
MH-1	68 ± 4	6.68 ± 0.25	5.59 ± 0.38	1	7.62 ± 0.51
MH-2	111 ± 5	13.77 ± 0.63	12.67 ± 0.69	1	6.19 ± 0.34
MH-3	96 ± 4	11.07 ± 0.50	9.97 ± 0.57	1	6.05 ± 0.35
EL-blank	7 ± 3	3.2 ± 1.2			
MH-blank-1	87 ± 6	9.21 ± 0.60			
MH-blank-2	10 ± 3	1.10 ± 0.28			

the above normalized production rates using neutron monitor data averaged over three solar cycles can be applied to the average production rates for the last four solar cycles.

The thicknesses of the water beds were 18.8 cm for Echo Lake and 21.8 cm for Mayer Hall. We have applied the following thickness correction

$$P_m = \frac{P_0}{d/\Lambda}(1-e^{-d/\Lambda}) \quad (3)$$

where P_m is the measured production rate in the water bed, P_0 is the production rate at the surface of the water bed, Λ is the $1/e$ attenuation length of cosmic ray interaction (165 g cm^{-2}), and d is the thickness of the water bed. This correction is of the order of 5-6% for both exposures. However, if we assume that the neutron flux at the air-solid interface is constant up to 12 g cm^{-2} as has been shown in a recent study [*Masarik and Reedy*, 1995], the correction decreases to 1-2%. The ^{7}Be and ^{10}Be production rates normalized with neutron monitor data and corrected using (3) are shown in Table 4. The normalized ^{7}Be production rates at Echo Lake are in good agreement for the three different exposure periods as are those for UCSD. These normalized production rates represent the average cosmic ray flux over the last four solar cycles. The ^{7}Be measurements at Echo Lake are the most reliable because the counting rates were relatively high. The excellent agreement of the three results demonstrates that the extraction method, snow cover correction, and ^{7}Be measurements at the two laboratories (UCSD and LLNL) were consistent and reliable. The ^{10}Be results for the Echo Lake samples are also in good agreement, but ^{10}Be in the UCSD samples scatters somewhat owing to the lower concentration of the nuclide produced in the water bed at this lower-elevation exposure.

Comparison With Short-Term Natural and Artificial Irradiation Measurements

The ^{7}Be production rates obtained correspond to 6.03 ± 0.07 x 10^{-6} atom g^{-1}·O s^{-1} at Echo Lake and 5.06 ± 0.20 x 10^{-7} atom g^{-1}·O s^{-1} at UCSD. *Lal et al.* [1960] earlier measured the ^{7}Be production rate in a small swimming pool set up in a room at Echo Lake. They obtained 9.0 x 10^{-6} atom g^{-1}·O s^{-1} which corresponds to 11.1 x 10^{-6} atom g^{-1}·O s^{-1} after normalization to the neutron monitor data and using an updated branching ratio for the ^{7}Be γ ray. Their production rate is nearly a factor of 2 higher than the production rate obtained in this work. The swimming pool used by *Lal et al.* [1960] (~1.5 m diameter) was open to the air, and over 6% of the water it contained evaporated during the 32-day exposure period. One possible explanation for the higher ^{7}Be measured in this earlier work is that part of the ^{7}Be in the swimming pool was actually contamination from dust or aerosol. *Nakamura et al.* [1972] also performed similar ^{7}Be experiments at three locations. Although Nakamura *et al.* provide insufficient information for a full comparison with our results, we are able to derive the following information. The ^{7}Be production rates were 2.2 ± 0.4 x 10^{-6} atom g^{-1}·O s^{-1} (recalculation of *Nakamura et al.* [1972, Table 1]) at Mount Fuji (geomagnetic latitude, 25.5°N, atmospheric thickness, 772 g cm^{-2}), 4.2 x 10^{-7} atom g^{-1}·O s^{-1} (from *Nakamura et al.* [1972, Figure 1 and text] data) at Tokyo (25°N; sea level), and 1.0 ± 0.2 x 10^{-6} atom g^{-1}·O s^{-1} at Gif-sur-Yvette (51°N, 1030 g cm^{-2}). Although we have not been able to ascertain the exact exposure dates for these experiments, neutron monitor data indicate that the neutron flux increased during 1971 and 1972 and that the neutron fluxes at this time were a few percent higher than the four solar cycle average.

