# OVERVIEW OF THE WORKSHOP ON SECULAR VARIATIONS IN PRODUCTION RATES OF COSMOGENIC NUCLIDES ON EARTH<sup>1</sup>

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#### INTRODUCTION

Measurements of cosmogenic nuclides made *in situ* in the Earth's surface are being used to help resolve a wide range of geologic and chronologic questions. Cosmogenic nuclides (<sup>3</sup>He, <sup>10</sup>Be, <sup>14</sup>C, <sup>21</sup>Ne, <sup>26</sup>Al, <sup>36</sup>Cl are presently used) can reveal rock exposure history information leading to estimates of timing of surface forming events, rates and styles of erosion, and timing and durations of episodes of burial. Depending on the problems being tackled, a significant source of error ( $\pm 10$ –25%) for any cosmogenic nuclide method is the present uncertainty in the spatial and temporal variability of the rates of production of these *in-situ* nuclides.

The Workshop on the Secular Variations in the Rates of Production of Cosmogenic Nuclides on Earth was hosted by Los Alamos National Laboratory on 2–5 February 1996 in Santa Fe, New Mexico. The workshop was a multi-disciplinary, international meeting of 38 scientists from seven countries (see below). The workshop gathered together not only geologists, chemists and physicists directly involved in the determination of *in-situ* production rates, but also workers in the related fields of paleomagnetism, nuclear cross sections, cosmic-ray transport and other branches of geochronology. This is a cross section of scientists that would never be found at any single societal meeting.

Bierman, P.	University of Vermont	
Brook, E.	University of Rhode Island	
Brown, E.	CNRS-IN2P3, France; University of Minnesota	
Caffee, M.	Lawrence Livermore National Laboratory	
Clapp, E.	University of Vermont	
Clark, D.	University of Washington	
Damon, P.	University of Arizona	
Dunne, A.	PRIME Lab, Purdue University	
Evenson, E.	Lehigh University	
Finkel, R.	Lawrence Livermore National Laboratory	
Flinsch, M.	New Mexico Tech	
Gillespie, A.	University of Washington	
Gosse, J.	Los Alamos National Laboratory	
Graham, I.	Institute of Geological and Nuclear Sciences, NZ	
Graf, Th.	University of California, San Diego; ETH Zürich	
Harrington, C.	Los Alamos National Laboratory	
Heisinger, B.	Technical University of Munich	

#### List of Attendees

<sup>1</sup>Note: Unless otherwise noted, cited references in all workshop abstracts can be found in the "Bibliography for *In-Situ* Production Rates" following this Overview.

<sup>2</sup>Geology and Geochemistry Group, EES-1, MS D462

<sup>3</sup>Astrophysics and Radiation Measurements Group, NIS-2, MS D436

<sup>4</sup>Geology and Geochemistry Group, EES-1, MS D462

<sup>5</sup>Chemical Science and Technology, CST-7, MS J514

Imamura, M.	Institute for Nuclear Study, University of Tokyo
Ivy-Ochs, S.	ETH Hönggerberg, Zürich
Jull, T.	University of Arizona
Klein, J.	University of Pennsylvania
Liu, B.	Los Alamos National Laboratory
Marti, K.	University of California, San Diego
Masarik, J.	Max-Planck Institute, Germany
McHargue, L.	University of Arizona
Nishiizumi, K.	University of California, Berkeley
O'Brien, K.	Northern Arizona University
Phillips, F.	New Mexico Tech
Phillips, W.	Los Alamos National Laboratory
Poths, J.	Los Alamos National Laboratory
Reedy, R.	Los Alamos National Laboratory
Stamatakos, J.	South West Research Institute, Texas
Sternberg, R.	Franklin and Marshall College
Stone, J.	Australian National University
Swanson, T.	University of Washington
Stuiver, M.	University of Washington
Verosub, K.	University of California, Davis
Zreda, M.	University of Arizona

The group recognized the limits and various sources of systematic uncertainties of the cosmogenic nuclides methods, especially the many ancillary data sets used, such as those used in adjustments for elevation and geomagnetic effects. There was a unique opportunity for open discussion and evaluation of the geological and chronological assumptions for the major calibration sites. Input from the fields of paleomagnetism, radiocarbon dating and cosmic-ray physics helped to address questions as they arose.

