

LA-UR- 95 - 3912

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

Title: Production Systematics of Cosmogenic Nuclides in the Earth.

Author(s): Robert C. Reedy

Submitted to:

Todai International Symposium on
Cosmochronology and Isotope Geology,
Tokyo, Japan, 17-20 January 1996.

RECEIVED
DEC 14 1995
OSTI

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

Los Alamos
NATIONAL LABORATORY

Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the University of California for the U.S. Department of Energy under contract W-7405-ENG-36. By acceptance of this article, the publisher recognizes that the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes. The Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy.



DISCLAIMER

**Portions of this document may be illegible
in electronic image products. Images are
produced from the best available original
document.**

PRODUCTION SYSTEMATICS OF COSMOGENIC NUCLIDES IN THE EARTH

Robert C. Reedy¹

¹ Astrophysics and Radiation Measurements Group, Mail Stop D436, Los Alamos National Laboratory, Los Alamos, NM 87545 USA.

The high-energy particles in the galactic cosmic rays (GCR) can produce nuclides deep in any object exposed to them. These cosmic-ray-produced (cosmogenic) nuclides have been extensively studied during the last four decades, mainly in meteorites and lunar samples (e.g., 1,2). In extraterrestrial matter, several approaches have been used to determine the production systematics of these cosmogenic nuclides. Production rates of most cosmogenic nuclides in the Earth are much lower, especially those nuclides made "in situ" in the Earth's surface. Many of these terrestrial cosmogenic nuclides are only now being measured because of improved techniques, such as accelerator mass spectrometry (AMS). There have been very few determinations of the production rates of nuclides made in the Earth by cosmic rays. The work being done for terrestrial cosmogenic nuclides is following the approaches used for studying the production of extraterrestrial nuclides.

For cosmogenic nuclides in extraterrestrial matter, initial studies used activities of radionuclides in meteorites to infer production rates. For a long, constant exposure to cosmic rays, a radionuclide's decay rate approaches equilibrium with its production rate. Irradiations of thick targets at high-energy accelerators were done to experimentally simulate GCR irradiations in space. While such simulations are limited because of the single energy at an accelerator instead of the wide range of energies in the GCR, such thick-target bombardments helped in understanding the nuclear processes involved. Measurements of radionuclides made in thick targets showed the build-up of the cascade of secondary particles and the removal of primary GCR particles (e.g., 3,4). Accelerators were also used with thin targets to measure cross sections for making these cosmogenic nuclides. For nuclides made mainly at high energies, such as nuclides with atomic numbers much less than 26 that are made in iron meteorites, ratios of their production cross sections were used for their production ratios by GCR particles.

Several models were developed for predicting the systematics of the production of cosmogenic nuclides, mainly semi-empirical fits to measured concentrations. Only recently have models based only on physics been used to study nuclear reactions in extraterrestrial matter (e.g., 5,6). Like laboratory simulations, these numerical simulations allow us to examine the details of how GCR particles interact with matter, such as the influence of the target's bulk composition on the production of secondary particles (7).

In determining the production systematics of nuclides made in the Earth's surface by cosmic rays, we are repeating the approaches used to study extraterrestrial cosmogenic nuclides. The terrestrial case is more complicated than the extraterrestrial ones because of the Earth's atmosphere and its strong geomagnetic field. Most cosmic-ray particles interact in the Earth's atmosphere. The most famous cosmogenic nuclide is one made in the Earth's atmosphere, ¹⁴C or radiocarbon. While ¹⁴C has been well studied, other nuclides made in the Earth's atmosphere, such as ³H, ⁷Be, ¹⁰Be, and ³⁶Cl, have not

been well studied.

The flux of energetic particles reaching the Earth's surface is attenuated by factors of $\sim 10^{-3}$ because of the Earth's thick (1033 g/cm^2) atmosphere. While some cosmogenic nuclides have been measured at great depths in a few iron meteorites (e.g., 8), the large difference in the composition and density of the Earth's atmosphere compared to extraterrestrial matter makes it hard to use data for cosmogenic nuclides in extraterrestrial matter for nuclides made deep in the Earth's atmosphere. One major difference is that pions made in dense extraterrestrial matter interact before they can decay, while many pions in the very-thin atmosphere decay to muons before interacting. These muons are weakly-interacting particles and can go much further than strong-interacting particles such as protons, neutrons, and pions. At depths below a few meters in the Earth's surface, muons are a major source of cosmogenic nuclides (9).

Another complication is that the production rates of a nuclide in the Earth's surface can vary much with the elevation of the sample and the geomagnetic latitude of the location (9). Many terrestrial samples are in dynamic environments, and production systematics can be complicated by material such as snow or dust covering a rock or by the erosion of the rock itself (10).