Star production in emulsions is widely used for calculation of cosmogenic nuclide production rates at different locations [*Lal and Peters*, 1967]. The star production was calculated to be 1.90 x 10^{-5} stars g^{-1} air s^{-1} at La Jolla, 21.2 x 10^{-5} stars g^{-1} air s^{-1} at Echo Lake, 1.36 x 10^{-5} stars g^{-1} air s^{-1} at Tokyo, 7.34 x 10^{-5} stars g^{-1} air s^{-1} at Mount Fuji, and 1.81 x 10^{-5} stars g^{-1} air s^{-1} at Gif-sur-Yvette [*Lal*, 1991; *Lal and Peters*, 1967]. Using our results and the data of *Nakamura et al.* [1972] the ratios of observed ^{7}Be production rates at different locations are 11.9 ± 0.5 for Echo Lake / La Jolla, 2.7 ± 0.5 for Echo Lake / Mount Fuji, 14.4 ± 1.8 for Echo Lake / Tokyo, and 6.0 ± 1.2 for Echo Lake / Gif-Sur-Yvette. The ratios of star production for the same location pairs are 11.1, 2.9, 15.6, and 11.7. Production rate and star production ratios agree very well except for the ^{7}Be data at Gif-Sur-Yvette, which are nearly

Table 4. Beryllium 7 and 10 Production Rates in Water Targets

	First Exposure, (1988-1989)	Second Exposure, (1989-1991)	Third Exposure, (1991-1993)	Average, (1988-1993)
Normalized ^{7}Be at Echo Lake, atom g^{-1}·H_2O yr^{-1}	169.3 ± 3.1	168.8 ± 3.4	168.6 ± 4.5	169.0 ± 2.0
Normalized ^{7}Be at UCSD, atom g^{-1}·H_2O yr^{-1}	14.70 ± 0.34	13.91 ± 0.28	13.59 ± 0.78	14.18 ± 0.57
Beryllium 7 production rate ratio, Echo Lake/UCSD	11.52 ± 0.34	12.14 ± 0.35	12.40 ± 0.78	11.9 ± 0.46
Normalized ^{10}Be at Echo Lake, atom g^{-1}·H_2O yr^{-1}	94.6 ± 3.9	85.1 ± 2.8	87.7 ± 3.7	88.1 ± 4.9
Normalized ^{10}Be at UCSD, atom g^{-1}·H_2O yr^{-1}	9.27 ± 0.62	7.72 ± 0.42	6.68 ± 0.38	7.51 ± 1.31
Beryllium 10 production rate, Echo Lake/UCSD	10.20 ± 0.80	11.02 ± 0.70	13.13 ± 0.93	11.3 ± 1.5
Beryllium 10/^{7}Be at Echo Lake	0.559 ± 0.025	0.504 ± 0.019	0.520 ± 0.026	0.523 ± 0.028
Beryllium 10/^{7}Be at UCSD	0.631 ± 0.045	0.555 ± 0.032	0.491 ± 0.040	0.554 ± 0.070

The production rates of ^{7}Be and ^{10}Be are referred to the surface of the target (depth = 0) assuming exponential decrease of cosmic ray interaction (see text).

a factor of 2 higher than expected. The consistency of our three extractions at each site and of our $^{10}Be/^{7}Be$ ratios at each site give increased confidence to the validity of our measurements.

Although the error is relatively large, the ^{10}Be production rate ratio was 11.3 ± 1.5 for Echo Lake / La Jolla, in good agreement with the ratio for star production. Beryllium 10 is produced by muons and protons at Earth's surface as well as by high-energy neutrons. Because muons have an absorption mean free path of about 250 g cm^{-2} compared with 165 g cm^{-2} for neutrons, muon production becomes progressively more important with depth. In fact, as discussed by *Nishiizumi et al.* [1989], we expect the ^{10}Be production ratio between Echo Lake and La Jolla to be about 10 if muon production is important at sea level. If spallation dominates, the ratio is expected to be about 11.1. Unfortunately, the low concentration of ^{10}Be in the low-elevation La Jolla samples limited the precision with which ^{10}Be could be determined, and the resulting uncertainty in the measured ratio between Echo Lake and La Jolla makes it impossible to determine the importance of muons in production of this nuclide from the present study. On the basis of our muon bombardment experiments at LAMPF (unpublished), the ^{7}Be production by muons is negligible compared with the production by neutrons, even at sea level.