#### Workshop Goals

The following goals and questions were addressed during the workshop:

- 1. To discuss the measurement of *in-situ* cosmogenic nuclide production rates. How accurate are the current measurements? How accurate do we need them to be and what are the limitations? How well do we know the relative production rates of cosmogenic nuclides? What should be involved in selection of calibration sites? What factors are important to consider? What are some suggestions for good calibration sites? What are the caveats involved in measuring production rates at calibration sites and what experiences can be shared to decrease the probabil-ity of repeating mistakes?
- 2. To discuss models relating cosmic-ray intensity and nuclear cross sections to production rates. What do the models tell us about spatial variability? What can they tell us about temporal variations? How good are the models? For example, if we accurately knew the Earth's magnetic dipole paleointensity, how accurately can we now calculate production rates on Earth? What are the uncertainties in the models and their magnitudes? Are these models worth pursuing? What is needed to improve the models? How strongly are cross section measurements needed?
- 3. To summarize the current state of knowledge concerning temporal and spatial variability of production rates. How well do we know the spatial and temporal dependencies? What literature exists? What needs to be done to improve our knowledge of the dependencies on altitude and latitude? Where should measurements of in-situ production rates be focused? What is the need for very low and very high latitude measurements? Are there problematic results published?

- 4. To discuss surrogate records of changes in parameters that affect production rates. What records can be used now? How good are they? What improvements are being attempted that will have an impact on production rate modeling within the next half decade? What models exist or are needed to calculate changes in production rates based on changes in such factors as the strength of the Earth's magnetic field? What records of prehistoric solar activity are available and how good are they?
- 5. To make suggestions that will help direct future efforts to calibrate cosmogenic nuclide surface exposure dating methods.
- 6. To compile a bibliography of current relevant publications (see below).

#### State of Production Rate Knowledge

The talks and discussions covered a wide range of topics and data. The reader is referred to the abstracts and bibliography to attain the most recent estimates of production rates. Estimates of the *in-situ* production rates of the cosmogenic nuclides currently used in exposure history applications were presented and evaluated at the workshop. Production rate estimates have been determined from measurements of the nuclides in geologic samples and artificial samples (such as water) and through numerical simulations of the reactions involved. It is clear that the uncertainty of a production rate for a given sample site is improving with the increasing number of empirically and experimentally derived production rates.

However, some important discrepancies are evident in the existing production rate estimates. The reasons for the discrepancies are complicated because multiple variables are involved, including 1) the calibration sites may have had uncertain exposure histories, *e.g.*, some sites may have been partially buried by till, snow, or ejecta; 2) there are inherent uncertainties in the calibration age for the site, due to either assumptions associated with the calibration age or the geomorphic relationship between the site for the calibration age and that for the cosmogenic nuclide sample (*cf.* abstract of Clark *et al.*); 3) the calibrations were done for different exposure durations and geographic locations—variations in the Earth's geomagnetic field strength with time and location can account for significant variability on a 20 kyr timescale and; 4) the uncertainties associated with the scaling that is used to normalize production rates to sea level and high latitude can cause additional deviations. It was generally agreed that multiple sites of calibration are needed to provide information and constraints on elevation, latitude and temporal controls on production rates. The need for cross calibration of the production rates of different cosmogenic nuclides was also emphasized.

New and previous empirical data, used to calculate or verify production rates for <sup>10</sup>Be and <sup>26</sup>Al in quartz, were presented by Nishiizumi *et al.*, Bierman *et al.*, Gosse and Klein, Ivy-Ochs *et al.*, Brown *et al.* and Brook *et al.* The rates normalized for production at sea level and high latitude are in fairly close agreement (all were within  $5.6 \pm 0.5$  atoms  $\cdot g^{-1} \cdot yr^{-1}$ ) for times ranging from the present to >10 Myr.

The rates for *in-situ* <sup>14</sup>C in quartz were reported by Jull and McHargue. The use of *in-situ* <sup>14</sup>C work is relatively recent and more production rate measurements are immediately needed. Considering the importance of <sup>14</sup>C in erosion rate and burial studies due to its short half-life, production rate refinement for this nuclide should get considerably more attention.

Data useful in constraining production rates of the cosmogenic noble gases <sup>3</sup>He and <sup>21</sup>Ne were presented by Brook *et al.*, Brown *et al.*, Poths *et al.*, Phillips, W. *et al.* and Marti and Graf. The current production rate estimates of these nuclides are in reasonable agreement for times >10 kyr. Discussions concerning the diffusion of the gases, such as <sup>3</sup>He in quartz and the non-cosmogenic compo-

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nents of the elements, addressed the present state of understanding and made suggestions for future noble gas experiments. Although M. Kurz and T. Cerling were unable to attend, references to their work in noble gas production rates are included in the bibliography.