Most production rates of nuclides made in the Earth are now based on a few measurements made with natural samples. Several summaries have been made of these production rates (e.g., 11,12). Generally, the production rate of a cosmogenic nuclide is converted from that determined at the location's elevation and geomagnetic latitude to the rate at sea level and a geomagnetic latitude ($\gtrsim 60^\circ$) with no geomagnetic cutoff of GCR particles. There are often uncertainties about the actual age of the sample, which is needed to convert the measured concentration of the nuclide to a production rate. Most measurements have been done on material with a simple composition, often quartz (SiO_2) or calcite (CaCO_3), so production rates for other target elements often have not been determined.

Several studies have been done at accelerators on production systematics of terrestrial cosmogenic nuclides. Many of these irradiations were done at the Los Alamos Meson Physics Facility (LAMPF). There are some locations near to, but not in, LAMPF's 800-MeV proton beam that have mainly neutrons with energy spectra similar to those in the Earth, such as behind the LAMPF beam-stop (13) and near the sides of a large cylinder filled with sand and other minerals (14). Targets of Si and SiO_2 irradiated near the LAMPF beam-stop were used to study the production ratios for several radionuclides (13). The $^{26}\text{Al}/^{10}\text{Be}$ production ratio in quartz (SiO_2) was measured to be 7.1, very similar to that measured in quartz from the Sierra Nevada mountains, 6.0 (15). Targets irradiated with such particles could be used to study production systematics for many targets, such as pure elements.

Some accelerators are also sources of muon beams. The $^{26}\text{Al}/^{10}\text{Be}$ ratio in quartz irradiated with stopping muons at LAMPF was 7.0, very similar to that in samples from the LAMPF beam-stop and in natural samples (16). Work is also being done on measuring absolute yields of nuclides for muons stopping in various targets.

Because terrestrial cosmogenic nuclides are made mainly by low-energy particles, such as neutrons, measurements of cross sections with thin targets can not be used by

themselves for production ratios. As the ratio of neutrons to protons at the Earth's surface is higher than in most meteorites and lunar samples, cross sections for the production of terrestrial cosmogenic nuclides by energetic neutrons are very important. The use of such neutron cross sections with some models is discussed below.

While several sets of calculations have been done for nuclides made in the Earth's atmosphere (e.g., 17), until recently very little was done for nuclides made by cosmic-ray particles in the Earth's surface. Neutron-transport codes, such as the Monte Carlo N-Particle (MCNP) code, can be used to study nuclides made by neutron-capture reactions. The MCNP code was used to show that rates for the $^{35}\text{Cl}(n, \gamma)^{36}\text{Cl}$ reaction depend on the composition of the rock and its water content (18).

The LAHET Code System (LCS) has been used very successfully for studying cosmogenic nuclides in extraterrestrial matter (e.g., 7). It has recently been used to study production of nuclides in the Earth (12). Rates calculated for the production of radionuclides in the Earth's atmosphere and of nuclides made in the Earth's surface agree well with measured values. LCS calculates the fluxes of cosmic-ray particles as a function of depth and energy. The product of these fluxes and cross sections for the relevant nuclear reactions are then integrated over energy to calculate the nuclide's production rates.

Terrestrial cosmogenic nuclides are made mainly by neutrons, but there are only a few cross sections measured for nuclide production by energetic ($\gtrsim 20$ MeV) neutrons. For many nuclides, cross sections measured with protons are used for neutron reactions. A problem with some nuclides, such as ^{10}Be and ^{14}C , is that using proton cross sections for neutron-induced reactions gives poor results. For these nuclides, the cross sections with incident GCR protons are different than those for incident neutrons (e.g., 19). More cross sections are needed for the production of cosmogenic nuclides by high-energy neutrons (e.g., 19,20).

For most nuclides made in the Earth's surface, cross sections for the important reactions have been tested by comparing measurements and calculations for extraterrestrial matter. Production rates calculated for several radionuclides at sea level and for no geomagnetic cutoff agreed well with rates determined from natural samples (12). The calculated variations in production rate with elevation agreed with those of Lal (9). A limitation of LCS is that it is not very good for incident GCR particles with energies above ~ 10 GeV. As geomagnetic cutoffs near the Earth's equator are ~ 17 GeV, the use of LCS for such regions is limited. Codes that can correctly handle incident GCR particles with energies above about 20 GeV are needed to calculate production rates near the equator.

