Rapid Communication



Calibration of the *in situ* cosmogenic ¹⁴C production rate in New Zealand's Southern Alps

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ABSTRACT: In situ cosmogenic ¹⁴C (in situ ¹⁴C) analysis from quartz-bearing rocks is a novel isotopic tool useful for quantifying recent surface exposure histories (up to ~25 ka). It is particularly powerful when combined with longer-lived cosmogenic isotopes such as ¹⁰Be. Recent advances in the extraction of *in situ* ¹⁴C from quartz now permit the routine application of this method. However, only a few experiments to calibrate the production rate of *in situ* ¹⁴C in quartz have been published to date. Here, we present a new *in situ* ¹⁴C production rate estimate derived from a well-dated debris flow deposit in the Southern Alps, New Zealand, previously used to calibrate ¹⁰Be production rates. For example, based on a geomagnetic implementation of the Lal/Stone scaling scheme we derive a spallogenic production rate of 11.4 ± 0.9 atoms ¹⁴C (g quartz)⁻¹ a⁻¹ and a ¹⁴C/¹⁰Be spallogenic production rate ratio of 3.0 ± 0.2. The results are comparable with production rates from previous calibrations in the northern hemisphere. Copyright © 2012 John Wiley & Sons, Ltd.

KEYWORDS: in situ ¹⁴C; New Zealand; production rate calibration; short-lived cosmogenic nuclide; Southern Alps.

Introduction

Surface exposure dating with *in situ* cosmogenic nuclides such as ¹⁰Be, ²⁶Al, ³⁶Cl, ³He and ²¹Ne is widely used to quantify landscape processes. The potential of the relatively short-lived *in situ* ¹⁴C (half-life 5730 years) to constrain recent and complex surface exposure histories has long been recognized (e.g. Lal, 1991), but its extraction from terrestrial silicates has proven challenging. Unlike the longer-lived (e.g. ¹⁰Be, half-life 1.39 Ma) and stable (³He and ²¹Ne) nuclides, *in situ* ¹⁴C is less sensitive to prior exposure, because components inherited from periods before ~25 ka ago have decayed below detection limits. Furthermore, pairing of short-lived *in situ* ¹⁴C in combination with a longer-lived nuclide allows investigation of complex surface exposure histories, including those involving erosion and surface burial.

First attempts to extract ¹⁴C from terrestrial whole rocks were reported over two decades ago (Jull *et al.*, 1989). However, at that time reliable measurements were limited by the difficulty of efficiently isolating the small amounts of *in situ* ¹⁴C from ubiquitous atmospheric ¹⁴C. It was the breakthrough achieved by Lifton *et al.* (2001), based on an improved protocol and the use of quartz separates, that allowed this tool to be applied successfully to problems of Earth-surface processes (Matmon *et al.*, 2005; Miller *et al.*, 2006; Anderson *et al.*, 2008; Goehring *et al.*, 2011; White *et al.*, 2011).

Successful application of cosmogenic nuclides for surface exposure dating requires that the production rate (the number of atoms produced per gram of target material per year) be well known. The production rate of a given nuclide can be determined by measuring the nuclide concentration in a surface with a simple, well-constrained exposure history (i.e. no intermittent shielding or significant erosion, and a reliable independently estimated exposure age). Because cosmogenic nuclide production rates depend on the altitude,

*Correspondence: I. Schimmelpfennig, as above. E-mail: schimmel@ldeo.columbia.edu geographic position and exposure duration, the resulting local production rate is, by convention, scaled to present day and to a 'sea level/high latitude' (SLHL) reference position by applying one of the several published scaling methods (e.g. Lal, 1991; Stone, 2000; Dunai, 2001; Desilets *et al.*, 2006; Lifton *et al.*, 2005).

The first experimental calibrations of *in situ* ¹⁴C production rates using the improved extraction procedure were conducted with samples from sites in the northern hemisphere by Lifton *et al.* (2001), followed by Pigati (2004; data in Miller *et al.*, 2006) and Dugan *et al.* (2008).