Observed $^{10}Be/^{7}Be$ production rate ratios at Echo Lake, 0.52 ± 0.03, and La Jolla, 0.55 ± 0.07, are very similar (see Table 4). SiO_2 and Si targets have been bombarded by high-energy neutrons at LAMPF [*Reedy et al.*, 1994]. The observed $^{10}Be/^{7}Be$ ratios, 0.624 ± 0.034 from O and 0.574 ± 0.029 from SiO_2, are slightly higher than the ratios observed from this work. Since the Si and SiO_2 targets were exposed to spallation neutrons (< 800 MeV), which were produced from a beam stop at LAMPF, the neutron spectrum was softer than that of the cosmic ray secondary neutrons. *Yokoyama et al.* [1977] calculated the production ratio in SiO_2 to be 0.13, which is a factor of 4 lower than the observed ratio, but these authors used proton-induced cross sections for ^{10}Be and ^{7}Be production in their calculation. The cross section of $O(n,x)^{10}Be$ is estimated to be much higher than that of $O(p,x)^{10}Be$ in the low-energy region based on production rates of ^{10}Be in lunar cores and meteorites [*Nishiizumi et al.*, 1984; *Tuniz et al.*, 1984]. Determination of high-energy neutron cross sections remains vital for knowledgeable application of cosmogenic nuclides in studies of terrestrial and extraterrestrial materials. On the basis of this work, we strongly suggest that future efforts to determine the global variation of production rates can be accomplished by neutron monitor and ^{7}Be measurements at different locations. The data can then be normalized to ^{7}Be and ^{10}Be results at one location, such as Echo Lake or at Climax station (neutron monitor). This would save the laborious work of measuring ^{10}Be in water at each location.

Comparison With Long-Term Geologically and Theoretically Determined Production Rates

The ^{10}Be production rate found at Echo Lake, 88.1 ± 4.9 atom $g^{-1}\cdot H_2O\ y^{-1}$, is equivalent to 55.0 ± 3.1 atom $g^{-1}\cdot SiO_2\ y^{-1}$. The latter value includes an estimate of ^{10}Be production from Si based on the irradiation experiments at LAMPF described above [*Reedy et al.*, 1994]. The ^{10}Be production rate in quartz from glacially polished rocks in the Sierra Nevada was found to be 61.9 ± 3.0 atom $g^{-1}\cdot SiO_2\ y^{-1}$ based on a 11,000-year exposure time [*Nishiizumi et al.*, 1989]. The Sierra Nevada result was normalized to 3340 m (685 g cm^{-2}) and geographic latitude of 38°N. A recent reassessment of the Sierra Nevada sites studied by *Nishiizumi et al.* [1989] indicates that about 13,000 years is a better estimate of the age of glacial retreat than the 11,000 years which was used by these authors [*Clark et al.*, 1995]. Furthermore, consideration of the migration of the geomagnetic dipole suggests that for samples with long exposure times, such as the Sierra Nevada rocks, geographic latitude would be preferred over geomagnetic latitude for making the latitude correction of cosmic ray intensity. This is true, even though we do not know the exact paleo-geomagnetic intensity at each site. The ^{10}Be production rate found at Echo Lake (3246 m, 48.7°N, present geomagnetic latitude) can be normalized to the Sierra Nevada location (3340 m, 44°N, present geomagnetic latitude) using star production (21.2 x 10^{-5} stars g^{-1} air s^{-1} at Echo Lake versus 20.5 x 10^{-5} stars g^{-1} air s^{-1} at Sierra Nevada). The normalized result is 53.2 ± 3.0 atom $g^{-1}\cdot SiO_2\ y^{-1}$, 86% of the Sierra Nevada value based on an 11,000-year deglaciation age and 102% based on a 13,000-year age. The excellent agreement with the 13,000-year deglaciation figure is fortuitous since as discussed below, both geomagnetic intensity and geomagnetic latitude changes must be considered in comparing long-term production rates with present-day values. In other words, the agreement does not indicate that the field intensity has necessarily been constant over the past 13,000 years.