Estimates of production rates of <sup>36</sup>Cl from various pathways (thermal neutron capture, muonic reactions and spallation reactions) were presented by F. Phillips *et al.*, Swanson *et al.* and Stone *et al.* The production rate estimates varied considerably, partly due to the factors mentioned above and partly due to the complications involved in the interactions of multiple cosmogenic and non-cosmogenic production pathways. Nevertheless, the production rates of <sup>36</sup>Cl are now sufficiently well known to yield ages that are within reasonably close agreement to ages calculated with cosmogenic <sup>10</sup>Be and <sup>26</sup>Al, based on data presented by Ivy-Ochs *et al.*, Gosse and Klein and Phillips, F. *et al.* This agreement is encouraging for our knowledge of the relative production rates of each nuclide.

Results of ongoing numerical simulations to calculate the production of various nuclides from different targets and at different energies was presented by O'Brien, Reedy and Masarik and Reedy. Production rates from the simulations show very good agreement with some measured production rates. Uncertainties in the fluxes of cosmic-ray particles (especially neutrons and muons), uncertainties in the reaction cross sections with various target elements and other sources of error were recognized when the simulation data were evaluated. Imamura reported on measurements of cross sections for producing these nuclides by energetic neutrons. Heisinger *et al.* presented work using accelerator-produced stopped negative muons and fast muons and comparisons with a measured depth profile to 250 m. A discrepancy between the contribution of muons that some models predict and the contribution that has been previously assumed (the assumed contribution may be too high) provided insight into the importance of the muonic reactions.

#### **Other Lessons Learned**

Much discussion centered on the effects of solar and Earth magnetic influences on the incident cosmic ray flux. There is a well-understood relationship between incident cosmic ray flux and the dipole moment of the Earth. Instantaneous *in-situ* cosmogenic nuclide production rates in rocks on Earth vary in a way similar to how production rates of the <sup>14</sup>C in the atmosphere varies: during periods of low field intensity, production rate is high and vice versa. The main difference is that the concentration of *in-situ* nuclides is the integral of the production rates over the entire exposure period whereas <sup>14</sup>C ratios record the production rate at small instances of time. Although short-lived highamplitude changes in paleointensity will affect the atmospheric <sup>14</sup>C time scale, the cosmogenic nuclide time scale will not feel the effects of a short paleointensity change unless the exposure time is of similar duration.

Several programs are being generated (Clapp *et al.*: COSMO-CALIBRATE; Dunne *et al.*) to correct cosmogenic nuclide production rates due to changes in the intensity of Earth's dipole field strength. These programs are useful to show the sensitivity of the integrated production rates of cosmogenic nuclides to changes in the geomagnetic field—however, they are only as good as the input data (see below). Using the existing paleointensity records, the programs predict that, depending on the latitude and elevation of the site, the time-integrated production rate for an exposure over the last 40 kyr is much higher (possibly  $\sim 20\%$ ) than the production rate over the last 5 kyr.

Unfortunately, existing absolute and relative paleointensity records are not sufficiently reliable to base adjustments of cosmogenic nuclide production rates. A presentation by K. Verosub and the ensuing discussions revealed the difficulties that paleomagnetists have had in interpreting the paleomagnetic data. Sources of uncertainties in the paleointensity records include poor age control, dif-

ficulty in evaluating the significance of non-dipole contributions and errors due to variations in magnetic grain size, multi-domain remanences and inclination shallowing. The uncertainty of many of the existing paleointensity records therefore probably approaches or exceeds 20%. Until more paleointensity records become available it may be difficult to adjust cosmogenic nuclide production rates with high degrees of uncertainty.

Other sources of variation in production rates were discussed at the workshop. R. Sternberg showed that significant changes in the average geomagnetic latitude of sampled sites (up to 4° over exposure durations longer than 2 kyr) are caused by secular variation of the dipole axis position. Only sites <~15 kyr are affected because the dipole axis position is approximately geocentric for longer exposures. The effects of a 2–4° uncertainty in geomagnetic latitude were modeled by Klein and Gosse, who showed that for high altitude mid-latitude sites (~30° latitude) the uncertainty could cause a discrepancy in production rate on the order of 10%.

The influence of solar modulation over a solar cycle was discussed by K. O'Brien. The model results he presented and results from simulation efforts by R. Reedy and J. Masarik suggest that solar modulation may play a significant role at high latitudes.