In this study, we present the first *in situ* ¹⁴C production rate from the southern middle latitudes, obtained from a debris flow deposited 9.7 ka ago in the Southern Alps of New Zealand. This site has a robust radiocarbon age and forms the basis for a ¹⁰Be production rate calibration (Putnam *et al.*, 2010). Four of the seven original ¹⁰Be calibration samples had sufficient material left for the *in situ* ¹⁴C production rate calibration. Using these four samples, we derive the local *in situ* ¹⁴C production rate in quartz at the Macaulay calibration site. We also present SLHL spallogenic production rates using the five scaling methods implemented in the CRONUS-Earth online calculator (Balco *et al.*, 2008) and compare and combine the results with previously published calibrations.

Calibration site

The geomorphology of the calibration site in the central Southern Alps of New Zealand is described in detail in Putnam *et al.* (2010). It consists of a bouldery debris-flow deposit that overran a vegetated alluvial terrace at Macaulay River on South Island, at 43.6°S latitude and 1030 m altitude. This burial event has been dated to 9690 ± 50 calendar years before 2008 Common Era (CE) based on ten radiocarbon dates on plant macrofossils from the soil horizon buried by the debris flow. Following Putnam *et al.* (2010), we interpret this date to represent the depositional age of the landform and thus the

Table 1.	Sample	data

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Sample	Latitude (°S)	Longitude (°E)	Altitude (m)	Thickness (cm)	Thickness correction factor	Shielding factor
MR-08-03	43.57452	170.60805	1029.4	2.55	0.979	0.988
MR-08-05	43.57435	170.60760	1032.1	2.39	0.980	0.991
MR-08-13	43.57751	170.60695	1027.6	1.41	0.988	0.991
MR-08-14	43.57787	170.60493	1032.0	2.35	0.980	0.991

beginning of cosmogenic nuclide production in the boulders exposed at its surface.

The altitudes of the four calibration samples range from 1028 to 1032 m. Sample data are summarized in Table 1.

In Situ ¹⁴C analysis, production rates and ${}^{14}C/{}^{10}Be$ ratio

The *in situ* ¹⁴C extraction procedure and details of the production rate calculations are given in the online Supporting information, Appendix S1. The blank-corrected ¹⁴C concentrations of the four samples, also corrected for topographic shielding and sample thickness effects, range between 180×10^3 and 214×10^3 atoms (g quartz)⁻¹ and give an arithmetic mean value and standard deviation of $(194 \pm 15) \times 10^3$ atoms (g quartz)⁻¹ (Tables 2a and 2b). While the 1σ accelerator mass spectrometry measurement uncertainties on the individual measurements are low (~2%), the scatter in the *in situ* ¹⁴C dataset (standard deviation ~8%) is higher than that in the corresponding ¹⁰Be dataset (standard deviation ~1%), and no correlation is observed between individual higher or lower *in situ* ¹⁴C and ¹⁰Be concentrations (Tables 2a and 2b). The scatter in the *in situ* ¹⁴C dataset therefore mostly reflects contributions from the extraction procedure.

The local time-integrated in situ ¹⁴C production rate determined from the four measurements at the site in New Zealand is 34.0 ± 2.7 atoms (g quartz)⁻¹ a⁻¹ (arithmetic mean \pm standard deviation, Tables 2a and 2b). This value represents the total in situ ¹⁴C production from spallation and muon capture. We calculated the spallogenic ¹⁴C production rate by first subtracting the muogenic ¹⁴C contribution for each sample estimated from Heisinger et al. (2002a,b) [3.8 atoms $(g quartz)^{-1} a^{-1}$ at SLHL], and then deriving the best-fit spallogenic production rates referenced to SLHL. To do this, we applied the chi-squared minimization approach of Balco et al. (2009) and the five production-rate scaling schemes incorporated in the CRONUS-Earth calculator (Balco et al., 2008), modified to utilize in situ ¹⁴C. Depending on the scaling scheme used, the time-integrated best-fit spallogenic pro a^{-1} duction rates range from 11.4 ± 0.9 atoms (g quartz)⁻¹ ('Lm': time-dependent adaptation of the method by Lal, 1991, accounting for paleomagnetic corrections) to 12.7 ± 1.0 atoms $(g quartz)^{-1} a^{-1}$ ('Li': method by Lifton *et al.*, 2005) (Table 3, Fig. 1).