As presented above, much progress has been made on determining production rates of nuclides made in the Earth's surface by cosmic-ray particles using natural samples, simulations at accelerators, and numerical calculations. Some work, such as cross-section measurements with neutrons, was noted above as being needed to better understand production systematics of terrestrial cosmogenic nuclides. However, there is also a need for much other work to improve our understanding of these production rates (e.g., 11). There are some uncertainties in the production rates as a function of geomagnetic latitude. Changes in nuclide production rates because of variations in the intensities of

GCR particles hitting the Earth, due to solar or geomagnetic variations, have not been studied much. Such changes need to be known for production rates as a function of time and location in the past. Production rates are needed for deep in the Earth, where muons and muon-produced neutrons make most nuclides. Some exposure dates used for natural samples are being questioned, and good ages are needed for many locations, including some over a range of elevations and latitudes. Artificial samples should be exposed to natural cosmic radiation because they will provide known exposure times. Many groups are now working together to do such work. In a few years, there should be an improved understanding of the production systematics of terrestrial cosmogenic nuclides.

References: (1) Reedy R.C., Arnold J.R., and Lal D. (1983) *Annu. Rev. Nucl. Part. Sci.* **33**, 505–537. (2) Vogt S., Herzog G.F., and Reedy R.C. (1990) *Rev. Geophys.* **28**, 253–275. (3) Honda M. (1962) *J. Geophys. Res.* **67**, 4847–4858. (4) Michel R., Lüpke M., Herpers U., Rösel R., Suter M., Dittrich-Hannen B., Kubik P.W., Filges D., and Cloth P. (1995) *Planet. Space Sci.* **43**, 557–572. (5) Michel R., Dragovitsch P., Cloth P., Dagge G., and Filges D. (1991) *Meteoritics* **26** 221–242. (6) Bhandari N., Mathew K.J., Rao M.N., Herpers U., Bremer K., Vogt S., Wölfli W., Hofmann H.J., Michel R., Bodemann R., and Lange H.-J. (1993) *Geochim. Cosmochim. Acta* **57**, 2361–2375. (7) Masarik J. and Reedy R.C. (1994) *Geochim. Cosmochim. Acta* **58**, 5307–5317. (8) Michlovich E.S., Vogt S., Masarik J., Reedy R.C., Elmore D., and Lipschutz M.E. (1994) *J. Geophys. Res.* **99**, 23,187–23,194. (9) Lal D. (1988) *Annu. Rev. Earth Planet. Sci.* **16**, 355–388. (10) Lal D. (1991) *Earth Planet. Sci. Lett.* **104**, 424–439. (11) Reedy R.C., Tuniz C., and Fink D. (1994) *Nucl. Instrum. Methods Phys. Res.* **B92**, 335–339. (12) Masarik J. and Reedy R.C. (1995) *Earth Planet. Sci. Lett.*, in press. (13) Reedy R.C., Nishiizumi K., Lal D., Arnold J.R., Englert P.A.J., Klein, J., Middleton R., Jull A.J.T., and Donahue D.J., (1994) *Nucl. Instrum. Methods Phys. Res.* **B92**, 297–300. (14) Drake D.M., Drosd M., Byrd R.C., Reedy R.C., Clark D.A., Englert P.A.J., Dempsey J.F., Bobias S.G., and Harris L. (1994) *Nucl. Instrum. Methods Phys. Res.* **B84**, 337–356. (15) Nishiizumi K., Winterer E.L., Kohl C.P., Klein J., Middleton R., Lal D., and Arnold J.R. (1989) *J. Geophys. Res.* **94**, 17,907–17,915. (16) Reedy R.C., Nishiizumi K., Klein J., Davis R., Middleton R., Lal D., Arnold J.R., Jull A.J.T., Englert P.A.J., and Elmore D (1994) in *Abstracts of the Eighth International Conference on Geochronology, Cosmochronology, and Isotope Geology*, U.S. Geological Survey Circular 1107, p. 262. (17) O'Brien K. (1979) *J. Geophys. Res.* **84**, 423–431. (18) Dep L., Elmore D., Fabryka-Martin J., Masarik J., and Reedy R.C. (1994) *Nucl. Instrum. Methods Phys. Res.* **B92**, 321–325. (19) Nakamura T., Sugita H., Imamura M., Uwamino Y., Shibata S., Nagai H., Takabatake M., and Kobayashi K. (1992) in *Nuclear Data for Science and Technology* (Springer-Verlag, Berlin), pp. 714–716. (20) Imamura M., Nagai H., Takabatake M., Shibata S., Kobayashi K., Yoshida K., Ohashi H., Uwamino Y., and Nakamura T. (1990) *Nucl. Instrum. Methods Phys. Res.* **B52**, 595–600. * This work supported by NASA and done under the auspices of the U.S. Department of Energy.