The cosmogenic nuclide production rate can be influenced by changes in the galactic cosmic ray intensity due to solar modulation, by changes in the cosmic ray flux in the interstellar medium, or by a change of Earth's dipole field. The latter effect is a mixture of change of the geomagnetic field intensity and change of the geomagnetic latitude at each site. Global effects of changes in the field intensity have been estimated from low-frequency changes in $^{14}C/^{12}C$ in tree rings (which reflect global changes in production) [*Bucha*, 1970; *Stuiver and Quay*, 1980; *Suess*, 1980], although climate effects have also been proposed [*Beer et al.*, 1988; *Lal*, 1985] for these variations. Previously [*Nishiizumi et al.*, 1989], we estimated the average production (past 10,000 years) for the nuclides ^{10}Be and ^{26}Al at the Sierra Nevada location to be 1.15 times higher than the present-day value at this site based on a semisinusoidal variation for the geomagnetic field intensity [*Bucha*, 1970]. No change in geomagnetic latitude was considered. Estimating the long-term production rate at a spot on Earth's surface becomes more complicated if we take into account the change of geomagnetic latitude as well as geomagnetic intensity. The observed ^{10}Be production rate derived above from our water bed results reflects the average cosmic ray intensity over the last four solar cycles at the Echo Lake and at the Sierra Nevada sites. On the other hand, the observed ^{10}Be concentration in quartz reflects changes of geomagnetic intensity and geomagnetic latitude at the Sierra Nevada site for the last 11,000 or 13,000 years. If we assume that the average geomagnetic latitude at the Sierra Nevada site for the last 11,000 to 13,000 years is similar to the geographic latitude, we find the average production rate to be 13% lower compared with the production rate at the present geomagnetic latitude. This latitude effect is opposite in direction to the 15% increase in production rate expected from the possible geomagnetic intensity changes described above. The resulting 2% different is not meaningful since neither

Table 5. Comparison of ^{10}Be Production Rate at Sea Level, >50°

Method	Timescale	Beryllium 10 Production Rate, atom $g^{-1}\cdot SiO_2 y^{-1}$	References
Water targets	4 solar cycles	5.21 ± 0.27	this work
Water targets	~10,000 years	6.0 ± 0.3*	this work
Glacially polished rocks at Sierra Nevada	11,000 years	6.03†	1
Glacially polished rocks at Sierra Nevada	13,000 years	5.11†	1
Glacially polished rocks at Sierra Nevada	11,000 years	6.84‡	1
Glacially polished rocks at Sierra Nevada	13,000 years	5.80‡	1
Antarctic rocks	≥ 4 M.y.	6.13	2
Antarctic rocks	≤ 2.5 M.y.	6.4	3
Theoretical calculation	> several M.y.	5.97	4

References: 1, *Nishiizumi et al.* [1989]; 2, *Nishiizumi et al.* [1991]; 3, *Brown et al.* [1991]; 4, *Masarik and Reedy* [1995].

* Long-term geomagnetic field correction is added.

† Values use present geomagnetic latitude.

‡ Values use geographic latitude (see text).

paleomagnetic pole movement nor geomagnetic intensity is well understood.

The observed production rate of ^{10}Be at Echo Lake, 55.0 ± 3.1 atom $g^{-1}\cdot SiO_2\ y^{-1}$, can be converted to 5.21 ± 0.29 atom $g^{-1}\cdot SiO_2\ y^{-1}$ at sea level, >50° using scaling factors obtained by *Lal* [1991] and *Nishiizumi et al.* [1989]. The comparison of ^{10}Be production rates (atom $g^{-1}\cdot SiO_2\ y^{-1}$ at sea level, >50°) is shown in Table 5. The production rate obtained from the Sierra Nevada results is shown using corrections from the current geomagnetic latitude and the current geographic latitude with 11,000-year and 13,000-year glaciation ages for comparison. It is important to note here that the production rate of ^{10}Be obtained from this work (four solar cycles average corrected by a factor of 1.15 to incorporate changes in the geomagnetic field), from the Sierra Nevada results (applying geographic latitude with 13,000-year average), from Antarctic rocks (million-year average) [*Nishiizumi et al.*, 1991], and from theoretical calculation using a cosmic ray intensity timescale of millions of years [*Masarik and Reedy*, 1995] all agree well, within a few percent (within experimental error). The present study indicates that the cosmic ray intensity on Earth for the last four solar cycles is 10-15% lower than the long-term average. Although current estimates of the contribution of muon-induced ^{10}Be [*Brown et al.*, 1995; *Nishiizumi et al.*, 1994] is somewhat smaller than our previous estimate [*Nishiizumi et al.*, 1989], we have used the same scaling factor. At the present time we prefer not to make any correction for the uncertainty in the muon contribution. When we have a better estimate for the muon effect, we can make suitable adjustments. The comparison of all the experimental data in Table 5 is only slightly effected by uncertainty in the muon contribution since all the measurements were performed at high altitude. Knowledge of the precise production rate for each nuclide over different time periods is essential not only for reliable application of cosmogenic nuclides to geochronology, but also to understand secular variation of cosmic ray intensity on Earth.

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