The ratio of ${}^{14}C/{}^{10}Be$ spallogenic production rates in our quartz samples is 3.0 ± 0.2 for all five scaling methods. This value is in good agreement with the ratio of ${}^{14}C$ and ${}^{10}Be$ spallogenic production rates in quartz of 3.12 derived from numerical modeling by Masarik and Reedy (1995).

We note that the muogenic *in situ* ¹⁴C contribution (for our samples ~24% of total ¹⁴C production at SLHL following Heisinger *et al.*, 2002a,b) is not yet well constrained (Balco *et al.*, 2008), and therefore our experimentally determined ¹⁴C spallogenic production rates and ¹⁴C/¹⁰Be spallogenic production rate ratio might change with future refinements of muogenic production rates.

Comparison with previous *in situ* ¹⁴C production rate calibrations

In situ ¹⁴C production rate calibrations were previously performed with samples from two locations in the northern hemisphere, the Bonneville shoreline at Promontory Point (Utah, USA; 41°N, 112°W, 1600 m altitude) and two landslide deposits in the north-western Scottish Highlands (Corrie nan arr and Maol-Chean-dearg, UK; 57°N, 5°W, 100-500 m altitude) (Lifton et al., 2001; Pigati, 2004; Miller et al., 2006; Dugan et al., 2008). For direct comparison with our results, we recalculated the *in situ*¹⁴C spallation production rates for these northern hemisphere sites using the same protocol as reported here for the Macaulay site. For Promontory Point we employed the exposure age of 17.4 ± 0.2 calka BP originally used in Lifton et al. (2001), based on the radiocarbon chronology of Lake Bonneville by Oviatt et al. (1992). It should be noted, however, that aspects of this chronology are continuing to evolve as additional age control becomes available (e.g. Miller et al., 2011). In Dugan et al. (2008), the exposure age at the Scottish calibration site is assumed to be 11.6 ± 0.2 ka based on the geomorphic relationship between landslide deposits and glacial landforms, which are assumed to date to the end of the Younger Dryas period ~11.6 ka ago. The recalculated best-fit in situ ¹⁴C spallation production rates from Promontory Point and Scotland are shown and compared with those from the Macaulay valley in Table 3 and Fig. 1. The combined in situ ¹⁴C spallation production rates from Promontory Point and Scotland range from 12.5 ± 0.9 to 14.1 ± 1.0 atoms (g $(quartz)^{-1}$ a^{-1} , depending on the scaling method used (Table 3), and agree within 1σ uncertainties with those from the Macaulay valley. The arithmetic means and standard deviations of the best-fit values from all three sites are between 12.1 ± 0.6 and 13.5 ± 0.7 atoms (g quartz)⁻¹ a⁻¹ (Table 3, Fig. 1).

Conclusions

In this study we present the first estimate of the *in situ* ¹⁴C production rate in the southern hemisphere, thus enhancing knowledge of the geographic distribution of *in situ* ¹⁴C production rates. The spallogenic production rates from this calibration site in New Zealand, calculated with five different scaling methods, agree within 1 σ with previous estimates from the northern hemisphere. The ¹⁴C/¹⁰Be spallogenic production rate agrees well with ¹⁴C/¹⁰Be ratios deduced from numerical spallation production rate simulations.