The uncertainty in the protocol for scaling production rates to different latitudes and altitudes was the theme of many discussions throughout the workshop. Existing data seem to suggest that the altitudinal correction of Lal (1991) is reliable to within 10%. However, J. Stone pointed out that elevation corrections may be complicated because atmospheric thickness above any site may change significantly due to a combination of factors such as eustatic sea level changes and glacial isostacy. Also, some special cases exist such as the effects on atmospheric thickness of the quasi-stable low atmospheric pressure and cold air over Antarctica. There was considerable disagreement on the reliability of the Lal (1991) latitudinal scaling. The debate over the relative significance of muon effects on latitude and altitude adjustments is not fully resolved. The effects of non-dipole field variations are presently ignored, mostly because insufficient data are available to model Quaternary non-dipole field paleointensities over the globe, according to J. Stamatakos. However, the spatial and temporal variations in non-dipole field strengths may be significant for Holocene measurements. I. Graham reported on plans that his group is making to measure production rates over a wide range of latitudes.

Discussions on the reliability of the dating methods against which the *in-situ* nuclide time scales are being calibrated centered on the <sup>14</sup>C method, and to a lesser extent, the <sup>40</sup>Ar/<sup>39</sup>Ar technique. The utility of surfaces assumed to be in saturation for <sup>10</sup>Be and <sup>26</sup>Al in Antarctica (and the reliability of the <sup>10</sup>Be and <sup>26</sup>Al half-lives) was also discussed. The majority of the production rate calibrations have been at <20 kyr, based on <sup>14</sup>C ages. The calibrated <sup>14</sup>C time for ages <~11 kyr is generally believed to be sufficiently reliable for calibration. Disagreement over the reliability of the U/Th coral calibration of older <sup>14</sup>C ages (to ~18 kyr) was not completely resolved, although M. Stuiver presented convincing isotopic data from the GISP core which supported the extended <sup>14</sup>C calibration curve. It is clear that continued improvement in calibration of the <sup>14</sup>C time scale is needed. Refinements in long term *in-situ* production rates will continue as improvements in the understanding of latitude and altitude effects enable reliable extrapolation of Antarctic production rates to other locations and by cautiously taking advantage of recent advances in Ar-Ar dating of whole rocks.

# Suggestions for Future Calibration of Cosmogenic Nuclide Time Scales

The following points reached general consensus among the group:

- 1. If possible, calibration against ages derived from other calibrated dating methods or other cosmogenic nuclide ages should be avoided. It was suggested that more emphasis should be placed on calibrating with ages based on the U and Ar systems.
- 2. It was pointed out that several existing production rates may be slightly low because the age assumed for the exposure duration was too high due to shielding by till or sediment (e.g., rocks at Meteor Crater may have been covered by ejecta). When selecting a site for calibration, the potential for partial or complete shielding due to burial must be considered. For young sites (<20 kyr) the effects of erosion are probably insignificant when compared to the effects of burial. For example, selection of sites with glacial polish to ensure no erosion has occurred since deglaciation may be counter-productive if the reason the polish has survived is because it was protected by a soil cover.
- 3. The group recommended that because suitable calibration sites are so difficult to find, no specific site latitude, elevation, or exposure duration should be particularly targeted.
- 4. It was also agreed that future proposed calibration sites should be peer evaluated so that any merits and problems of the site can be identified before the production rate data is generated. No official forum was suggested because it was felt that informal discussion should be sufficient.
- 5. An attempt should be made to measure multiple nuclides at calibration sites where funding and mineralogy/lithology permit. Also, multiple samples should be analyzed for at least one nuclide at each site to help minimize the effects of erosion, burial and pre-exposure.

Participants of the workshop have also agreed to conduct an inter-laboratory comparison of various aspects of the chemistry and isotopic measurements involved in the cosmogenic nuclide methods. Wide interest was expressed in an inter-laboratory standard, with one suggestion being Tabernacle Hill, Utah, where multiple lithologies co-exist with tight age constraints.

# Suggestions for Future Authors or Reviewers of Publications Containing Interpretations of Cosmogenic Nuclides

In order to calculate exposure ages or erosion rate information from the measured cosmogenic isotopic abundances, numerous other measurements and data must be considered and incorporated. The protocol is generally straightforward but can be rather involved. Representatives of several of the laboratories where measurements of the abundances of the cosmogenic nuclides are made have pointed out that cosmogenic nuclide dating is beginning to be carried out by scientists who have not been involved in the technique development over the last decade. We can only recommend that the researcher considers the question being addressed before selecting the nuclide systems being utilized and that before collecting the sample, the researcher makes himself aware of all of the caveats and information needed. Once an erroneous age makes it into the literature it is sometimes very difficult to remove it.