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Table 2a.	In situ ¹⁴ C \in	extraction results.									
Sample	Quartz weight (g)	V _{CO2} (cc STP)	V _{dilute} (cc STP)	CAMS number	$F_{ m m}$ measured	[¹⁴ C] blank corrected (10 ³ atoms g ⁻¹)	Blank correction proportion	<pre>[¹⁴C] (10³ atoms g⁻¹) shielding/ thickness-corrected</pre>	[¹⁰ Be] (10 ³ atoms g ⁻¹) shielding/ thickness-corrected	[¹⁴ C]/[¹⁰ Be]	Local ^{14}C production rate (atoms ^{14}C (g a) $^{-1}$)
MR-08-03 MR-08-05 MR-08-13 MR-08-14	5.03 5.01 5.01 5.01	$\begin{array}{c} 0.2061\pm0.0024\\ 0.1178\pm0.0013\\ 0.1812\pm0.0021\\ 0.0992\pm0.0011 \end{array}$	$\begin{array}{c} 1.573 \pm 0.018 \\ 1.456 \pm 0.017 \\ 1.573 \pm 0.018 \\ 1.492 \pm 0.017 \end{array}$	152042 152040 152043 152041	0.0259 ± 0.0002 0.0243 ± 0.0001 0.0244 ± 0.0001 0.0243 ± 0.0002	206.87 ± 9.44 174.73 ± 9.25 193.16 ± 9.34 179.81 ± 9.30	11% 12% 11% 12% Mean±SD	213.92 ± 9.76 179.90 ± 9.52 197.24 ± 9.54 185.07 ± 9.58 185.07 ± 9.58 194.03 ± 15.12 (7.8%)	$\begin{array}{c} 90.23\pm1.74\\ 91.71\pm2.50\\ 91.68\pm1.82\\ 90.47\pm1.85\\ 91.02\pm0.78\;(0.9\%)\end{array}$	$\begin{array}{c} 2.37\pm0.12\\ 1.96\pm0.12\\ 2.15\pm0.11\\ 2.15\pm0.11\\ 2.05\pm0.11\\ 2.13\pm0.18\ (8.3\%)\end{array}$	37.5 ± 1.7 31.5 ± 1.7 34.6 ± 1.7 32.4 ± 1.7 34.0 ± 2.7
Sample we $(F_m = \text{the}^{-1})^{-1}$ atoms of $^{-1}$ (Putnam <i>et</i> standard d.	eight, gas volt ⁴ C/ ¹³ C ratio o ¹ C in blank di <i>al.</i> , 2010), ¹⁴ , eviation.	ume after carbon ext of the sample vs. that vided by number of C/ ¹⁰ Be concentratio	traction from qua of a standard, bot atoms of ¹⁴ C in s in ratios and local	rtz (V _{CO2}) h correcteı ample), an time-inteε	and after addition o d to $\delta^{13}C = -25\% v$ V $d^{14}C$ concentration grated <i>in situ</i> ^{14}C pro	of a ¹⁴ C-free dilutio /PDB and to 1950 (is additionally corr oduction rates for e	n gas (V _{dilute}) (CE), blank-corr rected for topo, each sample, in	cc STP = cubic centimete ected ¹⁴ C concentrations graphic shielding and sam icluding all production m	ers at standard temperature with analytical uncertainti aple thickness effect (cf. Ta echanisms and corrected fo	e and pressure), meas les, blank correction p able 1). Also given ar or radioactive decay,	ured fraction modern rroportion (number of a ¹⁰ Be concentrations with mean value and
Table 2b.	<i>In situ</i> ¹⁴ C k	olank data producec	d with the sampl€	s in Table	, 2a.						
		V _{CO2}	(cc STP)		V _{dilutes} (cc STP	(CAMS nur	nber	F _m measured	No. of a	toms 14 C (10 ³ atoms)
Blank 5-11 Blank 6-1-	1-11	0.01571 0.01667	1 ± 0.00018 7 ± 0.00019		$\begin{array}{c} 1.346 \pm 0.015 \\ 1.379 \pm 0.016 \end{array}$		151903	3	0.0061 ± 0.0001 0.0053 ± 0.0001	150.16∃ 119.65∃	= 12.82 = 13.02
<i>In situ</i> ¹⁴ Cl standard d Table 3.	blank data prc eviation of th Best-fit spallo	oduced with the sam he nine blanks proce genic production ra	ples in (a). Blank ⁵ essed at LDEO si ates (PR) referenc	5-11-11 we nce the ye ed to SLH	is processed before $\dot{\epsilon}$ ar 2010 [(124.67 \pm L and present time (and Blank 6-11-11 :42.56) × 10 ³ aton (1950 CE) using fi	after the sampl as ¹⁴ C, Goehri ive different sc	les, respectively. For the bl ng <i>et al.</i> (2012)] were us :aling methods.	lank correction of the samp ed.	ole measurements, the	s arithmetic mean and
Scaling method	(at	NZ ${}^{14}C$ PR toms ${}^{14}C$ g^{-1} a^{-1})	(ator	NZ ¹⁰ Be P ns ¹⁰ Be g ⁻	R -1 a ⁻¹)	PPT 14 C PR (atoms 14 C g^{-1} a	a ⁻¹)	ScoHi 14 C PR (atoms 14 C g^{-1} a^{-1})	PPT + ScoHi ⁻¹ (atoms ¹⁴ C g ⁻	¹⁴ C PR ⁻¹ a ⁻¹)	$\begin{array}{l} \mbox{Mean} \ ^{14}\mbox{C PR} \\ \mbox{(NZ + PPT + ScoHi)} \\ \mbox{(atoms} \ ^{14}\mbox{C} \ g^{-1} \ a^{-1}) \end{array}$
St	11	1.7±0.9 (7.5%)		3.9 ± 0.1		12.9 ± 0.6		12.4 ± 1.6	12.8 ± 0	6.(12.3 ± 0.6
De		$1.8 \pm 0.9 \ (7.5\%)$		3.9 ± 0.1	_	13.1 ± 0.7		13.0 ± 1.7	13.1 ± 0	6.(12.6 ± 0.7
Du :		$1.8 \pm 0.9 \ (7.5\%)$		3.9 ± 0.1	_	13.1 ± 0.7		13.1 ± 1.7	13.1 ± 0	.9	12.7 ± 0.8
_	· 4 7	$2.7 \pm 1.0 (7.5\%)$		4.2 ± 0.1		14.1 ± 0.7		13.8 ± 1.8	14.1±1	0.	13.5 ± 0.7
Lm	11	$1.4 \pm 0.9 \ (7.5\%)$		3.8 ± 0.1		12.5 ± 0.6		12.4 ± 1.6	12.5 ± 0	.0	12.1 ± 0.6
St = Stone Putnam <i>et</i> 2008). PPT recirculatir best-fit val	(2000), Du = al., 2010). PP +ScoHi indi ng extraction ues from the t	Dunai (2001), De = ³ T denotes the Bonni icates the combinati systems at the Unive three calibration site	Desilets <i>et al.</i> (20) eville shoreline at ion of all measure ersity of Arizona. es. All uncertaint	06), Li = Li t Promontc ments fror Replicate I ies correst	fton <i>et al.</i> (2005), Lr Jry Point (Lifton <i>et a.</i> In Promontory Point measurements were sond to 1σ .	n = time-depender 1, 2001; Pigati, 20 and Scottish High averaged before t	nt version of Lal 04; Miller <i>et a</i> 11ands. The <i>in s</i> 1.the production	l (1991). NZ refers to the M II, 2006; Dugan <i>et al.</i> , 20 <i>situ</i> ¹⁴ C extractions from F rate calculations. The las	Aacaulay valley in New Zee 08). ScoHi refers to two sit PT and ScoHi samples we t column shows the arithm	aland (<i>in situ</i> ¹⁴ C fron tes in the Scottish Hig ere performed on the netic means and stand	n this study, ¹⁰ Be from shlands (Dugan <i>et al.</i> , flow-through and the dard deviations of the

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Figure 1. Comparison of the best-fit *in situ*¹⁴C spallogenic production rates determined at the three calibration sites, Macaulay valley in New Zealand (NZ), Promontory Point (PPT) and Scottish Highlands (ScoHi), shown for each of the five scaling methods, St, Du, De, Li and Lm (same references as in Table 3), used to reference the local production rates to SLHL and present time (1950 CE). The horizontal line and gray band show the arithmetic mean and standard deviation of the best-fit values from the three sites.

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Abbreviations. CE, Common Era; SLHL, sea level/high latitude.

Supporting information

Additional supporting information can be found in the online version of this article:

Appendix S1. Details of the *in situ* ¹⁴C extraction and production rate calculations.

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