There are several reasons why a single production rate for a particular nuclide cannot be suggested. The production rate for any nuclide at any time and location is not well known, mostly because of the very limited data set that exists but also because of small but significant discrepancies in the production rates that have been measured. Additionally, because the production rates probably change due to temporal fluctuations in the effective cosmic ray flux to the rock surface (due to past variations in geomagnetic field strength, dipole axis position, solar modulation, atmospheric pressure and other factors), a single time-independent production rate is not viable. Finally, there remain uncertainties in the scaling of production rates over different altitudes and latitudes.

Other factors that must be considered when calculating a site production rate based on the effective cosmic ray flux a rock receives includes the rock sample thickness and depth below the surface, surface geometry (addressed by A. Gillespie), amount of shielding of cosmic radiation (*e.g.*, by snow, water, sediment and topography with respect to the horizon) and erosion rate and style.

It is therefore crucial that authors consider including the following information so that as the science continues to develop we can re-evaluate the interpretations of the cosmogenic nuclide data (e.g., just as the calibration of the <sup>14</sup>C timescale is continually being improved). If page length restrictions limit the ability of the incorporation of these data, then the data should be made accessible elsewhere and that source be furnished. The list is not exhaustive but we hope reviewers of manuscripts will adhere to the suggestions (Table 1). Some other data may be necessary, depending on the technique and detailed descriptions of geometry and shielding should be included if applicable.

Meaningful estimates of the uncertainties in all calculated ages and erosion rates should be provided. Because the level of confidence or types of uncertainties used to describe the error limits may vary due to nature of the data or the style of the author, we recommend that some indication of what the error limit calculation involves also be included.

Data/parameter	Suggested unit or means
Site latitude and longitude	0
Site elevation	m
Effective attenuation length	$g \cdot cm^{-2}$
Density of the rock sampled	$g \cdot cm^{-3}$
Production rate(s) used	Atom $\cdot$ yr <sup>-1</sup> $\cdot$ g <sup>-1</sup> of material
Thickness/depth of the sample	cm
Chemical data	
Isotopic data	Atom $\cdot g^{-1}$ of material, or equivalent
Geometry/slope correction applied*,†	Dimensionless
Shielding correction applied*,†	Dimensionless
Density of the covering material*	g · cm <sup>−3</sup>
Thickness of the shielding cover*,†	cm
Duration of cover if known*	yr or kyr
Erosion rate correction*,†	Dimensionless, or mm $\cdot$ kyr <sup>-1</sup> or equivalent
Latitude and altitude scaling factor*,†	Dimensionless; provide ref. (e.g., Lal 1991)

TABLE 1. Information Suggested to Accompany Published Data

\*If applicable

†Requires explanation

## Utility of Cosmogenic Nuclide Exposure History Techniques

This workshop focused on how to improve our understanding of cosmogenic nuclide production rates and make recommendations for future calibrations. However, many unresolved questions involving Neogene chronologies, landform stability and tectonic, seismic, volcanic and climatic issues can be addressed with sufficient accuracy with the present uncertainty in the rates of production of cosmogenic nuclides. Some examples include 1) estimates of erosion rates to within 20% is generally of sufficient accuracy to address landform- or basin-scale questions; 2) relative sequences of events such as detailed deglaciation patterns or fan deposition can be attained from cosmogenic

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nuclides because the precision of the techniques approaches 3 to 5%; 3) similarly, cosmogenic nuclides can be used for correlation purposes (e.g., the existence of Younger Dryas moraines around the world) by comparing normalized concentrations ( $\pm 10\%$ ) of the same nuclide, thereby avoiding calibration to a different time scale; and 4) chronologic questions that do not require highly accurate exposure age assignments (better than 15%), such as 1) was this colluvial deposit formed during the early Quaternary, middle Quaternary or Late Quaternary, or 2) does this moraine correspond to a marine oxygen isotope stage II or an older ice volume advance?

Although many questions can be addressed with the existing uncertainties in production rates, it is clear that better estimates of production rates are direly needed to utilize the cosmogenic nuclide techniques to their full potential. A number of challenging questions that directly impact society will remain largely unanswered until the cosmogenic nuclide methods evolve further over the next decade.

#### **ACKNOWLEDGMENTS**

The workshop was sponsored by: National Science Foundation EAR-9526774; Office of Basic Energy Sciences–Geosciences and Engineering; Los Alamos National Laboratory: Earth and Environmental Sciences Division, Chemical Science and Technology Division, Institute of Geophysics and Planetary Physics and the Los Alamos National Laboratory Yucca Mountain Program Office. We thank Kim Thompson, Anthony Garcia, Lanny Piotrowski, Phyllis Auchampaugh, Carol LaDelfe, Grant Heiken, Scott Baldrich and Jim Aldrich for support and suggestions.

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